Magnetic excitations in helically ordered rare earth multilayers

1993 J. Phys.: Condens. Matter 5 3463

(http://iopscience.iop.org/0953-8984/5/21/010)

J. Phys.: Condens. Matter 5 (1993) 3463–3468. Printed in the UK

Magnetic excitations in helically ordered rare earth multilayers

Niu-Niu Chen and David Y K Ko
Department of Physics, Theoretical Physics, University of Oxford, 1 Keble Road, Oxford, OX1 3NP, UK

Received 11 December 1993

Abstract. Rare earth metals exhibit a rich spectrum of bulk magnetic order. When they are multilayered, these metals offer the possibility of producing a wide variety of tunable magnetic properties. We present an analysis of the magnetic excitations appropriate to the ferromagnetic, spiral and antiferromagnetic phases of such multilayers. We find that the interfaces play a very important role in determining the ground state of the system, and systems with identical spin equilibrium configurations can also have significantly different spin excitations, suggesting corresponding differences in their thermodynamic properties.

The fabrication of high quality magnetic multilayers have recently been successfully carried out for both rare earth [1] and transition metals [2], as well as for rare-earth–transition metals [3]. It has been found that in all these systems a long range RKKY based exchange interaction exists across magnetically inert layers such that a long range magnetic coherence remains [4]. In particular, experimental results on Ho/Y [5] multilayers have shown that the magnetic order extends over many multilayer periods and the spin order undergoes bulk transitions from a paramagnetic phase at high temperatures, through a spiral phase in intermediate temperatures to a spin slip phase at low temperatures. These and other experimental results [1] have shown that significant differences in the equilibrium spin configurations of the multilayers from the bulk configurations are common, and initial theoretical analysis [6] has indicated that these differences are due to a complex interplay between interface exchange and the bulk exchange behaviour. Also important in these novel spin systems is an understanding of the dynamical properties of the magnetic excitations which play a strong role in determining the thermodynamical as well as transport properties. A number of approaches have been developed for purely ferromagnetic or antiferromagnetic systems [7]. In this paper we present results on the excitations in the spiral phase of a rare earth magnetic multilayer. We find that firstly interface effects are strong even in the simple spiral phase, and secondly that two systems with identical static equilibrium spin configurations can have significantly different excitation spectra.

The system we consider consists of slabs of N atomic layers of a magnetic rare earth metal such as Dy or Ho separated by layers of a magnetically inert material, typically Y, repeated to form a multilayer. The materials have HCP symmetry with a long range RKKY exchange interaction between the localized 4f moments. In the bulk this leads to a rich magnetic phase diagram [8] which may be modelled readily by an n-nearest-neighbour Heisenberg model $H = -\frac{1}{2}\sum_{ij} J_{|i-j|} \boldsymbol{S}_i \cdot \boldsymbol{S}_j$ with $J_{|i-j|} \neq 0$ for $|i-j| \leqslant n$. For the spiral phase it is sufficient to have $n = 2$, though in real systems further neighbour interactions may be significant [9, 10]. The intra-plane exchange constant J_0 is usually positive, giving rise to ferromagnetically ordered layers, and a competing interaction exists between the nearest-neighbour inter-plane interaction J_1 and the next-nearest-neighbour inter-plane interaction J_2 leading to the development of a spiraling of the spin moment between adjacent layers. In a multilayer, the exchange across magnetically inert layers can be replaced by a set of effective interface exchange coupling constants $I_{|i-j|}$. Our starting point is therefore a Heisenberg Hamiltonian with second-nearest-neighbour interactions and a strong easy plane anisotropy so as to produce a planar spiral,

$$H = -\frac{1}{2}\sum_{i,j} J_{ij}\, S_i \cdot S_j + B\sum_i (S_i^z)^2. \tag{1}$$

The exchange constants J_{ij} take on values $J_{ij} = J_{|i-j|}^{\text{Bulk}}$ for i, j on the same side of an interface, and $J_{ij} = I_{|i-j|}$ for i, j on opposite sides of an interface, see figure 1. The c-axis of the hexagonal unit cell is in the plane of the paper, perpendicular to the interface. Since we are interested in the spiral phase, a natural way to proceed is to use the Villain representation [11]

$$S_i^{\pm} = \sqrt{S \pm S_i^z}\, e^{\pm i\phi_i} \sqrt{S \mp S_i^z} \tag{2}$$

where S_i^+ and S_i^- are spin raising and lowering operators and the operators S_i^z, ϕ_i denote the out-of-plane magnetization and the in-plane polar angular displacement respectively. They obey the commutation relation $[S_i^z, \phi_j] = -i\delta_{ij}$ with $\hbar = 1$. In this representation the Hamiltonian, equation (1), may be expanded in powers of $|S_i^z/S| \ll 1$ to give

$$H = -\frac{S(S+1)}{2}\sum_{i\neq j} J_{ij}\left\{\cos\phi_{ij} - \frac{S_i^z \cos\phi_{ij}\, S_i^z}{S(S+1)} + \frac{S_i^z S_j^z}{S(S+1)} + \frac{S_i^z S_j^z \cos\phi_{ij}\, S_j^z S_i^z}{4S^2(S+1)^2} - \frac{(S_i^z)^2 \cos\phi_{ij}\,(S_i^z)^2}{4S^2(S+1)^2}\right\} + B\sum_i (S_i^z)^2. \tag{3}$$

where $\phi_{ij} = \phi_i - \phi_j$ is the polar angle between spins i and j. The equilibrium configuration may be obtained by minimizing the expectation value of the first term with ϕ_{ij} replaced by the equilibrium angles ϕ_{ij}^0. To examine the excitation about the equilibrium configuration we first rewrite the displacement operators S_i^z and ϕ_i in terms of bosonic operators a_i and $a_i^\dagger$

$$S_i^z = \sqrt[4]{\tfrac{1}{4}S(S+1)}\,(a_i^\dagger + a_i) \qquad \phi_i = \phi_i^0 + \frac{1}{i}\frac{1}{\sqrt[4]{4S(S+1)}}(a_i^\dagger - a_i) \tag{4}$$

where $[a_i, a_j^\dagger] = \delta_{ij}$. To second order in the excitations the Hamiltonian, equation (2), becomes

$$H = -\frac{1}{2}\sqrt{S(S+1)}\left\{\sum_i\left[2B(a_i^\dagger a_i^\dagger + a_i a_i) + 4\left(\sum_j J_{ij}\cos\phi_{ij}^0 + B\right)a_i^\dagger a_i\right] + \sum_{i\neq j} J_{ij}\left[\left(\cos\phi_{ij}^0 - 1\right)(a_i^\dagger a_j^\dagger + a_i a_j) - 2\left(\cos\phi_{ij}^0 + 1\right)a_i^\dagger a_j\right]\right\}. \tag{5}$$

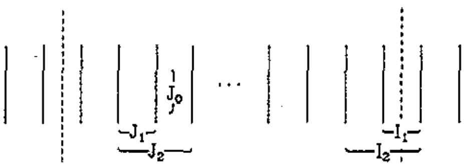

Figure 1. Schematic representation of a multilayer system with repeats of N layers of magnetic materials. The solid lines represent atomic planes of magnetic atoms, the dotted lines represent interfaces (magnetically inert spacers). The c-axis of the hexagonal unit cell is in the plane of the paper, perpendicular to the interface.

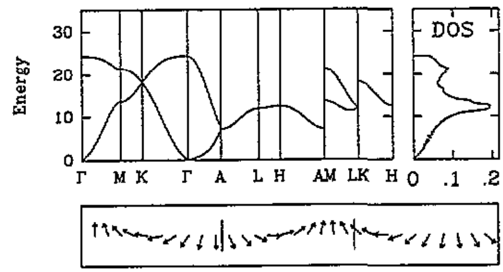

Figure 2. The energy dispersion (in units of $J_1\sqrt{(S(S+1))/2}$), density of states and equilibrium spin configuration for the bulk system.

For the ferromagnetic, $\phi_{ij}^0 = 0$, or antiferromagnetic, $\phi_{ij}^0 = \pi$, phase this Hamiltonian reduces to the Holstein–Primakoff spin–wave Hamiltonian such that the two limits are naturally represented in our approach. Treating the multilayer as a superlattice with an infinite number of repeats the quadratic Hamiltonian may be diagonalized by first Fourier transforming to give

$$H = \sum_{nm}\sum_{k}\left[A_{nm}(k)a_{n,-k}^{\dagger}a_{m,k} + B_{nm}(k)\left(a_{n,k}^{\dagger}a_{m,-k}^{\dagger} + a_{n,-k}a_{m,k}\right)\right]. \tag{6}$$

The matrices $A_{nm}(k)$ and $B_{nm}(k)$ are Hermitian and k is the three-dimensional wavevector in the superlattice. A Bogoliubov–Tyablikov transformation [12]

$$\begin{aligned}\xi_{n,k} &= \sum_m\left[u_{nm}(k)a_{m,k} + v_{nm}^*(k)a_{m,-k}^{\dagger}\right]\\ \xi_{n,-k}^{\dagger} &= \sum_m\left[u_{nm}^*(k)a_{m,-k}^{\dagger} + v_{nm}(k)a_{m,k}\right]\end{aligned} \tag{7}$$

where $[\xi_{n,k}, \xi_{m,q}^{\dagger}] = \delta_{nm}\delta(k-q)$ and the indices n, m run from 1 to N, the total number of magnetic layers in one multilayer repeat, reduces the Hamiltonian to diagonal form

$$H = \sum_{l,k}\epsilon_l(k)\,\xi_{l,k}^{\dagger}\xi_{l,k}. \tag{8}$$

The excitation energy $\epsilon_l(k)$ for the lth mode with wavevector k is given by the non-Hermitian eigenvalue equation

$$\begin{aligned}&\sum_m\left[(A_{nm} - \epsilon_l(k)\delta_{nm})\,u_{lm} - 2B_{nm}v_{lm}^*\right] = 0\\ &\sum_m\left[-2B_{nm}u_{lm} + (A_{nm} + \epsilon_l(k)\delta_{nm})\,v_{lm}^*\right] = 0.\end{aligned} \tag{9}$$

The excitation's creation operator may also be expressed in terms of the spin displacements operators using equation (3)

$$\xi_{n,-k}^{\dagger} = \sum_m\left\{\frac{1}{\sqrt[4]{4S(S+1)}}\left(u_{nm}^* + v_{nm}\right)S_{m,k}^z + \mathrm{i}\sqrt[4]{\tfrac{1}{4}S(S+1)}\left(u_{nm}^* - v_{nm}\right)\delta\phi_{m,k}\right\}.$$

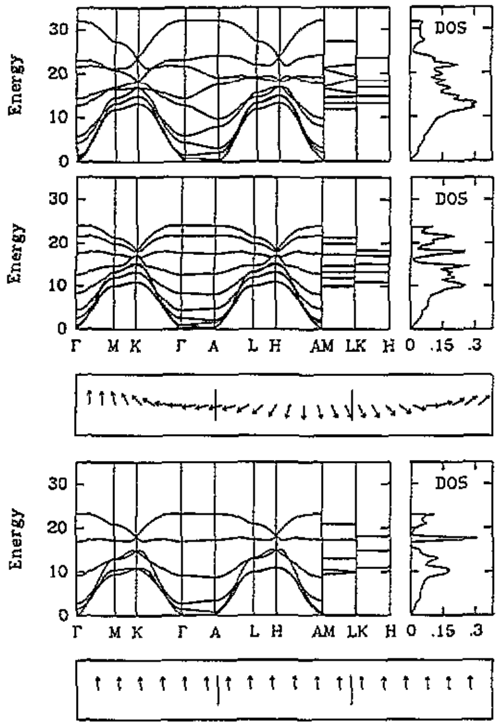

Figure 3. The energy dispersions (in units of $J_1\sqrt{(S(S+1))/2}$), densities of states and equilibrium spin configurations for the multilayer systems. The upper two sets of diagrams correspond to a strong (top set) and a weak (middle set) interface coupling with $N = 10$, the lowest set corresponds to a weak interface coupling with $N = 6$. Details of the parameters are given in the text.

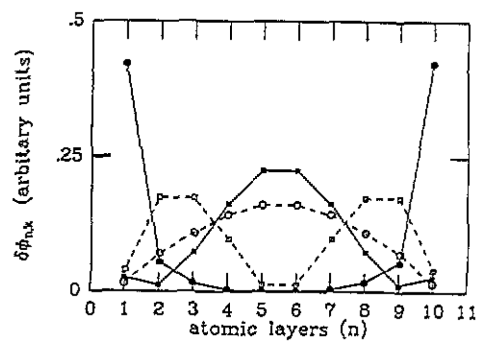

Figure 4. The amplitude of the angular displacements $\delta\phi_{n,k}$ for the highest energy (circle) and the second highest energy (square) state at the Γ point for the $N = 10$ multilayers. The filled symbols are for the strong interface system, and the open symbols are for the weak interface system.

In the following we shall consider a bulk system with $J_0/J_1 = 0.3$, $J_2/J_1 = -0.8$, $B/J_1 = 0.3$, multilayered with nearest-neighbour interface exchange only. The qualitative behaviour in the multilayers will be illustrated by considering the limits of a strong interface exchange, a weak interface exchange and a short multilayer period. Next-nearest-neighbour exchange across the interfaces will be neglected. The bulk behaviour with these parameters is shown in figure 2. The equilibrium spin configuration in this case consists of ferromagnetically ordered layers with a uniform inter-plane spiral angle of 24.4° between adjacent layers as shown in the bottom of figure 2. Here each arrow represents the orientation of the spins in the ferromagnetically ordered layers. The two vertical lines represent the positions of the interfaces of a multilayer system with $N = 10$, which will be considered below. From the energy dispersion we can see a strong dispersion along the c-direction (the Γ–A, M–L and the K–H lines, see, for example, reference [9] for the symmetry points of the Brillouin zone). The density of states for the bulk system is shown to the right of the energy dispersion. The results show the usual behaviour for a three-dimensional system with cusp-like van-Hove singularities at the band edges and a prominent peak in the density of states corresponding to the band edge at the H point.

The results for the multilayers are shown in figure 3. In the upper two sets of diagrams the energy dispersions and densities of states for two $N = 10$ multilayers are shown. The top and the second sets of figures correspond to the case of a strong interface exchange, $I_1/J_1 = 2$, and a weak interface exchange, $I_1/J_1 = 0.1$, respectively. For both systems we have set $I_2 = 0$ such that their equilibrium spin configurations, plotted below the dispersions, are identical. It is clear, however, that despite this similarity in their equilibrium configurations the magnetic excitation spectrum for the two systems are substantially different. In the system with the strong interface coupling, the top diagrams, there is a large dispersion along the c-axis (Γ–A line) which is noticeably missing in the weak interface multilayer. The strong dispersion also pushes the energy of highest band past the top of the energy spectrum of the pure system, leading to a band of states strongly localized to the interface. This is clearly demonstrated in figure 4, where the amplitude of the angular displacements $\delta\phi_{n,k}$ of the spins in the multilayers are plotted for the two highest energy states at the Γ point for both systems. For the weaker interface system we find that the displacements for the highest and the next highest energy states correspond well to what one expects from quantum well states with hard walls. Since only the amplitude is plotted the displacement in the highest energy state corresponds to that expected in the lowest energy quantum well state. In the strong interface coupling case, the highest energy state has displacement amplitudes which decay exponentially as one moves away from the interface. This behaviour is also reflected in the out-of-plane displacements $S^z_{n,k}$ and corresponding to an interfacially localized state. For states with other wavevectors in this band, this decay may be less rapid, but the localized nature remains. The presence of such a band of interfacial states should affect the high temperature thermodynamics of the multilayer and may be probed experimentally with optical techniques [13].

The density of states for both multilayer systems show substantial broadening of the peak found in the pure system, with many subsidiary peaks appearing over the broadened background. These peaks arise as a result of the mixing due to the folding of the bandstructure. In particular, the doubly degenerate A–L–H band in the pure system is folded into the Γ–M–K states and mixed, splitting and pushing the peak up in energy in the density of states at the H point. In the weaker coupling system there is, in addition, the development of a pseudogap in the spin excitations corresponding to a partial splitting of the degeneracy at the K point (the splitting is also seen in the H point of the superlattice since the two points fold into each other). In the strong interface system there is a similar pseudogap at a higher energy. However, in this case the states with energies above this belong to the band of interfacial states. We note further that the behaviour of the equilibrium spin configuration is significantly different from that of the bulk material. The inter-plane spiral angle in the multilayer varies continuously from 0° at the interface to 17° at the centre of the magnetic layers in contrast to the bulk value of 24.4°. It is easy to show that the effects of the interface may persist into much thicker multilayer systems.

Finally, the behaviour for an $N = 6$ superlattice with a weak interface coupling, $I_1/J_1 = 0.1$, is shown in the bottom diagrams of figure 3. The most striking feature of this system is the appearance of a fully ferromagnetically ordered equilibrium state despite the antiferromagnetic frustration. This is because for a given set of bulk and interface exchange parameters in the spiral phase there exists a critical number of magnetic layers below which the ferromagnetic phase is more stable. With the equilibrium configuration significantly modified in the present system the effects of the multilayering on the magnetic excitations is expected to be stronger. An examination of the dispersion reveals this to be indeed the case. Firstly, the excitations at long wavelengths now behave as $\epsilon(k) \sim k^2$ in contrast to the linear dispersion in the spiral phase. The effect of this on the density of states, however, is hidden by the ferromagnetic intra-plane excitations such that the implications on the thermodynamic behaviour is unclear. Secondly, the pseudogap in the previous system with a weak interface has now developed fully into a sizable gap in the middle of the bulk excitation spectrum, with a strong peak in the density of states lying just above it. This peak corresponds to a well defined quantum-well-like state and not to any interface state. That is, the amplitude of the spin displacements do not show any tendency to localization at the interfaces. The strength of the peak is due to the severe flattening of the bandstructure at that energy.

In conclusion, we have examined the spin excitations in magnetic multilayers in rare earth systems in the spiral phase. Our method spans naturally across the full range of ferromagnetic, spiral and antiferromagnetic phases and our results show that multilayering can lead to substantial modifications of the equilibrium spin configuration as well as in the magnetic excitations. The spin displacements in the multilayer take on quantum-well-like behaviour, and interfacial states, where the displacement amplitudes are limited to the interface region, may also be formed. The density of states show a complex splitting of the bulk structure, with a combination of two-dimensional singularities and steps due to saddle points and band edges, and three-dimensional cusp-like van Hove singularities.

We acknowledge the SERC for support under grants GR/H73028 and GR/G02727. We would also like to thank Bob Camley, Nick Constantinou, Mike Cottam, Roger Cowley, Dave Jehan, Desmond Morrel, Bob Stamps and David Tilley for useful discussions.

References

[1] Majkrzak C F, Kwo J, Hong M, Yafet Y, Gibbs D, Chein C L and Bohr J 1991 *Adv. Phys.* **40** 99
[2] Falicov L M, Pierce D T, Bader S D, Gronsky R, Hathaway K B, Hopster H J, Lambeth D N, Parkin S S P, Prinz G, Salamon M, Schuller I K and Vicotra R H 1990 *J. Mater. Res.* **5** 1299
[3] Taborelli M, Allenspach R, Boffa G and Landolt M 1986 *Phys. Rev. Lett.* **56** 2869
Weller D, Alvarado S F, Gudat W, Schroder K and Campagna M 1985 *Phys. Rev. Lett.* **54** 1555
[4] Yafet Y, Kwo J, Hong M, Majkrzak C F and O'Brien T 1988 *J. Appl. Phys.* **63** 3455
Fairbairn M and Yip S Y 1991 *J. Phys.: Condens. Matter* **2** 4197
Unguris J, Celotta R J and Pierce D T 1992 *Phys. Rev. Lett.* **69** 1125
Edwards D M, Mathon J, Muniz R B and Phan M S 1991 *Phys. Rev. Lett.* **67** 493
[5] Jehan D A, McMorrow D F, Cowley R A, Ward R C C, Wells M R, Hagmann N and Clausen K N 1993 *Phys. Rev.* B submitted
[6] Camley R E and Tilley D R 1988 *Phys. Rev.* B **37** 3413
Nortemann F C, Stamps R L, Carrico A S and Camley R E 1992 *Phys. Rev.* B **46** 10847
[7] Hinchey L L and Mills D L 1986 *Phys. Rev.* B **33** 3329
Qiu Z Q, Mattson J E, Sowers C H, Welp U, Bader S D, Tang H and Walker J C 1992 *Phys. Rev.* B **45** 2252
Martinez B and Camley R E 1992 *J. Phys.: Condens. Matter* **4** 5001
[8] Elliott R J 1961 *Phys. Rev.* **124** 146
Cooper B R 1968 *Solid State Physics* vol 21 (New York: Academic) p 393
[9] Jensen J and Mackintosh A R 1991 *Rare Earth Magnetism* (Oxford: Clarendon)
[10] McMorrow D F, Patterson C, Godfrin H and Jehan D A 1991 *Europhys. Lett.* **15** 541
[11] Villain J 1974 *J. Physique* **35** 27
Haldane D 1983 *Phys. Lett.* **93A** 464
[12] Tyablikov S V 1967 *Methods in the Quantum Theory of Magnetism* (New York: Plenum)
[13] Cottam M G and Lockwood D J 1986 *Light Scattering in Magnetic Solids* (New York: Wiley)