Spin wave free spectrum and magnetic field gradient of nanopatterned planes of ferromagnetic cobalt nanoparticles: key properties for magnetic resonance based quantum computing.

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Abstract. - We present a study by ferromagnetic resonance at microwave Q band of two sheets of cobalt nanoparticles obtained by annealing SiO$_2$ layers implanted with cobalt ions. This experimental study is performed as a function of the applied magnetic field orientation, temperature, and dose of implanted cobalt ions. We demonstrate that each of those magnetic sheet of cobalt nanoparticles can be well modelled by a nearly two dimensional ferromagnetic sheet having a reduced effective saturation magnetization, compared to a regular thin film of cobalt. The nanoparticles are found superparamagnetic above around 210 K and ferromagnetic below this blocking temperature. Magnetostatic calculations show that a strong magnetic field gradient of around 0.1 G/nm could be produced by a ferromagnetic nanostripe patterned in such magnetic sheet of cobalt nanoparticles. Such a strong magnetic field gradient combined with electron paramagnetic resonance may be relevant for implementing an intermediate scale quantum computer based on arrays of coupled electron spins, as previously reported (Eur. Phys. J. B (2014) 87, 183). However, this new approach only works if no additional spin decoherence is introduced by the spin waves excitations of the ferromagnetic nanostructure. We thus suggest theoretically some possible magnetic anisotropy engineering of cobalt nanoparticles that could allow to suppress the electron spin qubit decoherence induced by the collective magnetic excitation of those nanoparticles.

Magnetic nano-objects have many potential applications. Magnetic nanoparticles (NPs) can be used as contrasts agent in the diagnosis and treatment of cancer [1], magnetic nanostripes can be used as a medium for efficient classical data transmission and processing [2], and magnetic nanodots can be used as storage elements for high density magnetic data recording [3]. The present work reports on a new potential application of metal magnetic nanoparticles embedded in dielectric matrix. This is related to quantum information processing and electron paramagnetic resonance (EPR) spectroscopy. It was recently theoretically demonstrated [4] that the strong magnetic field gradient produced by a ferromagnetic nanostripe combined with the microwave pulses delivered by a pulsed electron paramagnetic resonance spectrometer, could constitute two of the three key elements constituting the hardware of a potential small scale spin based quantum computer, the third key ele-
Fig. 1: Ferromagnetic resonance (FMR) spectrum obtained at Q band (33.95 GHz) and $T = 297$ K, with the magnetic field applied in the plane (0°) of the cobalt magnetic nanoparticles, (1a) for sample I (1.10$^{17}$ cm$^{-2}$ Co$^+$ ions) and (1b) for sample II (0.510$^{17}$ cm$^{-2}$ Co$^+$ ions). The inset (1c) inside figure 1a shows the temperature dependence of the double integration (noted D.I.) of the FMR signal which is proportional to the effective magnetization of the cobalt nanoparticles detected by FMR spectroscopy at Q band. The modulation frequency was 100 kHz. The modulation amplitude was 4 G. The microwave power used was 2 mW. The narrow linewidth signals seen on the spectrum of the two samples below 0.8 Tesla (=8000G) are due to impurities in the Q band microwave cavity used.

In the first part of the article we present FMR experiments performed at microwave Q band on two sheets of cobalt nanoparticles obtained by annealing SiO$_2$ layers implanted with cobalt ions. These experiments are similar to previous ones performed at X band [5]. This FMR study allows us to demonstrate that each of those magnetic sheets of cobalt nanoparticles can be fairly modelled by a nearly two dimensional ferromagnetic plane having a reduced effective saturation magnetization, with respect to a true bulk-like thin film of cobalt. Subsequent calculations based on magnetostatics then demonstrate that a strong magnetic field gradient of around 0.1 G/nm can be produced by such a magnetic nanostripe made of cobalt nanoparticles. Since the key issue of the present work is quantum information processing of electron spin qubits, we show that a suitable engineering of the magnetic anisotropy of the cobalt nanoparticles may allow the individual microwave addressing and coherent manipulation of spin qubits state through EPR. This is done upon separating the spin resonance of the nanostripes from the spin resonances of the qubits. Two amorphous SiO$_2$ thin films (300 nm), obtained by
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Fig. 2: Full rotational pattern of the ferromagnetic resonance mode of sample I (a/), and of sample II(b/), measured at room temperature and at Q band. Solid lines are numerical simulations obtained using the Smit and Beljers formalism (see text).

oxidation of Si wafers, were implanted at room temperature (295 K) with 160 keV Co\(^+\) ions at two different doses, 1.10\(^{17}\) cm\(^{-2}\) (sample I) and 0.5.10\(^{17}\) cm\(^{-2}\) (sample II), and further annealed at 873 K under hydrogen flow, in order to produce the metallic cobalt nanoparticles inside the SiO\(_2\) matrix. Similar samples implanted at 160 keV with a nominal dose of 1.10\(^{17}\) cm\(^{-2}\) Co\(^+\) ions have been previously investigated by TEM and SQUID magnetometry [6]. This previous study has shown i) that the magnetic cobalt metal nanoparticles have mainly the hexagonal phase, with a c axis of magneto-crystalline anisotropy randomly oriented in the amorphous SiO\(_2\) matrix, ii) that the average diameter of those magnetic cobalt nanoparticles is around 4.5 nm, and iii) that they are superparamagnetic at room temperature. The ferromagnetic resonance study of sample I and II containing the magnetic cobalt nanoparticles is performed using an EMX Bruker continuous wave electron paramagnetic resonance spectrometer operating at Q band (microwave frequency around 34 GHz). Modulation coils are used to modulate the EPR signal at 100 kHz, which allows to perform a sensitive lock in detection of the microwave absorption signal. This modulation also produces a spectrum which appears as the derivative of a standard microwave absorption spectrum with gaussian or lorentzian lines. The cobalt implanted thin films of SiO\(_2\) can be rotated inside the microwave resonator in order to vary the direction of the applied static magnetic field, from in plane (angle: 0\(^o\)) to out of plane (angle: 90\(^o\)). The sample temperature can also be varied from 4 K to 300 K using an Oxford Helium flow cryostat with a temperature controller designed for EPR experiments.

The FMR spectra obtained for sample I and II are shown on figure 1. The FMR resonance at Q band for a magnetic field applied in the plane (0\(^o\)) occurs at around 10 600 G for sample I and at around 10 900 G for sample II. As expected, due to the random orientation of the c axis of the hexagonal cobalt nanoparticles and also due to their size distribution resulting from the implantation process, the FMR signal is broad (peak to peak linewidth \(\approx 2000\) G) for the two samples. Also, the intensity of the FMR signal of sample I is larger than the one of sample II, given the larger ion dose in this sample. The in plane (0\(^o\)) magnetization of sample I has also been recorded as a function of temperature (inset 1c of figure 1a). As it was previously shown by SQUID magnetometry [6] on similar samples, the cobalt nanoparticles of this sample are superparamagnetic at room temperature and ferromagnetic at low temperature. We found a transition between the two regimes occurring around a blocking temperature \(T_B \approx 210 K\). Experimentally, the sample was cooled below 100 K under zero applied magnetic field. The applied magnetic field is also set to zero before and between two successive FMR spectrum acquisitions. Due to the initial zero field cooling below \(T_B\), the initial average magnetization of the sample is negligible because the magnetization of each nanoparticle is then locked along the hexagonal c axis, which is random over the ensemble of cobalt nanoparticles. In this initial equilibrium state below the blocking temperature, the magnetization relaxation dynamics of the nanoparticles is extremely slow [7]. When recording a FMR spectrum, the magnetic field increases very rapidly, which produces a small and partial alignment of the magnetization vectors of the nanoparticles along...
the magnetic field direction, resulting from the slight decrease of the magnetization relaxation time expected in those experimental conditions [8]. For temperatures far below $T_B = 210K$, the slow relaxation dynamics of the magnetization thus leads to a small magnetization signal measured over the ensemble of cobalt nanoparticles. Around $T_B = 210K$ and above, the relaxation time of the magnetic nanoparticles of cobalt becomes much faster than the time scale required for the full FMR field sweep spectrum acquisition. As a consequence, around $T_B = 210K$ and above, the cobalt nanoparticles align rapidly their magnetic moment along the direction of the applied magnetic field, producing a net equilibrium magnetization. The magnetic relaxation time $\tau$ of an isolated superparamagnetic nanoparticle is given by a standard Arrhenius law expression [7]. It takes into account the temperature $T$ and the magnetic anisotropy energy $K$ of the nanoparticle of volume $V$. It is given by $\tau = \tau_0 \exp \left( \frac{K V}{k_B T} \right)$. The intrinsic time scale $\tau_0$ is generally in the range of $10^{-9} s \leq \tau_0 \leq 10^{-13} s$. Here, the measurement time considered is the time required for the acquisition of a full field sweep FMR spectrum over more than 8000 Gauss, which is around $\tau_{mes} = 100 s$ here. This measurement time roughly corresponds to the time window let to the nanoparticle to overcome the magnetic anisotropy barrier and thus to reorient itself in the direction of the applied magnetic field at the temperature $T$. Taking $\tau_0 = 10^{-9} s$, as in the previous analysis of similar samples investigated by SQUID magnetometry [6], and using $\tau_{mes} = 100 s$ for the FMR experiments, one obtains the following relation between the magnetic anisotropy energy $K$ of the nanoparticle of volume $V$, and its blocking temperature $T_B$ (blocking energy $k_B T_B$) : $K V \approx 25 k_B T_B$. Using the average diameter of $R = 4.5 \text{ nm}$ obtained by TEM analysis of similar samples implanted by 160 keV cobalt ions at a dose of $1.10^{17} \text{ cm}^{-2}$ [6], and using the standard value of the constant of anisotropy for cobalt, $K = 210^{6} \text{ erg cm}^{-3}$ [9], one estimates $T_B = 218K$. This shows that our experimental measurements by FMR spectroscopy are in very good agreement with well known cobalt properties and with previous data obtained by TEM and SQUID magnetometry on similar samples.

The room temperature equilibrium saturation magnetization of our two samples can be extracted from FMR experiments, either simply using the standard Kittel formula for the in plane and out of plane magnetic resonance fields [5, 10], or by performing the full rotational pattern analysis of the FMR signals measured, as it is shown here on figure 2a and 2b respectively for sample I and II. As a matter of fact, the observed rotational pattern symmetry is due here to the shape anisotropy of each magnetic film, which is related to its effective saturation magnetization. Numerical simulations of the rotational patterns, performed using the Smit and Beljers formalism [11] and taking into account shape anisotropy, thus confirm that the dipolar couplings between the cobalt nanoparticles can not be neglected in those two magnetic films. From this point of view, the two films of magnetic nanoparticles can thus be seen as mimicking bulk like magnetic thin films with a reduced effective saturation magnetization compared to the bulk material ($B_{sat,bulk} \approx 18400G$). The results of the simulation made for sample I gives $B_{sat, I} \approx 1500 G$ and the one made for sample II gives $B_{sat, II} \approx 790 G$. The ratio of the

Fig. 3: 3a: Nanofabrication process, based on etching through a mask, suggested to produce the diluted ferromagnetic nanostripe discussed in the text and shown on figure 3b. Figures 3c, 3d and 3e illustrate the physical concepts behind the magnetic anisotropy engineering of the nanoparticles discussed in the text.
effective saturation magnetizations of the two films is very close to 2. It nicely corresponds to the ratio of the dose of implanted Co ions between sample I and sample II. Also, one notes that the ratio between the effective saturation magnetization of each film and the bulk saturation magnetization, noted $f$, is given by $f_I = \frac{72}{108} \approx 0.66$ for sample I and $f_{II} = \frac{0.079}{0.081} \approx 0.98$ for sample II. This ratio $f$ should correspond to the averaged atomic fraction of cobalt in the $SiO_2$ matrix, as it can be measured by Rutherford BackScattering experiments (RBS) [12]. Those RBS measurements were previously performed [12] on similar samples obtained by implantation of $Co^+$ ions at 160 keV with a dose of $1.10^{17}$ cm$^{-2}$ and revealed an averaged atomic fraction of cobalt in the $SiO_2$ matrix of $f_{RBS,1.10^{17}} \approx 0.13$. The present FMR experiments give $f_{FMR,1.10^{17}} \approx 0.08$. The metallic cobalt to total cobalt ratio in the $SiO_2$ matrix, $R_{FMR/RBS} = \frac{f_{FMR,1.10^{17}}}{f_{RBS,1.10^{17}}} \approx 0.61$, is in good agreement with the ratio $R_{SQUID} \approx 0.72$ that was previously determined by SQUID magnetometry on similar samples [6]. SQUID measurements suggest that part of the cobalt NPs are not metallic nanoparticles but instead oxidized nanoparticles. An alternate assumption is that part of the metallic cobalt NPs has a too slow magnetization dynamic to be detected by FMR or SQUID, within the presently investigated temperature range.

The suggested application of the present work to quantum computing requires a strong magnetic field gradient [4]. The reported FMR experiments demonstrate that the two films of magnetic nanoparticles can be modelled as effective continuous magnetic thin films. Therefore, to get a strong magnetic field gradient one may elaborate nanostructures, e.g. nanostripes, within such nano-implanted thin films. This could be done by an etching process as shown on figures 3a and 3b, following either a deep UV optical lithography or electron beam lithography processing of a resist, depending on the targeted nanostripe dimensions. Such an isolated and magnetically diluted ferromagnetic nanostripe of cobalt nanoparticles could produce the required strong magnetic field gradient as shown in figure 4 by the theoretical simulations based on magnetostatic calculations [4]. The presented calculations assume a diluted ferromagnetic nanostripe of cobalt nanoparticles which is saturated along its width, applying a magnetic field of more than 1 Tesla in this direction, as required for Q band EPR experiments on spin qubits.

The dimensions of this nanostripe are: a width $W=1000$ nm along the $z$ axis, a thickness of $T=100$ nm along the $x$ axis, corresponding roughly to the halfwidth of the cobalt ion implantation profile in the $SiO_2$ matrix [12], and an infinite length along the $y$ axis (100 microns). Its effective saturation magnetization is $M_{sat, eff}$, with $M_{sat, eff} = B_{sat, eff} = 1840$ G $= \frac{M_{sat, bulk Co}}{10}$. The figure 4a plots the dipolar magnetic field produced by this nanostripe, $B_z(x, x_{optimal} = 290 nm)$, at the optimal position $x_{optimal}$ for the qubit and as a function of the in plane position $z$ of the qubit (see figure 3 for the definition of $x$ and $z$ axis, and see below for $x_{optimal}$). Figure 4a thus demonstrates the good in-plane homogeneity of the dipolar magnetic field produced by the nanostripe in a plane placed at the distance $x_{optimal} = 290 nm$ above (or below) this nanostripe and for $z$ values close to zero. This in-plane homogeneity is investigated more quantitatively on figure 4b using the homogeneity coefficient $C(x)$ previously introduced, $C(x) = \int_{-1}^{1} dz \left( B_z(x, z) - B_z(x, 0) \right)$ (see [4]). One

![Fig. 4: Magnetostatic properties of the dipolar magnetic field produced by a diluted ferromagnetic nanostripe of cobalt nanoparticles. See text for details concerning the parameters used for the calculations.](image-url)
Fig. 5: 5a/ Simulation of the three rotational patterns of the uniform FMR-like mode of the diluted superparamagnetic nanostripe expected in the three situations qualitatively depicted on figure 3c (blue dots: shape anisotropy alone, see the text for the nanostripe dimensions), 3d (black dots: shape anisotropy and magneto-cristalline (MC) anisotropy: effective MC anisotropy field $B_{A_{MC}} = 6826 \text{ G}$ (hexagonal cobalt), obtained from the K value in [9]) and 3e (violet: shape anisotropy, with magneto-cristalline anisotropy (hexagonal cobalt): $B_{A_{MC}} = 6826 \text{ G}$, and with exchange bias anisotropy due to the antiferromagnetic shells: effective exchange bias anisotropy field $B_{A\text{EB}} = 7400 \text{ G}$, see [14]). 5b/ Simulation of the overall ESR spectrum expected for the hybrid paramagnetic qubits-superparamagnetic nanoparticles nanodevice. This hybrid nanodevice is made of electron spin qubits placed at a distance $x_{\text{optima}} = 290 \text{ nm}$ above or below the diluted superparamagnetic nanostripe of cobalt NPs described on figure 3e. The linewidth of the uniform FMR-like mode was assumed to be around 1000 Gauss, and the one of the paramagnetic resonance of the qubits around 10 Gauss. $\mu_0 M_{\text{sat,eff}} = B_{\text{sat,eff}} = 1840 \text{ G}$.

defines the optimal position $x_{\text{optima}}$ by $C(x_{\text{optima}}) = 0$. For the diluted ferromagnetic nanostripe considered here, one finds $x_{\text{optima}} = 290 \text{ nm}$. The figure 4c then plots $B_z (z = 0, x)$, which allows to determine the shift of the resonant magnetic field of the paramagnetic qubits for a given position $x$. Figure 4d shows the gradient along $x$ of the dipolar magnetic field produced by the ferromagnetic nanostripe, $\frac{dB_z(x)}{dx}$ (assuming $z=0$ and $y=0$). Figure 4d shows that one could obtain a strong magnetic field gradient having a maximum strength of more than 0.1 G/nm at the optimal position $x_{\text{optima}} = 290 \text{ nm}$ above this nanostripe. This position is also the one where this nearly one dimensional magnetic field gradient has its maximum in-plane homogeneity, as it was discussed above. $x_{\text{optima}}$ is thus the position where many electron spin qubits should be placed nearby the nanostripe for quantum computing [4]. Note that the dipolar magnetic shift of the paramagnetic resonance lines of the qubits is expected to be much smaller at $x_{\text{optima}}$ in the case of such diluted ferromagnetic nanostripe of cobalt nanoparticles, than in the case of a true bulk like nanostripe (see figure 4c). As a consequence, and also due to the broad ferromagnetic resonance mode of the cobalt nanoparticles, a spectral overlap is expected between the broad FMR mode of cobalt nanoparticles and the weakly shifted paramagnetic resonances of the electron spin qubits of the quantum nanodevice ($g=2.00$ is assumed here for the qubits). This problem may be adressed by a careful engineering of the magnetic anisotropy of the cobalt nanoparticles, as illustrated on figure 3c, 3d and 3e. The optimal strategy is illustrated by figure 3e. Figure 3c shows hexagonal cobalt nanoparticles with randomly oriented $c$ axis in the amorphous $SiO_2$ matrix (this work and [5]). Figure 3d shows a similar assembly but with NPs having their hexagonal $c$ axis oriented along the in-plane $c$ axis of an anisotropic cristalline matrix, like $Al_2O_3$ [13]. Figure 3e shows another assembly similar to the one of figure 3d but here with all NPs surrounded and exchange coupled to a shell of antiferromagnetic material, whose spins are also directed along the $c$ axis common to the cobalt nanoparticles and to the anisotropic cristalline matrix [14, 15]. A controlled annealing of the sample under an oxygen atmosphere may allow to tune the properties of the antiferromagnetic shell around the metallic cobalt nanoparticles and thus to increase their resulting in-plane magnetic anisotropy. Ion implantation of cobalt ions directly inside an anisotropic antiferromagnetic cristalline matrix followed by the same nanofabrication process could be another way to produce this strongly anisotropic diluted ferromagnetic nanostripe with an in-plane easy axis of magnetization. The increase of the
uniaxial in-plane magnetic anisotropy, from case 3c to case 3e, is well demonstrated on figure 5, which shows the rotational pattern of the uniform FMR-like mode of such superparamagnetic implanted nanostructures in those three cases. It is observed (figure 5a), that increasing the in-plane uniaxial anisotropy shifts to lower magnetic field the uniform FMR-like mode of the diluted superparamagnetic nanostructure for an in-plane (0°) applied magnetic field. The paramagnetic resonance of the electron spin qubits placed at $x_{\text{opt}}$ above the nanostructure is quite insensitive to this magnetic anisotropy engineering, whereas the FMR-like mode is much shifted towards lower fields as previously discussed. This is summarized in figure 5b. Previous calculations [4] also showed that the inter-qubits distances inside spin chains have also to be properly selected in such kind of quantum processor. The weakening of the magnetic field gradient with respect to bulk nanostructures may lead to a reduced scalability of the nanodevice. This is due to the underlying requirement that the spin-spin coupling between successive spin qubits has to be much smaller than the difference between the Zeeman energies of successive spin qubits, but not too small to perform fast enough two qubits quantum gates. This difficulty may be overcome upon strengthening the total spin-spin coupling between successive spin qubits. This may be achieved by using e.g. the zinc oxide (ZnO) semiconductor as a host matrix, both for the electron spin qubits and for the cobalt NPs. This is argued as follows: i) ZnO has the wurtzite anisotropic structure (c axis of anisotropy), ii) ZnO is a semiconductor matrix with a very weak spin orbit coupling, which can be isotopically purified, iii) ZnO is a wide direct gap semiconductor with excellent optical properties, including a very large exciton binding energy. The first argument promotes ZnO as a good host matrix for the magnetic anisotropy engineering of the cobalt NPs. Indeed, the possibility to align the magnetic anisotropy axis of cobalt NPs with the c axis of ZnO has been recently demonstrated [16]. The second argument promotes ZnO as a good host matrix for electron spin qubits, as it was previously demonstrated by pulsed EPR experiments [17, 18] and theory [19]. The third argument promotes ZnO as a good host matrix for producing an effective optically induced long range exchange coupling between electron spin qubits [20, 21], as it is required here.

In conclusion, the present FMR study demonstrates that layers of Co nanoparticles implanted in SiO2 behave as quasi-2D ferromagnetic planes. Those have a reduced effective saturation magnetization with respect to bulky films of cobalt. Magnetostatic calculations have then shown that a strong magnetic field gradient of around 0.1 G/nm could be produced by a ferromagnetic nanostructure patterned in such magnetic plane of cobalt nanoparticles. This strong magnetic field gradient combined with electron paramagnetic resonance could be useful for implementing an intermediate scale quantum computer based on arrays of coupled electron spins as discussed in a previous work [4]. This is possible as far as the magnetic anisotropy engineering of the cobalt nanoparticles allows to overcome the problem of the spectral overlap between the narrow shifted paramagnetic resonances of electron spin qubits and the broad uniform FMR-like mode of the diluted superparamagnetic nanostrin. Such magnetic anisotropy engineering of the cobalt nanoparticles could thus suppress the spin qubit decoherence process due to unwanted spin waves excitations. The price to pay for using this new design is to create over large nanometer scale distances stronger spin-spin couplings between qubits than afforded by dipole interactions. We finally suggested that this new strategy for quantum computing may be particularly well suited for exciton-mediated exchange coupled electron spin qubits in wurtzite zinc oxide.

Contributions of the authors: B.K.: performed all the FMR experiments and extracted the experimental data curves from them, produced the figure 1 and 2, commented the manuscript; D.M.: designed and fabricated the two samples of nanoparticles by ion beam synthesis methods, and commented the manuscript; P.T.: wrote the manuscript; J.T.: proposed the FMR study and the new concept of diluted magnetic nanostructure made of magnetic nanoparticles for quantum computing applications, performed all theoretical calculations and numerical simulations of the data, produced figures 3,4,5 and wrote the manuscript.

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