Observation of cooperatively enhanced atomic dipole forces from NV centers in optically trapped nanodiamonds

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Since the early work by Ashkin in 1970 [1], optical trapping has become one of the most powerful tools for manipulating small particles, such as micron sized beads [2] or single atoms [3]. The optical trapping mechanism is based on the interaction energy of a dipole and the electric field of the laser light. In atom trapping, the dominant contribution typically comes from the allowed optical transition closest to the laser wavelength, whereas for mesoscopic particles it is given by the bulk polarizability of the material. These two different regimes of optical trapping have coexisted for decades without any direct link, resulting in two very different contexts of applications: one being the trapping of small objects mainly in biological settings [4], the other one being dipole traps for individual neutral atoms [5] in the field of quantum optics. Here we show that for nanoscale diamond crystals containing artificial atoms, so-called nitrogen vacancy (NV) color centers, both regimes of optical trapping can be observed at the same time even in a noisy liquid environment. For wavelengths in the vicinity of the zero-phonon line transition of the color centers, we observe a significant modification (10%) of the overall trapping strength. Most remarkably, our experimental findings suggest that owing to the large number of artificial atoms, collective effects greatly contribute to the observed trapping strength modification. Our approach adds the powerful atomic-physics toolbox to the field of nano-manipulation.

Whenever a polarizable particle is exposed to light, the electromagnetic field induces an optical dipole moment which in turn leads to an interaction energy that scales with the field intensity. This interaction energy results in optical forces that ultimately allow for spatial manipulation of the particle. Using a semi-classical approach the optical forces can be derived as [6]:

$$\mathbf{F}(\mathbf{r}) = \epsilon_0 \alpha' \frac{E_0(\mathbf{r})}{2} \nabla E_0(\mathbf{r}) - \epsilon_0 \alpha'' \frac{E_0^2(\mathbf{r})}{2} \nabla \phi(\mathbf{r})$$  \hspace{1cm} (1)

where the polarizability $\alpha$ of the particle has been split into real and imaginary parts $\alpha = \alpha' + i\alpha''$, $\epsilon_0$ is the dielectric permittivity of vacuum, and the incident time-averaged field amplitude and phase are $E_0$ and $\phi$, respectively. The real part of the polarizability gives rise to the so-called dipole force associated with a conservative trapping potential. In contrast, the imaginary part of $\alpha$ leads to dissipative resonant scattering forces proportional to the gradient of the phase of the field. Due to its dissipative nature, the resonant scattering force
plays a key role in atom cooling \[6\]. In the context of this work, we can neglect the resonant
scattering term by utilizing a Gaussian standing wave (GSW) trap \[7\].

For an isotropic and homogeneous object, the polarizability \(\alpha\) is directly related to the
refractive index of the material and is typically a slowly varying function of wavelength.
Conversely, in the case of a two-level quantum system (e.g., a single atom in free space) the
dipole force exhibits a strong dependence on the trapping wavelength through the resonant
nature of the polarizability. In particular, it changes sign at the transition frequency, \(\omega_0\),
and becomes repulsive for blue detunings (\(\omega > \omega_0\)). Experiments on ultracold neutral atoms
routinely exploit this strong detuning dependence for creating complex potential landscapes
\[5\]. In the following, we report on the observation of near-resonant optical forces from an
ensemble of artificial atoms embedded in a nanocrystal.

Nanodiamonds (NDs) containing color centers are excellent candidate nanoparticles for
observing atom-like trapping resonances. The color centers act like artificial atoms exhibiting
sharp optical transitions. In particular, the nitrogen-vacancy center (NV), consisting of a
nitrogen atom and an adjacent vacancy site (see Figure 1a left), has attracted a lot of
interest over the past decade. In its most stable form, the negatively charged NV\(^{−}\), it
exhibits outstanding spin-optical properties which persist even up to room-temperature \[8\].
Consequently, the NV\(^{−}\) has proven to be highly suitable as a solid-state spin qubit \[9\] and
nanoscale magnetic sensor \[10, 11\]. Here we are mainly interested in its optical properties.
The NV\(^{−}\) displays stable single-photon emission \[12, 13\] with a sharp zero-phonon line (ZPL)
in bulk at 637 nm followed by well-defined vibronic side bands \[14\]. At room temperature,
most photons are emitted into these sidebands and only a fraction into the ZPL (typically
4\% \[15\]). Note that, mainly due to strain, the ZPL position in NDs can shift considerably. In
our experiment, we measured the average ZPL to be 639 nm. Due to their strong and stable
fluorescence, NV\(^{−}\) centres hosted in nanodiamonds have been used as biolabels for high-
resolution, real-time and low-disruption imaging of living cells \[16\] and as carriers for drugs
and biomolecules \[17\]. Previous investigations on liquid trapping \[18, 19\] and levitating NDs
\[20\] used laser light at 1064 nm which is far away from the ZPL at 637 nm. None of these
experiments reported any effects due to the presence of the NV\(^{−}\) centers on the external
degrees of freedom of the nanodiamond in the optical trap.

In our experiment, we create a GSW trap near 639 nm by focusing a Gaussian laser
beam on a silver-coated mirror. The GSW provides a stronger trap along the direction of
FIG. 1: Trapping nanodiamonds. **a**, Highly-irradiated nanodiamonds contain many nitrogen vacancy centers which exhibit a strong optical transition at around 637 nm (left panel). The level scheme displays the relevant optical and non-radiative transitions (for spectroscopic notation see e.g. [8]). A single NV center trapped in the center of a focused Gaussian beam (numerical aperture 1.2, P= 4 mW) experiences a dispersive atom-like trapping potential due to the resonant dipole force (right panel). **b**, Experimental setup with the trapping laser focussed through a high numerical aperture objective into a micro-fluidic chamber producing a strong optical trap. The top of the micro-fluidic chamber consists of a mirror which results in a standing wave, improving the trapping efficiency and minimizing the scattering force. The light backscattered from the system is collected via a beam splitter (BS) and sent to a quadrant photo-detector (QPD) to track the position of the particle.

The standing wave compared to conventional focused Gaussian beam, and allows one to neglect scattering forces. The mirror forms the top of a static micro-fluidic chamber that contains the NDs suspended in deionized water (see Figure [b]). The laser sources are a set of temperature-stabilized laser diodes operating at different wavelengths detuned with respect to 639 nm (see Materials and Methods). In addition, a pulsed green laser (532 nm) serves as a weak re-pump to counteract resonant ionization to the neutral NV$^0$ state.
FIG. 2: **Measured relative trap stiffness.** a, Relative trap stiffness \((\kappa(\lambda)/\kappa(\lambda_{ref}))\) for NDs containing only few NVs. The symmetric spread in the measured trap stiffness for a given wavelength is due to experimental noise. b, Relative trap stiffness for NDs containing a large number of NV centers. The data scattering is strongly asymmetric with \(\kappa\)-ratios much lower than 1 for wavelengths below \(\lambda_{ref}\) and higher than 1 for wavelengths above \(\lambda_{ref}\). a, b The shaded areas are guides to the eye and indicate the range of data scattering.

The trap depth itself is hard to measure directly in optical tweezers in liquid. Instead, by measuring the corner frequency \(\omega_c\) (see Materials and Methods), we obtain the trap stiffness, \(\kappa\), which corresponds to the second derivative of the trapping potential. The nanodiamonds are in the strongly overdamped regime and their displacement from the trap center is small compared to the beam waist. As a result, the particles mainly probe the harmonic part of the potential near the trap minimum \(U \sim U(0) + \kappa x^2\). In order to obtain the stiffness experimentally, the position of the trapped particle is recorded using a quadrant photodiode. The stiffness can then be obtained from a Lorentzian fit of the power spectrum density of the signal (see Materials and Methods). Due to its dependence on the exact size of the trapped ND, \(\kappa\) is not a good observable when comparing the results obtained for different NDs. In order to circumvent this problem, we extract a relative value for \(\kappa\) normalized to the measured trapping stiffness at a reference wavelength (\(\lambda_{ref} = 639.13\) nm) for each ND separately (see Materials and Methods).

In order to obtain a significant resonant trapping effect, we use NDs with a high concentration of NV\(^-\) centers. We first characterized the NDs using a home-built combined confocal/AFM microscope setup. The investigated NV\(^-\) centers show a ZPL centered
at (639.08 ± 0.65) nm with an average spectral width of (2.09 ± 0.55) nm (see Extended Data Fig. 1). The ZPL width of individual nanodiamonds is a convolution of a Gaussian and a Lorentzian distributions (Voigt profile). Assuming a dephasing rate at room temperature of approximately $2\pi \times 1$ THz [25], we extracted the width of the underlying Gaussian distribution due to the variation of NV ZPL frequency within a ND to be $\sigma_{ZPL} = (1.82 \pm 0.55)$ nm.

We also measured the size distribution of the nanocrystals in liquid using dynamic light scattering and found an average size of (150 ± 23) nm. The expected average number of NVs per ND is $\langle NV \rangle \approx 9,500$ [23]. A sample of NDs with a similar size, (168 ± 31) nm, but with a much lower concentration of NV centers was used as a reference.

To characterize the contribution of the NVs to the trap stiffness, we measured the corner frequency on a number of different NDs for a given set of wavelengths for the two ND samples (low and high NV center concentration). We then extracted the trap stiffness for each ND separately and applied a statistical analysis in order to systematically discard unwanted events such as the trapping of multiple NDs or standing wave hopping (see Supplementary Material). The resulting trap stiffness ratios as a function of laser wavelength for the reference NDs are displayed in Figure 2a. This measurement serves as a reference as we do not expect the resonant trapping forces to have a measurable effect for this sample. The monotonic trend is attributed to chromatic aberrations which are aggravated by the standing wave trap. In contrast, Figure 2b displays the results for the ND sample with high NV− density. As in the case of the reference sample, the underlying monotonic trend is clearly visible. However, the high density data clearly shows a strongly asymmetrical distribution with $\kappa$-ratio values much lower than 1 for wavelengths below $\lambda_{ref}$ and values larger than 1 for wavelengths larger than $\lambda_{ref}$. The extreme values are attributed to NDs with larger number of NV centers leading to a significant contribution of the NVs to the trap stiffness.

Next, we extract the mean value from both data sets and plot the difference of the mean values as a function of wavelength (Figure 3a). Due to the choice of reference wavelength, this difference of mean values gives access to the ratio of the stiffness arising from the NV ensemble to the stiffness from the bare ND matrix (see Supplementary Material). The dispersive trend is clearly visible, corresponding to a stiffness ratio lower than 1 at wavelengths below the ZPL and larger than 1 for wavelengths above, with a magnitude of up to 10%.

Most remarkably, these experimental results cannot be accounted for only by assuming independent NVs: The dashed line in Figure 3b displays the expected trap stiffness from a
FIG. 3: Cooperative dipole force.  

**a**, Difference of the mean values from figures (2a) and (2b). This difference, $\Xi = \kappa(\lambda)/\kappa(\lambda_{ref})|_{\text{highNV}} - \kappa(\lambda)/\kappa(\lambda_{ref})|_{\text{lowNV}}$, is a close approximation of the ratio of the stiffness from the NV centres, $\kappa_{NV_s}(\lambda)$, over the stiffness from the diamond matrix, $\kappa_D(\lambda)$ (see Supplementary Information). The standard error is indicated in grey. The black curve shows the theoretically predicted stiffness ratio accounting for collective effects on the force obtained for collective sub-ensembles with a spectral width of 100 GHz. For comparison, the dashed line is obtained by considering independent NVs (magnified 55 times). In addition, our Monte Carlo calculation predicts that in the absence of noise, 90% of the experimental values for the difference ratio $\Xi$ should lie within the shaded area. (top panel) Experimental skewness (open squares) from high density NV sample measurements along with the simulation results from our Monte-Carlo including collective effects (solid stars). (bottom panel) experimental skewness from the low density NV sample (solid diamonds). Here the skewness displays no obvious deviation from zero.

**b**, Skewness as a function of wavelength. (top panel) Experimental skewness (open squares) from high density NV sample measurements along with the simulation results from our Monte-Carlo including collective effects (solid stars). (bottom panel) experimental skewness from the low density NV sample (solid diamonds). Here the skewness displays no obvious deviation from zero.
calculation that assumes independent NV centers and takes the average values for the ZPL width, ZPL position and number of NV centers per ND. Note that the curve has been magnified by 55 times for plotting. Clearly, assuming the NV centers to act independently does not reproduce our experimental findings. Besides the magnitude being off by almost two orders of magnitude, the shape of the curve also does not match the observations. In order to fully capture the experimental results, we therefore have to consider collective effects [26] between the NV centers within a single nanocrystal.

Cooperative effects between NV centers have a significant impact on the dipole forces by increasing the spontaneous decay rate and modifying the steady state population. The presence of collective effects in our high NV density NDs was independently confirmed through a set of scattering experiments where sub-nanosecond cooperative decay rates were observed [27]. Due to the large variation in ZPL position in a single ND, only sub-domains of NVs within narrow frequency windows are expected to act collectively. To model the collective forces, we applied the Dicke model [28] to each sub-domain. In this context, the $N_i$ NVs contained in the sub-domain $i$ can be represented as a superposition of $N_i + 1$ spin 1/2 states. The collective force can then be obtained by solving the Liouville equations for the collective spin operator $S_+ = \sum_{j=1}^{N_i} \langle e_j \rangle \langle g_j |$ where $|e_j \rangle$ and $|g_j \rangle$ are the excited and ground state of NV number $j$ respectively. In this simplified model, the single-spin dephasing rate is given by the spontaneous decay rate of the NV, and we used a collective spin dephasing rate at room temperature of $2\pi \times 1$ THz [25] (see Supplementary Material). The collective force calculated for our particular ND sample is presented in Figure 3a as the solid black line along with the experimental data points. The variability of the NV density was reproduced using a Monte Carlo approach, providing a confidence interval within which 90% of the experimental values should fall (shaded area in Fig. 3a). The number of sub-domains, and consequently their frequency width, has been used as the only adjustable parameter in our model and provides a number for NVs acting collectively. The measurements were best reproduced using a sub-domain size of approximately 100 GHz corresponding to an average domain size of 95 NV centers, or 1% of the total average number of centers (see Supplementary Material). Comparing with the force obtained for independent NVs, the overall impact of the collective effects is clearly apparent. We estimate an enhancement of the trap stiffness of around a factor 50, demonstrating the importance of collective effects in our analysis. In parallel to the average trap stiffness, we also modelled the expected skewness of the experimental
data points (Fig. 3b upper panel). Our model gives a good quantitative agreement with the skewness obtained from the experiment. As the model does not include any type of experimental noise, the skewness is consequently slightly over-estimated. For comparison, the skewness of the experimental data obtained for the reference ND sample is presented in the lower panel of Fig. 3b.

In conclusion, our observations open the door to a wealth of new research directions. The collective effects arising from the high number of NV centers in an individual nanocrystal provide a mechanism to significantly increase the optical forces. While our simplified model is in good agreement with the experiment, a more complete description accounting for dipole-dipole interactions [28] constitutes an interesting future research direction. In addition, the observed 10% change in trapping stiffness could be further increased by using defects such as silicon-vacancy centers which are characterized by higher densities [29] and stronger transition dipole moment [30, 31]. These centers could offer the opportunity to access a regime in which the resonant trapping forces dominate the dynamics of the system, with the nanocrystal essentially behaving like a very large atom, or superatom. In the context of quantum opto-mechanics, this could allow for single-photon strong coupling and side-band cooling at room temperature [32]. With all these exciting possibilities at hand, this work opens the door to applying the powerful quantum technologies developed for atom trapping and cooling to the manipulation of small nanoparticles introducing an unprecedented degree of control at the nanoscale.

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Materials and Methods

Nanodiamond (ND) Sample The two nanodiamond samples used in this work are synthetic type Ib diamond powders with a nitrogen concentration of 300 ppm, i.e. $3 \times 10^7$ nitrogen atoms per $\mu m^3$ (MSY <0.1 $\mu m$; Microdiamant). This ND powder was only chemically and mechanically processed to remove the sp$^2$ carbon-phase in excess. The NDs with no additional treatment were used as reference (low NV centers concentration) in the control experiment to determine the effect of chromatic aberrations in the standing wave trap. For high NV centers concentration, the ND powder was further treated to increase the concentration of NV centres (Academia Sinica, Taipei Taiwan) as follows: the nanodiamonds were purified by nitration in concentrated sulphuric and nitric acid ($H_2SO_4$-$HNO_3$), rinsed in deionized water, irradiated by a 3-MeV proton beam at a dose of $1 \times 10^6$ ions per cm$^2$ and annealed in vacuum at 700 degrees Celsius for 2 hours to induce the formation of NV centres. Prior to the trapping experiment, both nanodiamond samples were characterized by means of a lab-built confocal scanning fluorescence microscope (100x oil immersion objective UplanFL N, NA 1.3; Olympus) excited with a 532-nm CW diode-pumped solid-state laser (Compass 315-M100; Coherent Scientific) and combined with a commercial atomic force microscope (Ntegra; NT-MDT). For characterization, the diamond nanocrystals were dispersed on 170-$\mu m$ thick BK7 glass coverslips (BB022022A1; Menzel-Glaser) which were previously sonicated and rinsed in acetone (C3H6O, purity $\geq$ 99.5%; Sigma-Aldrich) for 10 min. The measured average size of the nanodiamonds is $(150.5 \pm 23.3)$ nm, determined by atomic force microscopy and confirmed by dynamic light scattering analysis (Zetasizer Nano-ZS; Malvern Instruments). The spectral interrogation of the NDs to identify emission from NV$^-$ centres was performed via a commercial spectrometer (Acton 2500i, Camera Pixis100 model 7515-0001; Princeton Instruments). While for the untreated sample the concentration of NV centres is extremely low (at most a few NVs per nanocrystals), for the irradiated one we estimate $\sim 10^4$ NV centres per nanodiamond. This was determined by correlating, for nanocrystals of different sizes, the average fluorescence intensity measured for each ND with its volume, and comparing this ratio with the one given by the sample provider.

Trapping Setup For the trapping experiment, the suspension of nanodiamonds in deionized water was inserted in a microfluidic chamber consisting of a BK7 glass coverslip...
(BB022022A1; Menzel-Glaser) and a protected silver mirror (PF10-03-P01; Thorlabs) using double sided tape for sealing (50 mm × 50 m, 0.14 mm-thick; 3M). The experiment involves five diode lasers, four used for the optical trapping itself and one for the re-pumping from NV$^0$ to NV$^-$. The re-pump laser is a 532-nm pulsed laser (LDH-P-FA-530B; PicoQuant) used at 40 MHz repetition rate with an average output power at the sample of 30 µW. The four other lasers are temperature-stabilized laser diodes combined through the same fibre to ensure that the focal spots of the different diodes are superimposed perfectly in lateral direction. One of these diodes is used to provide a stable conventional trap for the ND at a wavelength of 660 nm with 6 mW of power (660 nm/130 mW; Oclaro) in order to maintain the ND trapped during the measurement. The three remaining diodes provide the reference wavelength ($\lambda_{ref}$), the blue and the red wavelengths with 4 mW of power. The choice of diodes (Oclaro 633 nm/110 mW, Oclaro 637 nm/170 mW, Mitsubishi 638 nm/150 mW and Oclaro 642 nm/150 mW) provides an overall covered spectral range of 629-648 nm (see Extended Data Fig. 2a). The laser beams are switched using home-built electromechanical shutters controlled with a data acquisition system (NI-PCI 6289; National Instrument) in order to provide 50-s continuous time trace composed of 10-s segments with different wavelengths (see Extended Data Fig. 2b). The trapping beam at the output of the fiber is polarized using a Glan-Laser calcite polarizer (GL10-A; Thorlabs) and then focused into the chamber through a water-immersion objective (UPLSAPO 60XW, NA 1.2; Olympus) with a measured waist at 640 nm of $w_0=470$ nm. The position of the objective is set such that the reference laser diode (639.13 nm) is focussed on the mirror forming the top surface of the microfluidic chamber. Finally, the wavelength for the reference laser was chosen very close to the measured average NV ZPL position of 639.08 nm (see Extended Data Fig. 1b).

**Stiffness measurement** Using a quadrant photodiode, the position of the ND is tracked over time. A Fourier transform of the signal yields the corresponding power spectral density (PSD) in reciprocal space. From a Lorentzian fit to the PSD \( [2] \), the corner frequency \( f_c \) is extracted which can be directly related to the trap stiffness through \( \kappa = 2\pi\beta f_c \), where \( \beta \) is the drag coefficient of the ND. In the experiment, we measure \( f_c \) as a function of wavelength in the vicinity of the ZPL of the NV$^-$. The drag coefficient \( \beta \) is a function of the viscosity of the medium, the ND size and its distance to the surface \( [2] \). Consequently, referencing every measurement to a reference wavelength \( f_c(\lambda_{\text{measure}})/f_c(\lambda_{\text{ref}}) \) allows direct access to the ratio of the stiffnesses without explicit knowledge of the drag coefficient. In addition, we
subtracted to each measurements ($\lambda_{\text{red}}+660$-nm, $\lambda_{\text{blue}}+660$-nm or $\lambda_{\text{ref}}+660$-nm) the corner frequency obtained for the 660-nm laser in order to access the stiffness ratio for $\lambda_{\text{red}}$, $\lambda_{\text{blue}}$ or $\lambda_{\text{ref}}$ (see Supplementary Information).

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FIG. 1: Extended Data Fig. 1. ND properties. All the properties are extracted from photoluminescence (PL) spectra obtained on 40 different NDs. 

- **a**, Spectral width of the ZPL peak. The distribution was fitted by a normal distribution (in blue), giving an average value of $\sigma_{\text{spectrum}} = 2.09 \text{ nm}$. 
- **b**, Position of the ZPL peak. The distribution consists of two clearly distinct populations which we approximated by two normal distributions. The overall average ZPL position is $\langle ZPL \rangle = 639.08 \text{ nm}$. 
- **c**, Estimated number of NVs normalized to a 100 nm ND. The actual values were obtained by referencing the observed average photo-luminescence counts to the values obtained by the sample provider in Ref [23]. 

**a**, **b**, **c**, the experimental data is presented in orange and the model in blue.
FIG. 2: Extended Data Fig. 2. Laser sources and timing sequence. a, Spectra of the different laser diodes used in the experiment: shown in black is the output from the reference laser diode (\(\lambda_{\text{ref}}\)), in blue the output from the “blue” laser diodes (\(\lambda < \lambda_{\text{ref}}\)) and in red the output from the “red” laser diodes (\(\lambda > \lambda_{\text{ref}}\)). b, Timing sequence used during the 50-s acquisition interval. The 660-nm and 532-nm lasers are kept on all the time: the 660-nm laser provides efficient trapping, the 532-nm laser re-pumps NV centers from the NV\(^0\) to the NV\(^-\) state. The first and last 10 seconds of the acquisition window give the trap stiffness for the 660-nm trapping laser only. In the intermediate 30 seconds, there are three 10-s acquisition windows for the “blue”, reference and “red” lasers, respectively.
Supplemental Materials: Observation of cooperatively enhanced atomic dipole forces from NV centers in optically trapped nanodiamonds

Data acquisition and treatment

Data acquisition and sequencing

The data was obtained from an acquisition time trace of 50 s overall duration. The timing sequence for applying the different trapping lasers during these 50 s is displayed in the Extended Data Fig. 2. The time trace is partitioned in equal segments of 10 seconds, and the corner frequency during these 10 seconds is obtained from the power spectral density of each individual segment. During the first and last 10 s of acquisition, only the 660-nm laser is on corresponding to the corner frequency $f_{660}$. During each of the other 10 s intervals, the 660-nm laser is on plus one of the three following lasers at a time: the “blue” laser with $\lambda < \lambda_{\text{ref}}$ (giving the corner frequency $f_{\text{blue}}$), the reference laser with $\lambda = \lambda_{\text{ref}}$ ($f_{\text{ref}}$), and the “red” laser with $\lambda > \lambda_{\text{ref}}$ ($f_{\text{red}}$). Since the 660-nm laser is far detuned from the resonance the dipole force from the NV transition with this laser can be neglected. This point stands when using both the 660-nm laser and a laser closer to the NV transition (e.g. “red” laser): the dipole force from the NV due to the “red” laser does not depend on the 660-nm laser (for moderate power as such used in this work). Within the harmonic approximation, the overall corner frequency with two lasers on simultaneously (e.g. the 660-nm laser and the laser with $\lambda_{\text{ref}}$) can thus be written as: $f_{\text{ref}} = (\kappa_{660} + \kappa_{\text{ref}})/(2\pi\beta)$ with $\beta$ the drag coefficient of the particular nanodiamond. The observables of interest, $\kappa_{\text{blue}}/\kappa_{\text{ref}}$ and $\kappa_{\text{red}}/\kappa_{\text{ref}}$, are therefore determined by

$$\frac{\kappa_{\text{blue}}}{\kappa_{\text{ref}}} = \frac{f_{\text{blue}} - f_{660}}{f_{\text{ref}} - f_{660}} \quad \text{and} \quad \frac{\kappa_{\text{red}}}{\kappa_{\text{ref}}} = \frac{f_{\text{red}} - f_{660}}{f_{\text{ref}} - f_{660}}$$

(S1)

Normalizing the corner frequencies this way gives access to the trapping stiffness ratios without requiring knowledge of the actual size of the trapped NDs. In addition, the normalization avoids the dependence of the trapping stiffness on the volume. Hence, the normalized ratios can directly be compared for different NDs.
Data treatment

As explained previously, the first and last 10 s of the acquisition are necessary to extract the effect of the 660-nm trapping laser only. In addition, we compared the corner frequency for these two segments with the 660-nm trapping laser to identify anomalous events during the whole 50 s data acquisition. As a boundary condition for discarding anomalous time traces, we imposed a relative change in corner frequency smaller than 10% between the two segments. Based on this 10%-rule, we typically removed 20% of the data. The main cause for a dramatic change in 660-nm corner frequency at the beginning and end of the 50 s acquisition period would most likely be a second ND hopping into the trap due to the relatively high concentration of NDs in the solution.

The 10% selection rule was applied before calculating the trapping stiffness ratios and allowed us to discard clearly anomalous time traces. In addition, we also used a Local Outlier Factor (LOF) method to remove clear statistical outliers from the data. This method is based on calculating the local density of neighbors for each data point. Within the LOF method, (local) outliers are identified by their large LOF value. We rejected data points having a LOF larger than 5.7 using a LOF calculation based on the 6th-nearest neighbor. The same parameters were used for all the data and ultimately removed 3.4% of the remaining data points. We verified that this method would not impact the effect we wanted to underline. To do so, we used a Monte Carlo simulation to numerically reproduce the experiment accounting only for the variability of the ND properties (number of NVs, average ZPL transition and ZPL standard deviation). Applying the very same LOF selection rule to the numerical results only removed an average of 0.6% of the numerical data points. This confirms that even with the large variability in the NV density, the outlier method only marginally impacts the data.

Calculating the dipole force for an ensemble of NVs

Dipole force on a 2-level system

For a simple 2-level system with a dipole moment \( d_2l \) and transition \( \omega_{2l} \) in the presence of dephasing \( \gamma = \Gamma/2 + \gamma_c \) in an electric field with amplitude \( E_0(r) \), the optical dipole...
potential can be written as \[S2\]:

\[
U_{2l}(\omega, r) = -\frac{\hbar (\omega - \omega_{2l})}{2} \frac{\Gamma}{2\gamma} \log [1 + s_{2l}(\omega, r)],
\]

where \(s_{2l}(\omega, r) = \Omega(r)^2/\left[\Gamma \gamma (1 + (\omega - \omega_{2l})^2/\gamma^2)\right]\) is the saturation parameter and \(\Omega(r) = \sqrt{2/3} d_{2l} E_0(r)/\hbar\) the Rabi frequency. Here the prefactor \(\sqrt{2/3}\) comes from the time average of the electric field \((\sqrt{2})\) and the orientational average of the dipole moment \((1/\sqrt{3})\).

\[NV\ center\ and\ phonon\ sidebands\]

NV centers constitute good quantum emitters, yet their level structure is quite different from the ideal 2-level system usually considered in calculating the dipole forces. In particular, the large phonon sideband (PSB) needs to be taken into account when estimating the dipolar moment. From the spontaneous rate emission in vacuum of NVs, \(\Gamma_0\), at the transition frequency \(\omega_0\), the overall dipolar moment \(d\) can be determined via the Fermi Golden rule as \(d = \sqrt{\Gamma_0 3\pi \epsilon_0 c^3 \hbar/\omega_0^3}\). Here \(\epsilon_0\) is the vacuum permittivity, \(c\) the speed of light and \(\Gamma = n_1 \Gamma_0\) with \(n_1\) the refractive index of diamond and \(\Gamma\) the spontaneous rate emission of NVs in nanodiamonds \[S3\]. Decomposing the PSB in 7 distinct bands following \[S6\], the spontaneous rate emission can be written as \(\Gamma = \Gamma_{ZPL} + \sum_{i=1}^{7} \Gamma_i\) with \(\Gamma_{ZPL}\) the zero phonon line (ZPL) spontaneous rate emission and \(\sum_{i=1}^{7} \Gamma_i = \Gamma_{SB}\) the sum on the phonon sidebands \[S6\]. When studying the forces around the ZPL at \(\omega_0\), the different sidebands \((\omega_i)\) are far detuned enough to be ignored (\(\min_i |\omega_0 - \omega_i| > 10\) nm). As a consequence, we used \(d_{ZPL} = d \cdot \sqrt{\Gamma_{ZPL}/\Gamma}\) for the dipole moment:

\[
d_{ZPL} = \sqrt{\frac{\Gamma_{ZPL} 3\pi \epsilon_0 c^3 \hbar}{n_1 \omega_0^3}}
\]

(S3)

Consequently, due to the phonon sidebands the dipole moment is effectively reduced. Also we used a branching ratio, or Debye-Waller factor, of 0.04 \[S4, S15\].

In addition to reducing the dipole moment, we also took into account the sidebands calculating the dipole force. We made the following approximations in order to maintain a simplified and analytical approach: (i) the different sidebands have been accounted for as one extra channel with a decay rate going as the sum of the various bands, \(\Gamma_{SB} = \sum_{i=1}^{7} \Gamma_i\), (ii) the non-radiative phonon decay rate is assumed to be similar for all bands and equal to \(\Gamma_{Ph}/(2\pi) = 38\)GHz \[S7\], and (iii) the sidebands cannot be efficiently coherently driven due
their very large dephasing ($\gtrsim 60$THz) and large detuning from the laser resonant with the ZPL. Under these approximations, the relevant terms from the optical Bloch equation can be explicitly written as:

\[
\frac{\partial}{\partial t} \rho_{ee} = i \frac{\Omega}{2} (\tilde{\rho}_{eg} - \tilde{\rho}_{ge}) - (\Gamma_{ZPL} + \Gamma_{SB}) \rho_{ee} \\
\frac{\partial}{\partial t} \rho_{gg} = -i \frac{\Omega}{2} (\tilde{\rho}_{eg} - \tilde{\rho}_{ge}) + \Gamma_{ZPL} \rho_{ee} + \Gamma_{ph} \rho_{pp} \\
\frac{\partial}{\partial t} \rho_{pp} = \Gamma_{SB} \rho_{ee} - \Gamma_{ph} \rho_{pp} \\
\frac{\partial}{\partial t} \tilde{\rho}_{eg} = - (\gamma - i \Delta) \tilde{\rho}_{eg} + i \frac{\Omega}{2} (\rho_{ee} - \rho_{gg}),
\]

where $g$, $e$ and $p$ are the ground state, the excited state and the phonon sideband respectively, $\Omega(r) = \sqrt{2/3} d_{ZPL} E_0(r)/\hbar$ is the Rabi frequency and $\Delta$ the laser detuning from the transition $\Delta = \omega - \omega_0$. The density matrix element $\tilde{\rho}$ is the element $\rho$ in the rotating frame, $\tilde{\rho} = e^{-i\omega_0 t} \rho$. The decay rate from the excited state to the phonon sideband $e \rightarrow p$ is $\Gamma_{SB}$, from the excited state to the ground state $e \rightarrow g$ is $\Gamma_{ZPL}$, and from the phonon sideband to the ground state $p \rightarrow g$ is $\Gamma_{ph}$. At room temperature, the total linedwith of the NV is much larger than the lifetime limited linewidth. This linewidth (or transverse decay rate) is $\gamma = (\Gamma_{ZPL} + \Gamma_{SB})/2 + \gamma_c$, where $\gamma_c$ accounts for additional coherence decay (inhomogeneous broadening). The value typically observed at room temperature is $\gamma/(2\pi) \sim 1$ THz [S8].

In the Heisenberg-picture, the time averaged value of the dipole force can then be written as:

\[
\langle F_{dip} \rangle = -\frac{\hbar \Omega^* (r) \langle \sigma \rangle}{2} \nabla \log |\Omega (r)| + c.c,
\]

where $\langle \sigma \rangle = \tilde{\rho}_{eg}$ is the steady state coherence as obtained by solving the steady state optical Bloch equation for $\tilde{\rho}_{eg}$, and $\Omega^*$ is the complex conjugate of $\Omega$. The force can then be explicitly written as:

\[
F_{dip} (r) = -\frac{\hbar \Delta}{\eta} \frac{\Gamma}{2} \frac{\nabla s (r)}{1 + s (r)},
\]

with $\eta = (2\Gamma_{ph} + \Gamma_{SB})/2\Gamma_{ph}$ and the saturation parameter $s (r) = \eta \Omega (r)^2 / [\Gamma \gamma (1 + \Delta^2 / \gamma^2)]$. The impact of the phonon sidebands contained in the factor $\eta$ is clearly negligible if the phonon decay rate $\Gamma_{ph}$ (phonon sideband $\rightarrow$ ground) is the fastest decay. Intuitively, when this decay is fast the phonon sidebands are never populated (i.e. $\rho_{pp} = \rho_{ee} \Gamma_{SB}/\Gamma_{ph} \ll \rho_{ee}$) and the system behaves as a typical two-level system obtained for $\eta = 1$. Conversely, if this decay is slow enough to maintain population in the phonon sidebands both the saturation
parameter and the force are impacted. In terms of optical potential one obtains:

\[ U_q(\omega, \omega_0, \mathbf{r}) = -\frac{1}{\eta} \frac{\hbar (\omega - \omega_0)}{2} \frac{\Gamma}{2\gamma} \log \left[ 1 + s(\mathbf{r}) \right] \]  

(S7)

**Trapping stiffness**

The fitting of the experimental data is done on the trapping stiffness rather than the optical potential depth. For this reason, the position dependence of the potential has to be given explicitly. Along the measurement direction \( x \), we approximated the electromagnetic field to a Gaussian profile \( E(x) = E_0 \exp(-x^2/w_0^2) \), with the field amplitude \( E_0 = \sqrt{4P/(\pi w_0^2 n_2 \epsilon_0 c)} \).

Here the waist of the focus is defined by \( w_0 \) (\( w_0=470 \) nm, measured at 640 nm), the incident power by \( P \) and the refractive index of the medium (water) by \( n_2 \).

The optical potential for the force acting on the nanodiamond dielectric matrix is then given by [S9]:

\[ U_{cl}(x) = -2\pi \epsilon_0 n_2^2 R^3 \left( \frac{m^2 - 1}{m^2 + 2} \right) E_0^2 \exp \left[ -\frac{2x^2}{w_0^2} \right], \]  

(S8)

where \( m = n_1/n_2 \) is the refractive index of diamond (\( n_1 \)) relative to the medium (water, \( n_2 \)). Similarly, the optical potential acting on the NV center can be explicitly given by:

\[ U_q(x) = \frac{\hbar (\omega - \omega_0)}{2\eta} \frac{\Gamma}{2\gamma} \log \left[ 1 + \eta \frac{2d_{ZPL} E_0^2}{3\hbar^2} \frac{1/(\Gamma\gamma)}{1 + (\omega - \omega_0)^2/\gamma^2} \exp \left( -\frac{2x^2}{w_0^2} \right) \right] \]  

(S9)

For the sake of simplicity, in the following we refer to the first potential (force) as the "classical" potential (force) and to the second one as "quantum" potential (force).

In the overdamped regime, the variance in the position of the trapped ND can be obtained through the equipartition theorem \( \Delta x = \sqrt{k_B T (2\pi f_c \beta)}^{-1} \), with \( \beta \) being the drag coefficient of the ND. This variance is estimated to be around 40 nm for a 150 nm ND and an average corner frequency of \( f_c \approx 400 \) Hz. Such a small displacement allows neglecting any anharmonic effects such as for example a Duffing non-linearity [S10]. In this case the optical potential seen by the ND can then be well approximated by a simple harmonic potential of the form \( U \sim U(0) + \kappa x^2 \) where \( \kappa \) is related to the potential through \( \kappa = \frac{1}{2} \partial^2 U(x)/\partial x^2 \bigg|_{x=0} \).

For the simple case of a nanodiamond containing \( N \) identical NVs with transition frequencies \( \omega_0 \), the two components of the stiffness can thus be written as:

\[ \kappa_{cl} = \frac{4\pi \epsilon_0 n_2 R^3}{w_0^2} \left( \frac{m^2 - 1}{m^2 + 2} \right) E_0^2 \]  

(S10)
\[ \kappa_q = -N \frac{\hbar (\omega - \omega_0)}{2\eta} \frac{\Gamma}{2\gamma w_0^2} \left( \frac{s_0(\omega_0, \omega)}{1 + s_0(\omega_0, \omega)} \right), \quad \text{(S11)} \]

with \( s_0(\omega_0, \omega) \) the saturation parameter taken at \( x = 0 \) given by:

\[ s_0(\omega_0, \omega) = \frac{\eta}{\Gamma \gamma} \frac{2d_{ZPL}^2 E_0^2}{3\hbar^2} \frac{1}{1 + (\omega - \omega_0)^2/\gamma^2} \quad \text{(S12)} \]

Note that the waist of the Gaussian beam, \( w_0 \), is also a (monotonic) function of wavelength.

### Cooperative effects

When calculating the dipole force from an ensemble of NVs such as the one present in the nanodiamonds used in the experiment, this force is much smaller than the one observed experimentally. Due to the unusually high density of NVs in these samples, it is necessary to account for cooperative effects in order to fully capture the dipole force from the NV ensemble. In addition, the small size of the nanodiamonds (150 nm) allows for a simple description without finite size effects. Consequently, we used a simple model based on the Dicke model [S11].

Due to the dephazing and the slightly different transition frequency of each NV center, only sub-populations of NVs are expected to act cooperatively. This amounts to “coarse-grain” the ensemble of \( N \) NV centers into cooperative sub-ensembles. Within one such sub-ensemble \( i \) containing \( N_{\text{Coop}}^i \) NVs, we assume the system to be invariant under permutation in order to apply the Dicke model. In this context, the state space in each sub-domain is spanned by Dicke states \( \left| J, M \right> \) with \( J = N_{\text{Coop}}^i/2 \). The maximal angular momentum space is appropriate because the spin begin in the ground state \( \left| J, -J \right> \). We also assume that the PSB does not impact the excited and ground populations (\( i.e. \eta \approx 1 \)). The raising and lowering operators for the NV defect \( k \) are defined respectively as:

\[ S_k^+ \equiv \left| e \right> \left< g \right| \quad S_k^- \equiv \left| g \right> \left< e \right| \quad \text{(S13)} \]

end the diagonal operator as:

\[ S_k^z \equiv \frac{1}{2} \left[ \left| e \right> \left< e \right| - \left| g \right> \left< g \right| \right] \quad \text{(S14)} \]

For notational clarity we suppress the sub-domain label \( i \) on the collective spin operators but it is to be understood that the collective spin operators appearing in the equations to follow for the forces and stiffness ratios carry such an index.
Let us now consider one sub-domain $i$ containing $N_{\text{Coop}}^i$ NVs with a transition $\omega_i$. Using the collective operators for the collective sub-domain $i$, $S^\pm \equiv \sum_{k=1}^{N_{\text{Coop}}^i} S_k^\pm$ and $S^z \equiv \sum_{k=1}^{N_{\text{Coop}}^i} S_k^z$, the symmetrical state can be obtained by repeated action of the lowering operator on the fully excited state:

$$|JM\rangle = \sqrt{(J+M)! / N_{\text{Coop}}^i (J-M)!} \cdot (S^-)^{J-M} |e, e, e \ldots e\rangle$$

with $-J \leq M \leq J = N_{\text{Coop}}^i/2$.

This $|JM\rangle$ state represents the fully symmetrical collective state with $J+M$ atoms in the excited state $|e\rangle$ and $J-M$ in the ground state $|g\rangle$. The Hilbert space dimension of this collective spin state is then $N_{\text{Coop}}^i + 1$.

We then consider this system to be driven by a laser at frequency $\omega$ according to the Hamiltonian $H$ given, in the the rotating wave approximation, by:

$$H = - (\omega - \omega_i) S^z + \frac{\Omega}{2} (S^+ + S^-)$$

where $\omega_0$ is the spin transition frequency and $\Omega$ the Rabi frequency. The Louivillian describing the coupling of this system to the laser and the environment is then:

$$\dot{\rho}(t) = -i \frac{\hbar}{\hbar} [H, \rho(t)]$$

$$- \frac{\gamma_{\text{bare}}}{2} (\{S^+ S^-, \rho(t)\} - 2 S^- \rho(t) S^+) + \frac{\gamma_{\text{collective}}}{2} (\{S^z S^z, \rho(t)\} - 2 S^z \rho(t) S^z)$$

where the decay rate $\gamma_{\text{bare}}$ is the bare single spin decay rate, and $\gamma_{\text{collective}}$ is a collective dephasing rate. Since the observed inhomogeneous broadening of NV centers at room temperature has been attributed to phonon processes [S8], this dephasing mechanism is believed to act on the collective state as it will affect all spins indiscernibly. As a consequence, we used the spontaneous decay rate of the NVs, $\Gamma$, as the single spin decay rate and inhomogeneous broadening, $\gamma_c$ as the collective dephasing. In this context, the dipole force is given by:

$$F = -\Re [\hbar \nabla \Omega \langle \Sigma^+ \rangle]$$

where $\langle \Sigma^+ \rangle = \text{Tr} [S^+ \rho^{SS}]$ is the expectation value of $S^+$ in the steady state of the collective spin state $\rho^{SS} = \lim_{t \to \infty} \rho(t)$.

We can numerically compute the reactive force by finding the steady state of the collection of spins as follows. First we vectorize Eq. S17 by writing the density operator as a single
vector of dimension \((N^i_{Coop} + 1)^2\): \(\rho \rightarrow \vec{\rho}\), which is done by stacking the rows of \(\rho\) on top of one another (such that the state \(|0\rangle\) has a one in the first component and zeros otherwise). The dynamical equation is then

\[ \dot{\vec{\rho}}(t) = A\vec{\rho}(t) \]

where the Louvillian in the vectorized basis is

\[ A = i(\omega - \omega_i)(S^z \otimes \mathbb{1} - \mathbb{1} \otimes S^z) + i\frac{D}{2}((S^- + S^+) \otimes \mathbb{1} - \mathbb{1} \otimes (S^- + S^+)) \]

\[ -\frac{\Gamma}{2}(S^+ S^- \otimes \mathbb{1} + \mathbb{1} \otimes S^+ S^- - 2S^- \otimes S^-) \]

\[ -\frac{\gamma_c}{2}((S^z)^2 \otimes \mathbb{1} + \mathbb{1} \otimes (S^z)^2 - 2S^z \otimes S^z). \]

where \(\mathbb{1}\) is the unit vector of dimension \(N^i_{Coop} + 1\). The steady state is contained in the null space of \(A\) which for this case is one dimensional since the dynamical algebra is irreducible.

We also verified that the positional variance of the nano-diamond in the trap, \(\Delta x \approx 40\) nm, does not influence the steady-state population. Under the harmonic trap approximation, the stiffness along \(x\) for the sub-domain \(i\) can be obtained numerically as:

\[ \kappa_i(\omega, \omega_i, N^i_{Coop}) = -\hbar \frac{\partial^2 \Omega}{\partial x^2} \bigg|_{x=0} \Re \left[ \langle \Sigma^{+} \rangle \right] \]

Using this method, we calculated the total “quantum” stiffness \(\kappa_q\) as the sum of the stiffness \(\kappa_i\) from the different collective domains:

\[ \kappa_q(\omega) = \sum_{i=-N^{Grains}/2}^{N^{Grains}/2} \kappa_i(\omega, \omega_i, N^i_{Coop}) \]

As the size of the vectorized steady state is \((N^i_{Coop} + 1)^2\), the numerical solution of the steady state populations becomes difficult for large sub domain size. Consequently, we calculated the force obtained with \(N_{Coop} \in [1, 80]\) and extrapolated the force for larger sub-domain using polynomial fits.

Note the collective dephasing preserves the total angular momentum because the environment that induces it has interactions which are permutation-symmetric. Local dephasing would couple the system out of this fixed angular momentum subspace, reducing the number of spins in the collective state over time. In the context of this work focusing on the force, the steady state of the whole system constitute the most important aspect. The underlying assumption is that under continuous drive and in conjunction with local dephasing, the cooperative sub-domains will approach a steady state of fixed mean size. The size of these
sub-domains was not determined theoretically, but rather used as the only fitting parameter to reproduce our experimental data.

To define the size of sub-domains, the distribution of NV transition frequencies within a given nanodiamond has been modelled as a normal distribution centered at \( \omega_0 \) with a standard deviation \( \sigma_{ZPL} \) (see Extended Figure 1). The coarse-graining then consists in dividing this distribution in sub-ensembles of spectral width \( \Gamma_{Grains} \). Within each of these ensembles \( i \) (\( i \in \mathbb{Z} \)) the NVs are assumed to form a collective state with a transition frequency \( \omega_i \) and a size \( N_{Coop}^i \) defined as:

\[
\begin{align*}
\omega_{\text{start}}^i &= (i - \frac{1}{2}) \Gamma_{Grains} + \omega_0 \\
\omega_{\text{end}}^i &= (i + \frac{1}{2}) \Gamma_{Grains} + \omega_0 \\
N_{Coop}^i &= \int_{\omega_{\text{start}}^i}^{\omega_{\text{end}}^i} \frac{d\omega}{\sigma_{ZPL} \sqrt{2\pi}} \exp \left[ -\frac{(\omega_0 - \omega)^2}{2\sigma_{ZPL}^2} \right] \\
\omega_i &= \omega_0 + i \Gamma_{Grains}
\end{align*}
\]  

(S22)

As the normal distribution of NVs in the nanodiamonds has a finite standard deviation \( \sigma_{ZPL} \), only the sub-ensembles \( i \) such that \( \omega_i \in [\omega_0 - 4\sigma_{ZPL}, \omega_0 + 4\sigma_{ZPL}] \) are considered. Using this \( 4\sigma_{ZPL} \) interval allows one to account for 99.993\% of the NV population, it is also then possible to define the number of sub-ensembles \( N_{Grains} = 8\sigma_{ZPL}/\Gamma_{Grains} \). It is the spectral width of these sub-domains, \( \Gamma_{Grains} \), or equivalently the number sub-domains \( N_{Grains} \), that we used as the sole fitting parameter used to reproduce our experimental data.

**Stiffness ratio**

In the harmonic approximation, the measurement at a given wavelength yields \( \kappa_{\text{tot}}(\lambda) = \kappa_{\text{NVs}}(\lambda) + \kappa_{\text{Diamond}}(\lambda) \), where \( \kappa_{\text{NVs}} \) is the stiffness related to the NV centers and \( \kappa_{\text{Diamond}} \) the stiffness related to the diamond matrix. By normalizing with the reference wavelength, we obtain the following ratio:

\[
\text{Ratio}(\lambda) = \frac{\kappa_{\text{NVs}}(\lambda) + \kappa_{\text{Diamond}}(\lambda)}{\kappa_{\text{NVs}}(\lambda_{\text{ref}}) + \kappa_{\text{Diamond}}(\lambda_{\text{ref}})}
\]  

(S23)
In the case of the low NV-density NDs, the impact of the NVs on the total force force is neglected. The difference between the two ratios consequently gives:

$$\Xi(\lambda) = \left( \frac{\kappa_{NVs}(\lambda) + \kappa_{Diamond}(\lambda)}{\kappa_{NVs}(\lambda_{ref}) + \kappa_{Diamond}(\lambda_{ref})} \right)_{highNV} - \left( \frac{\kappa_{Diamond}(\lambda)}{\kappa_{Diamond}(\lambda_{ref})} \right)_{lowNV}$$

(S24)

The choice of the reference wavelength \(\lambda_{ref}\) at the average ZPL was such that the contribution of the NVs on the force is negligible (i.e. \(\kappa_{NVs}(\lambda_{ref}) + \kappa_{Diamond}(\lambda_{ref}) \approx \kappa_{Diamond}(\lambda_{ref})\)). With this approximation, the difference of ratios between high and low NV-density NDs simply yields the ratio of the stiffness from the NV centers to the stiffness from the diamond matrix at \(\lambda_{ref}\):

$$\Xi(\lambda) \approx \frac{\kappa_{NVs}(\lambda)}{\kappa_{Diamond}(\lambda_{ref})}$$

(S25)

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