

Anisotropy in ferromagnetic nanoparticles: Level-to-level fluctuations of a collective effect

G. USAJ¹ and H. U. BARANGER²

¹ *Instituto Balseiro and Centro Atómico Bariloche - (8400) Bariloche, Argentina*

² *Department of Physics, Duke University - Durham, NC 27708-0305, USA*

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Abstract. – We calculate the mesoscopic fluctuations of the magnetic anisotropy of ferromagnetic nanoparticles; that is, the effect of single-particle interference on the direction of the collective magnetic moment. A microscopic spin-orbit Hamiltonian considered as a perturbation of the much stronger exchange interaction first yields an explicit expression for the anisotropy tensor. Then, assuming a simple random matrix model for the spin-orbit coupling allows us to describe the fluctuation of such a tensor analytically. In the case of uniaxial anisotropy, we calculate the distribution of the anisotropy constant for a given number of electrons, and its variation upon increasing this number by one. The magnitude of the latter scales inversely with the number of atoms in the particle and is sufficient to account for the experimental data.

In a remarkable step toward a better understanding of ferromagnetism at the nanometer scale, individual electronic states within ferromagnetic nanoparticles were recently observed using electron tunneling spectroscopy [1,2]. Several features of the tunneling excitation spectra could not be explained within the simple electron-in-a-box picture, unlike the normal metal case [3]. Although this “failure” was anticipated due to the many-body character of ferromagnetism, several particular aspects were largely unexpected: 1) the non-linear magnetic-field dependence of the energy levels close to the switching field (B_{sw} , the field at which the magnetization suddenly changes its orientation), 2) very small g-factors, and 3) excitation energies much smaller than the mean level spacing associated with the finite particle size. While we briefly return to the latter two in closing, we focus on the first effect as representative of the peculiarities of nanoscale ferromagnetism.

A non-zero B_{sw} implies the presence of a magnetic-anisotropy energy barrier: the (large) magnetic moment of the particle points in a preferred direction. This preference originates from spin-orbit coupling and/or the shape of the grain. At B_{sw} , there is an abrupt change in the grain’s total energy, leading to “jumps” in the tunneling spectrum. References [1,2,4] show that such jumps can be described by a simple uniaxial anisotropy model: $\hat{H}_A = -k \hat{S}_z^2/S$, where k is the anisotropy constant—which is an *intensive* quantity in the bulk— S is the total spin and \hat{z} is the anisotropy axis. However, to account for the non-linear behavior and the magnitude of the jumps, it was crucial to *assume* that k depends on both the number of electrons in the nanoparticle *and* the particular many-body states involved in each electronic transition. A change of 1–3% upon adding one electron in a nanoparticle with roughly

1000 atoms was enough to account for the experiments. Although fluctuation of electronic properties is expected at the nanoscale, a microscopic connection to anisotropy is needed.

In the last years, several works have aimed to understand the origin of the additional resonances [2, 4–7] or the fluctuation of the anisotropy [8, 9]. While the former remains controversial, the latter seems to be accounted for (in nanoparticles with up to 260 atoms) by the self-consistent numerical calculation done in ref. [8]. One may wonder, however, if a simpler model, suitable for an analytic treatment of larger nanoparticles, can give similar results while providing additional physical insight.

In this work, we first derive an expression for the magnetic-anisotropy tensor in terms of the matrix elements of the spin-orbit (SO) Hamiltonian using a simple Hamiltonian model for the ferromagnetic nanoparticle. Then, by using random matrix theory (RMT) to account for the fluctuations of the SO matrix elements, we calculate the probability distribution of the magnetic-anisotropy constant for the case of uniaxial anisotropy. For nanoparticles with $N_a \simeq 1000$ atoms, we find that fluctuations of 1% are typical. Furthermore, the fluctuations scale inversely with N_a so that our results agree with those of ref. [8] for smaller nanoparticles.

Ferromagnetism originates in the electron-electron interaction —in particular, the exchange interaction. The simplest model Hamiltonian, then, reads

$$\hat{H} = \hat{H}_S + \hat{H}_Z + \hat{H}_{SO} = \sum_{m,\sigma} \varepsilon_m \hat{n}_{m\sigma} - J_S \vec{S}^2 + \hat{H}_Z + \hat{H}_{SO}, \quad (1)$$

where $\{\varepsilon_m\}$ are the single-particle energies, \vec{S} is the total spin operator, and J_S is the exchange constant. The latter produces the large spin of the nanoparticle (typically $S \approx 1000$ [1]); notice that this model assumes that the magnetic moment is coherent and uniform [8]. The second and third terms in eq. (1) describe the Zeeman energy, $H_Z = -g\mu_B \vec{B} \cdot \vec{S}$, and the SO coupling,

$$\hat{H}_{SO} = i \sum_{n,m} \sum_{\sigma,\sigma'} (\vec{A}_{nm} \cdot \vec{\sigma}_{\sigma\sigma'}) c_{n\sigma}^\dagger c_{m\sigma'}, \quad (2)$$

respectively. \hat{H}_{SO} is responsible for the magnetic anisotropy —we neglect the contribution of the shape anisotropy (magnetostatic) as its relative contribution decreases for smaller particles and so is very small at the nanoscale [10]. Here, $\vec{\sigma}$ is a vector of Pauli matrices, and

$$\vec{A}_{nm} = -i \frac{\xi}{2} \sum_j \sum_{\mu,\nu} \langle \mu | \vec{L}_j | \nu \rangle \psi_n(j, \mu) \psi_m(j, \nu), \quad (3)$$

where j labels the atomic sites and μ, ν the orbital bands (s, p, d). $\psi_n(j, \mu) = \langle j\mu | \psi_n \rangle$ is the n -th single-particle wave function, and \vec{L}_j is the angular momentum with respect to site j . The parameter ξ characterizes the SO magnitude; it is of order 80 meV for cobalt [8]. The SO matrix elements satisfy $\vec{A}_{nm} = -\vec{A}_{mn}$ due to time-reversal symmetry. We consider the case where both the exchange energy and the single-particle level spacing are large compared with the SO matrix elements (true for small enough nanoparticles) and treat \hat{H}_{SO} as a perturbation. In addition, because cobalt nanoparticles apparently have a fcc structure [10], we do not include here the crystalline magnetic anisotropy of bulk hcp cobalt.

We start by deriving an effective spin Hamiltonian for the nanoparticle. In the absence of SO coupling both the occupation numbers and the total spin are good quantum numbers. In its presence, only states with different occupation numbers are mixed (recall \hat{H}_{SO} is fully off-diagonal, $\vec{A}_{nn} = 0$). Therefore, it is natural to write an effective Hamiltonian restricted to a subspace of fixed occupation which includes virtual transitions out of such subspace due

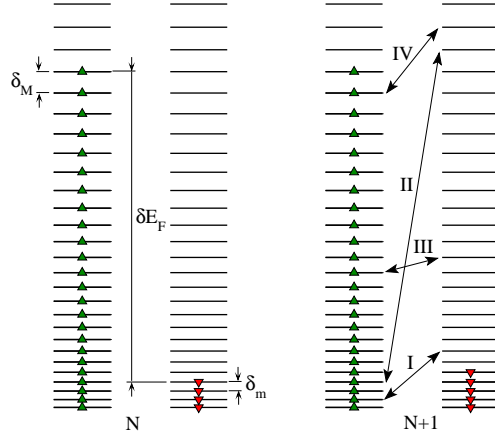


Fig. 1 – The single-particle energy levels of \hat{H}_S for N and $N + 1$ electrons. The arrows point out different classes of transitions induced by the SO coupling, labeled I, II, III, and IV. Here δ_m (δ_M) is the mean level spacing for the minority (majority) spins at the Fermi energy. δE_F is the single-particle energy difference between the highest occupied levels of the majority and minority spins.

to SO coupling (up to second order). This is done by a suitable canonical transformation, $\hat{H}_{\text{eff}} = \exp[-i\hat{U}]\hat{H}\exp[i\hat{U}]$. Let $\{|q\rangle\}$ denote the eigenstates of \hat{H}_S , $\hat{H}_S|q\rangle = E_q|q\rangle$. We choose \hat{U} such that $U_{q'q} = 0$ if $|q\rangle$ and $|q'\rangle$ have different occupation numbers and

$$U_{q'q} = i\frac{V_{q'q}}{(E_{q'} - E_q)} + B_{q'q} \quad (4)$$

if they do not [11]. Here $V_{q'q} \equiv (\hat{H}_{\text{SO}})_{q'q}$ and $B_{q'q}$ is proportional to \hat{H}_Z and \hat{H}_{SO} . As for the case of normal metal grains [12,13], the latter leads to a correction to the g-factor which we absorb into the Zeeman term. Then, to leading order

$$(\hat{H}_{\text{eff}})_{q'q} = E_q \delta_{q'q} + (\hat{H}'_Z)_{q'q} + \frac{1}{2} \sum_{q''} V_{q'q''} V_{q''q} \left(\frac{1}{E_q - E_{q''}} + \frac{1}{E_{q'} - E_{q''}} \right) \quad (5)$$

if $|q\rangle$ and $|q'\rangle$ have the *same* occupation numbers, and $(\hat{H}_{\text{eff}})_{q'q} = 0$ if not. Using the fact that the exchange interaction is very large ($\delta E_F \gg V_{q'q}$), eq. (5) can be further restricted to a subspace of a given total spin S . After using the Wigner-Eckart theorem [14] and some tedious algebra, we finally obtain the following spin Hamiltonian:

$$\hat{H}_{\text{eff}} = \hat{H}_S + \hat{H}'_Z + \sum_{\alpha, \beta=x,y,z} \hat{S}_\alpha \mathcal{K}_{\alpha\beta} \hat{S}_\beta / S, \quad (6)$$

with an explicit expression for the anisotropy tensor \mathcal{K} given below. Note that while \hat{H}_{eff} and \hat{H} have the same eigenenergies, they do not have the same eigenvectors — in fact, $|E\rangle = \exp[i\hat{U}]|E_{\text{eff}}\rangle$. Therefore, while the eigenstates of eq. (6) have well-defined spin and occupation numbers, this is *not* the case for the eigenstates of the physical Hamiltonian (1), as expected due to SO coupling. The spin selection rules for electron tunneling are thus less restrictive than for a state with well-defined spin.

We classify the transitions induced by the SO coupling in four types (see fig. 1): from doubly occupied levels to singly occupied (I) or empty (II) levels, and from singly occupied to

singly occupied (III) or empty (IV) levels. With this notation, the anisotropy tensor reads

$$\begin{aligned} \mathcal{K}_{\alpha\beta} = & \sum_{m>n} A_{nm}^\alpha A_{nm}^\beta \left(\frac{-2J_S(\delta_{\text{I}}+\delta_{\text{IV}})}{(\varepsilon_n-\varepsilon_m)(\varepsilon_n-\varepsilon_m-2J_S S)} - \left[\frac{4J_S S}{(\varepsilon_n-\varepsilon_m)^2-4J_S^2 S^2} \right] \frac{2\delta_{\text{III}}}{(2S-1)} + \right. \\ & \left. + \left[\frac{1}{\varepsilon_n-\varepsilon_m} \frac{2}{(S+1)} - \frac{1}{\varepsilon_n-\varepsilon_m-2J_S S} \frac{2}{(2S+1)} - \frac{1}{\varepsilon_n-\varepsilon_m+2J_S(S+1)} \frac{2S}{(S+1)(2S+1)} \right] \delta_{\text{II}} \right), \end{aligned} \quad (7)$$

where δ_X , $X = \text{I, II, III, IV}$, restricts the sum to the appropriate set of transitions. It is clear from this equation that states with different occupation numbers will have different anisotropy tensors, leading to level-to-level fluctuations of the anisotropy constant. *We then conclude that the addition of an electron to the nanoparticle will produce a different anisotropy tensor depending on the new occupied level.*

So far we have not specified any features of the SO coupling: eq. (7) is valid provided only that the SO coupling is small. A detailed treatment of the anisotropy tensor for a particular grain geometry would require the calculation of the SO matrix elements using, for instance, a non-interacting tight-binding model or a more sophisticated self-consistent Hartree-Fock approach [8]. However, our goal here is to calculate the generic features of the fluctuations of the anisotropy tensor. Since the shape of the grain is assumed to be irregular, we expect the SO matrix elements to fluctuate from level to level due to the fluctuations of the single-particle wave functions. The usual RMT approach of describing the single-particle fluctuations with the Gaussian unitary ensemble then implies that the $\{\bar{A}_{nm}\}$ are Gaussian uncorrelated random variables because of the sum over a large number of terms in their definition equation (3). This assumption of randomness of the SO matrix elements is consistent with the results of ref. [8]. On the other hand, we know from refs. [2,4] that a uniaxial anisotropy is needed to describe the main features of the non-linear behavior in the experiment. Therefore, we assume uniaxial anisotropy *on average*: $\langle A_x^2 \rangle = \langle A_y^2 \rangle$ and $\langle A_z^2 \rangle < \langle A_y^2 \rangle$. In a more detailed calculation, this asymmetry would probably come as a difference in the energy dependence of the SO matrix elements (this is the case in ref. [8]); in our model, however, it can only appear as a difference in the standard deviations. The case of pure ‘‘mesoscopic anisotropy’’ —*i.e.* when all $\langle A_i^2 \rangle$ are equal— was recently analyzed in ref. [9]; it was found that for $N_a > 100$ mesoscopics alone cannot explain the full magnitude of the anisotropy, thus indirectly supporting our insertion of an additional uniaxial anisotropy. Since we do not expect $\langle A_z^2 \rangle$ to be much smaller than $\langle A_x^2 \rangle$, we take $\langle A_z^2 \rangle = \langle A_x^2 \rangle / 2$; our results are only weakly sensitive to this choice (see below).

In the basis where \mathcal{K} is diagonal, the anisotropy Hamiltonian takes the form

$$\hat{H}_A = \frac{k_1 + k_2}{2S} \hat{S}^2 + \left(k_3 - \frac{k_1 + k_2}{2} \right) \hat{S}_3^2 / S, \quad (8)$$

where $\{k_1, k_2, k_3\}$ are the eigenvalues of \mathcal{K} and $\hat{S}_3 = \vec{S} \cdot \vec{k}_3$. The first term is a small correction to the exchange constant and can be neglected. The second term represents the uniaxial anisotropy. We have verified that for the values used below for the SO matrix elements, $\langle \hat{z} \cdot \vec{k}_3 \rangle \simeq 0.99$, so that $\hat{H}_A \simeq -k_N \hat{S}_z^2 / S$ with $k_N = (k_1 + k_2) / 2 - k_3$. The fluctuations of k_N are calculated by numerically diagonalizing \mathcal{K} for each realization of $\{A_{nm}^\alpha\}$. No fluctuation is introduced for the energy levels—they are taken to be locally equally spaced, in such a way that they reproduce the (hcp) Co density of states.

The distribution of k_N for a nanoparticle containing $N_a = 1200$ atoms is shown in fig. 2, where we have used the following values for the parameters: $\delta E_F = 2 \text{ eV}$, $\delta_M = 3.7 \text{ meV}$,

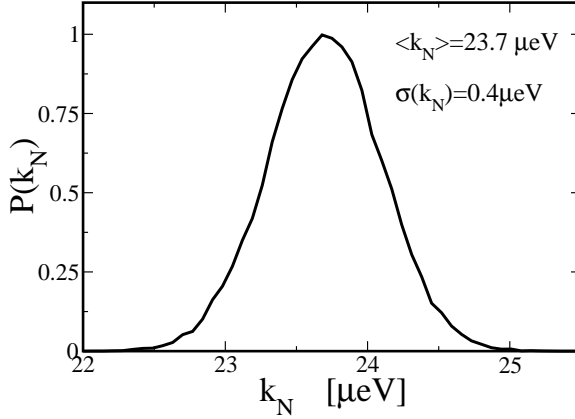


Fig. 2 – Probability distribution of the magnetic-anisotropy constant for a nanoparticle size of $N_a = 1200$. The distribution is Gaussian with $\langle k_N \rangle = 23.7 \mu\text{eV}$ and $\sigma(k_N) = 0.4 \mu\text{eV}$.

$\delta_m = 1.15 \text{ meV}$, $J_S = 0.95 \text{ meV}$, and $\sigma(A_x) = 0.32 \text{ meV}$. The latter is estimated from eq. (3) using random and uncorrelated wave functions, $\langle A_x^2 \rangle = [\xi^2/27N_a](N_s/N_a)$ —the additional factor N_s/N_a , with N_s the number of atoms at the surface, is discussed below. The distribution is Gaussian with a mean value $\langle k_N \rangle = 23.7 \mu\text{eV}$ and a standard deviation $\sigma(k_N) = 0.4 \mu\text{eV}$, *i.e.* $\sigma(k_N)/\langle k_N \rangle \simeq 2\%$. These values can be estimated from eq. (7) by replacing the sums by integrals and considering only the diagonal terms of \mathcal{K} . We then get

$$\langle k_N \rangle \simeq a [\langle A_x^2 \rangle - \langle A_z^2 \rangle] \frac{J_S}{\delta_m^2}, \quad (9)$$

$$\sigma(k_N) \simeq \sqrt{4b} \left[(\langle A_x^2 \rangle^2 + 2\langle A_z^2 \rangle^2) \ln \left(\frac{\delta E_F}{\delta_m} \right) \right]^{\frac{1}{2}} \frac{J_S}{\delta_m \delta E_F}, \quad (10)$$

where a and b are numerical factors that depend on the shape of the density of states. As expected, the fluctuations go to zero in the bulk limit, $\sigma(k_N)/\langle k_N \rangle \propto \delta_m/\delta E_F \propto N_a^{-1}$ up to a logarithmic factor. We also note that in bulk $\langle A_x^2 \rangle \propto N_a^{-1}$, so that $\langle k_N \rangle$ is independent of the system's size. However, since we assume the anisotropy comes from the breaking of symmetry at the surface of the nanoparticle, we take $\langle A_x^2 \rangle \propto N_s/N_a^2$, so that $\langle k_N \rangle \propto N_s/N_a$ [10]. For Co, we have $a \simeq 0.8$ and $b \simeq 1.6$. Evaluation of eqs. (9) and (10) gives $\langle k_N \rangle \simeq 29 \mu\text{eV}$ and $\sigma(k_N) \simeq 0.4 \mu\text{eV}$, in good agreement with the numerical data.

Let us now calculate the change of the anisotropy constant upon the addition of one electron with the minority spin (right panel in fig. 1). We calculate k_{N+1} as before but take into account that both k_N and k_{N+1} contain contributions from the *same* SO matrix elements. We obtain a Gaussian distribution for $\delta k_N = k_{N+1} - k_N$, with $\langle \delta k_N \rangle \simeq -0.03 \mu\text{eV}$ and $\sigma(\delta k_N) = 0.2 \mu\text{eV}$. Therefore, for a nanoparticle containing $N_a \simeq 1200$ atoms, $\sigma(\delta k_N)/\langle k_N \rangle \approx 1\%$. *This is of the order needed to account for the experimental data of ref. [2].*

Figure 3 shows $\sigma(\delta k_N)/\langle k_N \rangle$ as a function of N_a . The results for the experimental data and the self-consistent calculation are indicated by the square and the diamonds, respectively. It is important to emphasize that once the ratio $\langle A_z^2 \rangle/\langle A_x^2 \rangle$ is fixed, and so is $\langle k_N \rangle$, there are no free parameters left to fit the fluctuations. Thus, the fact that the relative fluctuation of the anisotropy constant agrees reasonably well with both the experimental data and the results of a self-consistent numerical study is quite remarkable and indicates that our simple model is able to capture the essential physics.

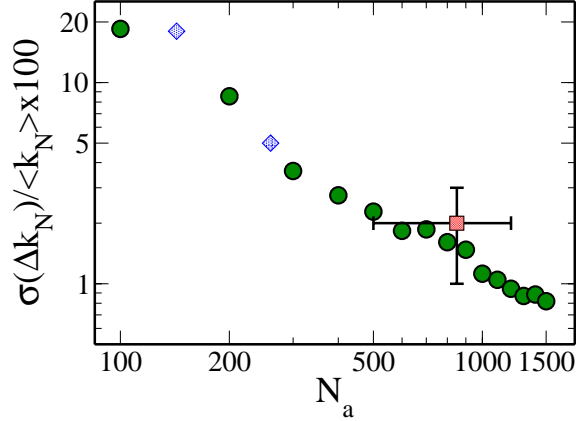


Fig. 3 – Relative fluctuation of the anisotropy constant upon the addition of one electron as a function of the numbers of atoms in the nanoparticles. Fluctuations are of the order of 1% for $N_a \simeq 1000$. Also shown are results of experiment (square, size of error bar corresponds to nanoparticles of 3–4 nm diameter) [2, 16] and of a self-consistent calculation (diamonds) [8].

Following the same procedure as above, we estimate

$$\langle \delta k_N \rangle \simeq c [\langle A_x^2 \rangle - \langle A_z^2 \rangle] \frac{J_S}{\delta_m \delta E_F}, \quad (11)$$

$$\sigma(\delta k_N) \simeq \frac{2J_S}{\delta_m \delta E_F} \left(2[\langle A_x^2 \rangle^2 + 2\langle A_z^2 \rangle^2] \frac{1.8\delta_m - 1.4J_S}{\delta_m - J_S} \right)^{\frac{1}{2}}, \quad (12)$$

where c contains logarithmic corrections in $\delta E_F/\delta_m$. In our case $c \simeq -3.5$, which gives $\langle \delta k_N \rangle \simeq -0.07 \mu\text{eV}$ and $\sigma(\delta k_N) \simeq 0.3 \mu\text{eV}$. Note that

$$\frac{\sigma(\delta k_N)}{\langle k_N \rangle} \propto \frac{\delta_m}{\delta E_F} \propto \frac{1}{N_a} \quad (13)$$

also decays to zero in the bulk limit. We notice that the ratio $\sigma(\delta k_N)/\langle k_N \rangle$ is *independent* of the value of $\langle A_x^2 \rangle$ and only weakly dependent (up to a factor 2) on the ratio $\langle A_z^2 \rangle/\langle A_x^2 \rangle$ if the latter is $\lesssim 0.5$. The same is true for the relative fluctuation of the ground-state anisotropy, $\sigma(k_N)/\langle k_N \rangle$. Consequently, our estimate of the magnitude of the fluctuations is quite robust, being largely controlled by the ratio $\delta_m/\delta E_F$.

It is straightforward to check from eqs. (11) and (12) that $\sigma(\delta k_N) \gg \langle \delta k_N \rangle$. This implies that k_{N+1} can be *larger or smaller* than k_N with roughly equal probability. While this is a key point for a correct account of the experimental data [2], it also has an interesting consequence if the dwell time for electrons in the nanoparticle is larger than the time it takes for the magnetization to switch. In that case, it should be possible to switch the magnetic moment of the nanoparticle by first tuning the magnetic field just below B_{sw} for the ground state and then increasing the bias voltage until a high-energy state with a smaller anisotropy constant is excited. The reverse procedure should present hysteresis. Gated nanoparticles should show similar effects as a function of the gate voltage.

Finally, let us briefly comment on the other two features of the experimental data mentioned in the introduction. First, as noted above, $B_{q'q}$ gives a correction to the g-factor (cf. eq. (4)). As for the magnetic anisotropy, this correction will depend on the particular

electronic state of the nanoparticle. We note that even very small fluctuations will strongly modify the effective g-factor,

$$g_{\text{eff}} \simeq \frac{|g_{N-1}S_{N-1} - g_N S_N|}{|\Delta S|} \approx g_N \left(1 + \frac{S}{\Delta S} \frac{\delta g_N}{g_N} \right) \quad (14)$$

with $\delta g_N = g_N - g_{N-1}$ and $\Delta S = S_N - S_{N-1}$. For instance, $\delta g_N/g_N \simeq 10^{-4}$, $S \simeq 1000$ and $\Delta S = -1/2$ can lead to the small g_{eff} observed experimentally (this requires $\delta g_N/\Delta S < 0$, which should be verified). Second, regarding the excitation spectrum, the experimental data [16] suggest that the low-energy spacing is roughly independent of the grain's size [17]. The only quantity with such scaling is the anisotropy constant. Then, resonances should be related to the excitation of collective modes (magnetization) [7] along an axis with a much larger anisotropy (hard axis) —this is crucial since the smallness of B_{sw} implies that the anisotropy along the easy axis is also small. We believe such resonances are described by a more general form of the anisotropy, such as $\hat{H}_A = -kS_z^2 + k'S_x^2$. Work in this direction is in progress.

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