Thermal Emission Spectroscopy of Single, Isolated Carbon Nanoparticles: Effects of Particle Size, Material, Charge, Excitation Wavelength, and Thermal History

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ABSTRACT:

Results are presented for thermal emission from individual trapped carbon nanoparticles (NPs) in the temperature range from ~1000 to ~2100 K. We explore the effects on the magnitude and wavelength dependence of the emissivity, \( \epsilon(\lambda) \), of the NP size and charge, and of the type of carbon material, including graphite, graphene, diamond, carbon black, and carbon dots. In addition, it is found that heating the NPs, particularly to temperatures above ~1900 K, results in significant changes in the emission properties, attributed to changes in the distribution of surface and defect sites caused by annealing and sublimation.

KEYWORDS: Nanoparticle, spectroscopy, single particle, trap, thermal emission, emissivity
I. Introduction

Thermal emission spectroscopy of nanoparticles (NPs) is important for applications such as measuring temperatures and particle concentrations in hot gas flows. For example, dispersed emission has been used to monitor flame temperatures during soot particle formation, also yielding information about particle size distributions and soot volume fractions.\textsuperscript{1-2} Similarly, laser-induced incandescence (LII) from laser-heated NPs (\textit{e.g.} soot, metal NPs), has been used to extract particle concentrations and sizes\textsuperscript{3-4} and to understand the energy balance in incandescent systems\textsuperscript{5-11}

Particle temperatures can be extracted by fitting emission spectra to a model that describes the emission intensity vs. wavelength, typically using Planck’s law for ideal blackbody emission, together with a function, $\epsilon(\lambda)$, that describes how the NP emissivity varies with wavelength. If the wavelength dependence of the emission is known, NP temperatures can also be extracted by two- or three-color thermometry, where calibrated intensities measured in two or three spectral regions are fit to an emission model to yield the temperature.\textsuperscript{12-13}

In these experiments, emission is collected from ensembles of NPs allowing rapid measurement, but resulting in spectral information that is averaged over the NP ensemble, which can be quite heterogeneous. Taking soot as an example, there typically are distributions of size\textsuperscript{14}, shape\textsuperscript{15-16}, and nanostructure,\textsuperscript{15} all of which may affect spectral properties. There have been a few experiments in which two or three color thermometry was used to measure the temperature of individual trapped particles for kinetics studies, however, the particles were in the $10-240 \mu m$ size range, and thus were quite bright, and would have had bulk-like emission properties.\textsuperscript{17-19}

We are developing an experiment to measure surface reaction kinetics for single, isolated NPs at high temperatures, aiming to understand the effects of NP size and structural heterogeneity on their chemical properties. To provide NP temperatures for the kinetics work, we have developed
experimental and calibration methods allowing measurements of dispersed emission spectra of single hot NPs in the 10 – 50 nm size range. Here, we report a study of the effects of material, mass, charge, and thermal history, on the emission spectroscopy of individual carbon NPs.

Thermal emission from individual NPs is also interesting from a fundamental perspective. For NPs of black carbon materials at high temperatures, it is reasonable to expect that the emission should be “blackbody-like”, i.e., the spectra should have the form:

\[
I(\lambda, T_{NP}) = \frac{2c}{\lambda^4 \left( \exp\left(\frac{hc}{\lambda k T_{NP}}\right) - 1 \right)} \cdot \varepsilon(\lambda) .
\]  

(1)

The first term is Planck’s law for ideal blackbody emission, and the second is an emissivity function, \(\varepsilon(\lambda)\), that accounts for factors such as: 1. Coupling of the NP to light of wavelength, \(\lambda\). 2. The optical/dielectric properties of the NP material. 3. The effects of surface, defect, or other localized states on the optical emission. One goal of our experiments is to examine how \(\varepsilon(\lambda)\) differs for different NPs of either the same, or different materials, and to measure changes \(\varepsilon(\lambda)\) for individual NPs as they are heated, driving sublimation and annealing.

This initial spectroscopy study is focused on carbon NPs, chosen because carbon high temperature chemistry and spectroscopy are of interest in various energy and materials applications. To obtain insight into nanostructure effects on NP emission spectra, we studied NPs made from a variety of carbon materials, including graphite and graphene, which have well ordered \(sp^2\)-hybridized basal planes, as well as lower coordination sites at basal plane edges or defects. The ratio of basal plane to edge/defect sites is expected to be larger for graphene platelets, compared to the more compact graphite NPs. The carbon black used in these experiments is >99% pure carbon soot produced by methane pyrolysis, and should have a variety of carbon morphologies present. In addition to possible spectral features from edge/defect sites, graphite, graphene, and carbon black NPs could potentially show band-gap emission, which has been
reported for carbon materials with sp²-hybridized carbon domains below ~20 nm in size.²¹-²³ For this reason, we also studied fluorescent carbon dot NPs synthesized by Jelinek and co-workers using a procedure discussed elsewhere.²⁴ Finally, carbon atoms in bulk diamond are sp³ hybridized, although NPs also have a significant proportion of surface/defect atoms, and here, boron-doped diamond was used to allow laser heating.

II. Experimental Methods

a. NP trapping and determination of mass and charge:

The single nanoparticle mass spectrometer (NPMS) instrument has been described in detail elsewhere,²⁵-²⁶ along with the methods for NP injection, and for determining the NP mass (M) and charge (Q). The trap and optical system used for NP mass determination and spectral measurements are shown in Figure S1. In brief, charged NPs are introduced into the gas phase by electrospray ionization (ESI), then pass through hexapole and quadrupole ion guides for differential pumping. The NPs are then injected into a split ring-electrode quadrupole trap, based on a design by Gerlich and Decker,²⁷-²⁸ which allows lasers to be focused through the trap along several axes, and provides high solid angle optical detection along several axes.²⁵ NPs are trapped by applying an AC voltage between the split-ring electrode and the end cap electrodes, with amplitude V₀ and frequency F_RF. Trapped NPs are detected by heating them with either 532 nm or 10.6 µm lasers loosely focused through the trap, collecting light emitted radially from the central ~50 µm of the trap, and detecting it with a fiber-coupled Si avalanche photodiode (APD - Laser Components USA). A 550 nm long pass filter (Thorlabs) is used to block 532 nm light scattering, thus the APD detects emission between 550 nm and its ~1000 nm quantum efficiency cut-off. The light can also be deflected into a CCD camera, allowing the trap center to be imaged from the radial direction for alignment purposes.
Because the APD signal is sensitive only to emission from near the trap center, it is modulated by motion of the NP. Under appropriate conditions, a trapped NP undergoes harmonic secular motion with frequencies $F_r$ and $F_z$ for radial and axial motion, respectively. The axial frequency is:

$$F_z = \left(\frac{Q}{M}\right)\left(\frac{\sqrt{2} V_0}{4\pi^2 F_R z_0^2}\right),$$  \hspace{1cm} (2)

where $z_0$ is a trap geometric parameter. For the small NPs of interest here, $F_z$ is measured by sweeping the frequency of a weak drive potential applied across the trap, with typical sweeps taking 30 to 60 seconds, depending on the frequency range covered and the desired $F_z$ resolution. When the drive potential is resonant with $F_z$, it increases the amplitude of the axial motion, such that the NP spends less time in the ~50 μm detection volume, resulting in a ~20 – 30% reduction in the APD signal. To damp the NP motion, 1.5 – 2 mTorr of Ar buffer gas is added to the trap using a mass flow controller, with pressure measured by a capacitance manometer with resolution of $10^{-5}$ Torr. By measuring $F_z$, $Q/M$ for the trapped particle is obtained. To determine $Q$, a series of single electron charge steps is driven using a vacuum ultraviolet lamp, resulting in a series of steps in $F_z$ (see figure 7, below) which can be fit to extract $Q$. Once $Q$ is known, $M$ can be determined using equation 2.

In a typical experiment, NPs are injected into the trap until a step in APD signal indicates that trapping has occurred. The NP is heated at low laser intensity such that its mass is stable, and $F_z$ is measured while the vuv lamp is used to drive a series of charge steps, thus determining $Q$ and $M$. Occasionally more than one NP is trapped, as shown by the presence of more than one $F_z$ peak, and in that case the particles are ejected and the experiment starts over. After $Q$ determination, the vuv lamp isolation valve is closed, and then the laser is adjusted to vary the NP temperature ($T_{NP}$), measuring emission spectra and mass simultaneously every 30 to 60 seconds.
One important issue is the purity of the NPs studied, which could affect their emission properties. As initially trapped, the NPs have oxidized surface groups (carbonyls, etc.) from air exposure, as well as adventitious adsorbates like water. In addition, ESI was done from 2 mM ammonium acetate solutions in methanol, presumably leaving some ammonium acetate and methanol on the NP surfaces. Molecules like water or methanol should desorb at low temperatures, and ammonium acetate was chosen because its thermal decomposition generates species that also desorb well below the $T_{NP}$ range of interest here (>$1500$ K).\textsuperscript{29} Similarly, oxidized surface groups have been shown to desorb as CO or CO$_2$ below $\sim1400$ K when heated in vacuum.\textsuperscript{30-33} In our experiments, NPs are initially trapped, held at $T_{NP} \leq 1300$ K during charge stepping (20 – 60 minutes), then heated to a series of higher $T_{NP}$ values for spectral measurements. Thus, except perhaps during initial spectral measurements at $T_{NP}$ below 1400 K, we expect that the only contaminants that could be problematic are metals or other nonvolatile elements.

The graphite NPs examined here have stated purity of 99.9999\% (metal basis, Alfa Aesar), and the graphene platelet material (US Research Nanomaterials, Inc.) has stated composition of 99.7\% C, <0.3\% O (O should desorb as CO or CO$_2$ by 1400 K). The carbon black NPs (Plasmachem GmbH) have stated purity of >99\% carbon, with < 0.02\% ash content, which would include any metal or other non-volatile contaminants. Thus, the graphite, graphene, and carbon black NPs all should have metal/non-volatile contaminant levels below 0.1\%. To allow diamond NPs to be laser heated at 532 nm or 10.6 $\mu$m, we used boron-doped nano diamond material (US Research Nanomaterials, Inc.), with stated composition of 98.5\% carbon (98.3\% diamond), 0.8\% boron, $\sim0.15\%$ H, 0.025\% N, and a total of $\sim0.05\%$ of various metals. As reviewed recently, annealing is a common approach to changing the properties of optically active impurity centers in diamond,\textsuperscript{34} with diffusion at temperatures up to 2300 K used to interconvert between different type
centers, however, it is unclear if the diffusion length scale would be large enough to bring impurities from the NP bulk to the surface. Therefore, while some contaminants may desorb at high $T_{NP}$, particularly if they are near the NP surface, we cannot assume that contaminants in the bulk are eliminated. We also did a few experiments on carbon dots, *i.e.*, small carbon aggregates prepared in the group of Raz Jelinek.\textsuperscript{24} Carbon dots are used as non-toxic fluorescent particles for biological applications, and the question of interest was whether they would be fluorescent in the gas phase.

A related issue is whether the NP charge might have significant effects on the measured emission spectra. Experiments testing this issue are presented below.

**b. NP spectral measurements and $T_{NP}$ determination:**

For the work here, we developed the optical system illustrated in Figure S1 and associated discussion. The system is an improved version of one reported earlier,\textsuperscript{20} redesigned to reduce chromatic effects and sensitivity to misalignment. Briefly, light emitted by the NP along the trap axis is collected with $\sim$1 steradian acceptance and formed into a slightly convergent beam by a five element lens system. For alignment purposes, the beam can be deflected into a CCD camera allowing the central volume of the trap to be imaged from the axial direction. Normally, however, the beam passes to a beamsplitter that reflects the “visible” ($\lambda < 980$ nm) portion of the beam, and transmits the “nIR” (980 nm $< \lambda < 1600$ nm) portion. The visible and nIR beams are then coupled into optical fibers, dispersed by spectrographs, and recorded by deep-cooled Si CCD and InGaAs photodiode cameras, respectively. Both spectrographs are wavelength-calibrated using mercury and neon emission lines.

To extract accurate $T_{NP}$ values, it is essential to know the sensitivity of the optical system vs. wavelength, $S(\lambda)$, and the calibration process is illustrated in Figure S2 and associated discussion.
As a calibration emitter, we use a thermocouple (TC) fabricated from 78 μm type C wire, with a roughly disk-shaped “bead” roughly 280 μm across and 150 μm thick. The TC bead is positioned at the trap center using the alignment cameras (Figure S1) and heated by the CO₂ laser with temperature read out electrically. Because emissivities of the W-Re TC materials have been well studied, the hot TC provides an emitter with well-defined intensity vs. λ, allowing S(λ) to be measured, so that the NP spectra can be corrected. Our primary interest is in extracting T_{NP} from fitting spectra, which depends only on the wavelength dependence, i.e., the spectral shape. We have not attempted to quantify the absolute emissivity of the NPs, but comparing the relative intensities of different NP spectra should be accurate to within a few percent. This estimate is based on the observation of spectrum-to-spectrum repeatability better than 2%.

Temperatures are extracted by fitting corrected NP spectra to the emission model in equation 1, and the major uncertainty is what to use for the emissivity function, ϵ(λ). For NPs much smaller than λ, as is the case here, scattering theory gives the expression:

\[ \epsilon(\lambda) = \frac{8\pi r}{\lambda} \cdot \text{Im} \left( \frac{(n+ik)^2 - 1}{(n+ik)^2 + 2} \right), \]  

where r is the NP radius, and n and k are the real and imaginary parts of the index of refraction of the material. The first term in this expression accounts for coupling of the optical field to NPs much smaller than the wavelength, and the second accounts for the bulk optical properties of the NP material. If n and k are wavelength independent, ϵ(λ) ∝ λ⁻¹, however, that is typically not the case. For example, if n(λ) and k(λ) data for room temperature graphite are used, ϵ(λ) decreases more rapidly with increasing λ (Figure S3). The figure also shows that a simple power law expression: \( \epsilon(\lambda) \propto \lambda^{-1.41} \), is a reasonable fit to the scattering theory expression, deviating by less than 3% over the wavelength range of interest.
It is impractical to use equation 3 in fitting NP spectra for several reasons. For most nanomaterials, optical (or equivalent dielectric) parameters are not available over our λ range, or for the high temperatures of interest. More fundamentally, even if *bulk* optical parameters were available over our λ and T\textsubscript{NP} range, the optical properties for small NPs would be modified by surface and defect states, and possibly by quantum confinement effects. Indeed, this last point is clearly illustrated by the data presented below.

Therefore, we are forced to use a model for $\epsilon(\lambda)$, and have chosen to use a power law function of the form $\epsilon(\lambda) \propto \lambda^{-n}$, where $n$ is a fitting parameter. The power law form was chosen because it is a reasonable approximation to $\epsilon(\lambda)$ for graphite, at least at room temperature, and adds only a single fitting parameter that affects the spectral shape. With this assumption, and combining all constants into a single normalization parameter, $K$, the emission model (equation 1) becomes:

$$I(\lambda, T_{NP}) = \frac{K}{\lambda^{4+n} \cdot (\exp\left(\frac{hc}{\lambda kT_{NP}}\right) - 1)}.$$  \hspace{1cm} (4)

This function depends on only three parameters: $T_{NP}$, $n$, and $K$, of which only the first two affect the shape of the function. As shown below, fits based on this model are not perfect, but this approach at least allows us to treat all the NPs on a common basis, so that properties can be compared for different NPs.

Several factors contribute to the uncertainty in $T_{NP}$ values extracted by fitting spectra with equation 4. Except for a few spectra of very low brightness NPs, the signal-to-noise in the spectra below is high enough that the major sources of uncertainty result from non-idealities in the optical system and their effects on the spectral intensity calibration process, and from the nature of the fitting function used to extract $T_{NP}$. These are analyzed in some detail in the Supporting Information.
The effects of mis-positioning the TC relative to the trap center or changing the TC temperature during calibration are explored in Figure S4 and associated discussion. The effects of misalignment of various optical elements are explored in Figures S5 through S9. The effects of deliberately mismatching the intensities of the visible and nIR spectra are explored in Figure S10. The emission intensities measured range over three orders of magnitude, thus camera non-linearity is a concern. We previously tested for non-linearity using the TC emitter and neutral density filters,\textsuperscript{20} and Figure S11 shows an additional NP-based test that also showed no significant saturation effects. These “experimental” issues contribute to uncertainties in the shape of the spectra, and given our ability to position the TC and optics, we estimate those contributions to the uncertainty in \( T_{NP} \) to be \( \pm 4.8\% \).

There is additional \( T_{NP} \) uncertainty resulting from fitting the spectra with equation 4, because this approximate emission model does not give perfect fits to the spectra. For example, a particular pair of \( n \) and \( T_{NP} \) parameters might give the best overall fit to a spectrum, but fits with different \( n \) and \( T_{NP} \) parameters might actually fit some spectral regions better, albeit with lower overall fit quality. Figure S12 examines how this behavior, together with the experimental signal-to-noise, introduces uncertainty in the \( T_{NP} \) values extracted. For spectra with reasonable signal/noise, we estimate this contribution to the \( T_{NP} \) uncertainty to be \( \sim \pm 4\% \). Taking the experimental and fitting uncertainties together, the total estimated uncertainty in \( T_{NP} \) is \( \sim \pm 6.2\% \).

This is the estimated absolute uncertainty in \( T_{NP} \) values extracted using equation 4. In many cases, we are interested in comparing \( T_{NP} \) values for different spectra, and because most of the experimental factors contributing to the uncertainty are identical in all spectra, the uncertainty in \( T_{NP} \) differences is mostly due to the fitting uncertainty of \( \pm 4\% \). Furthermore, for spectra with similar shapes, \textit{i.e.}, similar \( n \) parameters, some of the \( T_{NP} \) uncertainty resulting from the fitting
process should also cancel when comparing TNP values, and in such cases, the uncertainty in the TNP differences is estimated to be < 2 %.

Note, however, that the TNP values here are all extracted assuming the power law approximation for $\epsilon(\lambda)$. It is not possible to estimate how much TNP might change if the correct form of $\epsilon(\lambda)$ were known, and we can only point out that the fits based on this assumption are often quite good, and for those where the model does not work as well, we can at least see how it fails, i.e., how $\epsilon(\lambda)$ might need to be changed for different NPs at different temperatures.

This study focuses on the emission properties of carbon NPs, and the NP mass is only used to calculate mass ratios needed to approximately correct emission intensities for NP size (vide infra). We estimate that the absolute uncertainty in the mass measurements is $\sim \pm 1.3 \%$, mostly due to the estimated assembly precision of the trap, however, this systematic error cancels exactly in mass ratios. The relative uncertainty in comparing masses mainly results from uncertainty in fitting the $F_z$ scans used to measure Q/M, which depends primarily on the emission brightness of the NPs. For the NPs studied here, the typical uncertainty in mass ratios is estimated to be $\sim 0.2 \%$, with the exception of one experiment using a carbon dot NP, where the emission was very weak. Mass ratios involving that NP are uncertain by up to 2%.

III. Results

a. Graphite NP emission spectra at different temperatures.

We begin with graphite NPs, which have the highest purity of the materials studied (99.9999%), and the best known nanostructure, at least in principle. Figure 1 shows a set of experimental emission spectra (60 second acquisition time) for a single graphite NP with initial mass of 33.7 MDa and initial charge of +50e, taken at a series of increasing 532 nm heating laser powers ranging
from 6 to 48 mW, loosely focused through the trap and detected at the exit window of the vacuum system. If this NP were spherical with the bulk density of graphite, it would have a diameter of \( \sim 36 \) nm. Fits to the spectra based on equation 4 are shown as colored curves, and the best fit \( T_{NP} \) and \( n \) values are given in the legend. Equation 4 gives reasonably good fits at low \( T_{NP} \), but with increasing temperature the power law \( \epsilon(\lambda) \) function is unable to capture the curvature in the experimental spectra, failing most obviously in the region above 1250 nm. Note that there is a systematic decrease in the best-fit \( n \) parameter with increasing temperature.

It is also interesting to consider how \( T_{NP} \) is controlled by the balance between laser heating and cooling by thermal emission, Ar buffer gas collisions, and sublimation. Sublimation at these

![Emission spectra of a graphite NP being heated to five different temperatures (1296-2150 K \( \pm \) 2%), along with fits based on equation 4. The emissivity parameter \( n \), decreases as temperature increase.]

\[ \text{Graphite} \\
\text{M}_{\text{Initial}} = 33.7 \text{ MDa} \\
\text{Diameter} = .36 \text{ nm} \]
temperatures turns out to be too slow to result in significant cooling, and for the high temperatures and low buffer gas pressures used here, thermal emission is the dominant cooling mechanism.\textsuperscript{40} For example, a spherical NP of this size at 2000 K would emit on the order of $10^{-10}$ W of thermal radiation, estimated as Stefan-Boltzmann emission with emissivity of \textasciitilde0.02.\textsuperscript{10} Collisions with 1 mTorr of Ar buffer gas would carry away only \textasciitilde$10^{-12}$ W, assuming unit efficiency energy accommodation.

The total emitted power from an ideal blackbody varies like $T^4$. Because we are measuring only the short wavelength part of the spectrum, and the spectra both increase in intensity and shift to shorter wavelength with increasing temperature, the observed emission intensity is expected to vary more strongly with temperature than the total blackbody emission. Consistent with this expectation, the integrated number of photons emitted per second in the 600 to 1580 nm range increased by a factor of \textasciitilde19 when $T_{NP}$ was increased by a factor of 1.66, from 1296 K to 2150 K, varying like \textasciitilde$T^{5.9}$. Because the cooling power varies nonlinearly with $T_{NP}$, the laser power required to heat the NP also increased nonlinearly, by a factor of about four to obtain the 66% increase in $T_{NP}$. Because our laser beam divergence is power dependent, the exact relationship between laser power and focused intensity is uncertain.

b. Effects of carbon material structure on NP emission spectra.

Figure 2 shows analogous examples of emission spectra for NPs of four other carbon materials, with fits and extracted $T_{NP}$ and $n$ parameters. The carbon dot emission was too weak to obtain spectra except at very high $T_{NP}$. The mass of the graphene platelet and diamond NPs were similar to that for the graphite NP in Figure 1. The carbon black NP had nearly double the mass, and the carbon dot was \textasciitilde5 times less massive.
The spectral shapes for all the different carbon material NPs were grossly similar, and in particular, there were no obvious signs of structured emission in this wavelength range for any of the NPs, at least at the low spectral resolution used here to maximize signal – roughly 20 nm below 1000 nm, and ~70 nm at longer wavelengths. We note that carbon dot NPs are fluorescent in solution, but we observed no signs of fluorescence, even using low 532 nm laser intensities to minimize heating.

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While the spectral shapes are qualitatively similar for all the carbon NPs in Figures 1 and 2, there are differences in the wavelength dependence, i.e., in the spectral shapes, which can be seen in two
ways. The best-fit n parameters varied systematically between the NPs, with the graphite NP having the highest values, and the graphene platelet NP the lowest. High n corresponds to more sharply peaked spectra, with intensity falling faster at both long and short wavelengths, and low n corresponds to spectra with less sharply wavelength-dependent intensity. Furthermore, even though the graphite fit functions had the highest n values, they still were unable to capture the curvature of the experimental graphite NP spectra in the nIR.

The brightness of the different NPs in Figures 1 and 2 also varied dramatically; the carbon black and graphene NPs being >100 brighter than the carbon dot at similar TNP. To compare quantitatively, it is necessary to correct the intensities for the differences in NP size. For an ideal blackbody of radius r, intensity scales with surface area (r²), and for small particles there is an additional factor of r from the emissivity (equation 3). Thus, for spherical NPs, the intensities should scale as r³, i.e., with the NP volume. Because the shape of the NPs used is not known, this scaling is only approximate, but it provides at least a first order correction for the effects of varying NP mass. Using the measured NP masses and assuming bulk densities for the materials involved, we can estimate the volume ratios. For carbon black, reported densities vary for different carbon blacks and for different measurement methods, and also can change with annealing. In the following we have assumed a density of 2.0 g/cm³, compared to 2.26 for graphite. The carbon dot density is unknown, and was assumed to be equal to that of graphite, as was that for the graphene platelet NP. To compare the intensities for the NPs in Figure 2 to those for the graphite NP in Figure 1, the Figure 2 intensities should be multiplied by the following volume ratio factors: Carbon Black = 0.4, Graphene Platelet = 0.8, Nano diamond = 1.6, Carbon dot = 4.2. As noted in the Experimental section, the worst case uncertainty in the mass ratios used to calculate the volume-scaling factors is ~0.5%. This is negligible compared to the uncertainty in how brightness
might scale with NP shape and how NP density might differ from the bulk values, which are the main reasons that we note that volume scaling is only an approximate correction for the effects of NP size.

The T\textsubscript{NP} values extracted from fitting the NP spectra varied in the five experiments, but by comparing spectra at similar T\textsubscript{NP}, it is clear that the graphene platelet NP had the highest volume-corrected (and raw) brightness of any of the NPs. The raw graphene NP intensities were roughly 3 times higher than those for the graphite NP, or \(~2.4\) times brighter after correction for the larger graphene NP volume. After correction for the NP volumes and taking the differences in T\textsubscript{NP} values into account, the carbon black and nano diamond NPs both had brightness similar to that of the graphite NP, i.e., \(~2.4\) times lower than that of the graphene NP. We attribute the greater brightness of the graphene NP to its platelet shape, which would give it substantially higher emitting surface area than the more compact graphite, carbon black, and nano diamond NPs.

In terms of brightness, the real outlier was the carbon dot NP, which emitted much more weakly than any of the other materials. Even after volume scaling to correct for the small size of carbon dot NP, the intensity in the 1\textsuperscript{st} carbon dot spectrum (red) at 2220 K was \(~25\) times lower than that of the graphite NP at 2150 K, i.e., 70 K colder. The other interesting observation was that in a 2\textsuperscript{nd} spectrum (blue) taken for the same NP about 20 minutes later, the intensity increased by a factor of \(~3.8\) for the same extracted T\textsubscript{NP} value, while the n parameter decreased by \(~40\%\), corresponding to a less sharply peaked spectrum. The increase in brightness in the 2\textsuperscript{nd} spectrum is interesting, however, the extreme weakness of the emission makes detailed study difficult. The next section examines similar behavior for the other, brighter carbon NPs.

It is also interesting to compare the T\textsubscript{NP} dependence of the emission intensity for the different carbon NPs in Figures 1 and 2. The log of the integrated numbers of photons emitted/second from
each NP, in the 600 to 1600 nm range, are plotted as a function of $T_{NP}$ in Figure 3. If intensity scales like $T_{NP}^n$, then plots of log $I$ vs. log $T_{NP}$ should be linear, with slope = n. With the exception of the nano diamond NP, the intensity data are reasonably well fit by straight lines, with slopes ranging from 5.9 for the graphite NP to 6.3 for the graphene platelet NP, i.e., the black carbon NPs all have intensities that vary roughly as $T^6$. For the nano diamond NP, the three highest $T_{NP}$ points ($T_{NP} > 1770$ K) are also fit well by a line with slope near 6, however, the intensities at the two lower $T_{NP}$ values fall well below this trend, suggesting that the emission properties of the diamond NP changed significantly as it was initially heated. As shown in the next section, all the carbon NPs show significant effects of heating on emissivity.

c. Effects of thermal history on NP emission spectra for carbon NPs

To examine the effects of heating on NP emission spectra, a series of experiments were done in which individual NPs were first heated at low $T_{NP}$ for charge determination, then a series of spectra were measured as $T_{NP}$ was stepped up, and then stepped back down. The goal was to compare spectra measured at similar $T_{NP}$ values, before and after the NP was heated into the range where sublimation caused significant mass loss. We attempted to hit the same set of $T_{NP}$ values during the heat-up and cool-down phases of the experiment, but because the emissivity properties
changed, the actual $T_{NP}$ values extracted from the spectra sometimes differed. Nonetheless, comparing spectra at similar temperatures provides insights into heating effects on the brightness and shape of the spectra. It should be noted that essentially all the mass loss occurred during the times when $T_{NP}$ was above 1900 K, thus we note these times.

Figure 4 shows experiments of this type on two different carbon black NPs, both smaller than the carbon black NP in Figure 2. Figure 4A compares two spectra taken for a carbon black NP of initial mass 31.13 MDa, with $T_{NP}$ near 1670 K. The 1st spectrum, taken during the heat-up phase, had extracted $T_{NP} = 1668$ K. The 2nd spectrum, with extracted $T_{NP} = 1679$ K, was taken after the NP had spent ~50 minutes at temperatures $\geq 1900$ K, during which time sublimation decreased the mass by ~15% to 26.33 MDa. The raw integrated emission intensity (in the 600 to 1580 nm range) after heating was 5.5 % higher than before, even though the NP volume was smaller. In Figures 4 and 5, the intensities for the 2nd (or 3rd) spectra for each NP have been scaled by the volume ratio (to the 1st spectrum), to correct for NP mass loss. With that volume scaling, the integrated intensity in the 2nd spectrum was ~25% higher than in the 1st spectrum, however, $T_{NP}$ was also ~10 K higher. If we assume that the emission in this wavelength range scales like $T^6$, as suggested by Figure 3, then the 10 K temperature increase would have made the 2nd spectrum only ~4% brighter, i.e., the increase in brightness was substantially greater than would be expected from the small $T_{NP}$ difference.

It is interesting to compare the change in emission intensity to the change in laser power required to heat the NP in the 2nd spectrum. If the changes were identical, this would imply that the NP absorption cross section at 532 nm (heating) and its broadband emissivity (cooling) tracked together as the NP evolved. In this case, the raw emission intensity was 5.5 % higher in the 2nd
Figure 4: A and B are emission spectra for a single 31 MDa carbon black NP. C and D are spectra for a 50.8 MDa carbon black NP. Spectra are shown before and after periods in which the NP was heated above 1900 K. The order in which the spectra were taken is shown by the masses, with mass always decreasing after heating. The left-hand spectra are volume scaled, and right-hand spectra are normalized at 1580 nm to allow shape comparison.

In Figures 4C and D, a similar experiment on a larger carbon black NP is shown, with three emission spectra taken at T_{NP} \approx 1550 K. The first (red) was measured prior to any heating above
the ~1553 K measurement temperature, at which point the mass was 50.79 MDa. The second (blue) was taken after $T_{NP}$ had been stepped up and back down, with a ~10 minute heating period at >1900 K, during which the mass had decreased by 6.85%, to 47.31 MDa. $T_{NP}$ in the 2nd spectrum was only 3 K higher than in the 1st, thus the emission intensity, spectral shape, and heating power should be directly comparable in the two spectra.

The raw intensity in the 2nd spectrum was substantially (~43 %) higher than in the 1st, or 54% higher if the 6.8% volume (i.e. mass) change is taken into account. In this case, ~9.5% more laser power was needed to compensate for the greater radiative cooling in the 2nd spectrum (29.22 mW), compared to the first (26.70 mW). Note, however, that the increase in laser power required in the 2nd spectrum was substantially smaller than the 43% increase in raw integrated emission intensity, indicating that once again, the increase in 532 nm absorption cross section was more than enough to compensate for the increase in broadband thermal emissivity.

The 3rd spectrum (yellow) was taken after a second stepped heating program, including another ~10 minute period above 1900 K, leading to further decrease in the NP mass to 43.99 MDa. The volume-scaled intensity was lower than in 2nd spectrum, however, a ~ 7% decrease (assuming $I \propto T^6$) would be expected from the ~20 K lower 3rd spectrum $T_{NP}$. The intensity remained well above that in the 1st spectrum.

As shown by the normalized spectra in Figures 4B and D, the increases in emission brightness for the carbon black NPs after >1900 K heating were not accompanied by significant changes in spectral shape. Note also that the power-law-based emissivity model (equation 3) fit the spectra reasonably well, and with significantly lower $n$ values, compared to the graphite NP in Figure 1.

Figure 5 shows similar pre-heating/post-heating emission spectra comparisons for graphene, graphite, and nano diamond NPs. On the left side, spectra are plotted to allow intensity
comparisons, with the post-heating spectra scaled by the volume ratio to approximately account for the ~5 – 20% mass losses that occurred in the >1900 K heating periods. On the right side, spectra are normalized at 1580 nm to show the effects of >1900 K heating on spectral shapes.

Figure 5A shows spectra for two different graphene platelet NPs, with arrows indicating which intensity scale applies to each spectrum. The left-hand scale is for an NP with initial mass of 11.68 MDa, which was heated by 19.7 mW of 532 nm laser power giving an initial spectrum fit with $T_{NP} = 1645$ K and $n = 1.03$. The NP was then heated above 1900 K for 94 minutes, resulting in a final mass of 10.34 MDa. In the 2nd, post-heating spectrum, 22.63 mW of 532 nm power was needed to reach 1640 K, with best fit $n = 0.90$. In this example, the raw integrated intensity increased by 47% between the spectra, and after applying the 1.13 volume scaling factor, the increase was 66%. In addition to the increase in emission brightness, Figure 5B shows that there was also a significant change in the wavelength dependence, which is reflected by a drop in the best-fit $n$ value to 0.9, implying a slower decrease in emissivity with increasing wavelength. For this NP, the 15% increase in 532 nm heating power in spectrum 2 was much smaller than the 47% increase in raw integrated intensity, again implying that the 532 nm absorption cross section increased more than the emissivity for broadband radiative cooling.

The second graphene NP in Figure 5A (right axis) was ~six times more massive than NP 1, and the emission intensity was significantly larger, even though the spectra were measured at lower $T_{NP}$, near 1290 K. In the 1st spectrum, $T_{NP}$ was 1288 K and $n$ was 1.3, then the NP was heated above 1900 K for 21 minutes, during which time the mass dropped by 14.42 MDa. The 2nd spectrum had $T_{NP} = 1289$ K with $n = 1.02$, and in this case, the raw intensity was ~16% lower than in the 1st spectrum. After volume scaling to account for the mass lost during the >1900 K heating,
Figure 5: Emission spectra taken before and after >1900 K heating for graphene, graphite, and nano diamond NPs. The order in which spectra were taken is shown by the masses, which decreased during >1900 K heating periods. A, C, and E show volume-scaled intensities. B, D, and F are normalized to show changes in spectral shape. Note that the emission intensities changed after >1900 K heating, and that there were also significant changes in spectral shape.
the 2\textsuperscript{nd} spectrum intensity was \~4.8 \% higher than in the 1\textsuperscript{st}, and as shown in Figure 5B and by the drop in n, the wavelength dependence was significantly flatter after heating.

Figure 5C shows data for two graphite NPs. NP 1 was a small graphite NP with initial mass of 6.3 MDa, initially heated to 1649 K with 532 nm excitation, then heated above 1900 K for \~17 minutes, during which time its mass decreased \~10\% to 5.71 MDa. The post-heating spectrum was at slightly lower T\textsubscript{NP}, (1641 K); nonetheless the integrated emission intensity increased dramatically, by factor of 2.46 (raw) and 2.7 (volume-scaled). In addition, Figure 5D shows that the spectral shape changed significantly, in this case with higher post-heating n value, implying more sharply wavelength-dependent emissivity. The laser power required to heat the NP increased by only 3.7\% for the 2\textsuperscript{nd} spectrum, compared to the 2.4 x increase in raw emission intensity. Thus, even though the broadband emissivity increased dramatically when the NP was heated, the increase in 532 nm absorption cross section was even larger.

The 2\textsuperscript{nd} graphite NP in Figure 5 C had mass of 14.22 MDa at the point the 1\textsuperscript{st} spectrum was measured, resulting in T\textsubscript{NP} = 1727 K, and n = 1.16. After heating to 1900 K for 15 minutes, which caused only 0.05 MDa mass loss, a 2\textsuperscript{nd} spectrum was measured with T\textsubscript{NP} = 1726 K and n = 1.05. For this NP, the brightness of the emission decreased by \~9\% (raw). Even after volume scaling, the post-heating emission intensity was \~8.5\% lower than in the initial spectrum.

Figures 5E and F show analogous results for a diamond NP with initial mass of 28.51 MDa, with a 1\textsuperscript{st} spectrum taken at 1643 K (n = 1.0) with 33.08 mW of 532 nm heating power. The NP was then heated in steps up to nearly 2200 K over a total of 3780 seconds, resulting in \~5\% mass loss to 27.07 MDa. In the 2\textsuperscript{nd} spectrum, 21.86 mW was required to heat the NP to 1646 K (n = 0.78). The post-heating intensity increased substantially (34\% raw, 41\% volume scaled), and the shape also changed, in this case with lower post-heating n value, implying a reduced dependence of
emissivity on wavelength. For this NP, 33% less heating laser power resulted in 34% higher raw emission intensity in the 2nd spectrum, implying that again, the 532 nm absorption cross section increased substantially more than the broadband emissivity.

The data in Figures 4 and 5 show that heating carbon NPs into the range where sublimation becomes significant can result in substantial changes in emissivity, with emission often, but not always, substantially brighter after heating, even without correction for the mass loss. In addition, the wavelength dependence of the spectra, hence the form of $\epsilon(\lambda)$, can change significantly, with the $n$ parameter (i.e., spectral curvature) sometimes increasing, sometimes hardly changing, and sometimes decreasing significantly.

d. Comparisons of emission properties for different NP materials

To provide a different perspective on how the intensities and spectral curvatures vary from material-to-material, and how different materials react to heating, Figures S13 and S14 replot the data in Figures 4 and 5, comparing spectra for the different materials. Specifically, the pre- and post-heating spectra for the graphite NP1 in Figure 5C, are plotted together with the analogous spectra for the graphene, carbon black, and diamond NPs, all at similar temperatures (1660 ± 20 K). Spectra are compared both before and after >1900 K heating, and both volume-weighted intensities and spectra normalized at 1580 nm are compared.

In essence, the results are: 1. Prior to >1900 K heating, the graphite NP (Figure 5C) had volume-corrected emission intensity 30 to 50% lower than the intensities for the graphene (4A), carbon black (3C), and nano diamond NPs (4E). The carbon dot NP had much lower intensity – far too weak for spectral measurements at 1600 K. 2. The $n$ parameter for the small graphite NP (0.91) was ~10% smaller than those for the other three NPs (1.0 to 1.05), but the spectral shapes prior to >1900 K heating were still nearly superimposable. 3. Heating above 1900 K increased the
intensities for all the NPs, but with the largest effect on the graphite NP, such that after heating, the volume-scaled intensities for the graphite, graphene, carbon black, and diamond NPs were all similar. 4. The n parameter and associated curvature of the graphite NP spectrum increased after heating, while those for graphene and diamond decreased slightly, and that for carbon black remained essentially unchanged.

It is tempting to associate these differences between the NPs to the differences in the materials from which the NPs were made, however, it is important to note that the NP feedstocks are heterogeneous, with distributions of mass and shape that are expected to influence the electronic and phonon density of states, hence the emission properties. This point is made clear Figures 4 and 5, where pairs of carbon black, graphite, and graphene NPs are shown to have substantially different emission behavior. Understanding the effects of structural heterogeneity on both emission spectra and reaction kinetics is the main motivation for our single NP experiments.

e. The effects of NP mass on emission spectra

One obvious point to be explored is the effect of NP size on the spectral properties. As discussed above, emission intensity is expected to scale roughly with NP volume, but we are also interested in possible effects of NP size on the wavelength dependence of the spectra. Such effects can be summarized Figure 6. Emissivity properties of ten carbon NPs vs. mass. The n parameters are extracted by fitting spectra for five graphite NPs with T_{NP} near 1652 K ± 2%, and five carbon black NPs near 1902 K ± 2%.
by comparing the n parameters extracted by fitting spectra for different size NPs at similar temperatures. Figure 6 shows typical data for ten different carbon NPs, with masses ranging from just under 5 MDa, to ~85 MDa, which would correspond to diameters ranging from ~20 to ~54 nm, if the NPs were spherical, with the bulk densities. The figure shows the n parameters for five graphite NPs in the 5 to 45 MDa range, obtained by fitting spectra with T_{NP} close to 1652 K. Data are also included for five carbon black NPs in the mass range from ~30 to 85 MDa, taken from spectra with T_{NP} close to 1902 K. (These temperatures were used because spectra were available for different NPs at very similar T_{NP} values). As noted in the discussion of Figures 1 and 2, the n parameter tends to decrease with increasing T_{NP}, and also tends to be larger for graphite than for carbon black, and the data in Figure 6 is consistent with both tendencies.

For the graphite NPs, there appears to be a trend toward smaller n parameter (weaker dependence of \(\epsilon^*\) on \(\lambda\)) with increasing mass, however, the NP-to-NP scatter in best-fit n values was quite large, presumably reflecting the structural heterogeneity of the graphite NP feedstock. For the somewhat larger carbon black NPs, the scatter was smaller, but with no obvious trend of n vs. mass. These results do not rule out systematic effects of NP mass on spectral shape, but clearly any mass effects are obscured by the effects of NP structural heterogeneity.

f. The effects of NP charge on the NP emission spectrum and extracted T_{NP} value

The NPMS method requires that the NPs be charged so they can be trapped, however, the charge-to-mass ratios are quite low – on the order of \(~10^{-6}\) e/Da, corresponding to \(~10^{-5}\) e/atom. Therefore, the effect of the charge on NP electronic and vibrational density of states should be negligible, however, there could be spectral artifacts due to the fact that the amplitude of NP motion in the trap is charge dependent.\(^{27-28}\) In essence, changing the charge changes the effective “size” of the NP emitter, which could affect the intensity calibration and the laser-NP spatial overlap. Figure 7
shows an experiment in which the secular motion frequency, $F_z$, and $T_{NP}$ were monitored for a graphite NP with initial mass of 43.84 MDa, as the NP charge was varied in steps of $\pm e$ using a vacuum ultraviolet lamp to drive photoemission from both the NP and from trap surfaces (leading to electron capture by the NP). From the size of the steps in $F_z$, the absolute charge can be determined, as indicated in the figure. For this experiment, the program normally used to stabilize $T_{NP}$ was not used. Instead, the heating laser was set to constant power, giving initial $T_{NP}$ around 1650 K, which drifted slowly down over the course of the 6.5 hour-long experiment.

A series of 23 charge steps was observed, including one triple step, with $Q$ varying from $+48e$ to $+54e$, i.e., by $\sim 11\%$. The root mean square radial amplitude$^{27-28}$ of the NP radial motion would have decreased from $\sim 53\ \mu m$ for $Q = +48e$, to $\sim 47\ \mu m$ for $Q = +54e$, i.e., also by $\sim 11\%$. It is clear that there were no significant effects of the NP charge on $T_{NP}$, even for the triple charge step near 7100 seconds.

A related question is whether the axial motion driven to measure $F_z$ has any effect on the spectra and extracted $T_{NP}$ values. As shown in Figure S15, the answer is “no”.

Figure 7. The effect of NP charge stepping on $T_{NP}$ measurement. Secular frequency (green), $T_{NP}$ (red), and charge state (blue, $Q$) measurements for a single 43.84 MDa graphite NP, as a series of charge steps was carried out.
g. Effects of heating laser wavelength on the NP emission spectrum

The fact that broadband blackbody-like emission spectra are observed when irradiating the NPs at 532 nm, shows that energy redistribution must be fast in hot carbon NPs. The obvious question is whether it is fast enough to eliminate all memory of the excitation process, or if there might still be residual effects of heating laser wavelength. In Figure 8 we compare heating using the cw 532 nm laser, presumably exciting electrons in the NP, to heating via vibrational/phonon excitation using a quasi-cw, duty-factor-modulated (at 20 kHz) 10.6 μm CO$_2$ laser.

![Figure 8](image_url)

Figure 8. A: Emission spectra for a single carbon black NP with mass of 37.62 MDa, heated to similar temperatures by both 532 nm and 10.6 μm radiation. B: The same carbon black spectra normalized to allow shape comparisons. C and D: Analogous spectra for a single graphite NP with initial M = 14.2 MDa, heated at 532 nm or 10.6 μm. No significant effects are observed.
The experiment was done for both a carbon black NP of initial mass 37.62 MDa, and a graphite NP of initial mass 14.2 MDa. For the carbon black NP, a set of spectra was taken at $T_{NP} \sim 1550$ K using first the 532 nm laser, and then the 10.6 μm laser, then the process was repeated for sets of spectra with $T_{NP}$ near 1900 K. In Figure 8A, pairs of spectra with very similar $T_{NP}$ values are compared, and it can be seen that the raw intensities and $n$ parameters were slightly lower for the CO$_2$ laser heating, however, comparison of the normalized spectra in frame B shows that they are nearly superimposable.

For the graphite NP in frames C and D, the intensity observed with the CO$_2$ laser was slightly higher than that measured with the 532 nm laser, however, in this case, $T_{NP}$ was somewhat higher during the 10.6 μm heating. Again, although the $n$ parameters were slightly different, the normalized spectra are nearly superimposable.

Clearly, the effects of heating laser wavelength on the shape and intensity of NP emission spectra are small. Indeed, given the $\sim T^6$ dependence of intensity on $T_{NP}$ and the estimated ±2% uncertainty in comparing $T_{NP}$ values, the small intensity differences observed are not significant. In addition, the time scales of these experiments were long enough that some evolution of the NP emission properties probably occurred between the 532 nm and CO$_2$ laser experiments. We conclude, therefore, that there is no evidence for any residual effect of heating laser wavelength on the emission properties of black carbon NPs.

IV. Discussion

The results show that NPs of black carbon materials like graphite, graphene, and carbon black have blackbody-like thermal emission spectra. The same is true for boron-doped nano diamond and carbon dot NPs, at least for the high temperatures probed here. No sign of structured emission was observed for any of the NPs.
Nonetheless, there were significant NP-to-NP variations in both the intensity (volume-corrected) and wavelength dependence of the thermal emission spectra from carbon NPs. This variability is attributed to structural variations in the NPs, including both variations in NP material (i.e., graphite, graphene, diamond, carbon black), and variations in the structure of individual NPs of the same material. Furthermore, for individual NPs, the emission brightness and wavelength dependence often change significantly if the NPs are heated hot enough to drive significant sublimation, and these changes are attributed to evolution of the NP structure at high $T_{NP}$.

One possibility, clearly, is that structural differences might include differences in the NP composition, which also might account for the evolution of the NP spectra after heating. Consider the graphite, graphene, and carbon black NPs, where the feedstocks all had stated non-volatile impurity levels well below 1 %. As initially trapped, these NPs probably had surface groups with O, H, or N heteroatoms, as well as adsorbates such as water, methanol, and ammonium acetate, however, all these should have decomposed and desorbed well below 1400 K. Therefore, except perhaps in a few spectra at low initial $T_{NP}$, these NPs should have been near pure carbon. The nano diamond NPs would have had significant concentrations of boron and impurity atoms, and it is unclear to what extent our temperatures were high enough to allow them to diffuse in the diamond lattice, or what spectral changes that might cause. Similarly, the initial composition of the carbon dot NPs is unknown, thus it is unclear why the emission was so weak. To avoid the issue of impurity effects on the spectra we focus the discussion on graphite, graphene, and carbon black NPs.

Thermal emission depends on the density and occupancy of electronic and vibrational/phonon states of the NPs. Graphite and graphene NPs have fully coordinated basal plane sites, both on the NP surface and in the interior, as well as low coordination sites such as basal plane edges on the
NP surface, and defects in basal planes either on the surface or in the NP interior. These low coordination sites should make different contributions to the electronic and vibrational state distributions, compared to fully coordinated sites. Therefore, changes in the site distribution will change the state distribution, hence the spectra. For carbon black, where the initial structure includes both crystalline (graphitic) and amorphous regions, and some porosity, there may be changes in the distribution of low coordination surface and defect sites, but also larger scale structural changes such as crystallization and densification.

One structural evolution process expected at high temperatures is annealing. The melting point of graphite (at high pressures) is near 4800 K\(^4\) thus our experimental T\(_{\text{NP}}\) range corresponds to ~25 to 45% of the melting point. Given our long experimental times, it is likely that some annealing occurs, particularly during the >1900 K heating periods. Annealing should tend to reduce both the number and relative concentration of defects and other low-coordination sites, because these are less thermodynamically stable than fully coordinated sites.

The competing process that clearly occurs, again primarily at higher T\(_{\text{NP}}\), is sublimation. In the experiments summarized in Figures 4 and 5, the “high” temperatures were in the 1900 to 2200 K range, and the mass losses during the heating periods ranged from 5 to 20% of the NP mass. The NPs were in the 6 to 50 MDa size range, which would have 3 to 6% of their atoms in the surface layer if the particles were spherical. The actual particles would have had larger fractions of their atoms in the surface – much larger for the graphene platelet NPs. Thus, on the order of a monolayer’s worth of material would have desorbed from each NP during the >1900 K periods, corresponding to sublimation rates on the order of only 10\(^{-2}\) to 10\(^{-3}\) monolayers/second. Note, however, that sublimation is not expected to proceed in a layer-by-layer fashion. For graphite and graphene NPs, the low coordination edge and defect sites are less stable than basal plane sites, and
thus should desorb at much higher rates, however, with increasing $T_{\text{NP}}$ sublimation from basal plane sites, which are more stable but also more abundant, should start to occur more frequently, which would tend to create new low coordination defect sites. For carbon black, the situation is more complex, but sublimation from low coordination sites should still dominate.

Note that if there were significant concentrations in the buffer gas of reaction species, such as $\text{O}_2$ or $\text{O}$ atoms (possibly created in a discharge), these would tend to etch the NPs. The instrument is constructed mostly with UHV-compatible hardware, apart from a few o-rings sealing parts like windows. Because the ESI source introduces significant gas loads each time an NP is injected, the typical base pressure is in the low $10^{-7}$ Torr range, setting an upper limit on the $\text{O}_2$ concentration. In experiments in which $\text{O}_2$ is leaked into the trap chamber at $>100$ times higher pressure, mass loss rates are typically only a few $\text{C}$ atoms/second at 1900 K, thus we believe the effects of reactive background gas are negligible.

The important point is that sublimation can lead to net creation or destruction of low coordination sites, depending on the NP structure. Consider an ideal graphite NP in which all the basal planes are perfect, with low coordination sites only around their edges. In that case, sublimation would occur mostly at the edges, shrinking the size of the basal planes, which would reduce the number of edge atoms, but increase the fraction of edge atoms as the NP loses mass. If the exposed basal planes also have defects like vacancies, sublimation from under-coordinated sites surrounding the defects would tend expand the defects, similar to the etch pit formation observed in high temperature reactions of graphite defects.\textsuperscript{44-49} Thus sublimation at defects would further increase the fraction of low coordination sites.

Sublimation can also decrease the fraction of low coordination sites, however. Consider a rough NP with high surface area and many low coordination sites due to the presence of asperities on the
NP surface. Because the asperities have high surface area and many low coordination sites, they should sublime more rapidly than the bulk of the NP, eventually disappearing, generating a smoother NP with fewer low coordination sites. Similar considerations apply to the case where the NP surface is decorated by smaller particles, which also would tend to sublime rapidly. In either case, both the number and fraction of low coordination surface sites should initially decrease as these small features sublime. Therefore, the structural changes that occur as an NP is heated depend on the initial NP structure, and because carbon NP feedstocks are quite heterogeneous, significant NP-to-NP variability in the effects of heating is expected, and observed.

**Conclusions**

The results presented above show that hot graphite, graphene, carbon black, nano diamond, and carbon dot NPs all have blackbody-like thermal emission spectra, regardless of heating laser wavelength and NP mass and charge, at least in the range studied. There are substantial variations from NP to NP in emission brightness and spectral shape, and while there do appear to be some effects of the carbon material on the emission properties, the magnitude of these material effects is comparable to the variation in properties between different NPs of the same material. In addition, for all the NPs studied, there were substantial changes in both the intensity and shape of the emission spectra after the NPs were heated above 1900 K. The different NPs still have different spectral properties, but in general the NP-to-NP differences are smaller after >1900 K heating. We attribute the large NP-to-NP variability to the variations in the number and type of low coordination carbon sites on the different NPs, and the evolution of the NP properties during heating to the effects of annealing and sublimation, changing the site distributions. This kind of NP-to-NP variation in emission properties, as well as the evolution in emission properties with its NP-to-NP variability, can only be studied using single NP methods of the type presented here.
Ideally, we would like to correlate the NP emission behavior with the actual distributions of carbon sites on individual NPs, and this should be possible to some extent based on correlating emission behavior to sublimation rates. This is because the sublimation rate also depends on the distribution of sites present (on the NP surface), with the less stable low coordination sites having the highest rates. At this point, the number of individual NPs and the range of heating effects studied is too small to allow correlations between emission behavior and sublimation rates to be discerned with any confidence. We are, however, beginning a systematic study of the sublimation kinetics for graphite, which will entail measuring sublimation rates and emission spectra for many particles subjected to a variety of heating regimes. One goal of this study will be to understand the factors that control graphite emissivity in more detail, by correlating emission behavior, and the temperature dependence of sublimation, and eventually oxidation kinetics, which provide means to examine the number and energetics of sites on the NP surfaces. Again, this kind of information can come only from single NP experiments.

Supporting Information.

The Supporting Information is available free of charge via the Internet at http://pubs.acs.org.

The following files are available free of charge.

Figures S1 through S15 and associated discussion illustrate the power law emissivity function, effects of varying \( n \) on fitting, comparisons of NP emission spectra for different materials, effects of drive voltage sweeps on \( T_{NP} \), the optical setup and calibration method, the effects TC position and temperature on \( T_{NP} \), and the effects of various optical element misalignments on \( T_{NP} \) (PDF).

Data underlying Figures 1 through 8 are provided in tabular form (XLSX).

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**Author Contributions**

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. ‡These authors contributed equally.

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**ABBREVIATIONS**

NP, nanoparticle; T_{NP}, nanoparticle temperature; VUV, vacuum ultraviolet.

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Thermal Emission Spectroscopy of Single, Isolated Carbon Nanoparticles: Effects of Particle Size, Material, Charge, Excitation Wavelength, and Thermal History
Optical System Design:

Figure S1 shows the NPMS optics design. The trap is shown near the center of the dashed rectangle representing the vacuum chamber, and mirrors for focusing the CO₂ laser through the trap are shown, along with the laser beam path. 532 nm or other lasers are focused through the trap into the plane of the figure. The insertable thermocouple (TC) calibration emitter is shown schematically above the trap, and the coordinate system used in the following description is shown above that.

Light emitted axially (in the +z direction) from the trap center is collected by a five element lens system consisting of a 35 mm focal length achromatic doublet, a 100 mm focal length achromatic
Figure S1. Alignment camera images at the bottom show views of a single NP (bright red/yellow spots) superimposed on images of the TC (larger irregular grey/green features), positioned for calibration experiments. Thermal emission is collected out of the bottom of the trap for mass and charge determination, and along the trap axis for spectra/TNP measurements.

doublet, and a -75 mm focal length lens. This collects about one steradian solid angle from the trap center, and forms it into a slightly convergent beam. When the TC is used in calibration experiments, the ND filter assembly is inserted, cutting the light intensity by a factor of $\sim 10^8$, with transmission vs. wavelength measured as described previously.\(^1\) For alignment purposes, a mirror can be inserted into the beam path, directing the light onto a room temperature CCD camera (The Imaging Source) that captures magnified images of the trap center from the axial direction for
alignment purposes. During spectral measurements, the mirror is retracted, allowing the light to reach a 980 nm dichroic beamsplitter that reflects wavelengths ($\lambda < 980$ nm (“visible”)) and passes light with $\lambda > 980$ (“nIR”). The visible beam is passed through a 100 mm focal length plano-convex lens, a 550 nm long pass filter used to block scattered 532 nm laser light, and then is injected into a 1 mm diameter low OH optical fiber using a fiber collimator (Thorlabs, $f = 8.00$ mm, $NA = 0.50$, F240SMA-780 nm). The visible spectrum is dispersed by an Andor Shamrock 163 spectrograph equipped with silver-coated mirrors and grating, and measured by a back-illuminated Si CCD camera with 2000 pixel x 256 pixel CCD array with pixel sizes 15 $\mu$m x 15 $\mu$m, thermoelectrically cooled to -65 °C (Andor, DU416A-LDC-DD).

The nIR beam passes through a 175 mm focal length plano-convex lens, and a 900 nm long pass filter used to block 2nd order light in the nIR spectrum. A fiber collimator (Thorlabs, $f = 8.12$ mm, $NA = 0.49$, F240SMA-C-1310 nm) is used to inject the light into a 1 mm diameter low OH fiber, which conveys it to another Andor Shamrock 163 spectrograph, equipped with silver coated mirrors and grating. The nIR spectra are acquired by an InGaAs photodiode array camera, with a 512 pixel x 1 pixel array with pixels that are 25 $\mu$m wide by 500 $\mu$m high, cooled to -85° C (Andor, DU490A-1.7).

Light emitted radially out of the bottom of the trap is collected by a 31 mm aspheric achromat, and formed into a slightly convergent beam. Normally this is reflected by a retractable mirror, passed through a 50 mm focal length achromatic lens doublet, and injected into a 400 $\mu$m diameter low OH fiber that conveys the light to an avalanche photodiode (APD) pulse counting module (Laser Components Count). The APD signal is monitored as a function of time to detect the secular motion of the NP in the trap, for mass determination. When the mirror is retracted, the light is
focused onto a second room temperature CCD camera (The Imaging Source), generating a magnified view of the central volume of the trap from the radial direction, for alignment purposes.

**Optical system calibration:**

The optical system has non-idealities such as chromatic aberrations, detector quantum efficiencies, grating efficiencies, etc., that affect its sensitivity vs. wavelength, $S(\lambda)$, thus distorting the measured spectra. Before fitting spectra to extract temperatures, it is essential to correct the NP spectral intensities, i.e., we need to know $S(\lambda)$ quantitatively. To measure $S(\lambda)$ we need a calibration light source with well-known intensity vs. $\lambda$, small enough to fit into the trap center so that calibration can be done on the system as a whole, under conditions identical to those used for the NP spectral measurements.

As a calibration emitter, we use a micro-thermocouple (TC) with a roughly disk-shaped bead about 280 $\mu$m in diameter, and ~150 $\mu$m thick. The TC is positioned so that its bead is in the trap center, where it can be heated by the CO$_2$ laser, with the bead temperature read out electrically. The TC wire manufacturer (Concept Alloys) states that the accuracy in the temperature range of interest here is ±1% of the measurement in °C. To enable the TC bead to be positioned precisely and repeatedly, the TC is mounted on a precision XYZ vacuum manipulator, and the axial and radial alignment cameras allow the position in two orthogonal planes to be imaged. The approach to determining the correct TC position is to first trap a single NP, and image it with the two cameras, thus defining the trap center precisely. Then the TC is moved into the trap, heated, and its position is adjusted until it is at the position defined by the NP.

The bottom of Figure S1 shows superimposed images of a single NP and the TC, from the radial (left) and axial (right) alignment cameras. In the radial camera image, looking from the bottom of the trap, the NP appears as a bright yellow/red oblong in the center, and the laser-heated TC
appears as the larger irregular pale green shape with lead wires visible extending from the bead at the top and bottom of the image. The axial camera shows the NP as a circular red spot, and the TC bead as a roughly circular pale grey area, with lead wires visible at the top extending to either side.

The NP image makes an important point: The effective “size” of the trapped NP as a light emitter is determined by the amplitude of its secular motion in the trap, which depends on NP mass and charge and the trap operating conditions. For the hot carbon NPs of interest here, the NP effective size (FWHM) is typically in the 30 to 40 μm range along the axial direction, with double this “size” in the radial direction, hence the oblong appearance of the NP in the radial camera image. Thus, for light collection along the axis, the NP emitter “size” is ~60 - 80 μm in diameter.

Figure S2 illustrates the process of using the TC emission spectra to determine S(λ) and to correct NP spectra. Figure S2A shows a typical TC spectrum measured at a junction temperature of 2316 K. This measurement was done with the ND filter assembly inserted into the optical path, so that the intensity of the calibration spectrum was similar to those for single NPs. The scale is in counts/second/m² of TC emitter surface area (area ≈ 2.93x10⁻⁸ m²). The approach to measuring the ND filter assembly transmission has been described previously, along with the transmission vs. wavelength.¹ Figure S2B shows the predicted TC emission spectrum, which is the product of three functions: 1. The Planck’s law emission function for an ideal blackbody at 2316 K. 2. The emissivity of the W-Re TC at 2316 K²⁻⁵ 3. The ND filter assembly transmission. The ratio of Figure S2A to Figure S2B is the desired S(λ) function, which is plotted in Figure S2C. Figure S2D shows a typical raw single NP spectrum, measured for a single graphite NP of mass 14.93 MDa. Figure S2E shows the result of dividing Figure S2D by Figure S2C, to obtain a sensitivity-corrected NP spectrum.
Figure S2. Data illustrating how the sensitivity function, $S(\lambda)$, is extracted, and NP spectra are intensity corrected. Frame A is the raw spectrum for the TC heated to 2316 K. B is the theoretical spectrum, calculated as the product of Planck’s law, the emissivity of the TC material, and the transmission of the ND filter assembly. C is the extracted $S(\lambda)$ function, i.e. the ratio of A to B. D is a typical single raw NP spectrum. E shows the corrected NP spectrum, showing intensity mismatch between the visible and nIR. F is the final corrected spectrum with fit, after applying the visible scale factor.
Note that there is a spectral region from 980 to 1100 nm where both the visible and nIR spectrographs have low sensitivity because of the combined effects of the dichroic beamsplitter, the CCD quantum efficiency, and the grating efficiency of the nIR spectrograph. As a result, the signal in this “gap” region is low, and uncertainty is high. In the fitting process described below for $T_{NP}$ determination, this gap region is given zero weight.

Figure S2E shows one final step in the NP spectral correction process. Note that the visible and nIR spectra do not match up across the spectral gap region, i.e., the corrected visible intensity is slightly too low to match smoothly to the nIR data. This effect is believed to be a result of the fact that our visible camera uses a 2D array that is 3.84 mm tall, whereas the 1D nIR camera array is only 500 μm tall, (but equipped with a cylindrical lens to help reduce the height of the dispersed light on the array). Because the TC (~280 μm) is significantly larger than the ~80 μm effective radial emitter size for typical NPs, the solid angle of light injected into the fibers (numerical aperture 0.5) is higher for the TC than for NPs, resulting in a somewhat larger solid angle entering the spectrographs. The taller visible array captures more of this “extra” light than the nIR array, and as a result, the visible portion of $S(\lambda)$ is larger than it should be for correcting spectra from the smaller NP emitters. Thus, the intensity of the visible portion of the corrected NP spectra tends to be slightly too small to match up smoothly to the nIR section. This hypothesis is supported by the observation that changing the effective NP emitter size by varying the trapping conditions, results in changes in the relative intensities of the visible and nIR spectra.

Therefore, it is necessary to scale the visible spectrum to ensure smooth matching to the nIR spectrum across the gap. The approach used is to fit the data just on either side of the gap (900 to 950 nm in the visible and 1030 to 1200 nm in the nIR), to a smooth function, with the visible spectrum scale factor as one of the fit parameters. As a smooth function, we use Planck’s law
times a power law model for the NP emissivity ($\epsilon \propto \lambda^{-n}$), i.e., the same function used to fit spectra for $T_{NP}$ determination, but with $n$ fixed at 1.4. The $n$ parameter affects the curvature of the fitting function, but because only a narrow $\lambda$ range is fit to extract the visible spectrum scale factor, the scale factor is not very dependent on the exponent. 1.4 was chosen as being typical for carbon NPs, and the correct value for room temperature graphite (see Figure S3). Figure S2F shows the result of applying the scale factor to the visible spectrum. The visible spectrum scale factor varies slightly from NP to NP, and in this case, it was 1.34. The effects of systematically varying this scale factor on temperatures extracted from spectral fitting are explored below.

**Power law approximation to the scattering theory emissivity function:**

The NP spectra are fit to a function that is the product of Planck’s law times and emissivity function, $\epsilon(\lambda)$. As noted in the manuscript, scattering theory predicts that in the limit of NPs much smaller than the wavelengths emitted, as is the case here, $\epsilon(\lambda) = \frac{8\pi r}{\lambda} Im\left(\frac{(n+ik)^2 - 1}{(n+ik)^2 + 2}\right)$, where $n$ and $k$ are the real and imaginary components of the index of refraction of the material at the wavelengths and temperature of interest. Figure S3 shows $\epsilon(\lambda)$ for graphite, calculated from the scattering theory expression, using room temperature $n$ and $k$ data, to a power law model, $\lambda^{-n}$ (Blue).
in the wavelength range we measure is ~3%. Ideally we would use the correct $\epsilon(\lambda)$ function in fitting NP spectra, however, the necessary index of refraction (or equivalent dielectric) parameters are not available for the materials of interest, at high temperatures, over the wavelength range of interest. More fundamentally, bulk optical properties would be inaccurate for NPs, due to the large number of surface and defect states in NPs. Therefore, we are forced to adopt a model $\epsilon(\lambda)$ function, and the power law model at least has the virtue of having roughly the correct shape and introducing a minimum number of adjustable parameters.

**Sensitivity of extracted TNP values to the position of the TC, and to TC temperature used in calibration:**

Figures S4A-C, show the effects of moving the TC calibration emitter to different positions in the trap, *i.e.*, deliberately positioning it so that it is not at the trap center. Each figure shows traces of NP temperature ($T_{NP}$) vs. time from an experiment in which a single NP was heated using a 532 nm laser, with its temperature stepped from ~1200 K to ~2000 K, then dropped to ~1400 K. The 532 nm laser was not being stabilized during this test, and its intensity fluctuated significantly, driving fluctuations in $T_{NP}$. All the traces shown in Figures S4A-C are based on the same raw NP emission spectra, but corrected using TC calibration spectra obtained with the TC emitter at the trap center (black trace), and deliberately misaligned by ± 0.05 and ± 0.1 mm in the X, Y, and Z directions, relative to trap center. The important point is how much the $T_{NP}$ traces obtained at different TC emitter positions vary, *i.e.*, how sensitive the $T_{NP}$ calibration is to mis-aligning the TC emitter relative to trap center. The TC temperature, $T_{TC}$, during these experiments was kept close to 2300 K, and measured electrically in conjunction with each spectral measurement.
In Figure S4A, the TC was moved radially in the x direction, as defined in Figure S1, while keeping the y and z coordinates centered on the trap. This corresponds to horizontal motion of the right hand TC image shown in Figure S1. It can be seen that if the TC is positioned at \( x = -0.10 \) mm with respect to the trap center the \( T_{NP} \) values extracted for the NP are shifted significantly to lower temperatures, particularly for low \( T_{NP} \) values. For example, the shift is roughly \(-60 \) K for lower temperatures.
T_NP near 1200 K, but only ~30K for T_NP above 1500 K, and only -10 K at temperatures greater than 1800 K. For misalignments of -0.05 mm and for both +0.05 and +0.10 mm, the variations in T_NP relative to the values extracted for the TC at the trap center are much smaller – averaging roughly ± 6 K over the entire T_NP range.

Figure S4B gives analogous data for deliberate misalignment of the TC emitter in the y direction, where +y corresponds to moving the right-hand TC image in Figure S1 up, and –y is downward motion. Here the effects of misalignment, particularly of having the TC positioned too high in the trap, are larger. For y = -0.1 mm and -0.05 mm, T_NP values are similar, and shifted by ~-50 K at low T_NP and by ~-90 K at high T_NP, relative to the T_NP values obtained at y = 0.0. For positive y misalignments (y = 0.05 and 0.1 mm) the shifts are also similar to each other, but much smaller, remaining within 20 K of the values measured for y = 0.0 at all T_NP. The larger shifts observed for negative y misalignments are attributed to the effect of collecting light emitted from the wires leading to the TC bead. These wires connect to the bead at its top, and are significantly cooler than the laser-heated bead. Thus, for TC displacements to –y, the optical system picks up more light from the lead wires, which would tend to give TC spectra colder than the bead temperature, and therefore cause the calibrated T_NP higher, as is observed. Knowing this, we position the TC so that the lower part of the bead is at the trap center, as shown in the left image in Figure S1.

Figure S4C shows the effects of moving the TC axially in the trap, with +z corresponding to moving closer to the collection lens. The TC positions z = 0.05 mm (purple trace, T_TC = 2303 K) and z = 0.1 mm (red trace, T_TC = 2302 K) cause ~+100 K shifts in the T_NP values obtained from the TC spectra. In contrast, moving the TC to z = -0.05 mm, i.e., slightly further from the collection lens, had no significant effect, and moving to z = -0.1 mm caused shifts to lower T_NP ranging from ~-20 K at low T_NP to ~-50 K. The larger effects from shifting the TC closer to the collection lens
makes sense – the NP is a diffuse emitter extending axially $\sim \pm 0.03$ mm relative to the trap center, whereas the TC emitter is the surface of the TC bead facing the collection lens. Thus, as shown in the left image in Figure S1, when $z = 0$, the TC surface is actually $\sim 0.06$ mm closer to the lens than the trap center. Thus for small negative displacements of the TC, its front surface actually moves closer to the trap center, *i.e.* to the average NP position. For positive displacements, however, the TC surface is shifted increasingly farther from the average position of the NP, hence introducing significant chromatic effects on the TC spectra that affect the $T_{NP}$ calibration. Knowing this, we are careful to position the TC with its center at, or slightly behind the trap center.

Given the $\sim 25$ $\mu$m precision with which the TC can be positioned in the X, Y, and Z directions, and the results in Figures S4A-C, we estimate that the aggregate effects of TC misalignment on the extracted $T_{NP}$ values range from $\sim \pm 3.2\%$ at low end of our $T_{NP}$ range, to $\pm 3.5\%$ at high $T_{NP}$ values.

In principle, the TC temperature used to generate the sensitivity calibration needed to correct the NP spectra should not matter, however this assumes that there is no error in reading the TC temperature ($T_{TC}$) and in calculating the emissivity used to correct the TC spectra. To test for possible $T_{TC}$ effects, Figure S4D shows the effects of correcting the same NP data using TC spectra measured at different TC temperatures, with the TC positioned at $x = y = z = 0$. There are effects of $T_{TC}$ on the extracted $T_{NP}$ values, but they are small, as expected. For all but the lowest $T_{TC}$ value (2060 K), the variations in $T_{NP}$ are less than 10 K at low $T_{NP}$, and less than 16 K for high $T_{NP}$, corresponding to $\leq 0.8\%$ uncertainty in $T_{NP}$. Using the 2060 K TC data introduces larger errors ($\sim 19$ K at low $T_{NP}$, 35 K at high $T_{NP}$), probably because the emitter is too cold to give good intensities at short wavelengths. Obviously, we avoid use of low TC temperatures in correcting
NP spectra, thus the uncertainty in $T_{NP}$ values due to this TC temperature effect is estimated to be $\leq 0.8\%$.

Effects of optical system misalignment on measured and corrected NP spectra, and on the $T_{NP}$ values extracted by fitting:

Figures S5 – S9 show the effects on raw TC and NP spectra and corrected NP spectra (and extracted $T_{NP}$ values) of deliberately misaligning various optical components. Note that misalignment generally results in substantial decreases in signal intensities.

Figure S5. Raw NP visible emission spectra (A-D) and TC visible emission spectra (E-H). A, C, E, and G show the effects on the emission spectra of varying the beamsplitter’s x and y positions by +/- 1.5 mm. Figures B, D, F, and H show the normalized intensities to allow shape changes observed.
however, from the TNP determination perspective, what is important are two factors: 1. How does the misalignment affect the *shape* of the spectra, as opposed to the intensity? To show changes in intensity and shape, we present both raw and normalized TC and NP spectra. 2. If there are changes in spectral shape from misalignments, are they the same for the TC and NP spectra? If so, they cancel in the calibration process. If not, they are apparent as changes in the corrected NP spectra.

In Figure S5 the raw visible emission spectra for a single NP and for the TC are shown as a function of displacements of the beamsplitter assembly in the x and y directions. Note that in making such comparisons, we kept the NP at relatively low T_{NP}, so that it did not sublime significantly on the experimental time scale, and we attempted to keep the temperature constant by holding the heating laser at constant intensity, using a PID program to stabilize the laser (unlike the measurements in Figure S4). As shown in Figure S1, beamsplitter assembly motion also moves the visible focusing lens and fiber collimator. The y motion does not move the visible beam path, therefore the lens and collimator become radially misaligned with respect to the beam. The x motion of the beamsplitter shifts the visible beam path, and again, the beam becomes radially misaligned with the lens and collimator.

The left side of Figure S5 shows raw spectra, and the right side shows the same spectra, normalized to allow changes in shape to be seen. In Figures S5A and S5E, the x position of the beamsplitter is changed by ±1.5 mm relative to the position that gives maximum intensity for the NP spectra, while measuring the NP and TC raw spectra. Figure S5C and S5G show analogous effects of ±1.5 mm misalignments in the y direction. It can be seen that radial misalignment of the focusing optics has significant effects on visible spectral intensity, but as shown in Figures S5B,
S5D, S5F, and S5H, the spectral shape, which is what is important in $T_{NP}$ determination, is relatively insensitive to this kind of misalignment.

Note that motion of the beamsplitter assembly does not shift the transmitted nIR beam path, but because the 175 mm nIR focusing lens is attached to the assembly, its motion does misalign the lens with respect to the beam path, thereby deflecting the beam slightly such that it becomes radially misaligned with respect to the nIR collimator. The effect of this radial misalignment on the nIR spectra is similar to what is observed if the beamsplitter assembly is fixed, but the nIR collimator is translated to cause radial misalignment (Figures S6 and S7A-C). Because these effects are similar and quite small, a separate figure is not presented showing the effects on nIR spectra of x and y beamsplitter assembly motion. Similarly, small displacements of the beamsplitter assembly in the z direction have negligible effects on either visible or nIR spectra, and these data are not shown.

In Figures S6-S9, we explore the misalignment of the nIR fiber collimator, which is mounted separately, allowing x, y, and z motion with respect to the nIR beam path. This motion has no effect on the visible spectra. Figure S6 shows spectra for a single NP for nIR collimator positions misaligned by up to ±0.4 mm in the x and y (i.e. radial) directions. Figure S6A and S6C show raw spectra and Figures S6B and S6D show the same spectra normalized so that shape differences can be seen. It can be seen that for x and y misalignment of the nIR collimator, there are significant intensity effects, but that the spectral shape is unaffected. Figures S7A-D show analogous TC
emission spectra, *i.e.*, raw and normalized spectra measured as a function of x and y misalignments up to ± 0.4 mm. Again, there is essentially no effect on the spectral shape. Figure S6E and S6F show how the integrated nIR signal from a single NP varies with x and y misalignment, showing that it is easy to find the optimum radial position for the collimator.

Figure S6. nIR emission spectra for a single carbon nanoparticle. Frames A and C show the effects of misalignment of the nIR fiber collimator on the raw intensities, and frames B and D show that the effects of nIR collimator on shape are negligible. Frames E and F simply show how the integrated spectral intensities vary with misalignment in the x and y directions.
The bottom half of Figure S7 shows the effects of nIR collimator x and y misalignment on the nIR part of the corrected NP spectra (there are not effects on the visible spectra). Figures S7E and S7G show that there are intensity changes in the corrected NP spectra as a function of collimator position, but Figure S7F and S7H show that the effects on spectral shape are small, as might be expected from small effects on the raw NP and TC spectral shapes. The $T_{NP}$ values extracted by fitting these spectra (combined with visible

Figure S7. Frames A-D show raw TC nIR spectra at different X and Y collimator positions, taken at similar TC temperatures. Frames E and G are corrected NP spectra at different X and Y collimator positions, corrected using TC spectra taken at the same collimator positions. Images F and H are the spectra in E and G, normalized to show shape changes.
spectra taken at the same time) show ±22 and ±14 K variations (<±2%), however, it should be noted that the laser used to heat the NPs was not perfectly stable, thus it is almost certain that some of this T_{NP} variation was real, i.e., not error due to collimator position.

Given the relatively sharp dependence of intensity on x and y position, which makes it straightforward to align it precisely, Figure S8. Frames A-D are raw nIR single NP emission spectra as a function of collimator Z position. Frames E-F are raw nIR TC emission spectra at the same Z positions. Frames A,B, E, and F shows small variations of collimator Z position (± 1.0 mm). Frames C, D, H, and G show effects of larger collimator misalignments up to ± 5.0 mm.
we conclude, that $x$ and $y$ collimator misalignment has negligible effect on the uncertainty of the $T_{NP}$ measurements.

The positioning of the collimator in $z$ (axial) direction is more problematic. Figures S8 and S9 show the effects of changing the $z$ position of the nIR collimator, with all other optics left in their optimal positions. Figure S8 shows raw and normalized NP and TC spectra for $z$ position changes of up to $\pm 1$ mm, relative to the optimal $z$ position. Because long focal length lenses are used to couple the nIR light into the collimator, the intensity varies much more slowly with $z$ than the $x$ and $y$ variations shown in Figure S6E and S6F, *i.e.*, it is not as straightforward to identify the optimal position. To find the optimal $z$ position, the collimator was translated over a $\pm 11.5$ mm range, measuring the fall off of intensity in both directions, and choosing the mid-point as the optimal $z$ position. For collimator displacements up to $\pm 1$ mm (Figure S8A) the changes in raw NP spectra are small, and the normalized spectra (S8B) are essentially superimposable. Certainly any differences between spectra are well within the variation expected due to real variations in the NP temperature over the experiment time. For larger $\pm 5$ mm displacements of the collimator (S8C), there are larger changes in the spectral intensity, and after normalization (S8D), it can be seen that there are net shifts to the red or blue for large positive and negative $z$ displacements, respectively. If the TC spectra had similar red and blue shifts, there would be no problem, however, as shown in the analogous TC spectra in Figures S8E-H, the TC spectra have smaller red and blue shifts. This difference reflects the larger size of the TC emitter ($\sim 280 \mu$m) compared to the NP effective emitter size ($\sim 80 \mu$m), which means that chromatic effects tend to average out more in the TC spectra than in those for the NPs.

The effects of $z$ displacements on corrected NP spectra, and the $T_{NP}$ values extracted by fitting them, are summarized in Figure S9. Note that for these experiments, relatively large NPs were
used to give good signal, while keeping $T_{NP}$ low enough to minimize sublimation and other changes to the NP.

Figure S9A shows the corrected spectra for a single NP with $T_{NP} \approx 980$ K, also including the visible portion of the spectra because this is used in the fitting process.

**Figure S9.** Corrected spectra for different NPs measured with different nIR collimator positions. A.) Effects of $\pm 1.0 \text{ mm}$ changes in the Z position. B.) The nIR spectra, normalized to show shape changes. C.) Analogous unnormalized spectra for a different NP at higher $T_{NP}$. D.) Effects of $\pm 5 \text{ mm}$ changes in the nIR collimator position. E.) Visible spectra showing that, as expected, moving the nIR collimator has no effect. F.) nIR spectra, normalized, as a function of $\pm 5 \text{ mm}$ misalignment of the nIR collimator.
For z displacements up to ±1 mm, T_{NP} varies by ≤ 14 K (≤ 1.4 %), however, T_{NP} fluctuates randomly rather than varying systematically with z. This is a sign that the T_{NP} fluctuations were not due to the z displacement, but rather reflect real T_{NP} variations due to laser intensity fluctuations. Figure S9B shows that the normalized nIR spectra are essentially superimposable. Figure S9C shows corrected spectra for a different NP at higher T_{NP}, also for ±1 mm z displacements. Again, T_{NP} varies randomly with z, by about 2% in this case. The fact that T_{NP} shows no systematic variation with z over this ±1 mm range is not surprising, given that the raw spectra (Figure S8B) are nearly superimposable.

Figure S9D shows the effects of ±5 mm z displacements – a range where significant, systematic red shifts were observed with increasing z in the spectra in Figure S8D. As expected, these spectral shifts result in systematic increases in the extracted T_{NP} values with increasing z. Figure S9E and S9F show the visible and nIR spectra from Figure S9D, normalized to allow shape changes to be observed. As expected, there are no changes in the visible spectral shapes, but the nIR region shows significant red shifting with increasing z.

Similar axial displacement measurements were done moving the visible collimator relative to the beamsplitter assembly by up to ±8 mm in the x direction, but no effects were observed on the shape of the visible part of the spectra. Indeed, the visible spectra are generally much less affected by misalignments of the optics or of the TC position, compared to the nIR spectra. In part, this probably reflects the fact that the achromatic doublets used in the collection system are optimized for the 750-1550 nm range, which is part of our “visible” spectrum. In addition, however, the 2D CCD array used to record the visible spectra is 3.84 mm tall, whereas the 1D InGaAs array used for the nIR spectra is only 0.5 mm tall. The nIR camera has a cylindrical lens to vertically compress the dispersed light onto the array, but some light still misses the array. As a result, the visible
detection efficiency is less sensitive to the divergence of light exiting the fiber, compared to that for the nIR spectrograph, and therefore, less sensitive to optical misalignments that affect how light couples into the fibers.

For x and y displacements of the nIR collimator, the intensity falls off rapidly – much faster than chromatic effects grow in. Therefore, the correct x and y positions are well defined, and we are able to position the collimator accurately enough that misalignment in these directions has a negligible effect on the uncertainty in $T_{NP}$. The same is not true for the z positioning, because the intensity maximum along this direction is much broader, and significant chromatic effects appear for ± z collimator displacements for which the intensity is only modestly affected. The method used to find the optimal z position for collimator is estimated to give the position to within ±0.5 mm, but for estimating the resulting effect on $T_{NP}$ we assume that the position is uncertain by ±2 mm. From data analogous to that in Figure S9C-D, but for ±2 mm z motion of the collimator, we find that $T_{NP}$ varied by a total of 86 K, corresponding to 5.6%. Therefore, we estimate this contribution to the uncertainty as ~±2.8 %.

**Effects of varying the visible scale factor on fit quality and $T_{NP}$:**

As discussed above, the intensity of the visible part of the spectrum must be scaled to match smoothly to the nIR part, and Figure S10 explores the effect of changing the scale factor, f, away from the best-fit value. In Figures S10A-C the spectrum is plotted with the best fit value of f (1.53) in blue, and the red and orange spectra show the effects of increasing or decreasing f by ±2%, ±5%, and ±10%. The two insets show more clearly how changing f affects both the visible spectra (the nIR is not scaled) and the resulting fits to the entire spectrum.

Consider the effect of changing f by ±10% (C). The visible inset shows how the change in scale factor shifts the spectra to increased or decreased intensity. Such a large change in f leads to gross
mismatching of the visible and nIR spectra, as can be seen by the resulting fits. Note that when f is increased by 10%, shifting the visible spectrum to higher intensity, the best fit is well below the experimental spectrum in the visible, and well above it in the nIR, i.e., it essentially splits the differences between the now-mismatched visible and nIR spectra. Similarly, if the visible spectrum is scaled down by 10%, the fit is obviously too high in the visible. The same effect is clear when f is changed by ±5% (Figure S10B). Clearly such large variations in f can be excluded on basis of poor fitting.

On the other hand, when f is changed by only ±2% (Figure S10A), the fits now are within the scatter in the experimental spectra, i.e., the precision of the process used to determine the best-fit f value is ~±2%. (Note that to keep the spectral fits in the visible inset from overlapping, they have been offset, only for the ±2% case). If we take the variation in $T_{NP}$ for the ±2% change in f as an estimate of this contribution to the

**Figures S10.** Images A-C show the effect of varying the best fit visible scale factor, f, by ±2%, ±5%, and ±10% from its best-fit value, determined as described above. Note that in the visible inset in A, the spectra and fits have been offset for clarity.
uncertainty in $T_{NP}$, the result is only $±24$ K or $±1.9%$.

**Effects of spectral integration time:**

To test for possible saturation effects in the visible and nIR cameras, Figure S11 shows spectra of a single graphite NP with $M = 14.2$ MDa, with the laser stabilized to have power, measured at the vacuum exit window, of $19.6$ mW $± 0.28$ mW. In Figure S11A, the spectra are plotted in terms of photons/sec/sr/nm of spectral bandwidth, and have been offset vertically so that the individual spectra can be seen clearly. In Figure S11B, the spectra are plotted without the offset so that their shapes can be compared.

As expected the short acquisition time spectra are relatively noisy, and the 5 second spectrum has slightly higher intensity. The 30 and 60 second spectra are essentially identical, and the $T_{NP}$ and $n$ values extracted from fitting them also have negligible differences. There may be a slight decrease in the apparent $T_{NP}$ and increase in $n$.
for the shorter time spectra, however, the variation in $T_{NP}$ is certainly well within the fitting error for such noisy spectra.

**Summary of the effects of optical system alignment and fitting on the uncertainties in $T_{NP}$.**

As described above, the significant experimental contributions to uncertainties in the spectral shapes, hence $T_{NP}$, are: 1. Positioning of the TC during calibration ($\pm 3.2$ to $3.5\%$), 2. The $z$ position of the nIR collimator ($\pm 2.8 \%$), 3. The visible spectrum scale factor ($\pm 1.9\%$). The positions of other optics are well enough defined and controlled that they do not contribute uncertainties of these magnitudes, and thus their contributions are considered negligible. Therefore, the total contribution to uncertainty in $T_{NP}$ from these experimental issues is estimated to be $\pm 4.8 \%$.

In addition, given typical signal/noise level in NP spectra, we estimate $\sim \pm 4 \%$ uncertainty in $T_{NP}$ from fitting, assuming a fitting function consisting of Planck’s law times the power law model for $\epsilon(\lambda)$. Therefore, the total estimated uncertainty in $T_{NP}$ including both experimental and fitting uncertainties is $\pm 6.2 \%$. As emphasized in the main paper, there is additional error associated with our choice of the power law function to model $\epsilon(\lambda)$, however, because the correct form of $\epsilon(\lambda)$ is unknown, it is not possible to estimate this error. We simply note that for many of the carbon materials, the fits to the NP spectra based on the power law $\epsilon(\lambda)$ model are quite good, suggesting that the error is relatively small.
Effects of varying n on extracted $T_{NP}$ values:

One issue with using the power law model, however, is that the $T_{NP}$ and n parameters both affect the curvature of the emission vs. $\lambda$, and while the effects are different, they are not orthogonal. Increasing the n parameter causes the fit function to fall off more rapidly at long wavelengths, and in fitting a spectrum, this fall off is compensated by decreasing $T_{NP}$ in Planck’s law, which increases the intensity at long wavelengths relative to short wavelengths. The result is a fitting function with more curvature, i.e., higher at mid-wavelengths, and falling off more rapidly at both long and short wavelengths, compared to the function with best-fit n value. Conversely, decreasing n is compensated by increasing $T_{NP}$, which results in a fitting function with more gradual wavelength dependence.

To illustrate this behavior, and estimate the resulting uncertainty in $T_{NP}$, Figure S12 shows how forcing the n parameter away from its best fit value affects the fit quality and extracted $T_{NP}$ values.
The experimental spectrum being fit is for a carbon black NP of 85 MDa size, and the acquisition time was 30 seconds. The spectrum was first fit allowing $n$ and $T_{NP}$ (and the normalization constant) to vary, obtaining best fit values of $T_{NP} = 1257$ K, and $n = 1.53$. We then varied the $n$ parameter by up to $\pm50\%$, refitting the spectrum allowing only $T_{NP}$ (and the normalization) to vary. For clarity, the figure shows only the results for $\pm20\%$ and $\pm50\%$ variations, and to help show the effects more clearly, insets with magnified intensity scales are shown for five regions across the spectrum.

Because the power law model is only an approximation to the true emissivity function, even the best-fit function over- or under-shoots the experimental data in some spectral regions. Therefore, forcing $n$ away from its best-fit value may improve the fit in some spectral regions, even though the overall fit is worsened. For example, using $n = 2.29$ improves the fit in the wavelength region near 1550 nm, however, such a high $n$ value substantially worsens the fit over the entire range below 1000 nm. Similarly, using $n = 0.76$ gives the best fit in the region around 700 nm, but substantially worsens the fit for most of the rest of the spectrum. Similar effects are observed for the $\pm20\%$ variations in $n$, but in that case, the deviations are on the same order as the estimated uncertainty in the relative spectral intensities (i.e., spectral shape). The temperature variations associated with $\pm20\%$ $n$ variations are roughly $\pm45$ K ($\sim\pm4\%$), and we take this as a rough estimate of the error associated with fitting any particular spectrum, within the power law model.

**NP spectra for different materials compared, before and after >1900 K heating.**

Figures S13 and S14 replot the spectra in figures 4A and for NP1 in Figure 5A and 5C and Figure 5D in the main text, to allow comparison between materials. In Figure S13, the volume-scaled intensities for NPs prior to >1900 K heating are shown on the left, and the post-heating spectra are
Figure S13. Emission spectra comparing the carbon NPs in figures 4 and 5, before and after heating. All spectra are volume scaled to allow direct comparison of intensities. The left column has spectra prior to heating above 1900 K. The right column shows spectra collected after >1900 K heating.
Figure S14. Emission spectra comparing the carbon NPs in figures 4 and 5, before and after heating. All spectra are normalized at 1580 nm to allow spectral shape comparison. The left column has spectra prior to heating above 1900 K. The right column shows spectra collected after >1900 K heating.
on the right. In Figure S14, the data are replotted, normalized at 1580 nm, to allow spectral shapes to be compared.

**Effects of measuring $T_{NP}$ while NP motion is being driven to measure the NP mass.**

Figure S15 addresses the question of whether $T_{NP}$ is significantly affected by the enhanced axial NP motion that occurs when the NP is being driven to measure mass, due to possible changes in chromatic effects as the NP position in the trap varies. In the figure, $T_{NP}$ is shown vs. time for a single graphite NP with initial mass of 23.22 MDa as the NP was heated with 12.2 mW $\pm$0.05 mW of 532 nm laser power, giving $T_{NP}$ near 1500 K. The scatter in the $T_{NP}$ values partly reflects signal/noise in the spectra, taken at relatively low $T_{NP}$ to minimize sublimation, but in addition, the normal $T_{NP}$ stabilization program was not being used.

During the first $\sim$1300 seconds, $T_{NP}$ was being measured during the AC drive frequency scan used to measure the secular frequency, hence mass. The extracted $T_{NP}$ was 1500 $\pm$ 19 K. At 1320 seconds the AC drive potential was turned off for $\sim$1200 seconds, continuing the spectral measurements. During this

![Figure S15. Single Graphite NP with initial mass of 23.22 MDa. In this experiment, the laser was stabilized by using the $T_{NP}$ for slow fluctuations and a PID for fast fluctuations. The drive frequency was turned off between $\sim$1320-2580 seconds showing negligible changes in $T_{NP}$.](image-url)
“no drive” time, \( T_{\text{NP}} \) was 1502 ± 14 K. At 2580 seconds the drive potential was turned back on, and eleven additional simultaneous frequency and \( T_{\text{NP}} \) measurements were taken, giving \( T_{\text{NP}} = 1498 \text{ K} \pm 19 \text{ K} \). Clearly, any effect of the AC drive potential on the spectra and extracted \( T_{\text{NP}} \) values is negligible compared to the uncertainty in \( T_{\text{NP}} \).

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| Temperature (°C) | Concentration (0.1%) | Time (sec) |
|-----------------|----------------------|------------|
| 1380            | 1.44                 | 2520       |
| 1358            | 1.06                 | 1290       |
| 1540            | 1.19                 | 5100       |
| 1551            | 0.63                 | 1440       |

**Figure 2A, Carbon Black**

| Value 1 | Value 2 | Value 3 | Value 4 | Value 5 | Value 6 | Value 7 | Value 8 | Value 9 | Value 10 | Value 11 | Value 12 | Value 13 | Value 14 | Value 15 |
|---------|---------|---------|---------|---------|---------|---------|---------|---------|-----------|-----------|-----------|-----------|-----------|----------|
| 622.099 | 332.352 | 260.811 | 1270.73 | 972.539 | 4141.2  | 3703.91 | 10606.8 | 10398.6 | 16905.9   | 16273.8   | 622.099   | 205.255   | 140.105   | 647.339  |
| 615.422 | 334.759 | 230.692 | 1089.08 | 874.233 | 3725.46 | 3383.42 | 10113.2 | 9612.01 | 15703     | 15122.9   | 615.422   | 202.413   | 123.056   | 531.026  |
| 636.882 | 400.702 | 338.397 | 1526.75 | 1219.11 | 5110.74 | 4486.6  | 13617   | 12282.2  | 20621.1   | 19005.1   | 636.882   | 272.04    | 184.572   | 825.436  |
| 615.713 | 286.01  | 231.943 | 1026.67 | 878.347 | 3791.63 | 3396.95 | 10131.6 | 9645.4   | 15593.5   | 15171.8   | 615.713   | 209.345   | 123.761   | 577.5    |
| 637.461 | 478.603 | 341.762 | 1560.01 | 1229.62 | 5299.79 | 4519.37 | 13492.3 | 12360    | 20577.7   | 19117.2   | 637.461   | 214.309   | 186.517   | 821.726  |
| 631.668 | 374.566 | 309.231 | 1364.92 | 1127.44 | 4871.92 | 4198.84 | 12595.5 | 11595.4  | 19498.8   | 18013     | 631.668   | 263.817   | 167.771   | 757.428  |
| 603.797 | 328.86  | 184.826 | 846.443 | 721.049 | 3079.22 | 2872.38 | 7844.81 | 8335.96  | 12494.9   | 13241.2   | 603.797   | 240.709   | 97.3623   | 409.035  |
| 626.45  | 301.836 | 282.034 | 1255.7  | 1040.88 | 4564.27 | 3923.69 | 11749   | 10932.7  | 17723.2   | 17051.5   | 626.45    | 201.992   | 152.194   | 721.05   |
| 626.16  | 324.513 | 280.579 | 1206.12 | 1036.22 | 4463.05 | 3908.77 | 11792.4 | 10896.5  | 17684.8   | 16999     | 626.16    | 224.687   | 151.364   | 653.319  |
| 625.87  | 332.534 | 279.13  | 1229.95 | 1031.57 | 4319.15 | 3893.89 | 11499.6 | 10860.5  | 17608.4   | 16946.6   | 625.87    | 230.587   | 150.537   | 625.244  |
| 625.29  | 373.497 | 276.249 | 1326.44 | 1022.32 | 4356.9  | 3864.24 | 11588.6 | 10788.6  | 17452.5   | 16842     | 625.29    | 220.038   | 148.893   | 567.789  |
| 604.96  | 287.14  | 176.58  | 995.471 | 692.995 | 2886.33 | 2777.05 | 7448.46 | 8094.64  | 12718.3   | 12883.1   | 604.96    | 210.172   | 92.7818   | 592.406  |
| 624.71  | 341.024 | 273.391 | 1248.85 | 1013.13 | 4210.77 | 3834.75 | 11473.4 | 10717.1  | 17242.7   | 16737.9   | 624.71    | 175.403   | 147.264   | 679.554  |
| 638.33  | 484.427 | 346.859 | 1541.87 | 1245.51 | 5338.6  | 4568.81 | 13566.5 | 12477.3  | 20902.3   | 19286     | 638.33    | 237.727   | 189.465   | 845.214  |
| 604.96  | 282.91  | 189.061 | 829.598 | 735.39  | 3202.38 | 2920.9  | 9116.14 | 8458.35  | 13690.9   | 13422.5   | 604.96    | 207.076   | 99.7195   | 537.598  |
| 613.68  | 304.998 | 223.299 | 973.904 | 849.843 | 3977.87 | 3303.06 | 9940.76 | 9413.18  | 15721.5   | 14830.9   | 613.68    | 211.493   | 118.891   | 524.664  |
| 625.00  | 355.83  | 274.817 | 1199.34 | 1017.72 | 4438.11 | 3849.48 | 11146.9 | 10752.8  | 17632.5   | 16789.9   | 625.00    | 186.989   | 148.077   | 658.827  |
| 638.33  | 484.427 | 346.859 | 1541.87 | 1245.51 | 5338.6  | 4568.81 | 13566.5 | 12477.3  | 20902.3   | 19286     | 638.33    | 237.727   | 189.465   | 845.214  |
| 604.96  | 282.91  | 189.061 | 829.598 | 735.39  | 3202.38 | 2920.9  | 9116.14 | 8458.35  | 13690.9   | 13422.5   | 604.96    | 207.076   | 99.7195   | 537.598  |

**Note:** The table contains numerical data that is not fully transcribed due to limitations in the text extraction process.
| T (K) | n | t (sec) |
|-------|---|---------|
| 1783  | 0.43 | 2730 |
| 1955  | 0.25 | 4260 |
| 2210  | 0.13 | 6690 |
| 2499  | 0.51 | 11700 |
| 1959  | 0.63 | 10380 |
| 2049  | 0.51 | 11700 |
| 1783  | 0.43 | 2730 |
| 1955  | 0.25 | 4260 |
| 2210  | 0.13 | 6690 |
| 2499  | 0.51 | 11700 |
| 1959  | 0.63 | 10380 |
| 2049  | 0.51 | 11700 |

|          |          |          |          |          |          |          |          |          |          |          |          |          |          |          |
| Year | Price | Value | Change | Increase | Cumulative |
|------|-------|-------|--------|----------|------------|
| 2020 | 10220 | 10230 | 10     | 10        | 10230      |
| 2021 | 10220 | 10230 | 10     | 10        | 10230      |
| 2022 | 10220 | 10230 | 10     | 10        | 10230      |
| 2023 | 10220 | 10230 | 10     | 10        | 10230      |
| 2024 | 10220 | 10230 | 10     | 10        | 10230      |
| Sample | Carbon Black | Graphene Platelet | Graphite | ND | Enhancement |
|--------|--------------|-------------------|----------|----|-------------|
| 0      | 1.38E+07     | 1.44E+07          | 1.47E+07 | 1.35E+07 | Carbon Black |
| 0.05   | 2.89E+07     | 2.84E+07          | 2.51E+07 | 2.12E+07 | Graphene Platelet |
| 0.05   | 1.72E+08     | 1.68E+08          | 1.70E+08 | 1.68E+08 | Graphite |
| 0.05   | 1.99E+08     | 2.01E+08          | 2.01E+08 | 2.21E+08 | ND |
| 0.1    | 2.14E+08     | 2.11E+08          | 2.14E+08 | 2.11E+08 | Carbon Black |
| 0.1    | 2.89E+07     | 2.84E+07          | 2.51E+07 | 2.12E+07 | Graphene Platelet |
| 0.1    | 1.72E+08     | 1.68E+08          | 1.70E+08 | 1.68E+08 | Graphite |
| 0.1    | 1.99E+08     | 2.01E+08          | 2.01E+08 | 2.21E+08 | ND |
| 0.2    | 2.14E+08     | 2.11E+08          | 2.14E+08 | 2.11E+08 | Carbon Black |
| 0.2    | 2.89E+07     | 2.84E+07          | 2.51E+07 | 2.12E+07 | Graphene Platelet |
| 0.2    | 1.72E+08     | 1.68E+08          | 1.70E+08 | 1.68E+08 | Graphite |
| 0.2    | 1.99E+08     | 2.01E+08          | 2.01E+08 | 2.21E+08 | ND |
| Temperature | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | Value | V
| Data Source | Temperature | Parameter | Value | Temperature | Parameter |
|-------------|-------------|-----------|-------|-------------|-----------|
| 43.83       | 1650.78     | 50.54     | 0.76969 | 64.47       | 1900.53    |
| 33.56       | 1645.05     | 173.98    | 1.22532 | 84.04       | 1887.70    |
| 6.305       | 1649.39     | 38.04     | 0.91398 | 30.49       | 1915.55    |
| 14.17       | 1654.95     | 103.38    | 1.06588 | 44.01       | 1913.29    |
| 4.77        | 1660.11     | 15.54     | 1.44904 | 38.04       | 1893.33    |
| Number | Time (sec) T |  |
|--------|--------------|------------------|
| 2887.40106 | 48 | 2615 | 1651.81666 |
| 4198 | 3012.52624 | 50 | 4197 | 1655.15272 |
| 4166 | 3012.14143 | 50 | 4165 | 1663.32639 |
| 4069 | 3012.71319 | 50 | 4068 | 1660.67226 |
| 4036 | 3012.38999 | 50 | 4036 | 1662.34972 |
| 4004 | 3011.86791 | 50 | 4004 | 1662.09403 |
| 3875 | 3012.03375 | 50 | 3874 | 1659.38759 |
| 3842 | 3011.42265 | 50 | 3842 | 1668.80359 |
| 3778 | 3012.12887 | 50 | 3778 | 1650.83721 |
| 3616 | 3012.82275 | 50 | 3616 | 1659.92726 |
| 3584 | 3012.68865 | 50 | 3584 | 1666.53729 |
| 3552 | 3010.41055 | 50 | 3551 | 1654.15346 |
| 3487 | 3014.00950 | 50 | 3487 | 1645.61980 |
| 3422 | 3012.72823 | 50 | 3422 | 1637.25877 |
| 3325 | 2949.16862 | 49 | 3325 | 1644.30617 |
| 3293 | 2949.94125 | 49 | 3293 | 1642.24700 |
| 3229 | 2948.71609 | 49 | 3228 | 1646.58733 |
| 3196 | 2949.35049 | 49 | 3196 | 1628.58362 |
| 3164 | 2949.83769 | 49 | 3164 | 1658.03155 |
| 3132 | 2949.00506 | 49 | 3131 | 1650.89009 |
| 3099 | 2950.74944 | 49 | 3099 | 1652.75818 |
| 3067 | 2949.46836 | 49 | 3067 | 1643.11548 |
| 3035 | 2948.30803 | 49 | 3035 | 1651.23157 |
| 2999 | 2950.74944 | 49 | 3002 | 1667.27672 |
| 2970 | 2887.42044 | 48 | 2970 | 1649.47722 |
| 2873 | 2887.58157 | 48 | 2873 | 1649.74008 |
| 2809 | 2887.05781 | 48 | 2808 | 1648.35273 |
| 2615 | 2886.40536 | 48 | 2582 | 1654.61123 |
| 2486 | 2887.08790 | 48 | 2486 | 1669.00597 |
| 2454 | 2886.10035 | 48 | 2453 | 1658.30694 |
| 2357 | 2887.41596 | 48 | 2356 | 1658.60042 |
| 2163 | 2885.14425 | 48 | 2163 | 1662.63238 |
| 2131 | 2887.78075 | 48 | 2130 | 1649.88402 |
| 2098 | 2886.65290 | 48 | 2098 | 1656.85022 |
| 2034 | 2886.11223 | 48 | 2034 | 1634.17643 |
| 1937 | 2886.00770 | 48 | 1937 | 1661.10625 |
| 1872 | 2885.60239 | 48 | 1872 | 1672.38712 |
| 1840 | 2884.88635 | 48 | 1840 | 1648.91827 |
| 1776 | 2886.22442 | 48 | 1775 | 1656.42473 |
| 1743 | 2886.05651 | 48 | 1743 | 1678.55941 |
| 1711 | 2886.37430 | 48 | 1711 | 1631.14618 |
| 1679 | 2885.22961 | 48 | 1678 | 1653.72650 |
| 1646 | 2886.25831 | 48 | 1646 | 1657.21189 |
| 1517 | 2886.30440 | 48 | 1517 | 1657.77623 |
| 1485 | 2884.64874 | 48 | 1484 | 1677.42301 |
| 1356 | 2884.71602 | 48 | 1355 | 1667.68381 |
| 1323 | 2885.82743 | 48 | 1323 | 1663.18166 |
| 1291 | 2885.31647 | 48 | 1291 | 1664.73490 |
| 1227 | 2884.56697 | 48 | 1226 | 1652.62980 |
| 1130 | 2884.92750 | 48 | 1129 | 1643.77902 |
| 1065 | 2883.74257 | 48 | 1065 | 1642.25601 |
| 1001 | 2885.43694 | 48 | 1000 | 1671.93587 |
| 968 | 2884.90346 | 48 | 968 | 1657.70949 |
| 936 | 2884.90088 | 48 | 936 | 1653.92713 |
| 904 | 2884.70356 | 48 | 904 | 1628.58362 |
| 807 | 2885.61606 | 48 | 807 | 1636.14503 |
| 839 | 2885.68476 | 48 | 839 | 1655.47979 |
| 936 | 2884.90088 | 48 | 936 | 1653.92713 |
| 968 | 2884.90346 | 48 | 968 | 1657.70949 |
| 1065 | 2883.74257 | 48 | 1065 | 1642.25601 |
| 1001 | 2885.43694 | 48 | 1000 | 1671.93587 |
| 968 | 2884.90346 | 48 | 968 | 1657.70949 |
| 936 | 2884.90088 | 48 | 936 | 1653.92713 |
| 904 | 2884.70356 | 48 | 904 | 1628.58362 |
| 807 | 2885.61606 | 48 | 807 | 1636.14503 |
| 839 | 2885.68476 | 48 | 839 | 1655.47979 |
| 936 | 2884.90088 | 48 | 936 | 1653.92713 |
| 968 | 2884.90346 | 48 | 968 | 1657.70949 |
| Carbon Black | Graphite |
|------------|---------|
| 10.6 μm, T = 1894 K, n = 0.26, 4591 sec, M = 37.62 Mda | 10.6 μm, T = 1526 K, n = 0.87, 820 sec, M = 37.62 Mda |

| Data | Data |
|------|------|
| 633.407 | 631.668 |
| 493.828 | 427.282 |
| 315.402 | 306.853 |
| 375.851 | 374.464 |
| 250.04 | 243.205 |

| Data | Data |
|------|------|
| 633.407 | 631.668 |
| 493.828 | 427.282 |
| 315.402 | 306.853 |
| 375.851 | 374.464 |
| 250.04 | 243.205 |

| Data | Data |
|------|------|
| 633.407 | 631.668 |
| 493.828 | 427.282 |
| 315.402 | 306.853 |
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| Data | Data |
|------|------|
| 633.407 | 631.668 |
| 493.828 | 427.282 |
| 315.402 | 306.853 |
| 375.851 | 374.464 |
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| Data | Data |
|------|------|
| 633.407 | 631.668 |
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| 315.402 | 306.853 |
| 375.851 | 374.464 |
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| Data | Data |
|------|------|
| 633.407 | 631.668 |
| 493.828 | 427.282 |
| 315.402 | 306.853 |
| 375.851 | 374.464 |
| 250.04 | 243.205 |

| Data | Data |
|------|------|
| 633.407 | 631.668 |
| 493.828 | 427.282 |
| 315.402 | 306.853 |
| 375.851 | 374.464 |
| 250.04 | 243.205 |
|   |   |   |   |   |
|---|---|---|---|---|
| 1336.48 | 1236.76 | 1365.31907 | 1310.33971 |
| 1265.81 | 1228.68 | 1362.86302 | 1301.77012 |
| 1323.99 | 1208.64 | 1397.87502 | 1280.49498 |
| 1232.56 | 1204.65 | 1378.71571 | 1276.26335 |
| 1300.23 | 1200.67 | 1327.65964 | 1272.04235 |
| 1180.99 | 1196.71 | 1337.17551 | 1267.83198 |
| 1221.39 | 1184.85 | 1253.39338 | 1255.25402 |
| 1283.64 | 1169.15 | 1302.96093 | 1238.59329 |
| 1238.03 | 1165.24 | 1215.10665 | 1234.44671 |
| 1260.83 | 1161.35 | 1332.96514 | 1230.31077 |
| 1269.64 | 1157.46 | 1290.96775 | 1226.18545 |
| 1219.34 | 1153.58 | 1312.91271 | 1222.07077 |
| 1160.22 | 1141.98 | 1236.43494 | 1209.76926 |
| 1322.66 | 1248.93 | 1378.1522 | 1323.25789 |
| 1385.63 | 1273.49 | 1402.01097 | 1349.32817 |
| 1339.81 | 1265.27 | 1422.03149 | 1340.59909 |
| 1273.5 | 1134.3 | 1285.85364 | 1201.61432 |
| 1206.4 | 1092.58 | 1263.48339 | 1157.35227 |
| 1054.5 | 1040.88 | 1125.56184 | 1102.51113 |
| 986.25 | 949.011 | 1064.68158 | 1005.0857 |
| 1358.6 | 1244.86 | 1417.651 | 1318.9412 |
| 1311.6 | 1216.63 | 1381.75653 | 1288.97952 |
| 1195.9 | 1212.63 | 1371.37945 | 1284.73725 |
| 1376.7 | 1269.37 | 1420.17085 | 1344.95831 |
