Giant near-field radiative energy transfer due to residual charge modes

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We study the modifications of near-field radiative energy transfer caused by residual charges, which are common in MEMS/NEMS, colloid solutions and lunar dusts etc. Besides the well-known resonant modes (surface polariton and localized surface polariton), the residual charges give rise to an additional temperature-dependent mode providing a new heat flux channel. For low temperatures we find a giant charge-induced enhancement of the NFRET, whereas for temperatures larger than 200K the low-frequency charge-induced mode results in a weakening of the NFRET.

The energy transfer between two objects with a separation distance comparable to or less than the thermal wavelength $\lambda_T = \frac{hc}{k_BT}$ via thermal photons can exceed by several orders of magnitude the blackbody limit, which has been investigated widely from both theoretical and experimental side for many different geometrical configurations (e.g., planar structures1–9 and nanoparticles-involved structures10–18) as reviewed in Ref.19, for instance. In the aforementioned works, only electrically neutral objects are involved. However, electrically neutral objects can easily be charged by exchanging electrons or ions with their environment20–23. The presence of residual charges significantly affects the optical properties of nanostructures, e.g. the absorption and scattering cross sections of nanoparticles23–26, the reflection and transmission coefficients of planar structures27 in the infrared region. Since the optical properties of nanostructures in the infrared determine their ability to exchange heat with their environment by thermal radiation, it can be expected that thermal radiation of charged objects in general and near-field radiative energy transfer (NFRET) in particular will be affected by the presence of residual charges. Due to the lack of studies on this topic, it is largely unknown how the presence of residual charges will affect NFRET in different geometries.

In this letter, the NFRET transfer between charged objects, e.g. charged nanoparticles and charged planar slabs (see Fig. 1 (a) and (b)), is analyzed within the framework of fluctuational electrodynamics1,2,28, by introducing the residual charge modification. We will show that the presence of residual charges provides a new heat flux channel via an additional charge-induced mode which will typically enhance the NFRET in the temperature-dependent Planck window. This effect is particularly large for low temperatures around 10K, as considered for the planar slabs, because in this case the Planck window matches well with the charge-induced mode, which results in a giant charge-induced effect on NFRET. However, for room or higher temperatures as considered for the particle case, the Planck window is far away from the low-frequency charge-induced mode, so that NFRET is only weakly affected even for very high surface charge densities corresponding to high bias voltages between the particles.

The residual charges can be assumed to be localized at the surfaces of the nanostructures20,23,27. The surface conductivity due to the residual charges is given by 24.

$$\sigma_s = \frac{\eta e / m_e}{k_B T / h - i \omega},$$  

where $\eta$ is the surface charge density, $e$ is the electron charge, $m_e$ is the electron mass, $k_B$ is the Boltzmann constant, and $h$ is the reduced Planck constant. For the charged slabs, the surface charge density is $\eta = e N$ where $N$ [m$^{-2}$] is the surface charge number density, while for charged nanoparticles of the radius $a$ we have $\eta = e_0 \phi / a$ where $\phi$ [V] is the surface potential24,26 and $e_0$ is the vacuum permittivity. From Eq. (1) it can be seen that the surface conductivity induced by the residual charges is temperature-dependent. In particular, the surface conductivity decreases when increasing the temperature, because charge motion and collisions become more significant in that case.
Let’s start by considering NFRET between two identical charged nanoparticles. The expression for the exchanged power \( P \) due to NFRET between a particle 1 and particle 2 within the framework of fluctuational electrodynamics for small particles within the dipole approximation can be written in a Landauer form as \(^{28-30}\)

\[
P = 3 \int_0^{\infty} \frac{d\omega}{2\pi} [\Theta(\omega, T_1) - \Theta(\omega, T_2)] T_{12}(\omega),
\]

where \( \omega \) is the angular frequency, \( \Theta(\omega, T) = \frac{\hbar \omega}{\exp(\hbar \omega/k_B T) - 1} \) is the mean energy of the harmonic oscillator at temperature \( T \) and \( T_{12}(\omega) \) is the transmission coefficient between both particles \(^{28-30}\). The transmission coefficient \( T_{12}(\omega) \) depends on the optical properties of the two particles, i.e., their electric and magnetic dipolar polarizabilities \( \alpha_E = \frac{i\sigma}{\omega} a_1 \) and \( \alpha_H = \frac{i\sigma}{\omega} b_1 \), where \( k_0 = \omega/c \) with the vacuum light velocity \( c \), \( a_1 \) and \( b_1 \) are the dipolar scattering coefficients. Due to the presence of residual charges these scattering coefficients derived from Mie theory are modified such that \(^{24,26}\)

\[
a_1 = \frac{\sqrt{\epsilon} \Psi_1''(y) - \Psi_1''(y) + \frac{i\mu_0\omega\alpha_0}{k_0} \Psi_1''(y)}{\sqrt{\epsilon} \Psi_1''(y) - \Psi_1''(y) + \frac{i\mu_0\omega\alpha_0}{k_0} \Psi_1''(y)},
\]

\[
b_1 = \frac{\Psi_1''(y) - \sqrt{\epsilon} \Psi_1''(y) - \Psi_1''(y) + \frac{i\mu_0\omega\alpha_0}{k_0} \Psi_1''(y)}{\Psi_1''(y) - \sqrt{\epsilon} \Psi_1''(y) - \Psi_1''(y) + \frac{i\mu_0\omega\alpha_0}{k_0} \Psi_1''(y)},
\]

where \( x = k_0a, y = \sqrt{\epsilon}k_0a, \epsilon \) is the relative dielectric permittivity, \( \mu_0 \) is the vacuum permeability, \( \Psi_1''(y) = xj_1(x), \Psi_1''(y) = \chi_1(x), \Psi''(y) = \chi''(x), j_1(x), h_1^{(1)}(x) \) are the Riccati bessel functions and \( j_1(x), h_1^{(1)}(x) \) are the spherical Bessel and Hankel functions. For the neutral particles (\( \sigma_z = 0 \)), the coefficients \( a_1 \) and \( b_1 \) defined by the Eqs. (3) and (4) reduce to the well-known Mie scattering coefficients \(^{31}\).

Similarly, for two identical charged planar slabs with the surface conductivity \( \sigma_z \), fluctuational electrodynamics allows to determine the flux \( \Phi \) [W/m\(^2\)] between those slabs yielding again a Landauer form \(^{24,26}\)

\[
\Phi = \int_0^{\infty} \frac{d\omega}{2\pi} [\Theta(\omega, T_1) - \Theta(\omega, T_2)] \int_0^{\infty} \frac{d\beta}{2\pi} \sum_{s,p} T_I(\omega, \beta),
\]

where \( \beta \) is the magnitude of the in-plane wave vector and the \( T_I(\omega, \beta) \) \((i = s, p)\) is the energy transmission coefficient for the \( s \)- and \( p \)-polarized waves, which again strongly depends on the optical properties of the two slabs via their reflection coefficients \( r_I \) \((i = s, p)\). Hence, the impact of residual charges modifies the heat flux by modifying the reflection as follows \(^{37,33,34}\)

\[
r_s = \frac{k_s^{(0)} - k_z - \sigma_z \mu_0 \omega}{k_s^{(0)} + k_z + \sigma_z \mu_0 \omega},
\]

\[
r_p = \frac{k_p^{(0)} e^{-1} + k_z e^{(0)} + \sigma_z k_z^{(0)} k_z e^{-1}}{k_z^{(0)} e^{(1)} + k_z e^{(0)} + \sigma_z k_z^{(0)} k_z e^{-1}}.
\]

where \( k_s^{(0)} = \sqrt{\epsilon_0 k_0^2 - \beta^2} \) and \( k_z = \sqrt{\epsilon k_0^2 - \beta^2} \). For the neutral slabs (\( \sigma_0 = 0 \)), the above reflection coefficients Eqs. (6) and (7) reduce to the well-known Fresnel reflection coefficients \(^{3,33}\).

In order to study the impact of residual charges on the exchanged heat between two nanoparticles or two planar slabs, we choose for the numerical calculation the typical dielectric material SiC for both nanoparticles and slabs. The dielectric functions is described by the Lorentz-Drude model \( \epsilon(\omega) = \epsilon_{\infty}(\omega^2 - \omega_p^2 - i\gamma\omega) \) with \( \epsilon_{\infty} = 6.7, \omega_p = 1.827 \times 10^{14} \) rad/s\(^{-1}\), \( \omega_0 = 1.495 \times 10^{14} \) rad/s\(^{-1}\), and \( \gamma = 0.9 \times 10^{12} \) rad/s\(^{-1}\). The particle radius will be set to \( a = 50 \) nm. The separation edge to edge between two nanoparticles or slabs is labeled by \( d \). The minimum \( d_{\text{min}} \) between the nanoparticle is 2\( a \) guaranteeing the validity of the dipole approximation \(^{19,29}\). Below, we will seperately discuss the residual charge effect on NFRET for small particles and slabs.

To study the impact of residual charges we first define the relative deviation \( \Delta = (P - P_0)/P_0 \) of the exchanged power \( P \) with residual charges with respect to the exchanged power \( P_0 \) without residual charges. The dependence of \( \Delta \) on the separation \( d \) between the two charged nanoparticles is shown in Figs. 2 (a) and (b). In Figs. 2 (a), the two particles are fixed at 200 K and 0 K and the voltage (or amount of residual charges) is varied. The surface potentials used in this work are comparable to that of the Refs. \(^{24,26}\). It can be seen that in the near-field regime with separations \( d < 4 \mu m \) there is a clear enhancement of the exchanged power when increasing the amount of residual charges up to about 2.5\( P_0 \). That there is only a strong residual charge effect for such small distances is due to fact that the residual charges are strongly localized at the particle surfaces having a strongly decaying electric field. In Figs. 2 (b), again one particle (receiver) is fixed at 0 K and temperature of the other particle (emitter) is varied from 200 K to 600 K choosing \( \varphi = 50 \) V. It can be observed that for low temperatures the enhancement due to the residual charges is strong. When increasing the temperature \( \Delta \) gets smaller. For temperatures larger than 200K there is also a sign change at a given distance. When increasing the temperature this distance moves to smaller values such that at 400K we find negative \( \Delta \) for all shown distance. Hence, for relatively large temperature the exchanged power is diminished by the presence of the residual charges.

Since the NFRET between the nanoparticles strongly depends on their absorbities which are proportional to \( \text{Im}(\alpha_z) \), we can get some insight in the underlying mechanism behind the observed charge-induced effects by focusing on \( \text{Im}(\alpha_z) \) of the SiC nanoparticles as shown in Fig. 3. Besides the well-know Fröhlich resonance at \( \omega_r = 1.756 \times 10^{14} \) rad/s from the condition \( \text{Re}[\epsilon(\omega_r)] = -2 \), a new charge-induced resonance can be observed for \( \omega < \omega_r \). This resonance is consistent with the reported charge-induced resonance around wavelength of 1 \( \mu m \) of SiO\(_2\) and some other particles \(^{36}\). Due to the fact, that this resonance has such a small frequency, it is clear that it has a particularly large im-
Impact at low temperatures. Furthermore, the resonance peak monotonically increases when increasing the potential so that $\Delta$ will inevitably show a monotonically increasing behavior when increasing the potential due to this resonance as seen in Fig. 2(a). To understand why, for large temperatures $\Delta$ can change its sign as observed in Fig. 2(b), we show in Fig. 3 a close-up of the Fröhlich resonance peak. Obviously, the resonance peak is blue shifted due to the presence of the residual charges and by increasing the temperature it is becoming weaker. Hence, for larger temperatures where the Fröhlich resonance dominates the heat transfer rather than the charge-induced resonance, we find a smaller NFRET $P$ when the particles are charged compared to the neutral case $P_0$. By increasing the temperature $P$ becomes smaller and smaller compared to $P_0$ as observed in Fig. 2(b).

![Image](image.png)

**FIG. 2.** Dependence of the $(P−P_0)/P_0$ on the separation $d$ between two nanoparticles. $P$ is radiative heat flux with residual charges, $P_0$ is the radiative heat flux with no charge. (a) The two particles are fixed at 200 K and 0 K, $\phi = 25$ V, 50 V, 75 V and 100 V, respectively. The considered surface potentials are comparable to those used in the Refs. 24, 26. (b) $\phi = 50$ V. One particle is fixed at 0 K. The temperature of the other particle takes the following values, $T = 200$ K, 300 K, 400 K, 500 K and 600 K.

![Image](image.png)

**FIG. 3.** Charged SiC nanoparticle “absorptivity” Im($\alpha_2$): (a) for different potential (i.e., 25 V, 50 V, 75 V and 100 V) at $T = 200$ K and (b) at different temperatures (i.e., $T = 200$ K, 300 K, 400 K, 500 K and 600 K) and fixed potential $\phi = 50$ V. The polarizability of the neutral SiC nanoparticle ($\phi = 0$) is shown as reference. The region around the Fröhlich resonance peak at $\omega_c = 1.756 \times 10^{14}$ rad/s is enlarged.

**Residual charge effect on NFRET of slabs.**—From our analysis of the NFRET between nanoparticles we can expect that there is also an enhancement of NFRET for charged slabs which might be particularly significant at low temperatures.

Therefore, we will now focus on the NFRET between two charged slabs at 10K so that the Planck window matches well with the spectral position of the charge-induced resonance. We note that the NFRET between two neutral slabs has already been experimentally studied at the such low temperatures, recently. Instead of studying the heat flux as expressed in Eq. (5) we focus on the heat transfer coefficient (HTC, $h$ [W/(m$^2$K)]) defined by

$$h = \lim_{T_1 \to T_2} \frac{\Phi_{T_2}}{T_1 - T_2} = \int_0^\infty h_{\omega_2} d\omega_1$$

where $h_{\omega}$ [W/(m$^2$K-rad·s$^{-1}$)] is the spectral HTC.

In Fig. 4, we show the full HTC as function of separation distance $d$ for different surface number densities $N$. It can be seen that due to the presence of surface charges there is a strong near-field enhancement of about two to three orders of magnitude for $d = 100$ nm and the chosen values of $N$. This observation is radically different from the negligible electrostatic effect for the metal planar structures at room temperature reported recently in Ref. 39. Note that the charges induce a heat flux proportional to $1/d^3$, so that at distances below 100nm the enhancement would be even much larger. However, it has to be kept in mind that the charges also induce a force on the plates and in addition if the gap between the slabs is not filled with vacuum but a gas, for instance, there can be a discharge when the voltage reaches the breakdown voltage of the gas. For the largest values of $N = 10^{15}$ m$^{-2}$ and $3 \times 10^{14}$ m$^{-2}$ we find a pressure of $F/A = 1.4$ kPa and 130 Pa which is about 100 or 10 times the Casimir force at 100 nm. For the voltage we find 1.8 V or 0.5 V at 100 nm so that the value corresponds to 6 or 1.7 times the breakdown voltage of air. Hence, values of $N > 5 \times 10^{13}$ m$^{-2}$ can only be achieved under vacuum conditions as typically used in near-field experiments.

To understand the mechanism behind the enhancement of HTC, we show the corresponding spectral HTCs in Fig. 5. Two kind of peaks can be observed: (1) a broad peak due to the Planck spectrum (typically shown by the curve for $N = 0$) which is independent of the charge density; (2) a low frequency peak corresponding to the charge-induced resonance. Obviously, when increasing the charge number density this peak enhances the HTC by shifting towards the Planck window. This can be explained by increased charge collisions resulting in a blueshift of the charge induced resonance. Similarly to the case of two nanoparticles, when the separation $d$ increases the charge-induced enhancement effect decreases dramatically, which can again be attributed to the fact that the resonant mode due to residual charges is highly localized on the surfaces of the slabs. Note, that the surface phonon polariton resonance of SiC is at $\omega = 1.787 \times 10^{14}$ rad/s and therefore cannot be thermally excited at 10K. This also explains, why there the HTC does not show any $1/d^2$ dependence associated with the surface phonon polaritons in Fig. 4.

Finally, we show the transmission coefficient $T_\phi(\omega, \beta)$ of the $p$-polarized waves for $d = 100$ nm in Fig. 6 for $N = 3.0 \times 10^{14}$ m$^{-2}$ and $1.0 \times 10^{15}$ m$^{-2}$. The enhanced transmission due to the charge-induced mode can be nicely seen.
FIG. 4. Distance dependence of the HTC $h$ between two charged SiC slabs at 10 K considering the number densities $N = 0, 5 \times 10^{13} \text{m}^{-2}, 3 \times 10^{14} \text{m}^{-2}$ and $1 \times 10^{15} \text{m}^{-2}$. The line of $1/d^3$ is added for reference.

FIG. 5. Spectrum of the HTC $h$ at $T = 10$ K for $N = 0, 5 \times 10^{13} \text{m}^{-2}, 3 \times 10^{14} \text{m}^{-2}$ and $1 \times 10^{15} \text{m}^{-2}$ choosing the separation distance $d = 100 \text{nm}$. whereas the surface phonon polariton is outside the plotted frequency range. When increasing the charge number density [from Fig. 6 (a) to Fig. 6 (b)], the charge-induced mode blueshifts and approaches the Planck window at 10K. Hence, more and more energy can be transferred between the two slabs when increasing the charge density, which accounts for the increasing enhancement of HTC observed in Fig. 4. In Fig. 6, the dispersion relation of the charge-induced resonance is shown as dashed-symbol line. It is determined by the poles of the reflection coefficient $r_p$ in

Eq. (7). For the strong evanescent limit $\beta \gg k_0$ it is given by

$$\beta = \frac{(\varepsilon + 1)\varepsilon_0 \omega_m(e^{i k_0 T} + h + \omega)}{N \omega^2}.$$  

(9)

From this relation and Fig. 6 we can easily understand the blueshift of the charge-induced resonance. Because the evanescent waves with $\beta = 1/d$ dominate the heat flux between the slabs at a given distance $d$ we could chose a fixed value of $\beta = 1/d$ in the dispersion relation in Eq. (9). It is then clear that when increasing $N$ the numerator has to increase as well to obtain $1/d$ which can be achieved by increasing $\omega$ (assuming that dispersion can be neglected in that frequency range) explaining the observed blueshift in the spectral HTC in Fig. 5. Likewise, one can think of a vertical line in Fig. 6 at $\beta = 1/d$ which is about $30 \beta_0$ for $d = 100 \text{nm}$. The intersection point of that line with the dispersion relation marks the spectral position of the resonance. Then it is obvious from Fig. 6 that the resonances are blue-shifted when increasing $N$.

FIG. 6. The transmission coefficient $T_p(\omega, \beta)$ for $p$-polarized waves for two SiC slabs at separation $d = 100 \text{nm}$ with a charge number density $N$ given by (a) $3.0 \times 10^{14} \text{m}^{-2}$ and (b) $1.0 \times 10^{15} \text{m}^{-2}$. The frequencies and wave vectors are normalized to $\omega_0 = 1.0 \times 10^{14} \text{rad} \cdot \text{s}^{-1}$ and $\beta_0 = \omega_0/c$.

In summary, we have studied the effect of residual charges on the NFRET between two nanoparticles and two planar slabs made of SiC. The residual charges on the nanoparticles and slabs give rise to an additional charge-induced resonant mode in the low-frequency infrared range. This mode provides a new heat flux channel enhancing the NFRET. For temperatures as low as 10 K as considered for the two slabs the Planck window matches well with the charge-induced mode resonance resulting in a giant charge-induced enhancement of three orders of magnitude at $d = 100 \text{nm}$. Consequently, for temperatures around room temperature as considered for the nanoparticles, the Planck window is far away from the low-frequency charge-induced mode, which accounts for a much weaker residual charge effect on NFRET with an enhancement factor of 2.5. Furthermore, for temperatures larger than 200K we find that the NFRET is rather inhibited than enhanced due to the surface-charge effect. These observations pave the way for understanding NFRET between nanostructures under extreme electromagnetic conditions like in colloid solutions and lunar dusts or in nanostructures like MEMS/NEMS where residual charges are unavoidable.
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