Self-localization of a conduction electron in magnetically ordered crystals

V. D. Lakhno
Scientific-Research Computing Center, Pushchino, Moscow Province (Submitted November 16, 1981; resubmitted May 6, 1982) Fiz. Tverd. Tela (Leningrad) **24,** 2787-2793 (September 1982)

The conditions for the formation of self-localized states in antiferromagnets are analyzed. Criteria for the stability of such states are derived. Cases of weak and strong external magnetic fields are discussed.
PACS numbers: 75. 50. Ee, 71. 50. +t

The possibility of conduction electron self-localization in crystals with antiferromagnetic ordering of the lattice spins was first noted by de Gennes.[1] He put for ward the hypothesis that the self-localized state may be energetically more favorable than the band state of the free electron when the spin of one of the magnetic atoms in the antiferromagnetic lattice is reversed. In later work, it was shown that states may be even more favorable in which the electron self-localization takes place in a region where several spins of magnetic atoms are reversed.[2] The calculation of the optimal configurations of a spin system of magnetic atoms is evidently a complex and still unsolved problem.

Recently, however, it has become clear that the formation of self-localized states in magnetically ordered crystals requires the overcoming of the potential barrier arising from the short-range nature of the interaction in such systems. [3] It thus became of prime importance to discuss the conditions for self-localized states of this type to be possible in principle. In particular, the possibility of self-localized states on the surface of a sample has been discussed[4]; here, as the problem is quasi-two-dimensional, there is no potential barrier to be overcome. In a recent paper,[5] it has been shown that self-localized states can also occur within a sample subjected to a strong external magnetic field.

The present study analyzes, for a number of particular cases, the conditions under which localized states are formed in antiferromagnetic crystals. Stability criteria are derived in the general case, i.e., without reference to the configuration of the spin distribution and energy of such states. Some ways of creating these states in crystals are discussed.

1. FORMATION OF A SELF-LOCALIZED STATE IN CRYSTALS HAVING AN UNDEFORMED LATTICE

The interaction between the electron and the spins of magnetic atoms will be described by means of the s-f model[6]:

$$H_{e-f} = \sum_{f,S} A(f-g)(S_f, \sigma_g)$$

where y and S are the spin operators of the conduction electron and a magnetic atom, f the number of the atom, and A(f-g) the s-f exchange integral. We assume, as usual, that the conduction band width W is much greater than the s~f exchange energy AS/2. We shall take only the nearest-neighbor approximation.

In the continuum approximation (i. e., when all characteristic dimensions of the problem are much greater than the lattice constant), the magnetic moments of the atoms have a continuous distribution over directions, and the total energy functional becomes[4, 5]

$$F[\psi,\varphi]=\frac{\hbar^2}{2m^*}\int|\nabla\psi|^2 d\tau-\tilde{A}\int|\psi|^2\cos\varphi\, d\tau+L\int\cos 2\varphi\, d\tau \tag{1}$$

Here, A = AS/2, L = J/2a^3, J = ZIS^2, where Z is the number of nearest neighbors, I the Heisenber exchange integral, a the lattice constant, and 2φ is the angle between the spins of two neighboring atoms.

In the functional (1), the first two terms relate to the kinetic energy of the electron and the energy of its interaction with the spins of the magnetic atoms. The last term describes the Heisenberg exchange interaction between the atoms. This treatment is essentially analogous to the semiclassical description in the theory of a strongly bound polaron: the polarizable medium is here represented by the spin system, the spins being regarded as classical vectors. The model thus defined is self-consistent: the electron generates a spin distribution (specified by the angle φ) that it maintains by its localization therein.

Let us consider the conditions under which self-localization becomes favorable. Since it originates from a state with an undeformed antiferromagnetic lattice, we can take $\varphi \neq 0$ in the initial stage of the spin system deformation and the local state formation. Then, from Eq. (1),

$$\cos\varphi=\frac{\tilde{A}}{4L}|\psi|^2. \tag{2}$$

Substitution of this in Eq. (1) gives for the total energy functional[1)]

$$F[\psi]=\frac{\hbar^2}{2m^*}\int|\nabla\psi|^2 d\tau-\frac{\tilde{A}^2}{8L}\int|\psi|^4 d\tau. \tag{3}$$

This is in the same form as the functional given by the interaction of the electron with acoustic phonons.[7] It corresponds[7] to the case where the initial stage of the spin system deformation involves a loss of total energy, i.e., the self-localized state is separated from the free band electron state by a potential barrier. Using the same method as before,[7] let us consider the change in the total energy (3) when the wave function is deformed without change of normalization:

$$\psi_\xi=\xi^{3/2}\tilde{\psi}(\xi r), \tag{4}$$

where $\xi = 1$ corresponds to the wave function belonging to the extremum of the functional (3). Substitution of Eq. (4) in Eq. (2) gives

$$\max\ \cos\varphi=\frac{\tilde{A}}{4L}\max|\psi_\xi(r)|^2=\frac{A\xi^3}{4L}|\tilde{\psi}(0)|^2\le 1 \tag{5}$$

Thus, as ξ increases, a point is reached where the inequality (5) is no longer satisfied. This means that a region is formed in the crystal where the spins of the atoms are in parallel alignment (sin $\varphi = 0$). Let ξ_{cr} be the value of ξ for which $\varphi=1$. Three possibilities exist. If $\xi_{cr} < 1$, the self-localization is energetically unfavorable and the localized state is unstable. If $1 \le \xi_{cr}$ s$^3/_2$, it is again energetically unfavorable, but is separated from the continuous spectrum by a potential barrier. Such a state is metastable. Lastly, if $\xi_{cr} > {}^3/_2$, the self-localized state is energetically favorable. The value of ξ_{cr} can be calculated on a computer, and is

$$\xi_{cr}\cong 0.021(2m^*a^2/\hbar^2)\tilde{A}^{5/3}(2/J)^{2/3}. \tag{6}$$

The condition $\xi_{cr} \geq 3/2$ for the self-localized state to be energetically favorable may thus be written as

$$\widetilde{A} \geq 13(J/2)^{2/5}(\hbar^2/2m^*a^2)^{3/5}. \tag{7}$$

This condition defines the class of crystals in which the formation of self-localized states of the type in question is in principle possible.

For a numerical evaluation of the inequality (7), we choose parameter values typical of antiferromagnets: $m^* \approx m$, $J \approx 10^{-2}$ eV, $a \approx 3 \cdot 10^{-8}$ cm. We then find that the self-localized state is stable if $\widetilde{A} > 1$ eV. In the majority of crystals, the s -f exchange integral is usually[6] in the range $0.01 \leq \widetilde{A} \leq 1$ eV. In typical antiferromagnets, therefore, such states will usually not occur. There are, however, a number of crystals with extremely low Neel points (such as europium selenide and telluride) in which the condition (7) is satisfied and the formation of a state of the type under consideration is in principle possible.

2. FORMATION OF A SELF-LOCALIZED STATE IN CRYSTALS HAVING A DEFORMED LATTICE

The above results can be generalized to the case where lattice deformation (the condenson effect) takes place alongside deformation of the crystal spin system in the formation of a self-localized state. For an isotropic crystal, the total energy functional of the system is

$$\widetilde{F}[\psi,\varphi,u_{ik}] = F[\psi,\varphi] - G\int u_{ll}|\psi|^2 d\tau + \int\left[\mu\left(u_{ik} - \frac{1}{3}\delta_{ik}u_{ll}\right)^2 + \frac{K}{2}u_{ll}^2\right]d\tau, \tag{8}$$

where $F[\psi,\varphi]$ is given by Eq. (1), G is the deformation potential constant, K and μ the bulk modulus and shear modulus respectively. The variational parameters in Eq. (8) are the electron wave function ψ, the angle φ, and the strain vector **u**. Varying this functional with respect to **u** gives the Euler equation

$$\nabla^2 \mathrm{u} + \frac{1}{1-2\sigma}\,grad\ div\ \mathrm{u} = \frac{2(1+\sigma)}{E}\,grad|\psi|^2, \tag{9}$$

where ∇^2 is the Laplacian operator, σ Poisson's ratio, $\mu = E/2(1+\sigma)$, $K = E/3(1-2\sigma)$, and E is Young's modulus. The spherically symmetric solution of Eq. (9) which satisfies the conditions that the strain is finite at the origin and zero at infinity is

$$\mathrm{u} = \frac{Q}{4\pi}\frac{r}{r^3}\int_0^r |\psi|^2 d\tau, \qquad Q = \frac{(1+\sigma)(1-2\sigma)}{E(1-\sigma)}G. \tag{10}$$

Substitution of Eqs. (2) and (10) in the functional (8) gives the following expression for the total energy:

$$\widetilde{F}[\psi] = \frac{\hbar^2}{2m^*}\int|\nabla\psi|^2 d\tau - \frac{\widetilde{A}^2}{8L}(1+\kappa)\int|\psi|^4 d\tau, \tag{11}$$

$$\kappa = \frac{3}{2}\frac{8L}{\widetilde{A}^2}\frac{G^2}{3K+4\mu}. \tag{12}$$

In this case also, therefore, the formation of a self-localized state involves the overcoming of a potential barrier. It is important to note, moreover, that taking account of the lattice strain involves a further gain in the energy of the localized state, and therefore, a further stabilization of such states. According to the condition (7), the critical Neel point value above which the self-localized state does not exist becomes $(1 + \kappa)^{5/4}$ T^c_N, where T^c_N is the corresponding value with no lattice strain. To estimate κ, we take in Eq. (12) the same parameter values as in estimating the inequality (7). For typical values $G \approx 10$ eV, $K \approx \mu \approx 10^{10}$-$10^{11}$ N/m^2, κ can reach values ~1, i.e., the inclusion of the strain may raise the critical value of the Neel point by a factor of nearly 2.

3. FORMATION OF A SELF-LOCALIZED STATE IN THE PRESENCE OF A WEAK MAGNETIC FIELD

Let a weak (nonquantizing) external magnetic field be applied to an antiferromagnet. We take first the case of a crystal having a rigid (not deformable) lattice. The total energy functional becomes

$$\widetilde{F}[\psi,\varphi] = F[\psi,\varphi] - \widetilde{H}\int \cos\varphi\, d\tau, \tag{13}$$

where $\widetilde{H} = HS/a^3$,. H being the magnetic field in energy units. Varying this functional with respect to the angle φ gives the condition

$$\cos\varphi = \frac{\widetilde{A}}{4L}|\psi|^2 + \frac{\widetilde{H}}{4L}. \tag{14}$$

Substitution of Eq. (14) in Eq. (13) leads to the following expression for the total energy functional:

$$\widetilde{F}[\psi,\varphi] = F[\psi,\varphi] - \frac{\widetilde{A}\widetilde{H}}{4L}\int |\psi|^2 d\tau, \tag{15}$$

where the terms corresponding to the crystal energy without the electron have been omitted, and $F[\psi]$ is given by Eq. (3). It follows from Eq. (15) that in the case of a weak magnetic field a potential again has to be overcome.

Substitution of Eq. (4) in Eq. (14) gives

$$\max\cos\varphi = \frac{\widetilde{A}\xi^3}{4L}|\psi(0)|^2 + \frac{\widetilde{H}}{4L} \le 1. \tag{16}$$

From this, it follows that, as ξ increases, a point is reache at which the inequality (16) is no longer satisfied. Let $\xi_{cr}(H)$ denote the value of ξ for which cos ц = 1. As in Sec. 1, there are three possibilities. If $\xi_{cr}(H) < 1$, self-localization is energetically unfavorable. When $1 \le \xi_{cr}(3) \le {}^3/_2$, a metastable state can be formed. Lastly, when $\xi_{cr}(H) > {}^3/_2$, the self-localized state becomes energetically favorable. According to the relation (16), $\xi_{cr}(H)$ and $\xi_{cr}(O)$ are such that

$$\xi_{cr}(H) = (1 - \tilde{H}/4L)^{1/3}\xi_{cr}(0). \tag{17}$$

This shows that, if a crystal has a stable self-localized state, as the magnetic field increases the state first becomes metastable and then unstable, rapidly disappearing.

Let us now consider a crystal with a deformable lattice. Clearly, in cases where the formation of self-localized states depends mainly on deformation of the magnetic subsystem in the crystal ($\kappa \ll 1$), the external magnetic field is able to destroy the local state, as with a rigid lattice. In this case, the s—f exchange constant in the expressions (13) through (16) is replaced by its effective value $\tilde{\tilde{A}} = \sqrt{1+\kappa}\,\tilde{A}$. If the interaction of the electron with the lattice strain is predominant ($\kappa \gg 1$), the magnetic field does not destroy the local state. In the general case where interactions of both types are significant, there may be self-localized states of two kinds in the crystal, one due to the deformation of the spin subsystem and the other to the deformation of the lattice.

4. SELF-LOCALIZATION IN A STRONG MAGNETIC FIELD

We have discussed self-localized states in crystals having antiferromagnetic spin ordering, in the absence of an external magnetic field and in a weak magnetic field. In both cases, the formation of self-localized states involves the overcoming of a potential barrier. This impedes their formation within the crystal. It has been shown[5] that the situation is quite different if a strong magnetic field (not exceeding the critical field H_C for collapse of the sublattices in the antiferromagnet) is applied to the crystal. In this case, self-localization becomes favorable right from the initial state of polarization of the spin system, i.e., does not involve the overcoming of a potential barrier. Whereas, with no field or a weak field, there is a critical value of the Neel point above which the self-localized state is destroyed, in a strong field self-localization is possible for any value of the Heisenberg exchange integral. [2)] In this section, the previous results[5] will be generalized to the case of a deformable lattice.

The total energy functional in a strong magnetic field, with allowance for the lattice strain, is

$$F[\psi, \varphi, u_{ik}] = \frac{1}{2m^*}\int \psi^*\left(\mathrm{p} + \frac{e}{c}\mathrm{A}\right)^2 \psi\, d\tau - \tilde{A}\int |\psi|^2 \cos\varphi\, d\tau + L\int \cos 2\varphi\, d\tau$$
$$- G\int u_{ll}|\psi|^2 d\tau - \tilde{H}\int \cos\varphi\, d\tau + \int\left[\mu\left(u_{ik} - \frac{1}{3}\delta_{ik}u_{ll}\right)^2 + \frac{K}{2}u_{ll}^2\right]d\tau, \tag{18}$$

where $\mathbf{p} = -i\hbar\nabla$ and $\mathbf{A}$ is the vector potential of the field. The independent variation of the functional with respect to φ and $\mathbf{u}$ gives therelations (14) and (10) respectively. Substituting Eqs. (14) and (10) in Eq. (18), we get as the total energy functional

$$F[\psi] = \frac{1}{2m^*}\int \psi^*\left(\mathrm{p} + \frac{e}{c}\mathrm{A}\right)^2 \psi\, d\tau - \frac{\tilde{A}^2}{8L}(1+\kappa)\int |\psi|^4 d\tau - \frac{\tilde{A}\tilde{H}}{4L}\int |\psi|^2 d\tau, \tag{19}$$

which is the same in form as that considered previously.[5] The electron wave function and energy in the ground state are[5]

$$\psi = \pm\frac{1}{\sqrt{4\pi\rho_0^2 r_z}}\left(ch\frac{z-z_0}{r_z}\right)^{-1}\exp\left\{-\frac{\rho^2}{4\rho_0^2}\right\}, \qquad \rho_0 = \sqrt{\frac{2\mu_B c\hbar}{eH}},$$

$$E_e = -\frac{\hbar^2}{2m^* r_z^2} - \frac{\widetilde{A}\widetilde{H}}{4L}, \qquad r_z = \frac{\hbar^2}{2m^*} 8\pi\rho_0^2 \frac{8L}{\widetilde{A}^2} \frac{1}{(1+\kappa)}, \tag{20}$$

where r_z is the radius of the localized state and μ_B the Bohr magneton. The expressions (20) for the energy and the wave function, which take account of the lattice strain, differ from those for a rigid lattice by the presence of the factor $1 + \kappa$ in the expression for the radius r_z of the localized state. Including the lattice strain thus reduces the characteristic radius of the state, and therefore decreases the electron energy by a factor $(1 + \kappa)^2$. This is most important, since for parameter values typical of magnetic crystals with a rigid lattice the gain in the total energy ($=^1/_3$ $|\Delta E|$, $\Delta E = -\hbar^2/2m^* r^2_z$) by self-localization is only slight, and such states are thus destroyed above 3-4°K. Including the lattice strain makes such states more stable, the temperature range of stability being increased by a factor $(1 + \kappa)^2$.

The decrease in the radius of the localization region, due to the lattice strain, reduces the local field value at which a region with parallel spin orientation is first formed,

$$H_{loc} = \frac{32\pi^2\gamma^3 JS}{(1+\kappa)} \left[\sqrt{1 + \frac{1+\kappa}{8\pi^2\gamma^3 S^2}} - 1 \right], \qquad \gamma = \frac{\hbar^2}{ma^2\widetilde{A}} \left(\frac{m}{m^*} \right)^{1/3}. \tag{21}$$

In a field $H_{loc} < H < H_C$, the electron self-localization region contains a region of ferromagnetic spin ordering. The ferromagnetic microregion containing a localized electron, formed by a strong magnetic field, can itself act as a nucleus for the formation of a stable state (Sec. 1) when the field is removed. No potential barrier then needs to be overcome in order to form the self-localized state.

5. COMPARISON OF THEORY AND EXPERIMENT

Wachter[8] examined the effect of a magnetic field on luminescence in crystals having a low Neel point, and concluded that the experimental results can be explained by the formation of self-localized states in an antiferromagnet. Since this process involves the overcoming of a potential barrier (see above), a different explanation is more reasonable. Namely, the characteristic energy corresponding to the maximum of the observed absorption band corresponds rather to a transition from a local level of a defect such as a valence electron localized at a lattice vacancy. This appears the more probable in that the localization of the electron near the defect right from the initial stage makes a deformation of its spin environment favorable. At present, however, there is uncertainty on this point, to resolve which further studies, both experimental and theoretical, are needed.

There has apparently been no experimental study of self-localization in a strong magnetic field. To obtain such states, a low temperature and a strong magnetic field are necessary. The formation of these states should be accompanied by a considerable change in the magneto-resistance and by the appearance of further bands in the infrared.

[1)] We must emphasize again that the functional (3) corresponds only to the initial stage of deformation. The final state of the electron resulting from deformation of the spin system is found as the minimum of the functional (1).

[2)] Here, it is important that the exchange integral should not be too small, since otherwise the sublattices are reversed before the strong-field condition is satisfied. The conditions for the states discussed[5] to occur are therefore the opposite of those for the states considered above to become stable.

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Translated by J. B. Sykes