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MAGNETISM AND MAGNETOSTRICTION IN A DEGENERATE RIGID BAND *

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ABSTRACT

We investigate the influence of the spin-orbit coupling on the magnetic and magnetoelastic phenomena in ferromagnetic band systems. The description is within the Stoner model of a degenerate rigid band, for temperature $T = 0$.

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1. Introduction

The spin-orbit coupling and the orbital magnetism are often neglected in the theoretical description of band magnetism, which is a complex task itself. The orbital part of magnetic moments of atoms is known to be almost quenched in crystalline 3d metals. The spin-orbit coupling destroys, however, the degeneracy and mixes the states. This is particularly important if the interband hopping integrals vanish by the symmetry of the Brillouin zone and if the states are not far from the Fermi level [1,2]. As a result, a small contribution from orbitals to magnetism exists in pure metals. On the other hand, in disordered systems the spatial fluctuations of a local potential decouples orbitals. If any macroscopic atomic ordering occurs, this decoupling can be significant.

The magnetic anisotropy and the magnetoelastic effects are difficult to be obtained from quantitative calculations, as they are extremally sensitive to small variations of electronic structure [3]. The role of phenomenological models is then to serve as tools to interpret some essential features of experimental data. The aim of this report is the presentation of such a tool. Much physics is disregarded here, but some other part is taken into account, and we hope to make a good choice. Below, some information is collected on the results which are due to the Stoner condition, metamagnetic effect, transversal spin susceptibility, shape magnetostriction and its strain dependence in a degenerated rigid band. In particular, we are interested in the band filling dependences of these quantities.

Below, we use the model system which consists of $2l$ magnetic states, every state is characterized by a model wave function and a spin. The wave functions are choiced to be these of t_{2g} band of 3*d* metals. The shape of the density of states is assumed not to change while magnetizing process. This rigidity is reflected by the term "rigid band model". So, the only change of an electronic structure with magnetic field etc. are the mutual shifts of subbands. The energy of the system is to be found as the integral of the total density of states, multiplied by the energy variable *e*, up to the Fermi level. The latter is to be found self-consistently for a given number of electrons. Then, the appropriate derivatives of the total energy of the system can be calculated. These are: the magnetic susceptibility, the magnetoelastic tensor coefficients and its strain derivatives.

The results are described in the subsequent parts of the paper. At first, the general expression is derived for the magnetic susceptibility for any set of rigid bands (Section 2). In Sections 3-5, the two bands model is used, which describes the case of strong uniaxial orbital anisotropy. There, the direction of the external and/or effective magnetic field is assumed to be parallel to an easy orbital axis. In this case, the spin and orbital moments are up or down, what makes four magnetic states possible. The exception is Section 5, where the direction of magnetic field is perpendicular to an easy axis. In ever case, spin magnetic moments follow ther direction of external field.

In Section 6, we use the model of three bands. Therefore, three orbital states are available. To simplify the calculation, we confine our consideration to the case of one direction of spins only. The isotropic symmetry of real space is preserved, but not

this of spin space. This limitation does not change the results qualitatively.

In every case, a secular equation is solved for the hamiltonian. This is an easy task, as the only nondiagonal matrix elements are due to the spin-orbit interaction. The energy functional is calculated for the model shape of the density of states, which is assumed to be the same for all subbands. The subbands are centered around the eigenvalues of the hamiltonian.

The last Section contains the brief comments on the validity and the possible usefulness of the model.

2. Magnetic susceptibility

Let us consider the l-fold degenerate rigid band model, where the magnetic moment m_λ of a state λ ($\lambda=1,\ldots,2l$) is assumed to not depend on magnetic field. The one-subband susceptibility is then

$$\chi_\lambda = m_\lambda \, \rho_\lambda(\mu) \, \partial(\mu-E_\lambda)/\partial H \, , \qquad (1)$$

where E_λ and ρ_λ are the energy level and the density of states of λ-th subband, μ is the Fermi energy and is to be found self-consistently from the condition

$$\sum_\lambda \partial n_\lambda/\partial H = 0 \, , \qquad (2)$$

where n_λ is the average number of electrons in a λ-th state.

The Pauli susceptibility χ in an effective magnetic field

$$H_{eff} = H + I \sum_{\lambda} m_{\lambda} n_{\lambda} \tag{3}$$

is

$$\chi = \xi \ (1 + I \ \chi) \ , \tag{4}$$

where

$$\xi = \sum_{\lambda} \rho_{\lambda}(\mu) \ m_{\lambda} \ [m_{\lambda} - \sum_{\lambda'} \rho_{\lambda'}(\mu) \ m_{\lambda'} \ / \sum_{\lambda''} \rho_{\lambda''}(\mu) \ ] \ , \tag{5}$$

and I is the mean field ferromagnetic interaction constant.

3. The Stoner condition.

For the case $\rho_{\lambda}(\varepsilon)$=const($\lambda,\varepsilon$) the Eq.4 reproduces the known Stoner condition on the instability of paramagnetic state, $I\rho>1$. To extract the orbital contribution, let us consider the case of two magnetic orbitals, *i.e.* four magnetic states, in the presence of the spin-orbit interaction and of a magnetic field. The approach is equivalent to this of [4]. The hamiltonian is

$$\mathcal{H} = \sum_{\lambda} n_{\lambda} \ [\ A \ \nu(\lambda) \ \sigma(\lambda) \ - \mu_{B} \ H \ m_{\lambda} \ ] \ , \tag{6}$$

where $\nu(\lambda)$ and $\sigma(\lambda)$ are the orbital and spin quantum number, respectively, $\sigma=\pm 1/2$, $\nu=\pm 1$, and λ-th magnetic moment is

$$m_{\lambda} = 2 \ \sigma(\lambda) \ + \alpha \ \nu(\lambda) \ . \tag{7}$$

A is the one-electron spin-orbit coupling and it is always

positive. We use the variable α to separate the spin and the orbital contributions to the magnetic susceptibility. Its value can be interpreted as the ratio of the orbital moment to the spin one. Again, if $\rho_{\lambda}(\varepsilon)$=const($\lambda,\varepsilon$), we get

$$\chi = \frac{(1 + \alpha^2) \ \rho}{1 - \rho \ I \ (1 + \alpha^2)} \ . \tag{8}$$

The orbital contribution to χ is proportional to α^2, and we see that it extends the range of a ferromagnetic phase.

We also perform the numerical calculations of χ for the Lorentzian shape of $\rho(\varepsilon)$. There, the ratio A/Δ, Δ is the half-band width, is found to be relevant. As long as A<Δ, the spin orbit coupling does not change the results. Then, the susceptibility dependence on the number of electrons in a band remains symmetric with respect to the band centre. However, for A>Δ, the susceptibility is remarkably damped if the number of electrons n<2, and it is enhanced if a band is more than half-filled. Both the magnetic susceptibility and the magnetization follow the Slater-Pauling curve, if α=1 and A/Δ is of order of 5. The effect is even stronger if we introduce the ferromagnetic interaction I.

The results on the Stoner condition can be summarized as follows. The formation of local moments in metals for the band which is less than half-filled needs a stronger ferromagnetic interaction, than for a band more than half-filled. In the latter case, ferromagnetic moments are additionally enhanced by their orbital part.

4. Metamagnetic effect

The above formalism is applied to calculate the susceptibility in finite magnetic field. There, the metamagnetic effect is known to be present in certain narrow-band systems. In a finite value of magnetic field, magnetization increases abruptly, being relatively constant for the neighboring regions of field.

This effect can also be recovered within our simple picture. As before, the ratio A/Δ is found to be essential. For I=0, the value of A about 20Δ is needed to visualise a remarkable jump of the susceptibility. This arises when the magnetic field energy is equal to A, and the degeneracy arises at the Fermi level; the bands involved have different contributions to the magnetization. (The second peak of χ arises at $\mu_B H=2A$ for certain values of α and n.) The inclusion of the parameter I has two consequences: The first one is that the peak of χ is monotonically shifted towards smaller values of H, when I increases. The second consequence of I is that the width of the peak of χ is remarkably reduced. For I=A/2, we get the jump of magnetization which is practically infinitely abrupt. (Note that the peak is not perfectly symmetrical if I>0.) This ferromagnetic enhancement of the peak of the magnetic susceptibility allows us to extend the range of bandwidth, where the metamagnetic effect is possible. Also, the ferromagnetic coupling makes this effect possible for the range of external field less than the difference of the energy levels.

5. Transversal paramagnetic spin susceptibility

In order to omit the problem of the interaction of the orbital momenta with lattice, we assume that the anisotropy constant for the orbital magnetism is infinitely strong. Then we neglect the transversal orbital susceptibility, and we set $\alpha=0$ from now on. This is justified by the difference of scales of magnetic and electrostatic interactions.

The energy levels are [4]

$$E_\lambda = \pm (H^2 + A^2)^{1/2} . \tag{9}$$

Note that H is the absolute value. We see that for H=0, the transversal spin susceptibility $\chi_\perp=0$. For H>A, the change of sign differentiates the spin states. We have, then

$$\chi_\perp = (H^2 + A^2)^{-1/2} H \chi_{\perp o} , \tag{10}$$

where

$$\chi_{\perp o} = \sum_\lambda \sigma(\lambda) \rho_\lambda(\mu) [\sigma(\lambda) - \sum_{\lambda'} \rho_{\lambda'}(\mu) \sigma(\lambda') / \sum_{\lambda''} \rho_{\lambda''}(\mu)] . \tag{11}$$

If $H\to\infty$, $\chi_\perp$ tends to $\chi_{\perp o}$. Still, the latter depends on H through $\rho_\lambda(\mu)$. For A=0, we recover the isotropic case.

As we see, only some limit cases are considered here. For 0<H<A, the result $\chi=0$ means that the magnetic field is too weak to break the spin-orbit constant. We find the angle-dependent description to be easily applicable to the band systems.

6. Magnetostriction

It is convenient to discuss the system of three bands at the moment, to be free with the choice of the relative orientation of tensile stress and external magnetic field. The band is assumed to be p-like [5]. Instead, only one orientation of spin is allowed. This does not alter the results qualitatively.

The hamiltonian is completed by adding the elastic term and the mutual shifts of the energy levels when the tensile strain is applied. These shifts are due to the deformation of the crystal field. The proportionality coefficient b between the strain ε and the energy level shift remains unknown.

The secular equation is of second order. If strain is parallel to field, the eigenvalues are

$$E_1 = A - b\varepsilon ,$$
$$E_2 = 2b\varepsilon , \qquad (12)$$
$$E_3 = - A - b\varepsilon .$$

For the perpendicular orientation, the eigenvalues are

$$E_1 = [A^2 + (3b\varepsilon/2)^2]^{1/2} + b\varepsilon/2 ,$$
$$E_2 = - b\varepsilon , \qquad (13)$$
$$E_3 = - [A^2 + (3b\varepsilon/2)^2]^{1/2} + b\varepsilon/2 .$$

The energy of the system is expressed as

$$E = \sum_\lambda \int e \, \rho_\lambda(e) \, de . \qquad (14)$$

The coefficients of the magnetoelastic tensor B are defined as the derivatives of the energy E with respect to strain. Using the Voigt notation, we have

$$B_{11} = b \, (2n_2 - n_1 - n_3) , \qquad (15)$$
$$B_{12} = - 1/2 \, B_{11} . \qquad (16)$$

We can also calculate the strain derivatives of B. For example,

$$\partial B_{11}/\partial\varepsilon = - 6b^2 \rho_2(\mu) \, [\Omega - \rho_2(\mu)]/\Omega , \qquad (17)$$

where $\Omega = \sum_\lambda \rho_\lambda(\mu)$. The expressions for the magnetic corrections to the elastic constants (ΔE effect) can also be obtained.

The relations (15) and (16) fulfil the condition of the preserving of volume when tensile stress is applied. As the sequence of bands is *1,2,3* with increasing band filling, we observe the oscillations of the sign of B with the Fermi level. The application of this picture to thin films is described elsewhere [7].

7. Discussion

The above picture, although extremely simple, has some serious disadvantages. First of all, many different physical effects are interpreted within one model. It is obvious that various mechanisms contribute to these effects, and these mechanisms are not mentioned here. Secondly, remarkable problems arise if one tries to evaluate the physical parameters which are brought up to discuss the above phenomena. Example giving, to reproduce the Slater-Pauling curve, the ratio A/Δ is needed about two orders of magnitude greater, than it is estimated. On the other hand, the parameter α for, say, nickel is not greater [6] than 0.05. Further, the metamagnetic effects in RE's are commonly interpreted as due rather to crystal field or quadrupolar splitting than to the spin-orbit coupling. There, the latter is comparable to the magnetic energy of 10^3 T or more. Finally, it is not clear, should Δ be the width of a subband or should be due to the vicinity of some points in the Brillouin zone.

In our opinion, however, the discussion is still possible. We do not claim to explain the above phenomena, but rather suggest that the band degeneracy and the spin-orbit coupling should be included to their physical description. Also, we would like to remind some physical arguments to support conceptually our picture. These arguments are as follows:

- the formation of local magnetic moments in metals is known [8] to be very sensitive to the details: the density of conduction electrons and the local interaction. The orbital degeneracy can, in certains conditions, play an important role, and the orbital contribution to magnetization should be included to the discussion. As we have shown, the correction from the spin-orbit coupling can reflect the tendency of experimental data. We would like to stress that, in 3d metals, external magnetic field is enhanced by exchange interactions by at least three orders of magnitude, what can be deduced from the Mossbauer data [9].

- in RE's, the spin-orbit interaction modifies the structure of energy levels [10] and the effective splittings are possible much less than A itself. Also, it is reasonable to expect that the ferromagnetic exchange dependence of the metamagnetic effect does not depend on the nature of the splitting of energy levels. If so,

our simple calculation gives some actual information on the possibility of metamagnetic effect in metals, where the values of the parameters seemed to exclude such a possibility.

- the transversal susceptibility is expected [11] to increase with temperature up to certain value, still far from T_c, then it decreases. In this case, the ferromagnetic enhancement of an external field does not occur. The maximum is about some hundreds of kelvins, what corresponds to the evaluation of the spin-orbit coupling in 3d's. Our expression for $\chi_\perp$ is free from the singularity in H=0, which arises within the Heisenberg description [12].

- the oscillations of the sign of magnetoelastic tensor B with the number of electrons are known from both theory [13] and experiment. Up to now we have no had such a simple model to explain this effect. On the other hand, the negative slope of the magnetostriction with tensile stress, which is known from the experimental data (see, e.g. [14]) has not been explained up to now.

As we see, within this picture one is able to discuss a variety of magnetic effects. To the above list we can add anomalous Hall effect, collinear structures, nonlinear behaviour of magnetic orbital moments, de Haas-van Alphen effect and certainly some others. We believe that this advantage justifies the phenomenological and heuristic character of the model.

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