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Solid State Communications 119 (2001) 253–258

solid  
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communications

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# Quantum description of ferromagnet metal nanoparticles

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## Abstract

The quantum physics of normal metal nanoparticles can be understood simply by recognizing the discreteness of individual electron eigenenergies and the dominant role played by Coulomb interactions in determining the rate at which the chemical potential increases with electron number. Ferromagnetic metal nanoparticles, on the other hand, have collective magnetization degrees of freedom that are responsible for additional low energy excitations. We discuss some issues that arise in attempting to achieve a unified and consistent quantum description of both collective and quasiparticle physics in magnetic metal nanoparticles, especially when acknowledging the essential role of spin–orbit coupling and the magnetic anisotropy it produces. © 2001 Elsevier Science Ltd. All rights reserved.

PACS: 75.50.Mn; 75.30.Gw; 75.10.Lp

Keywords: A. Nanostructures; D. Spin–orbit effects; A. Metals

## 1. Introduction

Interest in the properties of nanometer scale magnetic particles has grown in recent years, in part because of advances in preparation techniques and in part because of their potential importance for future advances in magnetic information storage technology. It is now possible to reliably realize and study nanoparticles containing of order 1000 atoms using a variety of techniques [1–3]. Fine magnetic particles have usually been described using a classical theory in which the energy is a functional of the magnetization orientation distribution and the anisotropy barriers that separate different easy orientations play a key role. At the nanometer scale, however, the discreteness of the particle's quantum energy spectrum starts to become observable. In this article, we will discuss some issues that arise in attempting to develop a complete theory of the many-particle Hamiltonian spectrum of a ferromagnetic metal nanoparticle. For normal metal nanoparticles, there has already been considerable progress. Comparison with experiment has established a Fermi-liquid like picture in which the many-particle spectrum can be understood using a single-particle language, as illustrated schematically in Fig. 1. Interactions are essential only in understanding the

way in which the ground state energy  $E_0$  changes with total electron number  $N$ . Fig. 1 would suggest that  $[E_0(N+2) - E_0(N+1)] - [E_0(N+1) - E_0(N)] = E_0(N+2) + E_0(N) - 2E_0(N+1) \sim \delta$ , where  $\delta$  is the quasiparticle level spacing. In fact it turns out that  $E_0(N+2) + E_0(N) - 2E_0(N+1) \sim E_{cb}$ , where  $E_{cb}$  is known as the Coulomb blockade energy. Because of the long range of the Coulomb interaction  $E_{cb} \sim e^2/R$  (where  $R$  is the nanoparticle radius), much larger than  $\delta \sim 1/D(\epsilon_F)V$  (where  $V$  is the nanoparticle volume) for particles containing many atoms. Here  $D(\epsilon_F)$  is the bulk density-of-states per unit volume. The existence of the Coulomb blockade has many important consequences [4] and the physics of quasiparticle levels is interesting [5–7], but for the most part it is the physics of non-interacting electrons. The quantum physics of ferromagnetic metal nanoparticles includes that of normal metals, but is far more interesting because of the importance of the collective magnetization orientation degree of freedom. The first experimental studies [1] of the many-body spectrum of a ferromagnetic metal nanoparticle have yielded results that are not fully understood and have highlighted the lack of an adequate language for discussing this question. In this article, we discuss one possible approach. In Section 2, we briefly review elements of the classical theory of a fine ferromagnetic particle that will be important in the following discussion. Section 3 introduces a microscopic toy model for a ferromagnetic

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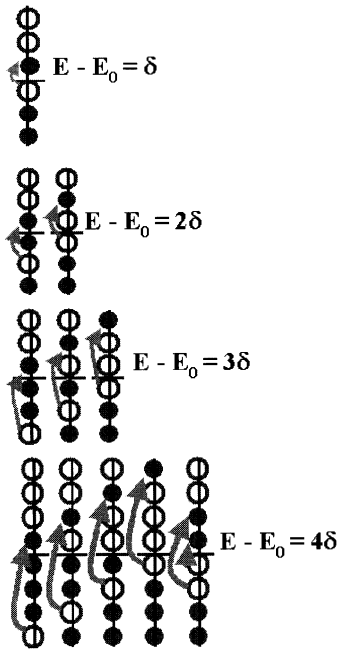


Fig. 1. Schematic illustration of the many-particle excitation spectrum at fixed particle number for the case of quasiparticles whose energy separation at the Fermi level is  $\delta$ . As in Landau's theory of a Fermi liquid, it is assumed that there is a one-to-one mapping between interacting and non-interacting electron states. The latter are labelled here by quasiparticle occupation numbers with full circles corresponding to occupied states and empty circles corresponding to empty states. The solid line separates quasiparticles that are occupied in the ground state, from those that are empty. For  $E - E_0 = \delta E \gg \delta$  the number of many-particle excitations grows rapidly.

metal nanoparticle that is not fully realistic, but can still be used to make some useful points about its quantum physics. The essential role of spin-orbit coupling is discussed in Section 4. Finally in Section 5, we sketch a path-integral approach that allows spin-orbit interactions to be incorporated. Finally in Section 6, we discuss how this language can be used to make the connection between classical and quantum physics more apparent and offer some speculations about the low-energy quantum spectrum.

## 2. Classical single-domain physics

A magnetic system is usually described classically by its micromagnetic energy functional, which relates energy to spatial variations in spontaneous magnetization orientation. The phenomenological functional includes an exchange term that favors spatially constant orientations, an anisotropy term that specifies the easy orientation axes and the barriers that separate them, and a magnetostatic term that originates microscopically from interactions between magnetic dipoles and is responsible for domain formation

in large magnetic particles. When applied to fine particles that are approximately spherical, the micromagnetic description simplifies because magnetostatic interactions become negligible [8] and the exchange energy cost of orientation variation inside the particle become prohibitive. The system is then described by a single global orientation degree of freedom. In the simplest version of this model [9,10], the energy of the nanoparticle in an external field  $H$  is

$$E = V[K \sin^2(\theta) - \mu_0 M_s H \cos(\vartheta - \theta)], \quad (1)$$

where  $\theta$  is the angle between the magnetization and the easy axis and  $\vartheta$  is the angle between the field direction and the easy axis. Here  $K$  is the single coefficient that appears in the simplest possible phenomenological model of a ferromagnet with uniaxial anisotropy, and  $M_s$  is the saturation magnetization of the material. For this model the easy magnetization directions are  $\theta = 0$  and  $\theta = \pi$ . For sufficiently weak fields  $H$ , there are two local minima in the orientation dependence of the energy. The energy barrier between these minima is proportional to the particle volume and is typically much larger than the thermal energy  $k_B T$ , enabling magnetic information storage. The limits  $\vartheta = 0$  and  $\vartheta = \pi/2$  (field along the easy axis and field perpendicular to easy axis) are particularly easy to understand. For fields along the easy axis, the  $\theta = \pi$  (magnetization down) configuration is metastable for fields smaller than  $H_a = 2K/\mu_0 M_s$ , while the  $\theta = 0$  (magnetization-up) configuration is metastable for  $H > -H_a$ . For  $|H| < H_a$ , both configurations are metastable and separated by an energy barrier. For the special case of fields along the hard-direction ( $\vartheta = \pi/2$ ) only one configuration is stable and the magnetization is gradually twisted from easy to hard directions. An elementary calculation shows that in this case,  $\sin(\theta) = H/H_a$  for  $|H| < H_a$  while the magnetization is along the field direction at stronger fields. Nanoparticle with volumes small enough that barriers are comparable to or smaller than  $k_B T$  are said to be superparamagnetic. Larger volume particles will stay in a metastable state for a long time.

This simple model captures the essence of hysteresis in ferromagnets, but is seldom fully realistic even for small particles. Even at a classical level, the physics of a typical magnetic nanoparticle is much more complex, often because of changes in bonding at its surface. Saturation moments, easy magnetization directions, and anisotropy coefficients all tend to be different at the surface than they are in bulk material [11]. We will ignore these sample specific complications entirely in the following. What we seek here is an understanding of how the quantum many-body spectrum of a ferromagnetic metal nanoparticle evolves with external field as the magnetic state of a fine particle is being manipulated by an external field, or at least a convenient language for discussing this issue. It is important to draw a clear distinction between ferromagnetic insulators, for which the only low-energy degrees of freedom is the collective spin-orientation, and ferromagnetic metals, which have

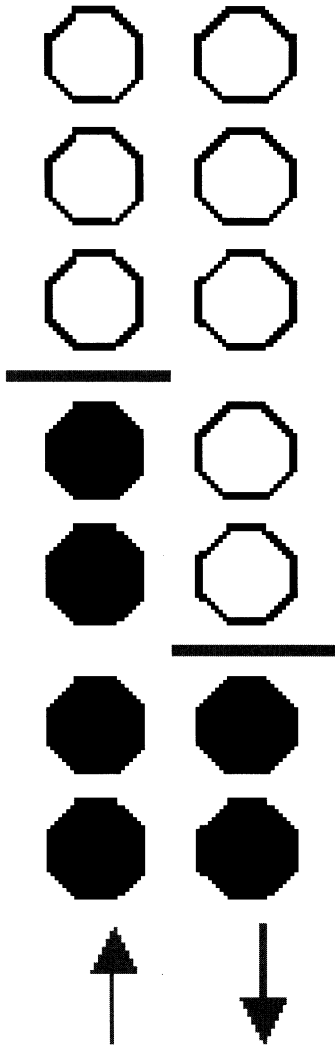


Fig. 2. Schematic illustration of the toy model ground state. States are specified by the list of doubly and singly occupied orbitals and by spin quantum numbers. In the ground state orbits go from doubly occupied to singly occupied to empty with increasing energy. Given a set of orbital occupations with  $N_s$  singly occupied orbitals, only the spin multiplet with  $S_0 = N_s/2$  is relevant at low energies. The degeneracy of this multiplet will be lifted by spin-orbit interactions. Other low energy multiplets are obtained by changing orbital occupation numbers near the double-to-single transition (minority spin Fermi energy) and the single-to-empty (majority spin Fermi energy) transitions indicated here by solid lines. These changes can include ones which alter  $N_s$  and  $S_0$ .

particle-hole excitations in addition. The situation in bulk metals is illustrated schematically in Fig. 3. The nanoparticle equivalent of these particle-hole continua is discrete excitation spectra like those illustrated in Fig. 2. The common practice of modeling a magnetic particle by a spin Hamiltonian, completely misses this aspect of metal physics. It will be necessary to include both types of spin-

excitations; unlike the bulk case, there is no wavevector conservation to assist with their separation. The idealized microscopic model of a ferromagnetic particle discussed in the next section will enable us to make some useful observations about many-particle spectra, but does not include spin-orbit interactions. Since spin-orbit interactions tend to dominate the magnetic anisotropy of small magnetic particles, and magnetic anisotropy is what controls the dependence of the system's state on external fields, the proper inclusion of spin-orbit interactions, addressed in the following sections, is at the heart of the problem we wish to address.

### 3. Toy model

Using Fermi statistics, interactions between electrons can be written quite generally [12] in a form which explicitly exhibits the ferromagnetic exchange interaction between their spins which leads to ferromagnetism. Our toy model [13] simplifies the physics of a ferromagnetic metal nanoparticle by including only these interactions, and by assuming identical exchange constants between all pairs of single-particle orbitals in the particle:

$$\mathcal{H} = \sum_{j,\sigma} c_{j,\sigma}^\dagger c_{j,\sigma} \epsilon_j - \frac{U}{8} \sum_{j,k} \sum_{s,s',t,t'} c_{j,s'}^\dagger \vec{\tau}_{s',s} c_{j,s} \cdot c_{k,t'}^\dagger \vec{\tau}_{t',t} c_{k,t}$$

$$= H_{1b} + V. \tag{2}$$

In Eq. (2),  $\epsilon_j$  is a nanoparticle orbital energy and  $\vec{\tau}$  is the Pauli spin matrix vector. The single particle orbitals will have an average spacing inversely proportional to the volume of the nanoparticle and are expected to exhibit spectral rigidity [6,7]. The one-body term in this Hamiltonian,  $H_{1b}$ , should be thought of as including a mean-field approximation to those portions of the interaction not captured by the exchange term  $V$ . The many-particle spectrum of this Hamiltonian follows readily from the following observations: (i) the total occupation of each orbital is a good quantum number, (ii) the interaction term is proportional to the square of the total electron spin operator  $\vec{S}_{tot}$ . The  $2^{N_s}$  states with a given set of  $N_s$  singly occupied orbitals have their band energy degeneracy lifted by the interaction energy  $-(U/2)S_{tot}(S_{tot} + 1)$ , where the total spin  $S_{tot}$  has a maximum value  $N_s/2$ . We show below that only this  $N_s + 1$  one-fold degenerate spin-multiplet is relevant to the low energy physics of a ferromagnetic nanoparticle. In the absence of spin-orbit interactions, spin coherent states [16] constructed in this manifold correspond to the classical magnetization configurations.

To understand the excitation spectrum of this model Hamiltonian, we start by considering the ground state of a ferromagnetic metal nanoparticle. The transition points at which orbitals go from being doubly occupied to being singly occupied and from singly occupied to empty are, in more conventional mean-field language, the majority and

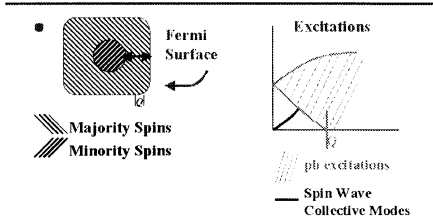


Fig. 3. Schematic Fermi surface and spin-flip excitation spectrum of a metallic ferromagnet. The majority spin Fermi surface has larger area than the minority spin Fermi surface. Gapless particle–hole excitations are possible around both Fermi surfaces. Gapless spin-flip particle–hole excitations are also possible for wavevectors exceeding the minimum  $k$ -space separation between majority and minority spin Fermi surfaces. The right panel schematically indicates the spin-flip excitation spectrum. The spin-wave collective modes are weakly damped at long-wavelengths.

minority spin quasiparticle Fermi energies, illustrated schematically in Fig. 3. We take these to have the values  $\epsilon_{Fa}$  and  $\epsilon_{Fi}$  at a reference total particle number and zero magnetic field. We are interested in how the nanoparticle state evolves as a function of external field and gate voltage. We assume, for simplicity, that the quasiparticle energy levels of majority and minority spins are equally spaced near their respective Fermi energies. It follows from the considerations of the previous paragraph that the total energy, relative to that of the reference state, is

$$\begin{aligned} \delta E = & [\epsilon_{Fa} - \Delta/2 + \delta_a/2]\delta N_a + [\epsilon_{Fi} + \Delta/2 + \delta_i/2]\delta N_i \\ & + E_{cb}(\delta N_a + \delta N_i)^2/2 + \frac{\delta N_a^2}{2}(\delta_a - \Delta/2(2S_0 + 1)) \\ & + \frac{\delta N_i^2}{2}(\delta_i - \Delta/2(2S_0 + 1)) + \delta N_a \delta N_i \frac{\Delta}{2(2S_0 + 1)} \\ & - (V_g + g\mu_B B/2)\delta N_a - (V_g - g\mu_B B/2)\delta N_i. \end{aligned} \quad (3)$$

In this energy expression, we have added by hand the electrostatic Coulomb blockade term, which depends only on the total number of particles in the grain. The terms proportional to  $\Delta$  in Eq. (3) originate from the interaction energy  $-US_{tot}(S_{tot} + 1)/2$ ; the notational change  $U \rightarrow \Delta/(S_0 + 1/2)$  is motivated by the identification, explained below, of  $\Delta$  as the spin-splitting energy of the quasiparticle bands. Here  $S_0 = (N_{a0} - N_{i0})/2$  is the ground state total spin of the reference state and  $S_{tot} = S_0 + (\delta N_a - \delta N_i)/2$ . Note that for a ferromagnetic particle  $S_0$  will be proportional to volume.

Stability of the reference system ground state requires that its energy increases at fixed total particle number  $N$  both for  $S_{tot} \rightarrow S_0 + 1$  ( $\delta N_a = 1$ ,  $\delta N_i = -1$ ) and  $S_{tot} \rightarrow S_0 - 1$  ( $\delta N_a = -1$ ,  $\delta N_i = +1$ ). From this it follows that

$$[\epsilon_{Fa} - \epsilon_{Fi} - \Delta + (\delta_a - \delta_i)/2] < [\delta_a + \delta_i - 2\Delta/(2S_0 + 1)]. \quad (4)$$

Since the RHS of Eq. (4)  $\sim N_A^{-1}$ , where  $N_A$  is the number of atoms in the grain, it follows that  $\Delta = \epsilon_{Fa} - \epsilon_{Fi}$  to within a fluctuating mesoscopic correction, i.e. that  $\Delta$  is the quasiparticle spin-splitting. The energy difference between the  $S_{tot} = (N_a - N_i)/2$  states retained in our considerations and the  $S_{tot} \leq (N_a - N_i)/2 - 1$  states we have discarded is  $\Delta 2S_0/(2S_0 + 1) \sim \Delta$ , well outside the energy range of interest. It follows that in the absence of spin–orbit coupling, the magnetization orientation degree of freedom appears quantum mechanically as a macroscopically degenerate spin multiplet associated with every orbital total-occupation-number configuration.

#### 4. Spin–orbit coupling: perturbation theory considerations

As mentioned above, the most interesting physics in ferromagnetic metal nanoparticles is due to spin–orbit interactions and the magnetic anisotropy they produce. Spin–orbit coupling is a relatively weak effect in the 3d transition metal ferromagnets. The energy scale [15]  $\xi_d$  that characterizes the coupling between spin and orbital degrees of freedom is in the range between 50 and 100 meV, less than 10% of the d-band width  $W_d$  in bulk materials. The expectation value of the single-particle spin–orbit interaction operator  $H_{so}$  in non-relativistic band states is zero, even in the case of ferromagnets, because [15] of angular momentum quenching. In an infinite perfect crystal,  $H_{so}$  only couples band states at the same Bloch wavevector so that the typical shift in the energy of a band state caused by spin–orbit coupling is  $\sim \xi_d^2/W_d$  in the range from 1 to 10 meV. The size of the anisotropy energy per atom is much smaller however, approximately 1/40th of a meV per atom in Cobalt, an order of magnitude smaller in Iron and a further order of magnitude smaller in Nickel. These relatively small anisotropies occur because of cancellations between different orbitals in evaluating the dependence of total band energy on the direction of spontaneous spin polarization. Net anisotropies per atom are generally larger in small magnetic particles because of the loss of symmetry at the surface. We note that the shifts in orbital energy due to spin–orbit coupling, expected on the basis of the estimates made here, are larger than the typical orbital level spacings in the particle size range of interest. As a consequence, we expect that level crossings between states with opposite spins, for example those that occur as a function of the magnitude or direction of the spin-splitting field discussed in the next section, will be strongly avoided at all spin-splitting fields. The dependence of the quasiparticle level spacing statistics on spin–orbit coupling strength, and its variation around a magnetic hysteresis loop, is an interesting open issue in the mesoscopic physics of small magnetic particles, one that is closely related to recent work [17,18] on  $g$ -factors in normal metal particles with spin–orbit coupling.

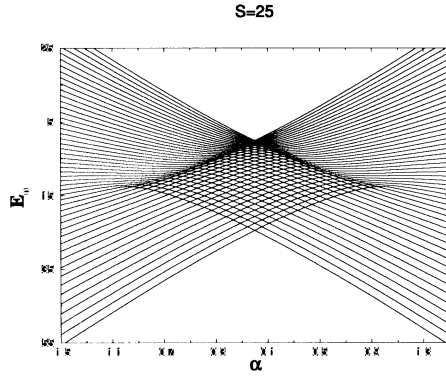


Fig. 4. Spectrum as a function of external field for a collective-spin-only quantum model of a ferromagnetic nanoparticle. These calculations are for a particle with ground state spin  $S = 25$ .  $\alpha$  is the external field in units of the anisotropy field which is oriented at an angle of  $45^\circ$  with respect to the easy axis. The energies are in arbitrary units.

The considerations in the preceding paragraph relate to the dependence of quasiparticle energies on magnetization orientation in the mean-field description of a ferromagnet. They do not, however, give us a language that permits us to make contact with the classical description of a magnetic particle discussed in Section 2 or one that tells us anything about the quantization of collective dynamics. Much better for these purposes, is the path integral approach described below.

### 5. Spin-orbit coupling: path integral approach

We now augment our toy model Hamiltonian by a spin-orbit coupling term:

$$\mathcal{H} = H_{1b} + H_{so} + V \quad (5)$$

where  $H_{so}$  is a one-body term with spin-orbit coupling that does not need to be specified precisely for this discussion. As we now show, we are able to make contact with the classical description of a fine magnetic particle by following some familiar steps [12,19]. We use a coherent state path integral representation of the interacting fermion partition function. Since the interaction term in our toy model is quadratic in the total electron spin, its contribution to the action can be replaced using Gaussian integral identities by a single auxiliary field that fluctuates in imaginary time. Taking  $T \rightarrow 0$  for the fermions, the partition function is  $Z = \int D[\vec{h}(\tau)] \exp(-S)$ , where the action

$$S = \int_0^\beta d\tau \left[ \frac{\vec{h} \cdot \dot{\vec{h}}}{2U} - \ln \langle \Phi_0(\vec{h}_N) | \exp(-\epsilon H_{\text{tot}}(\vec{h}_{N-1})) | \Phi(\vec{h}_{N-1}) \rangle \right. \\ \left. \times \dots \times \langle \Phi_0(\vec{h}_1) | \exp(-\epsilon H_{\text{tot}}(\vec{h}_N)) | \Phi_0(\vec{h}_N) \rangle \right], \quad (6)$$

$\beta = 1/k_B T$ , and the fluctuating single-body Hamiltonian

$$H_{\text{tot}}(\vec{h}) = H_{1b} + H_{so} - \frac{1}{2} \sum_{j,s',s} [\vec{h} + g\mu_B \vec{H}_{\text{ext}}] \cdot \vec{c}_{j,s'}^\dagger \vec{\tau}_{s',s} c_{j,s}, \quad (7)$$

has both spin-splitting and spin-orbit interaction terms. We have added an external magnetic field to Eq. (7), because of its important role in manipulating many-body states. Taking the limit  $\epsilon = \beta/N \rightarrow 0$  in Eq. (6) gives

$$S = \int_0^\beta d\tau \{ \langle \Phi_0(\vec{h}) | \partial \Phi_0 / \partial \vec{h} \cdot \frac{d\vec{h}}{d\tau} + E(\vec{h}) \}. \quad (8)$$

The second term in this action gives the energy of the magnetic particle as a function of the spin-splitting field; this is the quantum generalization of the phenomenological micromagnetic expression for the energy of a small magnetic particle. The dependence of this term on  $|h|$  will not be strongly altered by spin orbit interactions. It is easy to verify that in the absence of spin-orbit interactions and external fields, the value of  $h$  at which  $E(h)$  is minimized ( $h^*$ ) is just the spin-splitting field  $\Delta = \epsilon_{F_a} - \epsilon_{F_l}$  of Section 3. The first term in the action is a Berry phase term that captures the way in which spin-orbit interactions alter the quantization of collective excitations.

### 6. Discussion

Previous attempts [13,14] to develop a quantum description of ferromagnetic metal nanoparticles including spin-orbit coupling have adopted the physically appealing, but still ad hoc approach, simply replacing the magnetization vector  $\vec{M} = M_s \vec{\Omega}$  of the classical energy expression by a spin-operator acting in a space with total spin  $S = S_0$ . The evolution of the many-particle spectrum with an external field oriented at  $45^\circ$  from the easy axis for such a model is illustrated in Fig. 4. The lowest energy quantum states for  $H = 0$  are the two equivalent easy axis states with  $S_z = \pm S_0$ . For finite fields, states with definite  $S_z$  are no longer eigenstates. For weak fields of either sign, classical states with different spin orientation are stable, as evidenced by the weakly avoided crossing between groups of quantum states with opposite energy-vs-field slopes. These groups of states correspond to collective oscillations around the two classical extrema. The spread of energies at  $H = 0$  in this figure is the anisotropy energy scale of the particle. This type of spectrum is likely qualitatively correct for an insulating ferromagnetic nanoparticle. Although not fully realistic, primarily because of the simplified interaction model on which it is based, Eq. (8) can already be used to make some educated guesses on how this picture needs to be altered for a ferromagnetic metal. For example, if estimated from bulk anisotropy energy densities [9,10], the total anisotropy energy of a Cobalt nanoparticle containing  $\sim 1000$  atoms is  $\sim 25$  meV, much larger than the single particle level spacing which is  $\sim 1$  meV. Since we can (at least as a crude first approximation) expect particle-hole

and collective excitations to be independent; replicas of the collective excitation spectrum of Fig. 4 should occur for each low-energy majority spin or minority spin particle–hole excitation (the type of excitation illustrated in Fig. 1). Approximate replicas will also occur for each low-energy spin-flip quasiparticle excitations, like those discussed in Section 3. These interleaved excitation spectra will give rise to a complex pattern of avoided crossing gaps, whose properties might be usefully related to bulk quasiparticle–magnon interaction amplitudes. The quasiparticle states themselves will also change as the magnetization orientation is altered by an external field, however we do not expect quasiparticle levels to cross. Sorting out the interrelationships between all these effects may lead to a deeper understanding of current and future experiments.

### Acknowledgements

We are grateful to M. Deshmukh, S. Guéron, J. von Delft, and D. Ralph for helpful interactions. This work was supported by the Welch Foundation and by the Indiana 21st Century Fund. AHM acknowledges the helpful assistance of Lita Anglin.

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