Spin-mechanics with levitating ferromagnetic particles

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We propose and demonstrate first steps towards schemes where the librational mode of levitating ferromagnets is strongly coupled to the electronic spin of Nitrogen-Vacancy (NV) centers in diamond. Experimentally, we levitate ferromagnets in a Paul trap and employ magnetic fields to attain oscillation frequencies in the hundreds of kHz range with Q factors close to $10^4$. These librational frequencies largely exceed the decoherence rate of NV centers in typical CVD grown diamonds offering prospects for sideband resolved operation. We also prepare and levitate composite diamond-ferromagnet particles and demonstrate both coherent spin control of the NV centers and read-out of the particle libration using the NV spin. Our results will find applications in ultrasensitive gyroscopy and bring levitating objects a step closer to spin-mechanical experiments at the quantum level.

Spin-mechanical systems where electronic spins are entangled to the motion of individual atoms are now widely used for studying fundamental phenomena and for quantum information and metrological applications\cite{1}. Inspired by the coupling schemes developed for trapped atoms\cite{2} and single ions\cite{3,4}, new ideas emerged to extend the field to macroscopic systems\cite{5,6,8} with perspectives to test quantum mechanics on a large scale\cite{15}.

One long-standing goal in the field is the coupling of a macroscopic object to electronic spins. In this direction, the electronic spin of the NV center in diamond stands out as a promising solid state qu-bit with efficient optical initialization and read-out\cite{19} and long coherence times\cite{23,24}. Proposals for coupling NV spins to the motion of cantilevers at the quantum level through magnetic forces\cite{11,13,23,24} or lattice strain\cite{17,26} have been put forward. Although important experimental achievements have been made\cite{27,34}, the low spin-mechanical coupling rate caused by the large mass of the cantilever has for now not allowed coherent actuation and cooling of the cantilever. A levitating diamond provides a lighter mechanical oscillator and alternative schemes have been proposed to couple to its center of mass\cite{35} or librational modes\cite{9,10} to an NV spin. Under high vacuum, such platforms could take advantage of their record-high quality factor\cite{23,35}, but experimental progresses with optical tweezers\cite{39,41} or Paul\cite{11,31,47} traps have also been implemented. However, their low mechanical frequency put a severe road-block on the way towards recently proposed spin-entanglement schemes\cite{11,13} and limits the efficiency of the recently demonstrated spin-cooling of the motion of a trapped diamond\cite{48}.

In this letter, we propose and demonstrate first steps towards a platform for spin-mechanical coupling that leverages the aforementioned issues. Here, ferromagnets are levitated in a Paul trap and their librational mode is confined by an external magnetic field. Under modest magnetic fields (in the 0.1 to 1 T range), this levitated oscillator can reach frequencies ranging from 0.1 to 1 MHz, similar to the frequencies obtained using optical tweezers.

The proposed spin-mechanical coupling can be achieved in the two ways that are depicted in Fig. 1. First, the libration of a ferromagnet can be coupled to the spin of a distant (\textasciitilde1\mu m) NV center located in a fixed cold CVD grown nanopyramid (Fig. 1-a), inspired by magnetic force microscopy (MRFM)\cite{19,49} and single spin magnetometry\cite{28,29} platforms. Towards this aim, we assemble and levitate micron-sized particles of soft ferromagnetic materials. Under a magnetic field of 0.1 T, we observe librational frequencies exceeding 150 kHz and Q-factors close to $10^4$ at only $10^{-2}$ mbar of vacuum pressure. This scheme will also allow one to fully harness the properties of NV spins at cryogenic temperature, especially high fidelity initialization\cite{52}, or high fidelity projective read-out through heralding protocols\cite{25,53}.

In the second scheme, a diamond containing NV centers can be attached directly to the levitating ferromagnet

![FIG. 1. Schematics showing two proposed platforms for strong spin-mechanical coupling using the librational motion of ferromagnets levitating in a Paul trap. In the first setup (a), a nano-magnet is levitating at a distance $d$ from the electronic spin of an NV center within a bulk diamond at cryogenic temperature. In the second proposed set-up (b) a diamond is attached to a nano-magnet and coupled to the libration via the external static $B$ field. c) Potential energy of the magnetic field dependent harmonic libration resonantly coupled to the two-level spin system via a detuned microwave.](image-url)
FIG. 2. a) Experimental setup. A ferromagnetic rod is levitating in a ring Paul trap. A pair of permanent magnets generate a uniform magnetic field $B$ that confines the particle orientation. Magnetic coils are used to excite the particle librational motion via a transverse field $B_{\text{ext}}$. b) Scanning electron microscopy image of the levitating iron particles. c) Image of a levitating ferromagnetic rod showing the speckle pattern upon green laser excitation. d) Sequence used for excitation and ring-down measurements.

(FIG. 1-b)). Experimentally, we trap two types of hybrid particles composed of a nano-diamond attached to a soft ferromagnet and a micro-diamond with a ferromagnetic coating. Using these composite structures with large librational confinement, we show both efficient coherent manipulations of the NV centers’s spins and spin read-out of the particle libration, which are important steps towards spin-mechanical (SM) experiments in the quantum regime.

The levitating magnet. At the heart of these proposals, is the levitation of ferromagnetic particles in a Paul trap (see Fig. 2a). We first levitate spherical micron-sized particles made iron with 98 % purity, as shown in Fig.2-b), using similar ring traps and injection technique than in [2, 3]. For soft ferromagnets, best angular confinement are obtained for elongated bodies thanks to shape anisotropy [6]. In the experiments, we form elongated rods such as the one shown in Fig.2-c) by levitating simultaneously a few particles in the trap and applying a magnetic field to bind them together using the attractive magnetic forces (See SI, section I). Once a rod is formed, we apply a homogeneous magnetic field $B$ of up to 0.1 T using permanent magnets.

For a soft oblate ferromagnet, the librational motional frequency reads

$$\omega_\phi = B \sqrt{\frac{V |n_x - n_a|}{I_\phi \mu_0 n_a n_r}},$$

where $V$ is the particle volume and $n_x, n_a$ are geometrical factors (See SI, section I and [6]). We measure $\omega_\phi/2\pi$ using external coils that excite the librational mode and detect it using a laser beam that is retro-reflected off the particle surface (see Fig. 6-d) and SI). One bright spot in the speckle formed in the image plane is coupled to an optical fiber for efficient read-out of the libration. We show in Fig. 3-a) the optical signal under a small magnetic field $B$ of 10 mT. As expected, the librational confinement is found at a much higher frequency ($2\pi \times 20.7$ kHz) than the maximum Paul trap confinement (given by the trap frequency, here $2\pi \times 2.5$ kHz). In addition to this, a moderate vacuum of $4.5 \times 10^{-2}$ mbar already enables reaching a Q-factor of up to $9.3 \pm 0.8 \times 10^3$ (see Fig. 3-b)), and can be even larger under higher vacuum. The current limitation in our experiment is the locked rotation of the magnet around its main axis at lower vacuum pressure, which blurs the speckle pattern we use for detecting the libration. We could finally verify both the linear dependence of the quality factor as a function of $\omega_\phi$ (shown in Fig. 3-c)), and of $\omega_\phi$ with the strength of the external magnetic field $B$ (as shown in Fig. 3-d)). At the highest field of 0.1 T, we find $\omega_\phi = 2\pi \times (170 \pm 10)$ kHz in good agreement with theory (see SI, section I). Calculations also show that a 75 × 25 nm ellipsoidal soft ferromagnet would have $\omega_\phi/2\pi =24$ MHz under 0.1 T. One could even reach GHz frequencies by levitating permanent nano-magnets. These frequencies approach the relaxation rate of domain walls as given by the Landau-Lifshitz-Gilbert relaxation.
Several schemes have been proposed to couple an NV spin with a long coherence lifetime and orbital angular momentum [63], the Einstein-de Haas/Barnett effects [64]. Most importantly, the large librational mode is coupled to a distant NV magnet with the further advantage of high motional frequency. The energy of the spin states also changes and, as a result, a spin-dependent torque can be applied by the field onto the NV center quantization axis. We align it to the total magnetic field \( B \), generated by the micro-magnet at the NV position. In order to obtain a stronger SM coupling, \( \partial B_m/\partial \phi \) should be maximized and along the NV axis. We align it to the total magnetic field \( B = B_0 + B_m \) to avoid spin state mixing due to cross-magnetic fields [12]. We find (see section III of the SI) that a particular configuration \( \theta \sim \pi/4 \) maximizes \( \partial B_m/\partial \phi \) along the NV axis while the one perpendicular to it cancels. Fig. 4 (b) shows the SM coupling rate \( \lambda_\phi/2\pi \) as a function of the magnet radius \( R \) and distance \( d \) from the NV spin. The strong coupling condition \( \lambda_\phi/2\pi > 1/T_2^* \) can be reached with \( R \) and \( d \) in the 100 nm range and considering a bulk NV spin with a long coherence lifetime \( T_2^* \sim 500\mu s \). Using a diamond at cryogenic temperatures also offers additional advantages. Efficient ground state spin-cooling can for instance be achieved even in the regime where \( \lambda_\phi/2\pi < 1/T_2^* \). Generally, in the sideband resolved regime and under high vacuum (in the 10^{-2} mbar range), the limitation to efficient cooling is the longitudinal relaxation time \( T_1 \). Using the proposed scheme will bypass this limit since \( T_1(4K) \approx 100\ s \). Using sideband cooling, the mean final phonon number will thus be limited only by the spin initialization fidelity (which can also be very large at 4K [52]). Finally, dynamical decoupling could be used to mitigate the dephasing and eventually extend the coherence time towards the long spin lifetime [22] (see section III of the SI).

The other proposed scheme for coupling the ferromagnet librational motion to NV centers is to directly attach diamons to the ferromagnet. In this scheme (depicted in Fig. 1-a)), the projection of the external magnetic field onto the NV center quantization axis is changing as the hybrid particle rotates about its equilibrium position. The energy of the spin states also change and, as a result, a spin-dependent torque can be applied by the NV center onto the whole particle. Neglecting the magnetic field produced by the magnet (see SI), the interaction Hamiltonian describing the SM interaction is given by \( H_{\text{int}} = \lambda_\phi S_x (\hat{a}^\dagger + \hat{a}) \). Here, \( \hat{a}^\dagger \) and \( \hat{a} \) are the creation and annihilation operators of the phonon of the librational motion and \( \lambda_\phi \) is the SM coupling constant [9, 10]. A major difference between this scheme and the work of [9, 11], is that \( \omega_\phi \) is now controlled by the external B-field amplitude. Considering a composite system made of a spherical diamond and hard magnets with diameters of 40 nm and 20 nm respectively (see section II of the SI) a moderate homogeneous magnetic field of 30mT is enough to obtain a large confinement frequency.
\( \omega_0 = 2\pi \times 3 \text{ MHz} \) and \( \lambda_0 = 150 \text{ kHz} \). Using isotopically enriched diamonds, the strong coupling condition would be attained without the need for high AC voltages and surface charge control.

Experimentally, we present first steps towards coupling the librational mode of hybrid levitating structures to the electronic spin. In one experiment, we prepare particles consisting of 100nm fluorescent nano-diamonds (FNDs) containing many NV centers attached to the micron-sized iron particles (an SEM image is shown in Fig. 5-a-i)). In another experiment, we evaporate a thin nickel layer on top of diamond particles containing NV centers, which produces the particle depicted in Fig. 5-a-ii) (see SI for details of the fabrication in section II). Although the confinement frequency of both structures is not as high as with the ferromagnet rods, they can levitate stably with librational frequencies above the Paul trap librational confinement. As shown in Fig. 3 and in the section II of the SI, we could observe Electron-Spin-Resonances (ESR), spin-echoes (b) and Rabi oscillations (c) from the NV centers in the first structure (i), similar to what has been achieved recently with levitating micro-diamonds [3]. Operating under vacuum also revealed that laser heating was not as strong as what was observed in [2] with diamonds (see section II of the SI), due in part to the reflective property of the magnet. Let us turn to the second hybrid structure (ii). Using this structure, we could not only achieve spin control, but the electronic spin sensitivity to magnetic fields could also be exploited to detect the librational motion. To do this, parametric excitation of the motion was carried out similarly to in Fig. 3 and the microwave was detuned from the center of the ESR shown in Fig. 5-d) in order to benefit from the highest sensitivity. Fig. 5-e-i) shows ring-down measurements obtained by measuring the change in the photoluminescence (PL) rate as the diamond oscillates.

Using alternating blue and red microwave excitations of the spin, we could then extract the librational motion using the change of NV spins energy (Fig. 5-e-ii)). Interestingly, the evolution of the angle is slightly phase-shifted with respect to trace i). This can be attributed to the finite response time of the spin population dynamics, giving rise to a spin magnetisation that lags slightly behind the particle libration (see SI, section II). Note that this phase lag is at the root of SM cooling [31, 38], as in cavity opto-mechanical schemes [37]. Optimizing it will thus ensure efficient spin-cooling of this hybrid structure.

In conclusion, we proposed a method for quantum mechanical experiments with massive oscillators by coupling levitating magnets to NV centers. Towards this aim, we levitate magnets in a Paul trap and demonstrate large librational frequencies and Q-factors, which in itself offers the possibility to connect orbital momentum to the spin degree of freedom [61] and to implement ultra-sensitive torque magnetometry [68]. Importantly, it leverages the technical bound on the maximum librational mode frequency that can be attained using a Paul trap angular potential only. We demonstrate that it exceeds the spin transition linewidth of NV centers in CVD grown bulk diamonds, which means that sideband cooling and efficient motional state preparation are within reach. Last, we demonstrated coherent spin control of NV centers together with read-out of the mechanical motion using the spin of NV centers in hybrid ferromagnet-diamond particles, offering prospects for realizing single-ion-like protocols, such as entangling distant spins using the motional mode of the magnet as a bus [69].
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**Supplementary Material**

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I. LEVITATION OF FERROMAGNETIC PARTICLES IN PAUL TRAPS

Ferromagnetic particles are trapped using the same setup than for the trapping of diamonds, described in details in [1–3]. The experimental set-up is depicted in Fig.1. The trap and the objective are enclosed in a vacuum chamber. The trap is a ring Paul-Straubel trap [4, 5]. It consists in a small 25 µm thick tungsten wire with an inner radius of 200 µm. It is oriented so that the ring plane is perpendicular to the optical axis.

The particles that we mostly used are 98% pure iron particles that are spherical in shape (Goodfellow, ref. FE006045). While their diameters is rated to be from 1 to 6 µm, scanning electron microscopy images of our sample shows that diameters vary roughly from 0.5 to 3 µm with most of the particles having a diameter around 1 µm (see Fig.2). The particles come in the form of a dry powder and did not undergo specific surface processing. They are injected using a small metallic tip that is dipped into the powder and brought in the vicinity of the trap. With such micron-sized particles, we can operate the trap with a peak-to-peak voltage ranging from $V_{ac}=100$ V to 4000 V at driving frequencies in the kHz range. Particles are generally injected under ambient conditions at $V_{ac}=4000$ V and at a trap frequency of a few kHz.

Overall, the injection technique and trapping operations are essentially the same than for micro-diamonds. However, while levitating diamonds are always negatively charged, we found that both positively and negatively charged iron particles can be stably levitating in the trap. This indicates that the tribo-electricity, responsible for the charge acquired by the particles during injection, differs between iron and diamond. Given these observations, on one levitating iron particle, both positive and negative charge patches may be present simultaneously. This is an important observation with regards to the assembly operation that we describe next.
A. *In-situ* assembling of ferromagnetic particles

To benefit from the potentially large librational frequencies of the ferromagnetic particles offered by shape anisotropy, we form elongated rods by assembling few spherical iron particles. This is done *in-situ*, i.e. within the Paul trap, using the following procedure:

1- We inject several iron particles simultaneously in the Paul trap. Given the large size of the trap (200 $\mu$m diameter) compared to the size of the particles, tens of particles can be levitated simultaneously.

2- We lower the trap potential and eject some particles using air currents in order to then bind only 2 to 4 particles together (the optimum aspect ratio for large librational frequencies is calculated next).

3- We increase the Paul trap confinement by reducing the trap frequency in order to bring the particles as close as possible from each other. At this stage, the particles still repeal each other due to electrostatic forces, forming a so-called Coulomb crystal.

4- We then apply a magnetic field by bringing manually a permanent magnet next to the trap. Magnetic forces acting between the different particles which are then magnetized, are attractive. For sufficiently high magnetic field, of the order of few tens of Gauss, attractive magnetic forces overcome the repulsive electrostatic ones and the particles bind together, forming a rod aligned in the direction of the magnetic field. The eventual presence of charge patches of different sign on the particles, mentioned in the previous section, might also assist the binding process.

Once a rod is formed, particles remain bonded together even when the external magnetic field is nulled, thanks to remanent magnetization and/or Van-der-Waals forces.

As shown in Fig.3 we could image the levitating particles. For this we shine incoherent light onto the particles and use the objective to image the particles onto a CCD camera.
B. Librational mode excitation and optical detection

In the experiments, we use a pair of external coils in Helmholtz configuration in order to excite the librational mode of the levitating ferromagnet. The coils generate a magnetic field perpendicular to the field produced by the permanent magnets used to confine the orientation of the particle, allowing us to displace the particle from its equilibrium orientation. We can thus excite the librational mode by switching the current flowing through the coils. We typically run the coils with a current of 0.5A that is switched off in around 2 \( \mu \)s using a fast electronic switch (EDR83674/2 from company EDR). While the coils mainly excite the librational mode of motion, the center of mass motion is also weakly excited. To favor the excitation of the librational mode while minimizing the center of mass excitation, the coils current is switched ON and OFF, 3 times, at a frequency close to the librational frequency (See Fig.1d in the main text).

![Image of the particle speckle obtained at the levitating particle image plane under coherent illumination with green laser light.](image)

After excitation, we look at the particle motion using optical detection. With the objective inside the vacuum chamber, we focused a green laser onto the particle and collect the reflected light. As shown in Fig.4, at the particle image plane which is located few tens of centimeters away, an image of the particle is formed with an additional speckle feature coming from the coherent nature of the illumination. To detect our signal (shown in Fig. 2 in the main text), we focus a small area of this image onto a single-mode optical fibre and detect the photons transmitted through the fibre with a single photon avalanche photodiode. Thanks to the speckle feature, the detected signal is highly sensitive on the particle position and orientation. For a given levitating particle, we can optimize, in real time, the signal coming from the angular displacement of the particle by selecting the most favorable region of the particle image. To do this, we look at our optical signal while switching the excitation coils at a frequency of 1 Hz.

Our detection method is not intrinsically linear, i.e. the optical signal is not necessarily linear with the angular displacement, and indeed, harmonics of the confinement frequencies can be seen on the signal. However, quasi linearity can be obtained by finely adjusting the detection zone on the particle image. The data in Fig. 3-a) are well fitted by the sum of 3 exponentially damped sinusoids, indicating that our signal reproduces quite faithfully the particle motion. Quasi linearity is also seen in the spectrum of the time trace, shown in in the inset of Fig. 3-a) where we have highlighted the barely visible 2nd order harmonic features.

C. Magnetic confinement of ferromagnetic particles

We present here theoretical estimations of the torque applied to ferromagnetic particles by an external magnetic field and of the corresponding librational frequency.

We consider first the torque resulting from shape anisotropy applied on a prolate ellipsoidal soft magnetic body, which tends to align the long axis of the particle along the magnetic field direction (see Fig.6a). In [6], a model to calculate magnetic forces and torques applied on ideal soft ferromagnetic axially symmetrical bodies has been developed and tested experimentally with excellent quantitative agreement. The torque formula presented below is taken from there. We consider a particle with axial and radial dimensions 2a and 2b. The torque applied on the body by a weak external magnetic field \( B \) is given by
\[ T_{\text{soft}}(\phi) = \frac{V(n_r - n_a)}{2\mu_0 n_a n_r} B^2 \sin(2\phi) \]  

where \( \phi \) is the angle between the magnetic field and the body symmetry axis, \( V = \frac{4\pi}{3}ab^2 \) is the volume of the particle, \( \mu_0 \) is the vacuum magnetic permeability and \( n_r, n_a \in [0, 1] \) are the so-called demagnetisation factors. Those are purely geometrical and can be calculated analytically for ellipsoidal bodies. 

\[ n_a = \frac{1}{R^2 - 1} \left( \frac{R}{2\sqrt{R^2 - 1}} \ln \left( \frac{R + \sqrt{R^2 - 1}}{R - \sqrt{R^2 - 1}} \right) - 1 \right) \]
\[ n_r = \frac{1}{2}(1 - n_a) \]

where \( R = a/b \) is the aspect ratio of the particle. This expression of the torque is valid for a soft ferromagnetic material with large magnetic susceptibility \( (\chi \gg 1) \) and a magnetic field such as

\[ B < \mu_0 m_s \frac{n_a n_r \sqrt{2}}{\sqrt{n_a^2 + n_r^2}} \]

where \( m_s \) is the magnetisation at saturation of the material. It is quite remarkable that, under those conditions, the torque does not depend on the magnetic properties of the body but only on its geometry.

From kinematics principles, we find that the confinement frequency for the angle \( \phi \) resulting from the magnetic torque \( T_{\text{soft}} \) is given by

\[ \omega_\phi = \sqrt{\frac{V(n_r - n_a)}{I_\phi \mu_0 n_a n_r}} B \]

where \( I_\phi = \rho V(a^2 + b^2)/5 \) is the relevant component of the particle rotational inertia, \( \rho \) being the particle density.

The librational frequency is proportional to the applied magnetic field and, for a given aspect ratio, it is inversely proportional to the particle size. This latter property can be seen by writing

\[ \frac{V}{I_\phi} = \frac{1}{V^4} \rho \left( \frac{4\pi}{3} \right)^2 \frac{R^2}{R^2 + 1} \]

In Fig. 7, we plot \( \omega_\phi \) versus the particle aspect ratio for a fixed minor-axis size. Best confinement frequency is obtained for an aspect ratio of \( R \approx 2.606 \).

By approximating our experimentally trapped elongated rods with ellipsoids, we can compare our experimental measurement of \( \omega_\phi \) with theoretical calculations. In the experiment, with the particle shown in Fig. 6b, we measured \( \omega_\phi \approx 2 \pi \times 170 \text{kHz} \) at a field of 0.1T. Theoretically, for an ellipsoid of 5.4 \( \mu \text{m} \times 2.5 \mu \text{m} \) as drawn on top of the image, and taking an iron density of 7.86 g/cm\(^3\), we find \( \omega_\phi \approx 2 \pi \times 240 \text{kHz} \) at a field of 0.1T. This is in fair agreement with the experimental value given the rough approximation made on the particle shape. For this particle shape, the torque equation is valid for magnetic fields below 0.46T, assuming a magnetisation at saturation of iron of 2.2T.
FIG. 11. Magnetic confinement frequency versus aspect ratio for a soft ferromagnetic ellipsoid with fixed minor-axis. The ellipse drawn in the middle of the figure has an aspect ratio of $\approx 2.6$ which gives the highest magnetic confinement. The minor-axis is set to 25nm and the magnetic field to 0.1T.

confirms that a linear dependence of $\omega_\phi$ with the magnetic field is effectively expected for the range of fields used in the experiments (from 0 to 0.1T). Since $\omega_\phi$ increases with decreasing particle size, larger confinement frequencies could be obtained for levitating nano particles. We calculate that, for an ellipsoid of 75nmx25nm under a field of 0.1T, we would obtain $\omega_\phi \approx 2\pi \times 24$MHz.

Hard ferromagnetic materials (permanent magnets) could offer much stronger magnetic confinements than their soft ferromagnetic counterparts. For a magnet with uniform magnetisation $m$, as long as the external field does not modify the magnetisation of the material, the torque which tends to align the particle magnetisation direction along the external magnetic field is simply given by

$$T_{\text{hard}}(\phi) = VmB\sin(\phi)$$

where $\phi$ is now the angle between the magnetisation and magnetic field directions. This holds for any particle shape. Magnetic confinement frequencies are then given by $\omega_\phi = \sqrt{VmB/I_\phi}$. For a 5.4$\mu$mx2.5$\mu$m ellipsoidal neodynium particle, under a field of 0.1T, we find $\omega_\phi \approx 2\pi \times 18$MHz. We assume for this a magnetisation of 1.6T and a density of 7.4 g/cm$^3$. For a smaller 75nmx25nm ellipsoid, we have $\omega_\phi \approx 2\pi \times 1.3$GHz. Experimentally, trapping micro or nano-particles of a hard ferromagnetic material would be highly beneficial to boost confinement frequencies. However, because of the strong attractive magnetic forces, it is difficult to isolate the particles from each other.

D. Magnetic field calibration

We calibrated the magnetic field generated by the two permanent magnets as a function of their distance from the levitated particle using a Hall probe. We show in Fig. 8 the data taken for this calibration. We could also double check the calibration using, as magnetic sensors, NV centers contained in a micro-diamond deposited directly on the ring trap. One can indeed infer the magnetic field seen by NV centers by performing Electronic Spin Resonances (ESR) scans on the NV centers spins \[^8\]. For this, we calculate numerically the spin transitions energy of a NV center for all magnetic field strengths (up to 0.1T) and relative orientations with the NV axes. Experimental measurements of the spin transition frequencies is then compared with calculations to identify the magnetic field seen by the NV centers.

II. NV CENTERS IN DIAMONDS ATTACHED TO LEVITATING FERROMAGNETIC PARTICLES

Obtaining librational frequencies larger than 1 kHz is a relatively straightforward task with levitating ferromagnets as opposed to levitating diamonds, where the librational mode frequency depends on dipole or quadrupolar distribution
of charges on the diamond surface which are sample dependent. We thus propose a protocol that uses nanodiamonds attached to ferromagnets in order to increase the trapping frequency.

A. Preparation of the hybrid system

The employed nanodiamonds are bought in the form of a solution containing fluorescent nanodiamonds (brFND-100) from FND biotech, and contain a large fraction of NV centers (>1000 NV centers per particle are quoted by the manufacturer). We show in Fig.10 a confocal map of many FNDs nebulized on a quartz coverslip. Single FND are clearly seen and spin characterisation could be performed on this sample.
As mentioned in the main text, we then attach these NDs to the ferromagnets. This was done using the following procedure:

1. Several iron particles are cast on a quartz coverslip.

2. A solution containing nanodiamonds (100 nm in diameter) with a large concentration of NV centers is prepared and injected in the reservoir of ultrasonic nebulizer and the solution is nebulized on top of the quartz coverslip supporting the iron particles. A SME image of the mixture is shown in figure [13] and an image showing a single iron particle is shown in the main text. Their concentration has been chosen so that single nanodiamonds could be excited optically once in the trap.

3. The prepared sample is then scrapped using a small metallic wire and brought close to the trap for injection and the PL rate, ESR, echo signals are monitored in the same way as demonstrated in [3].

![Figure 14](image)

**FIG. 14.** a) ESR spectrum from NV centers in a nanodiamond attached to a levitating ferromagnet. b) ESR spectra taken without a magnetic field at three different vacuum pressures: 0.5, 0.07 and 0.01 mbar for traces i), ii), iii) respectively.

We encounter several issues with the characterization of the hybrid structure when observing the ESR. One subtle point is that, if some ESR lines overlap (which is a problem when one wishes to perform efficient Rabi oscillations), tuning the external magnetic field orientation with respect to the diamond axis is not enough to lift this degeneracy. The magnet main axis follows the B field angle so the NDs that are attached to it are also aligned in the very same way as the B field is rotated. If this situation occurs, another ND on the particle or another sample must be chosen. This effect does not impact the spin-mechanical coupling mechanism since the magnet inertia will let the NV feel a varying magnetic field.

Another problem that was encountered is that the ESR contrast, already in the absence of magnetic field, was often reduced compared to the optimum (10%) that is typically observed. Spin control was thus not possible for all levitating hybrid particles. The chemical properties of the hybrid particles or the presence of charges patches close to the nano-diamonds could be responsible for this, but further investigations would be needed to nail down this issue.

One promising feature however is that most hybrid particle are less sensitive to the laser torque. Less noisy ESR spectra can thus be obtained. Fig. [14]a) shows one such spectra, taken at room temperature, which displays similar properties (contrast and width) than ESRs typically observed with MSY micro-diamonds [3]. Further, the temperature of the assembly can be kept rather low even at significant vacuum levels even when laser light is shone to the particle. Fig. 14b) shows ESRs taken at three different vacuum levels. The temperature of the particle can be estimated using a similar procedure than in [2], and we found a temperature of around 400 K for trace ii), which was obtained at under 0.07 mbars. Compared to the experiment performed in [2], at least an order of magnitude in pressure could thus be gained, when using similar green laser powers (100µW). As mentioned in the main text, the reason is likely to be that iron reflects most of the impinging light instead of absorbing it. Here this could also be because the ND dissipates heat more efficiently than micro-diamonds due to the larger surface to mass ratio of the NDs compared to the micro-diamonds. The fast drop of the ESR contrast in trace iii) is unexpectedly large, which may also stem from chemical reactions or patch potentials in combination with the normal contrast drop due to the large temperature.

**B. Magnetic field generated by the ferromagnetic micro-particles on the attached NV centers**

As mentioned in the main text, we have been able to measure the magnetic field produced by a ferromagnetic particle, while magnetized by an external field, using the nano-diamonds attached to it. To do this, both a micro-diamond containing NV centers and an hybrid particle composed of fluorescent nano-diamonds (FND) attached to
a cobalt micro-particle are deposited on the ring trap electrode. We perform ESR spectrum in the presence of a magnetic field. The use of the ring trap is for us an easy way to drive the spin with microwaves.

The corresponding ESR spectra is shown in Fig. 9. For the hybrid particle, one can identify the presence of 2 FNDs. An extra 10\% field is seen by the FNDs attached to the ferromagnetic particle compare to the micro-diamond alone. Depending on the position of the FND around the ferromagnet, one can expect different magnetic fields to be seen. In our measurements, the two FNDs sense the same magnetic field indicating that they are likely to be aggregated.
C. Spin-mechanical protocol

The magnet + diamond structure will oscillate about the mean angle which will still modulate the spin-mechanical coupling. The diamond will also exert a torque to the whole structure, effectively changing the mean angle of the composite particle. For a quantum protocol with strong coupling, the mechanism is the same as outlined in [9].

The coupling strength is given by \( \lambda = \gamma B \phi_0 \), where
\[
\phi_0 = \sqrt{\frac{\hbar}{2I\omega}}
\]
where \( \omega \) is in turn defined by the magnetic field angle and magnitude applied at the location of the hybrid structure, i.e. by equation (3) and the inertia momentum is the one of the whole composite particle [9].

The shift of the diamond position from the center of mass (as shown in the figure 1 of the main text) will couple the center of mass and the librational modes together, but the frequency of the center of mass mode will be orders of magnitude lower so the coupling will be inefficient.

We estimated that with a nanodiamond and a ferromagnet both ellipsoidal with long axes of 80nm and short axis of 40 nm, a coupling of \( \lambda \sim 100 \text{kHz} \) can be obtained at an external field of 30mT with an optimum angle of 55° [10] while the oscillator reaches a frequency of \( \omega \sim 2\pi \times 3 \text{ MHz} \).

D. Spin read-out of the angular motion of a levitating hybrid particle

As presented in the main text, we could perform spin read-out of a levitating particle angular motion. For this, we use a hybrid particle composed of a micro diamond (MSY 8-12µm) with a 200 nm thick magnetic nickel coating on one side of the diamond. This was realized using simple sputtering of nickel atoms from an oven onto a layer of MSY cast on a quartz coverslip. The injection into the trap was done by carefully scratching the sample with a tip and the injection was done in the same manner as with the previous particles.

We stably trap the hybrid particle in the Paul trap. Applying a uniform magnetic field of around 140G then yields angular confinement of around 4.2 kHz for one of the librational mode. This mode could be parametrically excited and optically detected using the method described in the section I B. Ring down of the oscillator motion is shown in Fig. 3-e) in the main text.

To read the angular motion of the particle using the embedded NV centers spins, we exploit the angular dependency of the NVs ESR transitions frequencies. When the orientation of particle changes with respect to the external magnetic field, the frequencies of the ESR transition change and, by applying a microwave field tuned to the side of one ESR transition, so does the population in the excited spin state. Spin state population can be read out optically by collecting the NV centers Photo-Luminescence (PL) under green excitation.

Experimentally, we run the same excitation/detection sequence as for the direct detection of the particle motion, looking this time at the PL signal and applying a microwave field slightly detuned from one ESR transition. The measurement is performed for two different microwave detunings, one on each side of the ESR transition, for which the slope of the ESR signal as an opposite sign. The corresponding ESR spectrum is shown in Fig.4-d) in the main text. Fig.18 shows the PL signal versus time after parametric excitation of the librational mode for both microwave detunings.

Regardless of the spin states, the amount of collected PL changes with the particle angular position. This is primarily due to the fact that the green laser illumination conditions changes with the particle angular position. The PL rate thus changes when the particle rotates. Oscillations that are in phase with the particle motion are indeed clearly visible for both microwave detunings. To unambiguously extract the signal coming from the spin states, we plot the difference between the two curves. This allows to remove the spin-independent part of the signal which, again, is the same for both detunings, while keeping the spin-dependent part which has an opposite phase for both detunings.

The resulting signal is plotted in Fig.4-e) in the main text. We see that the PL signal reproduces well the direct optical measurement demonstrating a spin read-out of the particle angular motion. We note that, in our experimental conditions, direct optical measurement of the particle motion using the retro-reflected light is much more sensitive than the spin read-out. This is seen in the much better signal-to-noise ratio despite a much shorter total acquisition time for the optical (2mins) than for the spin (50mins) measurements. Interestingly however, the spin signal is slightly delayed from the optical one. This can be attributed to the finite response time of the spins population dynamics giving rise to the spins population lagging slightly behind the particle motion.

To be quantitative, we plot in Fig.15 the PL count rate versus time after switching ON and OFF a resonant microwave field. This allows to measure the rate at which the spins population are excited by the microwave field.
and polarised by the green light. By fitting the curves with an exponential decay, we found an excitation rate of 28\(\mu s\) and a polarisation rate of 34\(\mu s\).

 Those figures can be compared to the apparent delay between the spin read-out and direct optical measurement of the particle motion. In Fig. 14, we plot the squared difference between the two curves while adding a temporal delay of variable length between them. We found that the two curves coincide best for a delay of 32\(\mu s\) which is in perfect agreement with the measured rate of evolution of the spins population.

### III. COUPLING THE LIBRATIONAL MODE OF LEVITATING MAGNETS TO A DISTANT NV CENTER

Here we present the calculations involving the librational mode of levitating magnet coupled to a distant NV centers in details. This corresponds to the proposal a) of the Fig. 1 in the main text. Note that this scheme is largely inspired by the proposals to couple an NV spin to a magnetized cantilever [11] or to the librational mode of a levitating diamond [9, 10]. We therefore only show how a similar coupling can be achieved in the specific case of the librational mode of a levitating magnet coupled to a distant NV spin.
FIG. 19. Configuration of the NV center spin relative to the levitating magnet. The only degrees of freedom except $d$ is $\theta$ as the magnet is on average aligned with the magnetic field ($\phi = 0$) and the NV center orientation is set along the total field $B_T$.

A. NV configuration in the field generated by the micro-magnet

As explained in the main text, the SM coupling is obtained through the field $B_m$ generated by the micro-magnet at the NV position. This field varies with $\phi$, the angle between the moment of the micro-magnet $M$ and its equilibrium position along the external magnetic field $B_0$ which can give rise to the SM coupling. To obtain a strong enough coupling, the NV center must be placed in close vicinity with the magnet (inferior to 1 $\mu$m), but one can rotate the NV around the magnet and around itself.

With this regards, there are two main constraints to obtain a better coupling:

- The NV center axis must be aligned with the total magnetic field $B_T = B_0 + B_m$ to avoid spin state mixing by the transverse magnetic field. This would indeed degrade spin initialization and read-out efficiency [12].

- To enhance the SM coupling, the first order variation of $B_m$ in $\phi$ should be maximal and along the NV axis.

Let us first note that the external magnetic field $B_0$ orientation and amplitude is already determined as we need it to confine the angular degree of freedom of the micro-magnet with a certain frequency $\omega_\phi$. Given a certain position of the NV center around the micro-magnet, it thus fixes the orientation of the NV axis along the total field $B_T$. The only degree of freedom left is then to rotate the NV position around the magnet.

With a spherical levitating magnet, the field generated by the magnet at a point given by the polar coordinates $(d, \theta)$ described in figure 19 will be:

$$B_m(\theta) = \frac{MR^3}{3(d+R)^3} (2 \cos(\theta - \phi) e_r + \sin(\theta - \phi) e_\theta)$$

(5)

where $M$ is the magnetization of the magnet material, $R$ the radius of the magnet and $\phi$ is the angle between the magnet moment and the external field.

Since the mean orientation of the magnet is aligned with the external magnetic field $B_0$ (i.e. $\phi = 0$) and the NV center is aligned along the total field $B_0 + B_m$, one can calculate the derivative of the field along the NV axis and perpendicular to it as a function of $\theta$.

We find that for a particular angle $\theta_{op}$ the contribution along the NV axis is maximal and the one perpendicular to it cancels, with

$$\theta_{op} = \frac{1}{2} \arccos \left( -\frac{3D_\phi}{6B_0 + D_\phi} \right) = \arccos \sqrt{\frac{9B_0}{6B_0 + D_\phi} - 1}$$

(6)

where $D_\phi = MR^3/(d + r)^3$.

At this optimal value we have $\partial B_t/\partial \phi = D_\phi u_{NV}$.

Note that this angle is actually close to $\pi/4$ since the homogeneous magnetic field tends to be stronger at a reasonable distance from the levitating magnet.

B. Spin-mechanical coupling scheme

In the optimum configuration the total field at the NV center is longitudinal up to the first order in $\phi$ (see fig 1-a and SI for details) and its first derivative is $D_\phi$. In order to obtain a resonant SM interaction, a resonant microwave
FIG. 20. a) Experimental scheme proposed: a magnet is levitated at a distance $d$ from an NV spin within a bulk diamond at cryogenic temperature. b) Sequence for preparation of a Fock state. $|\Psi\rangle$ is an arbitrary mechanical state, for instance the ground state. c) Level diagram for the hybrid SM system with and without microwave. Colored points show the populated levels during a sequence for creating a 1 phonon Fock state from the ground state. Here $|\psi\rangle$ is the $n = 0$ phonon state. d) State of the hybrid system during a sequence that increases the phonon number by one.

is added to drive the $m_s = 0 \rightarrow 1$ spin transition of the NV center at a Rabi frequency $\Omega_R = \omega_\phi$. The spin-only part of the Hamiltonian is then diagonal in the $|\pm\rangle = (|0\rangle \pm |1\rangle) / \sqrt{2}$ basis and the full Hamiltonian can be written:

$$\hat{H}/\hbar = \omega_\phi \hat{S}_z + \gamma D_\phi \phi_0 \hat{S}_x (\hat{a}^\dagger + \hat{a}) + \omega_\phi \hat{a}^\dagger \hat{a}$$

(7)

where $\hat{S}_\mu$ are the Pauli matrix operators in the $|\pm\rangle$ basis, $\gamma$ is the gyromagnetic ratio of the NV electron spin, $\hat{a}^\dagger$ and $\hat{a}$ are the creation and annihilation operators for the phonon of the angular motion, and $\phi_0 = \sqrt{\hbar/(2I_\phi \omega_\phi)}$ its zero-point motion.

This is a Jaynes-Cummings Hamiltonian with a coupling $\lambda_\phi = \gamma D_\phi \phi_0$. Under the so-called strong coupling regime where the decoherence of both the spin and the MO are small compared to $\lambda_\phi$, it allows a coherent exchange between a phonon and the spin state at a rate $\lambda_\phi/2\pi$.

This coupling and the high degree of control of the spin can be used to generate non-classical mechanical states such as the zero phonon ground state or any arbitrary superposition states $|\pm\rangle$. A key point here is that the NV spin excellent initialization fidelity. At cryogenic temperature (4K), it can be initialized in the $|0\rangle$ or $|1\rangle$ state by using resonant optical excitation: after a number of optical cycle, the NV spin has a high probability to relax in the spin state for which the optical excitation is non-resonant. Initialization fidelity can be as high as 0.998 $|1\rangle$ for the $|m_s = 1\rangle$ state.

As an example, figure 20 depicts the sequence that one can use to prepare a one phonon Fock state from the ground state. Here, we assume perfect spin initialization and neglect the decoherences sources. The spin is first initialized in the $|0\rangle$ state and rotated to the $|+\rangle$ state by a $\pi/2$ microwave pulse polarized along the x axis. A resonant microwave is then turned on to achieve the spin-mechanical resonance, with a Rabi frequency $\Omega_R = \omega_\phi$ and a polarization along the y axis (or with a $\pi/2$ dephasing compared to the first microwave pulse). After a time $\tau = 1/(2\lambda_\phi)$, the spin is flipped due to coherent exchange with the mechanical oscillator: a subsequent $\pi/2$ pulse (x polarization) maps it unto the $|1\rangle$ state while the phonon number has increased by one.

In order to first cool the oscillator to the ground state, one only needs to apply this sequence but with an opposite rotation of the spin so it is in the $|-\rangle$ state before the SM coupling. The fidelity of the state obtained in the stationary
regime usually depends on the $T_1$ of the oscillator. However, at low enough pressure heating from gas molecules will be negligible (see below) so the mean phonon number will be solely limited by the spin initialization fidelity (0.998) to $\sim 0.002$.

Once in the ground state, sequences similar to the one described above can be applied to generate any desired state \cite{11,16}. To obtain a high fidelity, one is however limited to the strong coupling regime as the decoherence will damp the SM Rabi oscillations and result in imperfect SM pulses.

C. Decoherence sources

1. Mechanical oscillator

Since the scheme does not requires shining light on the levitating magnet the main mechanical decoherence is expected to come from gas collision with the levitating particle. The decoherence of the mechanical oscillator will limit the time during which the whole experiment sequence (cooling, mechanical state preparation, measurement) can be carried out. Although novel theoretical tools are being developed to estimate the decoherence rate \cite{17,18} of librational states, it will highly depend on the shape of the levitating particle, its roughness and eventually the potential governing the scattering of molecules impacting it. Note that it can be strongly mitigated if one works with an isotropic particle \cite{17,18} or under high enough vacuum. To give a rough estimate of the heating rate and hence of the lifetime of a mechanical state, we calculate the damping rate of the levitating particle $\Gamma_{gas}$ due to air molecules in the classical regime.

In the Knudsen regime, when the mean free path of the gas molecule is higher than the size of the levitating particle, one gets \cite{19} :

$$\Gamma_{gas} = \sigma_{eff} \frac{10\pi P}{a \rho \overline{c}}$$

with $\sigma_{eff} \sim 1.1$ the accommodation coefficient, $a$ the radius of the particle, $P$ the pressure in Pa, $\rho$ its density and $\overline{c}$ the molecular velocity of the gas molecules considered. We obtain a relatively low heating rate : for a 1µm radius sphere, it is of about 1Hz at $P = 10^{-3}$mbar.

2. NV spin decoherence

The NV spin itself has an excellent lifetime at cryogenic temperature. It can reach up to a hundred seconds \cite{20}. However NV spins are coupled to the fluctuating nuclear spin bath of the diamond crystal. This results in fluctuations of the frequency of the NV spin and considerably reduces the $T_2^*$. Although this effect can be mitigated using isotopically purified diamond to remove $^{13}$C spins \cite{21,22}, the best linewidth as far as we know was as low as 0.7 kHz ($T_2^* = 460 \mu s$) \cite{23}.

Those coherence times are however obtained with NV center deep deeply buried inside the diamond crystal: when the NV spin is closer to the surface, its properties will suffer from the presence of surface impurities. The magnetic noise from those impurities lowers both the lifetime \cite{12} and coherence time \cite{24} of shallow NV spins. It should however be noted that dynamical decoupling could be integrated within a spin-mechanical experiment to solve this issue. The sequence described in figure \cite{20} is actually similar to a spin-locking decoupling sequence \cite{25}, with a decoupling rate $\Omega_R = \omega_\phi$. We expect this will protect the NV spin from decoherence caused by the spin bath, as already mentioned in \cite{11}. It is difficult to predict the coherence time one can obtain with this scheme, but using a Hahn echo decoupling sequence one can reach coherence time up to 200 $\mu s$ for 5-nm-deep NV spin and of 800 $\mu s$ for 50-nm-deep NV spin \cite{26}. The impurities themselves could also be driven to decouple them from NV spins \cite{27}.

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