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Probing magnetic 2D crystals with quantum sensors

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Ferromagnetic two dimensional (2D) crystals are a new addition to the fascinating world of 2D crystals. An absolute measurement of their magnetic moment has been impossible so far, as their micron scale areas and atomic scale thickness lead to a total magnetic moment that goes under the radar of conventional magnetometry. Taking advantage of the outstanding detection performance of quantum sensors based on NV center scanning magnetometry [1], Thiel *et al.* [2] have mapped the magnetization of CrI_3, a recently discovered ferromagnetic 2D crystal [3] , with a resolution of a few tens of nm, and have provided very important insight on the nature of its anomalous interlayer spin interactions.

The discovery of graphene [4], back in 2004, created a new paradigm in Materials Science: atomically thin 2D crystals can are obtained by mechanical exfoliation of layered compounds. In the ensuing years, many other interesting 2D crystals have been isolated and studied [5]. This includes semiconductors, insulators, superconductors, and topological insulators. The discovery of ferromagnetic order in monolayers of two different materials, CrI_3 [6] and $Cr_2Ge_2Te_6$ [7], reported in 2017, added ferromagnets to the list.

Exploration of magnetic 2D crystals faces an important obstacle: probing their magnetism is particularly challenging, on account of the their small volume. Magneto-optical methods. [5,6], such as Kerr effect and magnetic circular dichroism, are the techniques of choice to detect ferromagnetic order in these 2D magnetic semiconductor. However, these probes do not provide an absolute measurement of the magnetic moment density and have a very limited spatial resolution. In a recent paper by Thiel and coworkers [2], a radically different method is used: few-layer CrI_3 samples are scanned with a NV center quantum sensor magnetometer [1].

Bulk CrI_3 is a relatively well know ferromagnetic semiconductor with a layered crystal [7]. At every layer Cr atoms arrange in a honeycomb lattice, and are surrounded by an octahedra of I atoms. In this environment Cr atoms have a. spin $S=3/2$. In bulk, both coupling intralayer and interlayer exchange are ferromagnetic, so that the crystal orders ferromagnetically at 61 Kelvin [7]. The first experiments in few-layer CrI_3, using both magnetooptical [3] and tunneling electrons [8,9,10] conclusively show that interlayer exchange is antiferromagnetic, at odds with the case of bulk CrI_3. This unexpected surprise comes as a blessing, as a small magnetic field can off-set the antiferromagnetic interlayer coupling, aligning the layers, permitting a simple way to control these systems.

NV center quantum sensors are one of the most mature quantum technologies of the second quantum revolution. [1]. NV centers are atomic scale defects in diamond, consisting on a missing carbon atom, or vacancy (V), that lies next to a nitrogen (N) substitutional impurity. NV centers have spin $S=1$ and therefore can be in 3 possible quantum states, with different spin projections $S_z= +1,0,-1$. NV centers also turn out to be very emissive single photon emitters. Crucially, their photon yield depends on the spin projection S_z. The combination of these features makes it possible to use them to carry out optically detected single spin magnetic resonance experiment that permit to read out magnetic fields.

The optically detected magnetic resonance experiment is carried out as follows [1]. A static external magnetic field B_0 produces a Zeeman shift of the NV center energy levels. The NV center is pumped both optically with a laser and with a microwave, with frequency ω in the GHz range. The magnetic field of the microwave excites transitions between the spin levels of the NV center. When the frequency ω matches the transition frequency of the spin levels, that depends on B_0, a large variation of the occupation of the emissive states occurs, leading to a variation in the photon count. This permits an optical readout of the resonance, and thereby the magnetic field B_0, with a very large precision, on account of the large spin coherence time. In addition, placing the NV center in a nanodiamond attached to the tip of a scanning probe[1], makes it possible to image the magnetic field.

The magnetic field image, obtained using scanning NV center magnetometry, permits to infer the magnetization field across the CrI_3 sample. At any given point, the magnetic field probed by the NV center is the sum of the external field, which is known, and the so called stray field created by the magnetization gradients of the CrI_3 sample, that occur predominantly at the boundaries and at domain walls. Thiel et al [2] determine the magnetization field at the sample, given the measurement of the stray fields, solving the inverse magnetostatic problem, which is not completely straightforward and requires some noise filtering. This process permits to obtain very valuable information. For starters, it permits to determine the average magnetization density for samples with different number of layers. Monolayers have average magnetization densities consistent with complete spin polarization of the Cr atoms. Stacks with an even number of layers come out with a vanishing total magnetization, whereas stacks with an odd number of layers have a magnetization very similar to the one of the monolayer, consistent with the antiferromagnetic alignment between monolayers observed with other methods [3,8-10].

The scanning probe nature of the experiment permits to map the magnetization, observing different lateral changes of magnetization, coming both from thickness fluctuations as well as magnetic domains. The last, but not the least, an accidental contact of the scanning tip with the sample , that created a puncture on the non-magnetic capping layer, led to a 9.7 fold increase of the magnetization of a 9-layer sample, restoring the ferromagnetic interlayer exchange. Further Raman experiments confirmed a structural transition of the CrI_3 thin film, providing a strong indication that interlayer exchange is related to the stacking structure, as suggested in previous work [7,10,11].

The success of the approach Thiel and coworkers, together with the outstanding capabilities of NV center scanning magnetometry [1,12] pave the way towards a widespread use of this technique to explore many interesting phenomena, such as spin waves, or to look for skyrmions in magnetic 2D crystals.

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