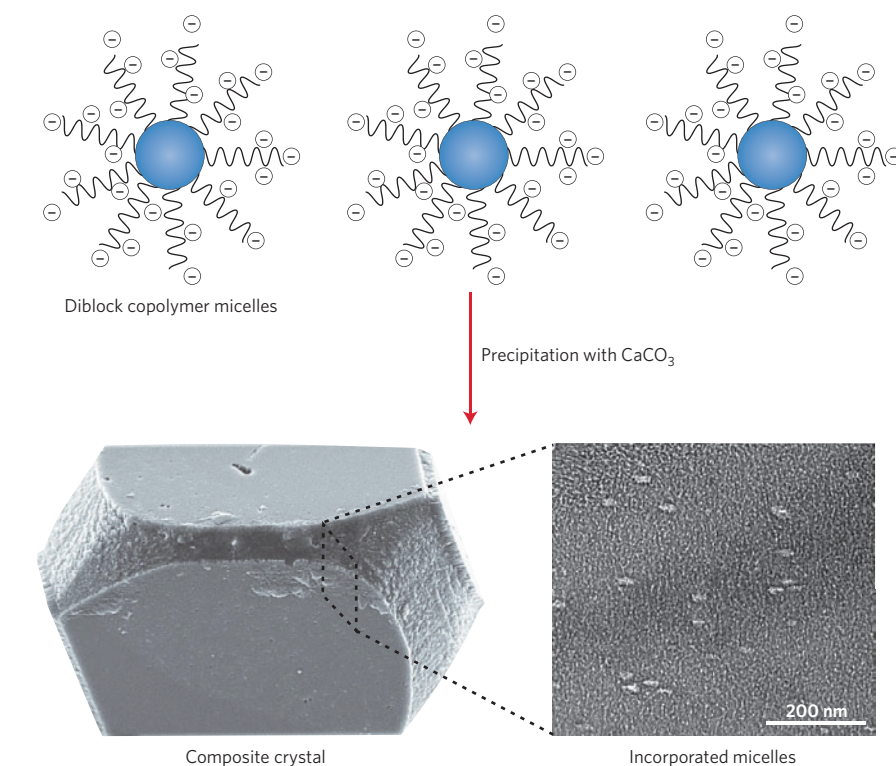


the underlying mechanisms leading to the observed morphology, including various instabilities in the crystal growth front when macromolecules bind to step edges<sup>10</sup>. However, as the size of the additive increases beyond the dimensions of the unit cell of the host crystal, the role of surface chemistry seems to diminish and incorporation becomes significantly less dependent on the specific crystal lattice. For example, when either carboxylate-functionalized polymer particles (200–250 nm) or neutral agarose hydrogel fibres (10–20 nm) are trapped within single crystals, incorporation is random along the crystallographic directions<sup>2–4</sup>. In the context of those studies, the inclusion of 20-nm anionic micelles achieved by Kim and colleagues<sup>8</sup> is surprising. Despite the fact that they are similar in size to the hydrogel fibres and in surface chemistry to the polymer particles, the micelles selectively adsorb on specific crystal facets. This result shows that the right combination of surface chemistry, size and rigidity can lead to the oriented incorporation of objects larger than previously thought possible. However, further understanding of the interplay of the relevant factors involved in the mechanism of incorporation, including the kinetic competition between crystal growth and adsorption of organic material<sup>10</sup>, is needed to determine design criteria for the pairing of host crystals with occlusion materials that achieve a desired degree of inclusion and crystallographic selectivity.

In the future, bio-inspired synthetic approaches such as that of Kim and co-authors<sup>8</sup> could be used to tune the mechanical properties of inherently brittle inorganic single crystals and to obtain a better understanding of the microscopic origin of the enhanced mechanical properties of the organic–inorganic composites. We foresee the application of synthetic



**Figure 1** | Inclusive crystallization of micelles within calcite single crystals<sup>8</sup>. When calcite crystals are grown in the presence of anionic diblock copolymer micelles, the micelles become trapped within the final structure. On incorporation, the originally spherical micelles become ovoid (transmission electron microscopy image, bottom right) and align along a specific crystallographic direction.

approaches to a variety of host crystals and occlusion materials with electronic, photonic or magnetic properties, eventually leading to functional, nanostructured, single-crystal components.

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## QUANTUM INFORMATION

# Noisy neighbours under control

The ability to control the nuclear spins in a semiconductor quantum dot is an important step towards a long-lived and controllable electron spin qubit.

Guido Burkard

The solid state has been the arena for numerous demonstrations of quantum mechanical effects. Lately, coherent control of single quantum systems has been achieved in a variety of systems, including single atoms and photons, as well

as solid-state systems. One example of a two-state quantum system is the spin of an electron, which constitutes an interesting realization of a quantum bit (qubit)<sup>1</sup>. In solid state, the interaction between two qubits can typically be made strong rather

easily, which is a big advantage if one aims to construct a quantum information processor. The disadvantage, however, is that solid-state qubits tend to interact with a high number of other particles in their environment, which essentially results in

noise. For example, the above-mentioned electron spin qubits are affected by the noise created by the interaction with surrounding nuclear spins of the atoms in the solid. This noise can effectively destroy the quantum coherence of the electron spin, which is essential for any quantum information protocol. Various ways to increase the coherence by acting directly on the electron spin and its interaction have been explored in the past. Now, writing in *Nature Materials*<sup>2</sup>, Maxim Makhonin and colleagues demonstrate a different route whereby they directly control the nuclear spins in a semiconductor quantum dot.

Nuclear spins are harder to detect and manipulate than electron spins because their magnetic moment is almost 2,000 times smaller. The magnetic moment determines the strength of the coupling with an external magnetic field. Despite this, nuclear magnetic resonance (NMR) is a vastly successful method to access the nuclear spins, with many applications reaching from materials science to medicine. Interestingly, as the spin of the atomic nucleus obeys quantum laws, it has been considered an efficient qubit itself, for example it was used in NMR-based quantum computation experiments<sup>3</sup>.

Usually, the number of nuclear spins addressed with NMR is macroscopic, that is, on the order of  $10^{23}$ . However, NMR in much smaller samples has been detected in semiconductor quantum wells<sup>4</sup> and quantum dots<sup>5</sup> down to  $10^8$ , whereas in colour centres in diamond<sup>6</sup> and using magnetic resonance force microscopy<sup>7</sup> it is even possible to detect a single nuclear spin.

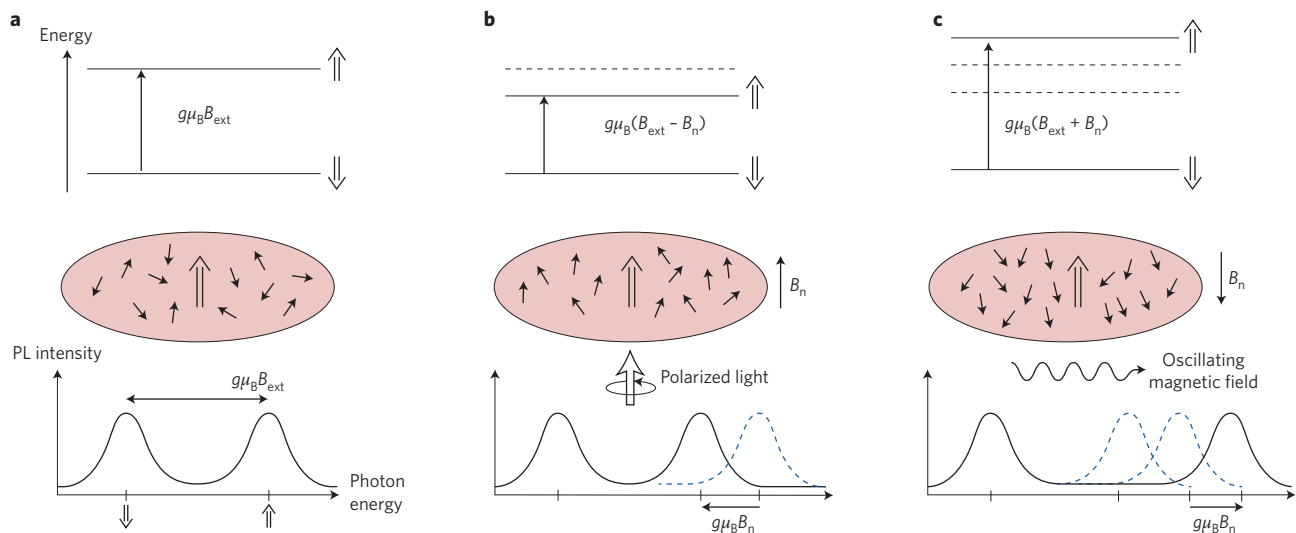
This allows for the use of the nuclear spin of a nitrogen-vacancy centre in diamond as a qubit memory<sup>8</sup>. Because the magnetic signal of a small number of nuclear spins is too faint for direct magnetic detection, alternative detection schemes need to be employed. Makhonin *et al.* use optically detected NMR where the energy shift of an electron interacting with  $10^5$ – $10^6$  nuclear spins present in a quantum dot is detected in the photoluminescence (PL) signal.

Obtaining control over the nuclear environment in a semiconductor quantum dot is important because the nuclear spins play a pivotal role for the quantum phase coherence of the spin of an electron located in the dot. The nuclear spins play the role of a fluctuating magnetic field, in which the electron spin loses its phase coherence. If no action on the environment of the spin qubit is taken in an unprepared sample, this decoherence process takes place on a rather fast timescale of ten nanoseconds, but it has been possible to extend that time beyond one microsecond using Hahn spin echo<sup>9</sup> and more recently to hundreds of microseconds with more elaborate pulse sequences<sup>10</sup>. This form of control is applied to the electron spin, that is, the system that one attempts to protect from decoherence.

The experiment by Makhonin and co-workers is different: here, the nuclear spin environment itself is being controlled directly. Control of the environment instead of the qubit has recently been used to decouple a defect spin in diamond from its environment consisting of electron spins<sup>11</sup>. Compared with such electron spin resonance, the NMR rotations in a

semiconductor quantum dot produced by Makhonin *et al.* are much slower. However, the effective nuclear magnetic field produced by the nuclear spins was built up to about half a Tesla by partially polarizing the nuclear spins optically. This magnetic field could then be rotated by  $180^\circ$  within  $15 \mu\text{s}$ , something that would be very hard if not impossible to achieve by reversing a magnetic field produced in an external coil.

In the experiment<sup>2</sup>, the controlled rotation of the nuclear spins and their detection were achieved as follows. An external magnetic field splits the energy of the spin-up and spin-down states of an electron sitting in the quantum dot (Fig. 1a). This Zeeman splitting can be detected optically by observing two distinct peaks in the intensity of the light emitted from the quantum dot under continuous laser excitation (PL). By using properly polarized light it was possible to orient the electron spins so that through the hyperfine interaction some of the nuclear spins would be aligned opposite to the external field (Fig. 1b). This nuclear polarization is seen by the electron spin as a magnetic field that is subtracted from the externally applied field, therefore decreasing the splitting observed in the PL. In a second step, a small oscillating magnetic field is applied perpendicular to the constant field for a short time (Fig. 1c). If the frequency of this oscillating field matches the energy splitting of the nuclear spin in the external field (divided by Planck's constant), NMR occurs, and the nuclear spins are rotated from antiparallel to parallel to the external field. After the rotation, the nuclear field adds



**Figure 1** | Electron energy levels (top), nuclear spin configuration in the quantum dot (centre) and PL intensity (bottom). These are shown schematically **a**, before nuclear spin polarization, **b**, after nuclear polarization with a circularly polarized laser and **c**, after nuclear spin rotation using NMR with an oscillating magnetic field. The symbols  $B_{\text{ext}}$ ,  $B_n$ ,  $g$  and  $\mu_B$  denote the external and nuclear magnetic fields, the electron  $g$ -factor and the Bohr magneton. The double arrows denote the electron spins and the single arrows the nuclear spins.

to the external field and, instead of being reduced, the splitting of the two lines in the PL is now increased. In fact, Makhonin *et al.* could observe the nuclear spin while rotating coherently by varying the length of the NMR pulse. By further controlling the phase of the oscillating field, rotations about different axes were achieved, thus allowing full control of the nuclear spin.

The reconfigurable and strong local field that was achieved is useful for performing quantum gate operations for quantum computation<sup>12</sup>. The rotations of the collective nuclear spin can be carried out on timescales shorter than the nuclear decoherence time and therefore in a coherent manner. Thus, one can

foresee the creation of non-classical states of the nuclear environment<sup>13</sup>, as well as entanglement between an electron spin and an entire collection of nuclear spins. Of course, once these goals are reached, the nuclear spins will have been promoted from mere environment to part of the actual system. Finally, whereas a single nuclear spin is clearly a microscopic object ruled by quantum mechanics, a collection of a million nuclear spins lies somewhere between the known quantum and classical regimes. Hence, controlling these nuclear spins coherently and observing their behaviour may also help our fundamental understanding of the boundary between the quantum and classical worlds. □

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## X-RAY IMAGING

# A coherent look at stress

Molecular ligands are widely used to functionalize gold nanoparticles, but their influence on the particle structure has been difficult to probe. Coherent X-ray diffraction has now reached sufficient sensitivity to resolve adsorption-induced near-surface stress in a single nanocrystal.

Frank Schreiber

Gold nanoparticles have already been used — unconsciously, of course — in ancient times. One of their first applications in coloured glass exploited their optical properties, which show a change of apparent colour from gold to red as their size is reduced down to the nanometre scale. Recently, interest in these materials has been exploding, as, although occasionally overestimated or mystified, the opportunities they offer are indeed breathtaking. Possible applications of gold nanoparticles (AuNPs) and gold nanostructures in general range from their use in sensors to exploiting near-field optical effects, such as antenna effects for field enhancement<sup>1</sup> or microscopy beyond the diffraction limit<sup>2</sup>.

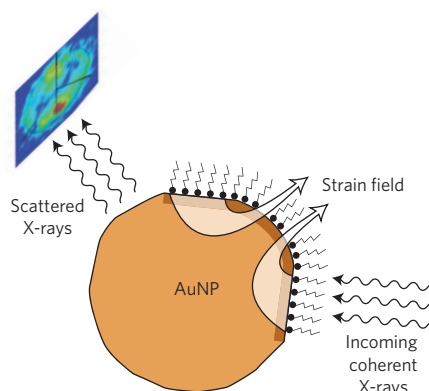
In many cases, in particular if the particles are to be dissolved or processed without aggregating, or if a specific surface functionality is desired, they are coated with an organic layer<sup>3</sup>. For AuNPs, the use of thiols, which incorporate sulphur as a linking group, is the most common and most successful strategy for functionalization. For extended gold surfaces (bulk crystals or thin films) this scheme has been employed in a vast number of studies<sup>4</sup>. By virtue of the thiol anchor, a broad variety of functionalities can be attached to a gold surface to render it sticky or non-sticky, hydrophilic or hydrophobic, to give just a few examples.

Although this recipe has been employed widely, the structure of many AuNPs and their magic coatings has remained surprisingly elusive even today. Writing in *Nature Materials*, Moyu Watari *et al.* now report that they have successfully resolved the structure and the surface strain in a single AuNP using coherent X-ray scattering<sup>5</sup>.

When it comes to the crystallographic characterization of bulk crystals and their surfaces, which, interestingly, Watari's

co-author Ian Robinson pioneered among others, we are spoiled by the possibility to almost routinely solve such questions with a mouse click, provided the materials are sufficiently well defined and extended<sup>6</sup>. However, some AuNPs are not crystalline at all, and even for those that are it is not obvious how conventional X-ray diffraction techniques would reveal this structure, let alone subtleties such as surface strain. A conventional diffraction approach on an ensemble of unaligned AuNPs would face difficulties to resolve the issues addressed by Watari *et al.*, and it lacks the required surface sensitivity for these systems.

The approach developed by Watari *et al.* circumvents part of the classical phase problem of crystallography. In a previous project the authors studied already bare, that is, uncoated, NPs using coherent X-ray scattering<sup>7</sup>. This technique has become possible with the development of modern highly coherent X-ray sources, that is, synchrotrons, and now also X-ray lasers. The idea is that if the sample is bathed in a coherent beam of X-rays, the usual (incoherent) averaging is avoided, and more information, including the relative scattering phase of rather distant regions within the sample, can be extracted. In essence, this is the result of the oversampling of the diffraction pattern, which can be further processed and inverted to obtain



**Figure 1** | Schematic of the experiment by Watari *et al.*<sup>5</sup>. The particle is bathed in a coherent beam and the relative scattering phase of rather distant regions within the sample is measured. The results can be analysed to obtain an extremely detailed real-space picture, including surface strain.