Magnetostriction ferrons

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A study is made of the self-localization of conduction electrons within ferromagnetic regions created by the conduction electrons in metamagnetic materials (such as EuSe) with a strong volume dependence of the exchange integral. In contrast to the standard ferron states, the creation of a ferromagnetic region is accompanied by the change of sign of the exchange integral between magnetic atoms in the electron self-localization region. The energy loss due to the creation of such a region is governed essentially by lattice strain. It is shown that an external magnetic field increases the magnetic moment of a ferron and, for $h = h_{sf}$ (h_{sf} is the spin-flop field), the ferron moment is approximately twice as large as the moment in the absence of a field. The ferron magnetic moment increases also in the absence of a magnetic field in a sample subjected to an applied stress creating a strain in a crystal.
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It was shown in Refs. 1 and 2 that, for certain semiconductor crystals, a reduction in the total energy is achieved when a conduction electron creates a microscopic region which would not be stable under normal circumstances but can be stabilized by the localization of the conduction electron. Most favorable conditions for the appearance of such states are created in antiferromagnetic semiconductors. The region of localization of the conduction electron represents a nucleation ferromagnetic center in the antiferromagnetic matrix. The total energy of such a state is lower because the ferromagnetic ordering leads to a minimum in the conduction electron energy. The self-localized states of carriers in antiferromagnetic semiconductors were called ferrons and were observed in EuSe and EuTe crystals.[3] The estimates of the reduction in the electron energy due to the self-localization that were made in Refs. 1 and 2 indicate that the energy lowering is of the order of 0.1-0.2 eV.

The estimates of the ferron energy made in Refs. 1 and 2 were based on the assumption that the crystalline lattice was completely rigid. However, the electron self-localization should lead to displacements of the atoms in the lattice that should influence considerably the ferron energy. It was shown in Ref. 4 that the polarization of the Ionic cores due to ferrons increases considerably the stability of ferron states. An increase in the ferron stability can be also expected in crystals subjected to elastic compression or shearing not accompanied by the creation of dipole moments. This problem will be studied in the present paper.

A lattice strain changes both the energy of the electron localization (the condenson effect[5]) and the exchange energy between magnetic atoms (the magnetostriction effect). The former effect is usually small. It is of the order of CΔV/V, where C is the deformation potential constant and ΔV/V is the relative volume change. For typical values C ~ 10 eV and ΔV/V $\leq$ 10^{-3}, the aforementioned effect is of the order of few hundredths of an electron-volt. However, the magnetostrictive deformation of the lattice due to the formation of ferrons plays an important role in magnetic materials in which the Heisenberg exchange integral depends strongly on the volume. The situation when the lattice constant is close to the value a_c for which the exchange integral $J(a_c)$ vanishes is of special interest.

Such a situation is realized, for example, for EuSe in which the ferron state was observed.[3] The Neel point of EuSe is low ($T_N \approx 4.6°$K) and a comparatively small pressure of 1 kbar leads to a shift of T_N by 1 deg (see Ref. 6). Moreover, the transition to a disordered state is of first order in EuSe, which is typical of materials with a strong volume dependence of the exchange integral.[7] Finally, a strong volume dependence of the exchange integral can explain the observed

metamagnetism of EuSe, which manifests itself by a discontinuous transition from the antiferromagnetic to ferromagnetic state in weak magnetic fields $\sim 10^2$-10^3 Oe (the standard explanation of the metamagnetism, which is due to a strong magnetic anisotropy, does not apply to EuSe since this material is isotropic and the anisotropy field quoted in Ref. 8 is only about 100 Oe). Since the magnetostriction in EuSe is strong, the external magnetic field in which the anti-ferromagnet-ferromagnet transition occurs under pressure is lowered.[6]

The magnetostriction of EuSe should influence the formation of ferron states much more strongly than the lattice polarization. The lattice polarization energy is inversely proportional[4] to the radius of a ferromagnetic microdomain and, since the energy loss due to the formation of ferromagnetic regions in a metamagnet is small, the radius in question is large for EuSe (~ 10 a). Therefore, we shall neglect the effects related to the polarization. We shall consider only the case when the exchange between the carriers and localized magnetic moments AS/2 Is small compared with the width of the carrier energy band W (which is satisfied for europium chalcogenides[9]).

The formation of a ferromagnetic microdomain by an electron in an antiferromagnet should lead to a distortion of the microregion and of the whole antiferromagnetic matrix. The created strain influences the exchange integral and, when the exchange integral depends strongly on the lattice parameter, even the sign of the exchange integral can change. The dependence of the exchange integral on the lattice constant a can be written as an expansion in powers of (a-a_0), where a_0 is the value of the lattice constant in the absence of exchange

$$J = J_0 + \frac{\partial J}{\partial a}\left(a - a_0\right) = J_0 + J_1 u_{ll}, \quad \text{where} \quad J_1 = \frac{1}{3}\frac{\partial J}{\partial a} \tag{1}$$

In the nearest-neighbor approximation, we obtain $J=ZS^2I/2a^3$, where S is the spin of a magnetic atom; Z is the number of neighbors; I is the exchange integral; u_{ll} is the sum of the diagonal components of the strain tensor. The problem of self-localization of electron states can be solved by the variational method. We shall assume that a ferromagnetic state is stable within a sphere of radius R and the ordering outside this sphere is anti-ferromagnetic. The radius of the aforementioned region R is treated as a trial parameter. We shall also assume that the sample is subjected to a pressure P.

The total energy functional written in the first approximation in AS/W has the following form:

$$E = \frac{\hbar^2}{2m^*}\int\left(\nabla\psi\right)^2 d^3r - \frac{AS}{2}\int\Theta_1\left(r\right)\psi^2 d^3r - \int\left(J_0 + J_1 u_{ll}\right)\Theta_2\left(r\right)d^3r$$
$$+ \int\left[\mu\left(u_{ik} - \frac{1}{3}\delta_{ik}u_{ll}\right)^2 + \frac{K}{2}u_{ll}^2\right]d^3r \tag{2}$$

where ψ is the electron wave function; u_{ik} Is the strain tensor; K and μ are the bulk and shear moduli. The first two terms in Eq. (2) represent the energy of an electron interacting with magnetic atoms via the s-f exchange, the third term corresponds to the exchange energy of magnetic atoms, and the fourth term to the deformation energy; since the pressure acts on the crystal surface, it can be taken into account in the equilibrium equation as a boundary condition.[10] The functions $\Theta_1(r)$ and $\Theta_2(r)$ are determined by the spin ordering of the lattice and are given by:

$$\Theta_1\left(r\right) = \begin{cases} 1, & r < R; \\ 0, & r > R; \end{cases} \qquad \Theta_2\left(r\right) = \begin{cases} 1, & r < R, \\ -1, & r > R. \end{cases}$$

The lattice deformation necessarily results in a lowering of the energy of the ferron state. This is due to the fact that the deformations are governed by equilibrium equations corresponding to the minimization of the functional (2) with respect to the displacements u(r). Using Eq. (2), we shall evaluate the stress tensor σ_{ik} from the general expression

$$\sigma_{ik} = \frac{\delta E}{\delta u_{ik}} = K u_{ll}\delta_{ik} + 2\mu\left(u_{ik} - \frac{1}{3}\delta_{ik}u_{ll}\right) - J_1\delta_{ik}\Theta_2(r) \tag{3}$$

Then, we can apply Eq. (3) to transform the equilibrium equation for the stress tensor $\partial\sigma_{ik}/\partial x_k = 0$ to the following form:

$$\Delta u + \frac{1}{1-2\sigma}\, grad\ div\ \mathbf{u} = 0 \tag{4}$$

where $\mathbf{u}$ is the strain vector; Δ is the Laplace operator; σ is the Polsson ratio; $\mu = E/2(1+\sigma)$, $K=E/3(1 - 2\sigma)$, E is the Young modulus. Since the strain must be finite at $r = 0$, we can seek the solution of Eq. (4) in the following form:

$$\mathbf{u} = \begin{cases} C_1 r, & r < R; \\ C_2 r + C_3\dfrac{r}{r^3}, & r > R, \end{cases} \tag{5}$$

where C_1, C_2, and C_3 are unknown constants which can be evaluated from the continuity of displacements and stresses

$$\left.\begin{aligned} &u(R-0) = u(R+0); \\ &\frac{E}{(1+\sigma)(1-2\sigma)}\left[u_{rr}(1-\sigma)+\sigma\left(u_{\theta\theta}+u_{\varphi\varphi}\right)\right]_{R+0} + J_1 = \frac{E}{(1+\sigma)(1-2\sigma)} \\ &\qquad\times\left[u_{rr}(1-\sigma)+\sigma\left(u_{\theta\theta}+u_{\varphi\varphi}\right)\right]_{R-0} - J_1; \\ &\sigma_{rr} = \frac{E}{(1+\sigma)(1-2\sigma)}\left[(1-\sigma)u_{rr}+\sigma\left(u_{\theta\theta}+u_{\varphi\varphi}\right)\right]+J_1 = -P, \quad r\to\infty \end{aligned}\right\} \tag{6}$$

It follows from Eq. (5) that the equalities

$$\left.\begin{aligned} &u_{\theta\theta} = u_{\varphi\varphi} = u_{rr} = C_2 + \frac{C_3}{R^3}, \quad r < R; \\ &u_{\theta\theta} = u_{\varphi\varphi} = C_2 + \frac{C_3}{r^3}; \quad u_{rr} = C_2 - \frac{2C_3}{r^3}; \quad r > R \end{aligned}\right\} \tag{6'}$$

are satisfied. Using Eqs. (6) and (6`), we obtain the following expressions for the unknown coefficients:

$$C_1 = \frac{J_1}{3K}\frac{3K-4\mu}{3K+4\mu} - \frac{P}{3K}, \quad C_2 = -\frac{J_1}{3K} - \frac{P}{3K}, \quad C_3 = \frac{2J_1}{3K+4\mu}R^3. \tag{7}$$

Substituting Eqs. (6') and (7) in Eq. (2), we find the total energy functional:

$$E = \frac{\hbar^2}{2m^*} \int (\nabla \psi)^2 \, d^3r - \frac{AS}{2} \int \Theta_1(r) \psi^2 d^3r$$
$$+ \frac{4\pi R^3}{3} \left[-2J_0 + \frac{2J_1}{K} \frac{4\mu}{3K + 4\mu} - \frac{2PJ_1}{K} \right] + E_A, \tag{8}$$

where E_A is the energy of the antiferromagnet in the absence of an electron. To proceed further, the actual form of the electron trial wave function will have to be specified. For $R \gg a$, the trial wave function reduces to the wave function of a particle in an infinitely deep spherical potential well of radius R (with an accuracy up to terms $\sim a/R$)[1]

$$\psi = \frac{1}{\sqrt{2\pi R}}, \quad \frac{\sin \zeta r}{r} \zeta = \sqrt{2m^* \left(\frac{AS}{2} - |E_e| \right)}, \quad r < R \tag{9}$$

where E_e is the electron energy. Substituting Eq. (9) in Eq. (8), we obtain

$$E \approx \frac{\hbar^2 \pi^2}{m^* R^2} + \frac{8\pi}{3} R^3 \left(-J_0 + \frac{J_1^2}{K} \frac{4\mu}{3K + 4\mu} - \frac{PJ_1}{K} \right) - \frac{AS}{2} + E_A \tag{10}$$

The equilibrium value of the radius R can be obtained from the requirement that e should have a minimum: $\partial \varepsilon / \partial R = 0$. It follows from Eq. (10) that the equilibrium radius is given by the following expression that holds not only for metamagnetic materials but also for Heisenberg antiferromagnets with a strong volume dependence of me exchange Integral:

$$R = \left[\frac{\hbar^2 \pi}{8m^* \left| -J_0 + \frac{J_1^2}{K} \frac{4\mu}{4\mu + 3K} - \frac{PJ_1}{K} \right|} \right]^{1/3} \tag{11}$$

For metamagnetic materials, it is necessary to assume that the lattice constant in the absence of pressure ($P = 0$) is close to the value a_c for which the exchange integral changes its sign,[7] i.e.,

$$J = L(a - a_e), \quad J_0 = L(a_0 - a_e), \quad J_1 = \frac{1}{3} La_0 \tag{12}$$

Since $u_{ll} = 3[(a - a_0)/a]$, the energy of a crystal in the absence of conduction electrons is given by[7] (per magnetic atom)

$$E = -La^3(a - a_e) \cos 2\varphi + \frac{9K}{2a^2} (a - a_e)^2 - hS \cos \varphi \tag{13}$$

in accordance with Eq. (2). we have

$$a_0(P) = a_0(0) \left[1 - \frac{P}{3K} \right],$$

where h is the external magnetic field (in energy units) and φ is the angle between the magnetic field and the magnetic moment. The minimization of Eq. (13) with respect to the lattice constant a and the angle φ indicates[7] that, for $h = 0$, the energy change corresponding to the ferromagnet-antiferromagnet transition is given by

$$E_{AF} - E_F = 2La^3 \left[a_0 P - a_e \right] \equiv 2J_0(P) \tag{14}$$

The lattice constants corresponding to the ferromagnetic and antiferromagnetic orderings are given by

$$a_F(P) = a_0(P) + \frac{La_0^5}{9K}, \qquad a_{AF} = a_0(P) - \frac{La_0^5}{9K}. \tag{15}$$

In an external magnetic field $h < h_{sf}$ (h_{sf} is the spinflop field), the orientations of the moments of both sublattices of an antiferromagnet are practically unchanged. Therefore, Eq. (14) assumes the form

$$E_{AF}(h) - E_F(h) \cong 2J_0 + hS = 2J_0 \left(1 - \frac{h}{h_{sf}} \right); \quad Sh = 2|J_0|. \tag{16}$$

Replacing in Eq. (11) J_0 by $J_0(1 - h/h_{sf})$, we obtain the following expression for the ferron radius [see Eq. (15)]:

$$R = \left[\frac{\hbar^2 \pi}{8m^* \left| |J_0| \left(1 - \frac{h}{h_{sf}} \right) + \frac{9}{4} K \left(\frac{a_F(P) - a_{AF}(P)}{a_F(P)} \right)^2 \frac{4\mu}{4\mu + 3K} \right|} \right]^{1/6}. \tag{17}$$

It follows from Eq. (17) that the ferron moment increases with increasing h but the ferron radius remains finite at $h = h_{sf}$. Assuming that m^* is the free electron mass and the elastic constants K and m are of the order of 10^{10}-10^{11} N/m^2, we find that, for $|(a_F - a_{AF})/ a_{AF}| \approx 10^{-3}$ (for our estimate, we use the magnetostriction in EuO in the ferromagnetic state induced by a magnetic field[11] above T_c) and $h = h_{sf}$, the radius of the ferromagnetic region is given by $R \approx 9a$, where $a \approx 3A$. The quantity J_0 can be estimated using Eq. (16) and the critical field for the dissociation of ferron states $h_{sf} \approx 10^3$ Oe (see Ref. 9). Therefore, the ratio of the ferron moments for $h = h_{sf}$ and $h = 0$ is given by [Eq. (17)]

$$\frac{M(h_{sf})}{M(0)} = \frac{R^3(h_{sf})}{R^3(0)} \approx 2$$

This fact should be taken into account in the experimental observation of ferrons from the change of the magnetic susceptibility of a sample due to illumination.

The antiferromagnetic-ferromagnetic phase transition under pressure was studied in EuSe and EuTe in Refs. 6 and 12. It follows from Refs. 6 and 12 that the antiferromagnetic ordering corresponds to a larger lattice constant than the ferromagnetic ordering: $a_{AF} > a_F$.

Therefore, the antiferromagnet-ferromagnet phase transition takes place under pressure.[6,12] Therefore, it is necessary to assume in Eq. (12) that a_0 is greater than a_c and $L < 0$. Equation (17)

also describes the pressure dependence $P_c = 3K(1 - a_c/a_0)$ of the ferron moment up to Its critical value for which $a_0(P)$ reaches the value a_c. The ferron moment increases as a function of the pressure and, in the absence of a field h, its value for $P = P_c$ is approximately twice as large as the value corresponding to $P = 0$ (the estimates apply to the same numerical values of the parameters).

To avoid misunderstanding, we would like to stress that the ferron radius remains finite at the phase transition point regarded as a function of P. Moreover, the ferron moment remains finite even in the region in which the antiferromagnetic state is metastable and increases as a function of the distance from P_c and tends to infinity underpressure $P_2 = 3K(1 - ac/a0 + |J_1| 4\mu/(4\mu + 3K)$. The ferron moment remains finite for $P_2 > P > P_c$ because of strains Induced in the metastable phase created by a heterogeneous nucleation center. The situation here is similar to the nucleation centers that appear in a first-order phase transition.

Finally, we would like to point out that the model of a metamagnetic material proposed in Ref. 7 simplifies somewhat the actual situation in EuSe. In fact, the anti-ferromagnet-ferromagnet phase transition in EuSe in a magnetic field is preceded by a antiferromagnet-ferromagnet phase transition with $h_{sf} \sim 100$ Oe. However, ferrons can occur even in the ferrimagnetic phase since its moment is small.[13] In fact, our theory can essentially be applied also to metamagnetic transitions of the type ferrimagnet-ferromagnet.

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