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OPTICAL DIPOLE FORCES

Working together

Strength lies in numbers and in teamwork: tens of thousands of artificial atoms tightly packed in a nanodiamond act cooperatively, enhancing the optical trapping forces beyond the expected classical bulk polarizability contribution.

Clarice D. Aiello

Conservative optical dipole forces — used to levitate and confine particles and atoms in three dimensions — arise when dipoles interact with a gradient of electric field intensity such as that in a standing wave of laser light. These forces can be well described either by classical electrodynamics or by quantum physics. In the classical picture, depending on the refractive index of the material, a particle is attracted to — or repelled from — regions with large electric field gradients¹. Similarly, in the quantum mechanical description, light that is red- or blue-detuned with respect to an atomic transition will push an isolated atom towards or away from regions with large electric field gradients. Macroscopically, the magnitude of the optical dipole force depends on the bulk polarizability, which measures how easily internal charges in the material are displaced by an electric field. Microscopically, in the quantum case, the relevant property is the polarizability of the atom. Writing in *Nature Physics*², Mathieu Juan and co-workers show that, contingent on the presence of a sufficiently high density of artificial atoms inside a solid, quantum effects need to be taken into account to explain the magnitude of the macroscopic optical dipole force acting on the bulk.

Juan *et al.* use a laser standing wave to trap several different nanodiamonds, each containing approximately 10,000 nitrogen–vacancy (NV) centres (Fig. 1). NV centres are defects arising when a nitrogen substitutional impurity is adjacent to a carbon lattice vacancy. Even at room temperature, they are stable and their fluorescence is robust. Because the ensemble of NV centres is tightly packed in a messy environment (full of local lattice imperfections), it exhibits inhomogeneous broadening of its spectroscopic lines. Measuring how strongly each nanodiamond is confined — in other words, the optical trap stiffness — the authors can compare the magnitude of the optical dipole force for laser frequencies above and below the NV centres energetic transition. Statistically, the trapping force decreases noticeably

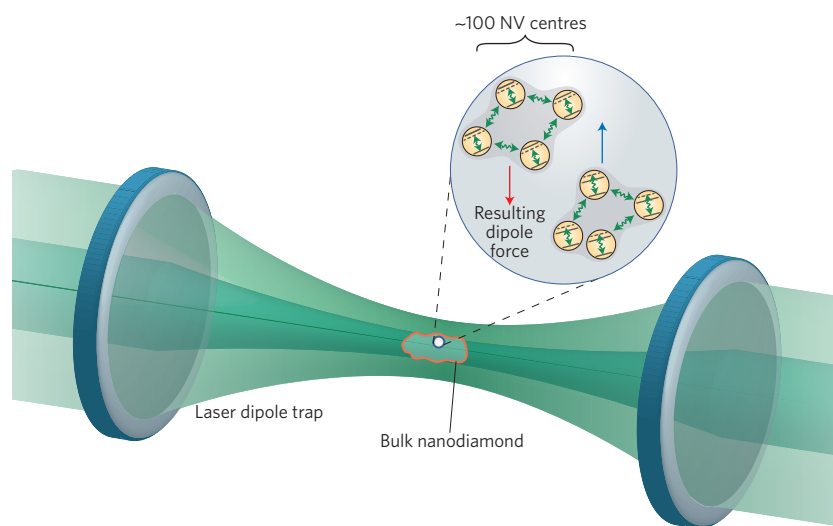


Figure 1 | Quantum effects can alter the dipole forces in the bulk if the embedded nanoemitters are close enough to act cooperatively. Using a laser, the authors trap nanodiamonds containing thousands of artificial emitters known as NV centres. The trapping force acting on the bulk material is altered by a quantum mechanical effect whereby sub-ensembles of NV centres act cooperatively as a ‘superatom’. As depicted in the magnified region, this superatom interacts with the laser dipole trap in the same way that an isolated atom would: when near-resonantly excited with red-detuned light, it experiences a force towards regions of stronger electric field gradients — that is, towards the trap. The inverse is true for blue-detuned light. NV centres are depicted as two-level systems, each with a slightly different energetic transition. Green arrows represent the laser dipole trap photons.

when sweeping the laser frequency from red-detuned to blue-detuned. The classical contribution to this change in the dipole force strength is, however, negligible, because the diamond index of refraction only changes by less than 0.1% for the probed wavelengths. So quantum effects might be playing a role: each of the nanoemitters could be reacting to the laser field in the same way as an isolated atom would. In other words, NV centres excited with blue-detuned light, for example, would experience a force pointing away from the trap centre, which would decrease, even if only slightly, the overall trap stiffness. Yet, such an effect for isolated NV centres should be almost 50 times weaker than the measured one. What’s going on?

It has been known since Dicke’s paper on super-radiance³ that cooperative effects arise

when spectrally indistinguishable two-level systems are near-resonantly excited, while confined to length scales much shorter than their transition wavelengths. Intuitively, any excitation photon must be ‘shared’ since its wavelength is much larger than the mean distance between the spatially indistinguishable emitters. Alternatively, one can depict the emitters working as a giant, collective dipolar antenna, radiating together in phase with the driving field, and thereby exchanging excitations among themselves.

Cooperative phenomena are weakened for two-level systems that are not spectrally indistinguishable, even if they are spatially indistinguishable. Nevertheless, Juan and colleagues show that a theoretical model considering quantum cooperative interactions among the NV centres is consistent with their experimental data. As a

result of the inhomogeneous broadening in the bulk, the cooperation is limited to small sub-ensembles of emitters (on the order of hundreds). Each of these sub-ensembles functions as a 'superatom' interacting with the electric field gradient of the laser, sufficiently strongly to modify the trapping force on their host crystal.

The effect measured by Juan and colleagues is not exactly super-radiance, despite the fact that the system is described by the same collective (Dicke) eigenstates that are necessary — but not sufficient — to sustain super-radiance. Super-radiance in the solid state — as it has previously been observed in quantum dots⁴ — would entail

confirming two things. First, both the dipole force enhancement and the intensity of emitted radiation upon a pulsed excitation should have a quadratic scaling with the number of cooperating nanoemitters. Second, the collective quantum state should be resilient to global environmental effects, such as interactions with lattice phonons.

These results suggest that the quantum properties of artificial atoms could drive the development of new laser-cooling and trapping techniques for macroscopic objects. The technology to engineer such quantum properties is already within reach. Moreover, understanding cooperative quantum behaviour could be relevant for

more complex phenomena — for instance, excitation transfer in photosynthetic light-harvesting molecules may rely on such super-radiant effects⁵. □

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PHOTOEMISSION DELAY

The White Rabbit's clock

Without a very precise timer one can never catch up with the electron released in photoemission. Attosecond streaking spectroscopy allows such a chronometer clock to be set to zero and reveals the role of electron correlations.

Francesca Calegari

Photoemission is not an instantaneous process. Following the absorption of a sufficiently high-energy photon, the electron can leave the atom only after a few tens of attoseconds¹. Why does the electron hesitate before leaving the atom? The first reason is the interaction of the ionized electronic wave packet with the ionic potential. This can be seen as a half-scattering mechanism leading to a phase shift of the wave packet, which ultimately translates to a time delay². The second reason is the influence of the remaining electrons — the so-called electronic correlation. For all quantum mechanical systems except the hydrogen atom — the only one-electron system — photoemission must be modelled as a many-body process because the action of the fellow electrons on the ionized one is not negligible. And since quantum effects are never obvious, this action can result in either the retardation or advance of the electronic wave packet.

Recall the White Rabbit in *Alice in Wonderland*: constantly checking his clock in a race against time. Similarly, in photoemission delay experiments, without a sufficiently precise clock we can never be in time to catch the released electron. Now, writing in *Nature Physics*, Marcus Ossiander and co-workers³ show how to set the absolute zero of time in photoemission with sub-attosecond accuracy, and reveal the presence of a correlation delay in helium,

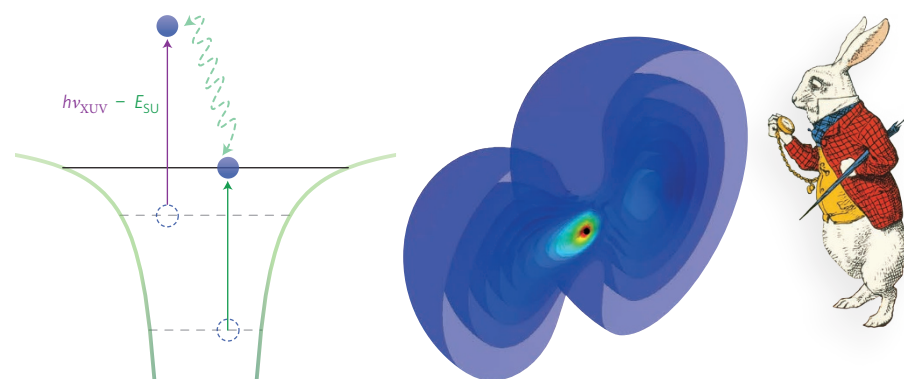


Figure 1 | Correlation delay in photoemission. Upon the absorption of an XUV photon with energy $h\nu_{\text{XUV}}$ one electron is emitted, a process accompanied by the excitation of the second electron in a shake-up (SU) state. Due to electronic correlation (symbolically represented by the light green double arrow), the kinetic energy of the electron is reduced by an amount equal to the excitation energy E_{SU} . The asymmetric charge density distribution of the helium ionic state retrieved from the measurement of the correlation delay (measured as six attoseconds on the White Rabbit's clock) is shown on the right. Reproduced from ref. 3, NPG. White Rabbit illustration © Granger Historical Picture Archive / Alamy Stock Photo.

from which crucial information about the ionized system can be obtained (Fig. 1).

The presence of only two electrons makes the helium atom the ideal test system for studying electron correlations. When the helium atom absorbs photons with the energy above 24.6 eV, one electron is released with kinetic energy corresponding to the difference between the photon energy and the ionization potential of the

atom (direct photoemission). However, if the photon energy exceeds 65.5 eV, the electron emission can be accompanied by the excitation of the remaining electron into ionic states with higher energy (shake-up) or even a second ionization event (shake-off). Shake-up and shake-off lend themselves well to investigations of correlation delay thanks to the presence of a strong electron–electron interaction.