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Cooperative effects between color centers in diamond: applications to optical tweezers and optomechanics

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ABSTRACT

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] Interestingly, both an atom and a lump of dielectric material can be manipulated through the same mechanism: the interaction energy of a dipole and the electric field of the laser light. In the case of atom trapping, the dominant contribution typically comes from the allowed optical transition closest to the laser wavelength while it is given by the bulk polarisability for mesoscopic particles. This difference lead to 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. In this context, solid state artificial atoms present the interesting opportunity to combine these two aspects of optical manipulation. We are particularly interested in nanodiamonds as they constitute a bulk dielectric object by themselves, but also contain artificial atoms such as nitrogen-vacancy (NV) or silicon-vacancy (SiV) colour centers. With this system, both regimes of optical trapping can be observed at the same time even at room temperature. In this work, we demonstrate that the resonant force from the optical transition of NV centres at 637 nm can be measured in a nanodiamond trapped in water. This additional contribution to the total force is significant, reaching up to 10%. In addition, due to the very large density of NV centres in a sub-wavelength crystal, collective effects between centres have an important effect on the magnitude of the resonant force.[6] The possibility to observe such cooperatively enhanced optical force at room temperature is also theoretically confirmed.[7] This approach may enable the study of cooperativity in various nanoscale solid-state systems and the use of atomic physics techniques in the field of nano-manipulation and opto-mechanics.

Keywords: Optomechanics, cooperativity, colour centers, optical force, near-field levitation

1. INTRODUCTION

The possibility to manipulate single atoms with light is central to many cold-atom experiments. Their sharp electronic transitions lead to poles in the complex atomic polarizability that enables for detuning-dependent optical forces. In particular, for the dipole force (or reactive force) the sign of the detuning determines whether the force is repulsive or attractive (*i.e.* low-field or high-field seeker respectively). Using this dependence, complex optical near-field traps for cold atoms have been demonstrated.[8] Introducing detuning-dependent optical manipulation techniques in the field of nano-manipulation by embedding artificial atoms into the nano-objects would therefore open up a new range of possibilities. While resonant forces have been studied for micro-/nanoparticles exhibiting plasmonic resonances or Mie resonances, they are usually specatrally wide (tens

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of nanometres).[9] Alternatively, to obtain sharp Mie resonances,[10] large particles are required along with either near-field coupling or complex propagating beams[11] to efficiently excite them. Our approach is more directly inspired from cold-atom physics and relies on using embedded artificial atoms in the nanoparticle. In particular, we choose to use nanodiamonds as they provide good optical quality and contain colour centres such as nitrogen-vacancy (NV) centres.[12] Such artificial atoms provide spectrally sharp resonances, even at room temperature. In the context of levitated optomechanics,[13,14] these resonant forces could enable novel cooling techniques such as Doppler cooling. In order to apply these methods from cold-atoms physics to nanoparticles, one important effect needs to be taken into account: the coupling to the environment. This coupling typically constitutes an important decoherence mechanism leading to reduced optical forces and is indeed present in our experiment. Yet, we show that this detrimental effect is compensated by cooperative effects between the embedded artificial atoms. While cooperativity has been theoretically studied in the context of optical forces,[15,16] experimental evidence has been limited to dissipative forces.[17,18] We show in our experiment that the cooperative effects enhance the dipole force by more than one order of magnitude compared with the force that would be obtained considering independent artificial atoms.

2. EXPERIMENTAL RESULTS

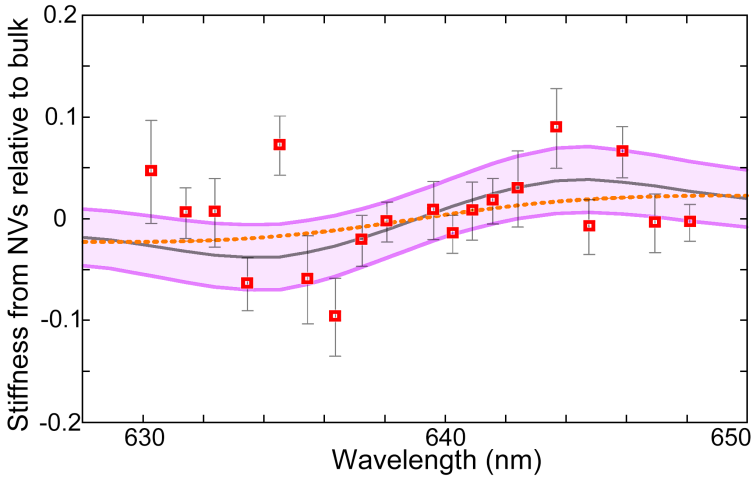

Figure 1. Trap stiffness related to the NV centres. The difference from the measurements of two ND samples (low and high NV centres densities) provides the ratio of the stiffness component form the NV centres to the stiffness form the bare ND (solid squares with the standard error). The black curve shows the theoretically predicted stiffness ratio accounting for cooperative effects. For comparison, the dashed line showing the theoretical prediction for independent NVs was magnified 40 times to reach comparable magnitude. Ou Monte Carlo model was used to obtain the 1σ interval where 68.2% of the experimental value should lie.

In our experiment, we create a Gaussian standing wave (GSW) trap[19] near 639 nm by focussing a Gaussian laser beam on a silver-coated mirror. The GSW provides a stronger trap along the direction of the standing wave compared to a conventional focused Gaussian beam, and also allows to neglect scattering forces. The mirror forms the top of a static micro-fluidic chamber that contains the nanodiamonds (NDs) suspended in deionized water. The laser sources are a set of laser diodes operating at different wavelengths detuned with respect to 639 nm. In order to characterise the effect of the NV centres on the optical force, we measure the trap stiffness as a function of the incident wavelength (see details in Ref.[6]). Since the measurement of the trap stiffness gives both the component from the ND and from the NVs, it is necessary to use a reference. For this, we measured two ND samples: NDs containing a large number of NV centres ($\langle NV \rangle \approx 9500$) and a size of (150 ± 23) nm, and NDs containing only a few NV centres with a similar size, (168 ± 31) nm. By obtaining the stiffness as a function of the wavelength for these two ND samples, it is possible to extract the stiffness arising from the NV ensemble normalised to the stiffness from the bare nanodiamond matrix (see Fig. 1). The dispersive trend expected for

atom trapping is visible with stiffness ratio lower than 0 at wavelengths below the NV transition and larger than 0 for wavelengths above, with amagnitude of up to 10%. Most remarkably, these experimental results cannot be accounted for only by assuming independent NVs: the dashed line in Fig. 1, displaying the expected trap stiffness assuming independent NV centres, was magnified 40 times to be comparable in magnitude with the experimental values. To fully capture the experimental results, we included cooperative effects[20] between the NV centres within a single nanocrystal. The presence of such cooperative effects was also confirmed in lifetime measurements.[21]

3. THEORY OF COOPERATIVELY ENHANCED DIPOLE FORE

Cooperative effects between NV centres are expected to have a significant impact on the force by modifying both the spontaneous decay rate and the steady-state population. Due to the large variation of the optical properties of each NV centre within one given ND, only sub-domains (*i.e.* smaller ensembles) of NVs are expected to act cooperatively. To theoretically describe the cooperatively enhanced forces, we apply the Dicke model[22] to each of these cooperative sub-domains. For this first model we assume the emission to be perfectly collective, the dephasing (inevitable at room temperature) to be collective, and the dipole-dipole was not taken into account as it would require knowledge of the relative position and orientation of the NV centres. The results from this model, presented in Fig. 1, is in good agreement with the experiment when using sub-domain size corresponding to 95 NV centres in average (or 1% of the expected total number of NV centres).

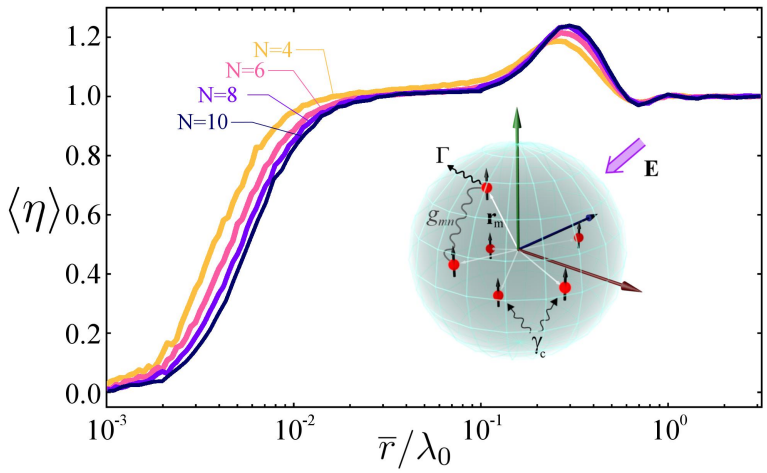

Figure 2. Theoretical prediction for the enhancement of the dipole force due to cooperative effects. The ratio η is defined as the ratio the force obtained form the TLE ensemble taking into account collective effects with the force obtained for the same ensemble considering the emitters to be independent. The detuning of the drive relative to the TLE transition is taken such that the force the independent TLE ensemble is maximum. We consider between 4 and 10 TLEs distributed randomly (with the same dipole orientation) in background matrix as a function of the average separation between the emitters. The values of the enhancement are the mean over 1000 random distribution of the TLEs.

To confirms these results, we also developed a more refined model for cooperatively enhanced forces.[7] For this model we consider a closely packed ensemble of two-level emitters (TLEs) subjected to strong coherent drive and dephasing. The TLEs are positioned randomly in a background matrix and driven by a classical electromagnetic field. Apart from the dynamics induced by the interaction with the external drive, we assume the TLEs to experience collective dephasing as well as dipole-dipole interaction and spontaneous emission due to the interaction with free electromagnetic field modes in the vacuum state (more details can be found in Ref.[7]). For parameter comparable with the experiment using NDs, we show that the dipole force on such an ensemble can be significantly larger than on an equivalent one where each TLE spontaneously emits independently (see Fig. 2). For the emitter separations that we consider here, the dipole-dipole interaction can be larger than the linewidth of the individual emitters and the spontaneous emission is not perfectly collective. In this situation,

one typically expects cooperative effects to be suppressed. Remarkably, cooperative effects can be restored by a combination of strong driving and large collective dephasing even in the presence of dipole interaction shifts and non-collective spontaneous emission.

4. CONCLUSION

To conclude, our observations open the door to a wealth of new research directions. As shown both experimentally and theoretically, cooperativity provides a mechanism allowing to significantly increase optical forces. While such effect had already been studied, a more surprising feature reside in the role played by collective dephasing in such solid states systems. In diamond, the underlying mechanism for the large dephasing at room temperature is mediated via phonon interactions[23] and consequently changes rapidly with the temperature of the lattice. Since the presence of large collective dephasing plays an important in restoring cooperative effects, this raises the tantalising prospect of repeating optical force or lifetime measurements at lower temperatures. At lower temperature, the dephasing is expected to be reduced, ultimately leading to a reduction of the collective effects. This effect would be counter-intuitive to the study of collective effects in atomic systems where temperature has to be low. In connection to the proposal for levitated optomechanics with nano-diamonds[24] it is interesting to explore if collective effects in dense ensembles lead to polarizabilities comparable or greater than the bulk polarizability of the embedding medium. A promising direction for further research is to explore other scenarios where large collective dephasing restores cooperative effects. Remarkably, in systems such as superconducting qubits where the collective dephasing can be externally controlled, this could allow to observe cooperative effects even in the presence of inhomogeneities and dipole shifts.

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