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A possible mechanism for quasi-two-dimensional ferromagnetism: the spectrum gap induced by s–d exchange interaction

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Abstract. A quasi-two-dimensional magnetic system is studied in which magnetic atoms are confined in the two-dimensional plane but the conduction electrons are in fact three-dimensional. By virtue of spin-wave theory, we find that a spectrum gap can be formed at the bottom of the spin-wave spectrum, induced by the s–d exchange interaction between conduction electrons and magnetic localized electrons. Thus, a new possible mechanism is proposed for two-dimensional ferromagnetic order at finite temperature.

1. Introduction

Whether or not two-dimensional (2D) ferromagnets exist has been a controversial problem for several decades. The usual spin-wave theory implies that no ferromagnetism can exist in 2D if the dispersion relation between the energy spectrum and wave vector $\omega_q \sim q^2$ holds. The reason for this lies in the logarithmical divergence of the thermal excitation spin waves. The argument, first proposed by Bloch [1], was later rigorously proved by Mermin and Wagner [2], who showed that no 2D long-range magnetic order can exist at finite temperature provided that the interactions between spins are isotropic and short ranged (more precisely, the intersection must decrease more rapidly than $1/r^3$, where r is the distance between atoms).

The old problem of 2D ferromagnetic order has attracted renewed interest in recent years due to the development of ultrahigh-vacuum technology, via which clean monolayers of high structural quality can be made and where long-range ferromagnetic order is found experimentally. Since an ideal 2D system cannot in practice exist, these phenomena are usually referred to as quasi-2D ferromagnetism. Evidence was first reported by Flevaris *et al* [3] in 1980, and verified in many later studies [4–5]. Moreover, ultrathin magnetic layers and multilayers are subjects of intense current research, because of their novel properties, such as in the problems of surface magnetization [6–8] and especially the interlayer exchange coupling and giant magnetoresistance [9–11].

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In order to give an interpretation of the experimental results theoretically, many investigations have been carried out as a result of the fact that the Mermin–Wagner theorem cannot be fulfilled by a real physical system. First, a two-dimensional system is intrinsically anisotropic. Due to uniaxial anisotropy, the logarithmic divergence could be removed by opening a gap at the bottom of the spin-wave spectrum, and therefore stabilizing the long-range ferromagnetic order. This mechanism was carefully studied by Döring [12] and Corciovei [13]. Secondly, magnetic dipole–dipole interactions (which decrease like r^{-3}) always exist. The effect of this on the magnetic long-range character was discussed by Maleev [14], and quite recently by Yafet *et al* [15]. Furthermore, by taking into account both the anisotropy and the dipolar interactions, Bruno [16] has recently investigated the 2D magnetic behaviour using spin-wave theory.

However, both the above mechanisms seem to encounter difficulties in sustaining the long-range magnetic order at finite temperature. In a case where a uniaxial anisotropy favours an easy-magnetization plane rather than an easy-magnetization axis, the spins would be freely rotated in the easy plane and no spectrum gap would appear [16]. Additionally the dipolar interaction alone seems too weak to stabilize the magnetic order at finite temperature. Thus, it is worthwhile to find out whether there exist other possible mechanisms that may overcome the difficulties. In this paper, we will suggest a new possible mechanism, the s–d exchange interaction, which also leads to a spectrum gap in the 2D spin wave. This differs from the crystalline anisotropy approach in that the bottom of the spectrum gap induced by s–d interaction is independent of the magnetic anisotropy.

2. The Hamiltonian and the general formula for quasi-two-dimensional magnetic systems

We first give the physical picture of the system. For simplicity, we consider the magnetic monolayer to be a simple square lattice, and the set of magnetic atom position vectors is confined in $x0y$ -plane and can be represented as

$$\mathbf{r}_n = a(n_x \mathbf{e}_x + n_y \mathbf{e}_y) \quad (1)$$

where $n_x, n_y = 0, \pm 1, \pm 2, \dots$ and a is the lattice constant. Note that in a practical system, the magnetic monolayer is grown on a substrate and covered by a protective layer (usually of a noble metal, such as Ag—see, e.g., [9]) with the thickness of a few nm. We consider that not all of these nonmagnetic substrate and covered materials can be neglected, because they may contribute conduction electrons (we will call these electrons s electrons and assume that their energy band has the free-electron structure), and these conduction electrons may have exchange interactions with the localized magnetic electrons (they are called d electrons hereafter). This is the well known s–d exchange interaction, and in this paper we will suggest that the interaction is one of the main contributions to the quasi-two-dimensional ferromagnetic order at finite temperature. Thus, both for simplicity and to obtain a clearer physical picture, the crystalline anisotropy and dipole–dipole interactions are not considered in our following discussions.

Three numerical parameters, N_e , N_m and N_v , are important in our further discussions and must not be confused; N_e is the number of conduction s electrons which can move freely in 3D rather than 2D, N_m is the number of magnetic unit cells and also the number of localized d electrons with nearest-neighbour Heisenberg interactions, and N_v is the total number of unit cells, including not only those of the magnetic monolayer but also those of the substrate and covered materials—which we have simply assumed to have the same unit-cell volume as the magnetic layer.

We can write down the Hamiltonian of the system as

$$\hat{\mathcal{H}}_{tot} = \hat{\mathcal{H}}_s + \hat{\mathcal{H}}_m + \hat{\mathcal{H}}_{sd} \quad (2)$$

which indicates the sum of the Hamiltonians of conduction s electrons, localized magnetic electrons and s–d exchange interactions, respectively. In this paper, we set $\hat{\mathcal{H}}_0 = \hat{\mathcal{H}}_s + \hat{\mathcal{H}}_m$ as the unperturbed Hamiltonian; it is chosen as the sum of the free band electrons and the 2D Heisenberg model:

$$\hat{\mathcal{H}}_0 = \hat{\mathcal{H}}_s + \hat{\mathcal{H}}_m = \sum_{\mathbf{k}, \sigma} \epsilon_{\mathbf{k}} \hat{a}_{\mathbf{k}\sigma}^+ \hat{a}_{\mathbf{k}\sigma} - J_m \sum_{\langle i, j \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \quad (3)$$

where $\epsilon_{\mathbf{k}} = k^2/2m$ and we have chosen $\hbar = 1$ for simplicity. \mathbf{k} and m are the wave vector and effective mass of s electrons. $\hat{a}_{\mathbf{k}\sigma}^+$ and $\hat{a}_{\mathbf{k}\sigma}$ are creation and annihilation operators. J_m is the exchange integral describing exchange between magnetic electrons, and $\hat{\mathbf{S}}_i, \hat{\mathbf{S}}_j$ are the spin operators at sites i, j . The sum denoted in $\langle \ \rangle$ is restricted to the nearest-neighbour pairs.

The perturbation Hamiltonian of the exchange interaction $\hat{\mathcal{H}}_{sd}$ can by virtue of the spin-operator representation for magnetic electrons and the single-occupation condition, be given as [17]

$$\hat{\mathcal{H}}_{sd} = -\frac{1}{N_v} \sum_{\mathbf{k}, \mathbf{k}', n} e^{i(\mathbf{k}' - \mathbf{k}) \cdot \mathbf{r}_n} J(\mathbf{k}', \mathbf{k}) \left\{ \left(\hat{a}_{\mathbf{k}\uparrow}^+ \hat{a}_{\mathbf{k}'\uparrow} - \hat{a}_{\mathbf{k}\downarrow}^+ \hat{a}_{\mathbf{k}'\downarrow} \right) \hat{S}_n^z + \hat{a}_{\mathbf{k}\uparrow}^+ \hat{a}_{\mathbf{k}'\downarrow} \hat{S}_n^- + \hat{a}_{\mathbf{k}\downarrow}^+ \hat{a}_{\mathbf{k}'\uparrow} \hat{S}_n^+ \right\} \quad (4)$$

in which N_v is the total number of unit cells as defined above, and \uparrow and \downarrow represent the spin directions of s electrons parallel and antiparallel to the magnetization direction. From symmetry considerations it is reasonable to assume that the magnetization direction would be perpendicular to the magnetic layer plane, i.e., in the same direction as \mathbf{e}_z . Hereafter, we will make the usual approximation $|J(\mathbf{k}, \mathbf{k}')| \approx \text{constant}$, and denote $J(\mathbf{k}', \mathbf{k})$ as J_{sd} . Physically, the first term in (4) corresponds the conduction electron's polarization due to the s–d exchange interaction and the other two terms correspond to the spin-flip processes.

Suppose that the ground-state magnetic axis of the system has all spins up, in the \mathbf{e}_z -direction; the Holstein–Primakoff transformation then gives (the constant term is omitted)

$$\hat{\mathcal{H}}_m \approx \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{b}_{\mathbf{q}}^+ \hat{b}_{\mathbf{q}} \quad (5)$$

where the spin-wave vector $\mathbf{q} \in \Omega^*$, and Ω^* is the inverse wave-vector space of the magnetic layer. $\hat{b}_{\mathbf{q}}^+$ and $\hat{b}_{\mathbf{q}}$ are creation and annihilation operators in \mathbf{k} -space. $\omega_{\mathbf{q}} = zJ_m(1 - \gamma_{\mathbf{q}})$ and $\gamma_{\mathbf{q}} = z^{-1} \sum_{\mathbf{a}} e^{i\mathbf{q} \cdot \mathbf{a}}$, in which z is the number of nearest neighbours and \mathbf{a} is the nearest-lattice-position vector. The interactions of four boson interaction are neglected.

The above treatment will also work for $\hat{\mathcal{H}}_{sd}$. For simplicity, we will omit the polarization term, i.e., the first term of $\hat{\mathcal{H}}_{sd}$ in equation (4); this is permissible for the following two reasons. First, it is obvious that the first-order perturbation contribution is zero, and a second-order treatment will lead to fourth-order boson interactions—which are negligible since the spin-wave interactions of four bosons have already been neglected in the Holstein–Primakoff transformation. Secondly, we have noticed some early work by Kim and Nagaoka [18] from 1963, which reinforces our point made above. In their work, they discussed the effect of s–d exchange interaction on the localized magnetic moment. According to their calculations, they find that the effect of the \hat{S}_n^z -term is mainly to shift the total energy level (i.e., it provides a constant term in the Hamiltonian), whereas the \hat{S}_n^{\pm} -term will affect the localized magnetism. Now, using the notation $\mathbf{u} = (\mathbf{k}' - \mathbf{k})_{\parallel}$ and $\mathbf{v} = v\mathbf{e}_z = (\mathbf{k}' - \mathbf{k})_{\perp}$

(i.e., the compound vectors in the $x0y$ -plane and e_z -direction respectively), in \mathbf{k} -space one gets

$$\hat{\mathcal{H}}_{sd} \approx -\frac{N_m^{1/2}}{N_v} \sum_{\kappa_m, \mathbf{q}} \sum_{\mathbf{k}, \mathbf{u}, \mathbf{v}} \left(J_{sd} \hat{a}_{\mathbf{k}\uparrow}^+ \hat{a}_{\mathbf{k}+\mathbf{u}+\mathbf{v}\downarrow} \hat{b}_{\mathbf{q}}^+ + \text{HC} \right) \delta(\mathbf{u}, \kappa_m + \mathbf{q}) \quad (6)$$

where $\kappa_m = 2\pi(m_x e_x + m_y e_y)/a$, $m_x, m_y = 0, \pm 1, \pm 2, \dots$, is the reciprocal-lattice vector in the square magnetic plane. The factor $\delta(\mathbf{u}, \mathbf{q} + \kappa_m)$ arises because the spin-wave vector \mathbf{q} is confined in Ω^* whereas \mathbf{u} , corresponding to the in-plane compound vector difference between the position of the conduction electron before and after scattering, is in fact unrestricted.

To find the eigenvalue spectrum of $\hat{\mathcal{H}}_{tot}$, we will use the technique of canonical transformation:

$$\hat{\mathcal{H}}_{eff} = \exp\{-\hat{\Theta}\} \hat{\mathcal{H}}_{tot} \exp\{\hat{\Theta}\} \quad (7)$$

where the rotation operator in Hilbert space is chosen as

$$\hat{\Theta} = -\frac{N_m^{1/2}}{N_v} \sum_{\kappa_m, \mathbf{q}} \sum_{\mathbf{k}, \mathbf{u}, \mathbf{v}} \frac{J_{sd} \hat{a}_{\mathbf{k}\uparrow}^+ \hat{a}_{\mathbf{k}+\mathbf{u}+\mathbf{v}\downarrow} \hat{b}_{\mathbf{q}}^+ - \text{HC}}{\epsilon_{\mathbf{k}+\mathbf{u}+\mathbf{v}} - \epsilon_{\mathbf{k}} - \omega_{\mathbf{q}}} \delta(\mathbf{u}, \kappa_m + \mathbf{q}). \quad (8)$$

Substituting (8) in (7), expanding the exponential term and averaging $\hat{\mathcal{H}}_{eff}$ in the conduction electron's state, we find the first-order boson term is removed. Now, the two-dimensional effective-boson spin-wave interaction will be given as

$$\langle \hat{\mathcal{H}}_{eff} \rangle_{bos} \approx \sum_{\mathbf{q}} (\alpha_{\mathbf{q}} + \omega_{\mathbf{q}}) \hat{b}_{\mathbf{q}}^+ \hat{b}_{\mathbf{q}} \quad (9)$$

where the conduction electron term and the constant term are omitted, and $\alpha_{\mathbf{q}}$ can be expressed as

$$\begin{aligned} \alpha_{\mathbf{q}} &= \frac{2N_m}{N_v^2} \sum_{\kappa_m, \mathbf{v}} \sum_{\mathbf{k}} |J_{sd}|^2 \frac{\langle n_{\mathbf{k}} \rangle - \langle n_{\mathbf{k}+\mathbf{u}+\mathbf{v}} \rangle}{\epsilon_{\mathbf{k}+\mathbf{u}+\mathbf{v}} - \epsilon_{\mathbf{k}} - \omega_{\mathbf{q}}} \delta(\mathbf{u}, \kappa_m + \mathbf{q}) \\ &= \frac{2N_m}{N_v^2} \sum_{\kappa_m, \mathbf{v}} \sum_{\mathbf{k}} |J_{sd}|^2 \left[\frac{f(\epsilon_{\mathbf{k}}, T)}{\epsilon_{\mathbf{k}+\mathbf{u}+\mathbf{v}} - \epsilon_{\mathbf{k}} - \omega_{\mathbf{q}}} + \frac{f(\epsilon_{\mathbf{k}}, T)}{\epsilon_{\mathbf{k}-\mathbf{u}-\mathbf{v}} - \epsilon_{\mathbf{k}} + \omega_{\mathbf{q}}} \right] \delta(\mathbf{u}, \kappa_m + \mathbf{q}) \end{aligned} \quad (10)$$

in which $f(\epsilon_{\mathbf{k}}, T)$ is the Fermi distribution of conduction electrons. Usually the temperature T is not very high; then $f(\epsilon_{\mathbf{k}}, T) \approx \theta(\mathbf{k}_F - \mathbf{k})$ holds, in which \mathbf{k}_F is the Fermi wave vector. In this case, the sum over \mathbf{k} can be calculated analytically, similarly to in the calculations on RKKY interactions [17]. One obtains

$$\alpha_{\mathbf{q}} = \sum_{\mathbf{v}} |J_{sd}|^2 \left[B(\omega_{\mathbf{q}}, \sqrt{q^2 + v^2}) + B(-\omega_{\mathbf{q}}, \sqrt{q^2 + v^2}) \right] \quad (11)$$

with

$$B(\omega, x) = \frac{3N_e N_m (x^2 + 2m_e \omega)}{16N_v^2 \epsilon_F x^2} \left[1 + \frac{4k_F^2 x^2 - (x^2 + 2m_e \omega)^2}{4k_F x (x^2 + 2m_e \omega)} \ln \left| \frac{x^2 + 2m_e \omega + 2k_F x}{x^2 + 2m_e \omega - 2k_F x} \right| \right] \quad (12)$$

in which ϵ_F is the conduction electron's Fermi energy. Since we can easily check that the function $B(\omega, x)$ decreases dramatically as x increases, both for simplicity and reasonableness, we here only concern ourselves with the contribution of the $\kappa_m = 0$ term—or 'Umklapp process' of (10)—in (11), and so we set $\mathbf{q} = \mathbf{u}$.

An energy gap will appear at $\mathbf{q} = 0$, the bottom of the spin-wave spectrum, for $\alpha_0 \neq 0$. From the assumption that $|J_{sd}|$ is constant, the integral of (11) can be exactly calculated at $\mathbf{q} = 0$. Similarly to in the treatment in [16], we can introduce an equivalent magnetic field $H_{sd} = \alpha_0/\mu_B$. One has

$$\alpha_0 = \mu_B H_{sd} = \frac{3\eta\pi k_F a |J_{sd}|^2}{16\epsilon_F} \quad (13)$$

where $\eta = N_e/N_v$ is the number of conduction electrons per lattice cell. For a square lattice, we have $\omega_{\mathbf{q}} \approx J_m a^2 q^2$. Usually the exchange energy J_m for the exchange between magnetic electrons is considered to be far larger than J_{sd} , and thus the contribution of the s-d exchange interaction can be neglected at $\mathbf{q} \neq 0$. Now, the energy spectrum of (9) is $\omega'_{\mathbf{q}} \approx \alpha_0 + \omega_{\mathbf{q}} = \mu_B H_{sd} + J_m a^2 q^2$. The dispersion relation is very similar to the form of the anisotropy contribution in the case of a perpendicular easy axis, which has already been well discussed by Döring [12]. Following an expression from [16], we can write the relative magnetic moment variation $\Delta M(T)/M(0)$ as

$$\frac{\Delta M(T)}{M(0)} = \frac{1}{N_m} \sum_{\mathbf{q}} \langle b_{\mathbf{q}}^+ b_{\mathbf{q}} \rangle = -\frac{k_B T}{4\pi J_m} \ln \left[1 - \exp \left(-\frac{\mu_B H_{sd}}{k_B T} \right) \right]. \quad (14)$$

Equations (13) and (14) are the major results of this work.

3. Calculations and discussions

Because ferromagnetic order exists at finite temperature only when $\Delta M(T)/M(0) < 1$, it can be seen clearly from (13) and (14) that the systems with high J_m and high J_{sd} will favour long-range order at finite T . This is, of course, physically reasonable.

Since (14) has the same form as the *effective anisotropy approximation* [12, 13, 15, 16], for which numerical calculations and comparisons with experimental results have already been well discussed by Bruno [16], we will not discuss it again here. However, it is worth emphasizing that the parameter H_K^{eff} (the effective magnetic field of the anisotropy) given phenomenologically in [16] surprisingly coincides numerically with our H_{sd} . On choosing the usual values: $k_F a \sim 3-10$, $\eta \sim 1-3$, $\epsilon_F \sim 3-10$ eV, $J_{sd} \sim 10^{-2}$ eV, equation (13) gives $H_{sd} \sim 10^0-10^2$ kOe. In [16], in order to compare theoretical results with the experimental ferromagnetic properties of Co fcc(001) monolayers [5], H_K^{eff} is chosen as 2, 14, and 100 kOe. Obviously, these values lie in the range of our H_{sd} . Thus, we conclude that we can indeed explain the origin of quasi-two-dimensional ferromagnets via a new possible mechanism—the gap induced by s-d exchange interactions.

According to the *effective anisotropy approximation*, problems may arise in the case of easy-plane anisotropy, in which the ferromagnetic order cannot be sustained unless much weaker dipolar interactions are considered to provide a key contribution. However, in our *s-d exchange interaction approach*, this difficulty does not exist, since the induced bottom gap always exists and is independent of the direction of the anisotropy. Nevertheless, we feel that it would be interesting and meaningful to consider the contributions of both the s-d exchange interaction and the anisotropy to the quasi-two-dimensional ferromagnetism, and such a consideration may appear in a future publication.

To make the salient points of our treatment and the physical picture of our result clearer, it is necessary to give a more exact theoretical discussion. First, the physical picture in our model is that the s-d exchange interaction does indeed provide an effective long-range coupling among magnetic atoms, i.e., the coupling falls off as the cube of the distance between magnetic atoms. Thus, in this quasi-two-dimensional model it is

theoretically possible to stabilize the ferromagnetic order at finite temperature—due to the condition of short-ranged interaction of the Mermin–Wagner theorem [2] not being satisfied. Secondly, we must point out here that it is not only because of physical reasonableness (as discussed above) that our treatment of s electrons should be in 3D rather than 2D, but also because going to three dimensions is a prerequisite for generating a spectrum gap in such a Hamiltonian. In fact, from the symmetric point of view, if s electrons are also confined in 2D, then the total Hamiltonian will have full rotational symmetry both in spin space and in real space. In this case, the Goldstone theorem [19] must be satisfied, which tells us that the excitation spectrum of such a system should be gapless!

The above theoretical analysis is supported by our calculations. In fact, the spectrum gap induced by three-dimensional s electrons and local d electrons is comparable with the crystalline anisotropy approach [13, 16], as shown above. However, if the s electron is also confined in 2D, the calculation of equation (10) indicates that α_q (as $q \rightarrow 0$) in 2D will be two orders of magnitude less than in the 3D case—this is not surprising, since the value zero is expected, and the reason for the value being slightly greater than zero arises only from the perturbation approximation error, we believe.

Another interesting conclusion is that a high density of conduction electrons, i.e., a large value of η in (13), will favour the quasi-two-dimensional ferromagnetic order, according to our approach. Experimentally, the prediction can be easily checked by making measurements on the same monolayer magnetic materials grown on different substrates and covered by different protective layers.

4. Conclusion

To conclude, we point out that the contribution of the s – d exchange interaction cannot be negligible in the problem of quasi-two-dimensional ferromagnetism. By virtue of spin-wave theory, we find that a gap may be induced at the bottom of the spin-wave spectrum due to the s – d exchange interaction and thus provide a new mechanism for inducing 2D magnetic order at finite temperature. The main difference from theories advanced up to now is that our approach is independent of the magnetocrystalline direction. Since our theory indicates that a higher density of the conduction electrons of the system may be favourable for sustaining magnetic order, we suggest that the magnetic moment of the magnetic monolayer may depend on the substrate and covering layer. However, this should be checked experimentally. Moreover, although the magnetocrystalline anisotropy in thin films has already become the subject of current research [20], it seems that no attention has been paid to the surface s – d interaction. We feel that a detailed study on 2D ferromagnetic order is needed to compare the contributions of the above two mechanisms.

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