New type of self-trapped carrier state in antiferromagnetic semiconductors and the HTSC problem

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A new type of self-trapped charge-carrier state in antiferromagnetic (AFM) semiconductors, which can be realized in materials with high Néel points, is proposed. The carrier produces a microscopic normally unstable AFM region, in which it is trapped. Its energy in this region is lower than in the stable one. An additional energy gain is produced by the canting of the moments of the sublattices in the carrier trapping region. Self-trapped carriers can become paired to form zero-spin complexes that can lead to high-temperature superconductivity of degenerate semiconductors. Calculation of the self-trapping of carriers in a chessboard-order antiferromagnetic within regions with layered AFM ordering show that this trapping can take place even far from the boundary of these two phases. The possibility of self-trapping of carriers at $T = 0$ in a spin-fluid microregion is discussed.

INTRODUCTION

It is common knowledge that the HTSC problem is closely connected with the problem of magnetic self-trapped states in antiferromagnetic (AFM) semiconductors, as are almost all high-temperature superconductors. The same interactions that lead to self-trapping can lead also to pairing of charge carriers.1)

Three different models have been proposed to date for self-trapping in AFM semiconductors. In the de Gennes model[1] it was assumed that the spin of one of the magnetic atoms is skewed away from its initial position and this skew is sustained by a trapped nearby carrier. A subsequent analysis has shown, however, that such a state is possible at utterly fantastic, at the very least tenfold and even tens of times larger, values of the magnetic-atom spin S [see Ref. 2 and the latest survey of magnetic semiconductors[3]].

A self-trapping model proposed in Ref. 4 has become very popular most recently. The term "quasioscillator" proposed in it is replaced now by the term "magnetic string." This model is suitable only for AFM semiconductors with $S = 1/2$, if they are inadequately described by the Hubbard model in its semiconductor limit. A carrier in a string state oscillates about a certain center. A carrier moving away from the center produces on its trajectory an antiphase AFM ordering that increases the exchange energy of the magnetic atoms. If, however, the carrier returns along the trajectory, it replaces the antiphase ordering by a normal one.

It must be noted that recent authors state that Refs. 4 correspond to the Ising rather than the Heisenberg model. This statement, however, is unfounded: in fact, use is made in Refs. 4 of the perfectly natural inequality $W \gg T_N$, where W is the width of the energy band of the carrier and T_N is the Néel temperature (W is of the first order and T_N of second order of smallness with respect to the overlap of neighboring-atom orbits). It is just this inequality which makes it possible to regard the magnetic-order disruption by the interatomic transition as basic, and the quantum oscillations of the magnetic-atom spins as corrections to it. In particular, as indicated in Refs. 4, allowance for quantum oscillations is essential for a description of translational motion of a string through a crystal, but it has little effect on the energy of a string at rest.

In the third type—ferron[5]—of self-trapping the carrier produces in the AFM semiconductor a microscopic region of ferromagnetic (FM) phase and stabilizes it by its own inner self-trapping. The reason why phase trapping the carrier was regarded as FM was that just in FM ordering is where the carrier energy is minimal. A survey of many subsequent ferron studies (following Ref. 5, the ferron was rediscovered many times by other workers) is contained in Refs. 2 and 3. Real ferrons can be produced only when the energy loss for production of an FM region is small, i.e., in materials with sufficiently low T_N.

A new type of self-trapped carrier in an AFM semiconductor is proposed in the present paper. Its idea is based on the fact that a potential well for a carrier can be not only an FM, but also some other phase, for example an AFM but of a type different from that realized in a crystal. In particular, the carrier energy in a layered AFM phase is lower than in a chessboard AFM phase, and their difference can reach several times ten eV. A carrier therefore becomes self-trapped in an antiferromagnetic with chessboard ordering in a microregion with a layered AFM ordering. The energy of the "carrier + microregion of another AFM phase" quasiparticles (dubbed "afmon" hereafter2)) can be additionally lowered if the moments of the sublattices in the self-trapping region are skewed and the quasiparticle acquires an additional magnetic moment.3) Large skews of the moments, however, are excluded by the condition that the ferrons become energywise unfavored so that, strictly speaking, afmons become possible.

Naturally, afmon self-trapping does not postulate low Néel points. On the contrary, it is possible only in materials with sufficiently high T_N, where ferrons are impossible. The optimal conditions for the onset of afmons obtain near the boundary of two AFM phases. It will be shown below, however, that they can be realized also far enough from the phase boundary.

If the carrier density is high enough, cooperative effects among the afmons become substantial. The simplest of them

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is afmon pairing, which can have the most direct bearing on HTSC. It should be pointed out that the existence of HTSC was explained earlier as being due to the possibility of pairing of strings[6,7] and ferrons.[8] Since paired strings have zero spin and finite mobility, they can play a role in superconductivity. Similar arguments hold also for biferrons. The question of afmon pairing will be discussed in Sec. 5.

In contrast to biferrons, which have nonzero but integer moments, afmon pairing should lead the quasiparticles with zero spin; placement of two carriers with opposite spins in an orbital ground state suppresses the skewing of biafmon sublattice moments. At the same time, in contrast to the ferrons, pairing of afmons becomes energywise pronounced since formation of a triafmon calls for placing a third carrier in a higher orbital state separated by a gap from the ground state. The conditions for the existence of a triafmon are therefore much more difficult to satisfy than those for a biafmon.

Still higher carrier densities permit more complicated afmonic states corresponding to breakup of a degenerate semiconductor into two phases. One is the normally stable ("old") AFM phase, from which two carriers go off. The second is a normally unstable ("new") AFM phase stabilized by receiving all the carriers in the crystal. Such a state is the analog of the cooperative ferron state first investigated in Ref. 9, in which the crystal breaks up into AFM and FM phases, with all the conduction electrons concentrated in the FM phase. It is natural to expect the structure of the two-phase state in both cases to be the same. At relatively low carrier densities the new FM phase forms an aggregate of old-phase drops insulated from one another, and each such drop contains several carriers. With increase of the carrier density the number and dimensions of the new-phase drops increase, and at some density percolation sets in: the drops of the new phase come in contact with one another. The crystal changes instantaneously from insulating (all the carriers are trapped each in its own drop) into highly conducting. After the percolation the new-phase region changes from multiply to singly connected, and the opposition transition takes place in the region of the old phase.

Obviously, a singly connected region cannot exhibit long-range AFM order—only a short-range order is possible within the confines of one and the same drop. Long-range order, however, can also be absent from a singly connected phase, since the dimensions and locations of the drops of the second phase have a certain randomness due to fluctuations of the electroactive defects. It may be energywise easier for the singly connected phase to break up into domains that ensure a minimum energy of the boundary between the two phases.

As the temperature rises, the two-phase state becomes single-phase, ordered or paramagnetic, in full analogy with melting, investigated in Ref. 10, of a cooperative ferron state. Particular interest attaches to the first case, when a long-range AFM order first appear when the temperature rises but then vanishes with further rise. Such a reversing AFM long-range order recalls the one usually observed in HTSC. It is natural to assume that in real HTSC it is also due to phase separation.

Strictly speaking, one cannot guarantee the existence of some other magnetic phases in which carrier trapping is even more favored energywise than in the new AFM phase. In particular, the estimates that follow show that at certain values of the parameters self-trapping is more favored not in the new AFM phase but in regions which are fully disorderd at $T = 0$ ("spin fluid"). It has not been able, however, to prove rigorously that at the same parameters there will not be an energywise even more favored self-trapping in FM regions. If self-trapping is indeed feasible in a spin fluid, it would be possible to attribute in a natural manner, for degenerate semiconductors, the vanishing of long-range AFM order when the temperature is lowered, to formation of spin-fluid regions in which charge carriers are localized.

At any rate, if there exists some other self-trapping mechanism more effective than the afmonic, this only reinforces our main statement: self-trapping of a carrier in an altered-phase region is possible also in AFM semiconductors with high T_N.

1. DEPENDENCE OF CARRIER ENERGY ON THE TYPE OF THE AFM ORDERING

The analysis that follows is based on the *s-d* (*s-f*) model. According to Ref. 2, it describes adequately not only situations in which the carriers and the localized magnetic moments of the atoms correspond to orbital states of different type, but also when they are of the same type. For example, the $3D$ model is equivalent to the Hubbard model in the semiconductor limit, when the *d*-atom spin S is 1/2, and the *s-d* exchange integral A tends to $-\infty$. The results of the *t-J* model are also duplicated in the *s-d* model.

The *s-d* model Hamiltonian is taken here in the form

$$H=B\sum a_{\mathbf{g}\sigma}^{*}a_{\mathbf{g}+\Delta\sigma}-A\sum(\mathbf{S}_{\mathbf{g}}\mathbf{s})_{\sigma\sigma'}a_{\mathbf{g}\sigma}^{*}a_{\mathbf{g}\sigma'}$$

$$-\frac{1}{2}\sum J(\mathbf{h})(\mathbf{S}_{\mathbf{g}}\mathbf{S}_{\mathbf{g}+\mathbf{h}}). \qquad (1)$$

where $a_{\mathbf{g}\sigma}^{*}$ and $a_{\mathbf{g}\sigma}$ are operators of an *s*-electron stimulating the charge carrier (conduction electron or hole), $S_{\mathbf{g}}$ are the *d*-spin operators, $s_{\sigma\sigma'}$ is a Pauli matrix, and B is the Bloch integral. The lattice of magnetic atoms will be regarded as simple quadratic in the two-dimensional case ($D = 2$) and simple cubic in the three-dimensional case ($D = 3$), with a constant a. The vector $\mathbf{\Delta}$ joins nearest neighbors.

In the absence of an *s*-electron the energy of a magnetic structure with vector $\mathbf{Q}$ is given by

$$E_{\mathbf{Q}}^{M}=-J(\mathbf{Q})S^{2}/2, \qquad (2)$$

where exchange to neighbors of third order in distance inclusive is taken into account.[4)]

$$J(\mathbf{Q})=2J_1(\cos Q_1+\cos Q_2+\cos Q_3)$$
$$+4J_2(\cos Q_1\cos Q_2+\cos Q_2\cos Q_3$$
$$+\cos Q_3\cos Q_1)+8J_3\cos Q_1\cos Q_2\cos Q_3. \qquad (3)$$

For $D = 2$ it suffices to take into account exchange up to neighbors of second order in distance:

$$J(\mathbf{Q})=2J_1(\cos Q_1+\cos Q_2)+4J_2\cos Q_1\cos Q_2 \qquad (4)$$

($\mathbf{Q}$ is measured in reciprocal lattice constants).

According to (3) and (4), depending on the values of the exchange integral J_i between neighbors of *i*th order in distance, the following structures are possible in an insulating crystal in the three-dimensional case: FM (F) with $\mathbf{Q}_F = (0, 0, 0)$, Néel AFM structure (N) with $\mathbf{Q}_N = (\pi, \pi, \pi)$, and two layered AFM structures with $\mathbf{Q}_1 = (\pi, 0, 0)$ and $\mathbf{Q}_2 = (\pi, \pi, 0)$. The triple point for AFM structure is

$$J_1=2J_2=4J_3. \tag{5}$$

In the two-dimensional case it is possible to have besides the FM structure also a Néel structure with $\mathbf{Q}_N = (\pi, \pi)$ and a layered structure (L) with $\mathbf{Q}_L = (\pi, 0)$. The boundary of the AFM phases is the straight line

$$J_1=2J_2. \tag{6}$$

It will be assumed in the calculation of the s-electron energy that the magnetic ordering in the crystal is that of a skewed AFM structure. In the simplest variant the magnetic-atom spins are assumed to be too large to be treated as classical vectors. The energy of an electron with quasimomentum $\mathbf{k}$ is then given by an expression easily obtained from (1) by "entangling" the operators $a^*_{\mathbf{k}\sigma}$ and $a^*_{\mathbf{k}+\mathbf{Q},-\sigma}$:

$$E_\mathbf{k} = \frac{1}{2}\{E_\mathbf{k}{}^0+E^0_{\mathbf{k}+\mathbf{Q}} - [(E_\mathbf{k}{}^0-E^0_{\mathbf{k}+\mathbf{Q}}-AS\cos\varphi)^2+A^2S^2\sin^2\varphi]^{1/2}\}, \tag{7}$$

where the bare energy $E^0_\mathbf{k}$ is described in case (1) by a simple cosinusoidal dispersion law

$$E_\mathbf{k}{}^0=2B\sum_{i=1}^{D}\cos k_i, \tag{8}$$

and φ is the angle between the moment of the sublattice and the total moment of the crystal ($\varphi = \pi/2$ in the absence of skewing).

2. INFLUENCE OF SPIN QUANTUM FLUCTUATIONS ON CARRIER ENERGY

Equation (7) alone does not identify the spins that can be regarded as classical vectors and whose quantum fluctuations can be neglected. We also consider therefore separately the limiting case $AS \ll W$ and its converse, in each of which it is possible to take into account the influence of the fluctuations on the carrier ($W \equiv W_3 \simeq 12|B|$ is the width of the s-band in the three-dimensional case).

In the first of them, confining ourselves to the two-spin approximation, we obtain from (1), with allowance for the inequality $W \gg T_N$ discussed in the Introduction, from the known commutation relations for spin operators:

$$E_{\mathbf{k}\sigma}=E_\mathbf{k}{}^0-A\langle S^z\rangle\sigma + \frac{A^2}{4N^2}\sum_{\mathbf{g}\mathbf{f}\mathbf{q}\neq 0}\frac{[\langle \mathbf{S}_\mathbf{g}\mathbf{S}_\mathbf{f}\rangle-2\sigma\langle S^z\rangle\delta_{\mathbf{g}\mathbf{f}}]}{E_{\mathbf{k}\sigma}-E^0_{\mathbf{k}+\mathbf{q}}-i0}\exp[i\mathbf{q}(\mathbf{g}-\mathbf{f})], \tag{9}$$

where N is the number of magnetic atoms. For the spin mean values and the correlators in a skewed structure we have

$$\langle S_\mathbf{f}{}^z\rangle=S\cos\varphi, \tag{10a}$$

$$\langle \mathbf{S}_\mathbf{g}\mathbf{S}_\mathbf{f}\rangle=S^2\{\cos^2\varphi+\sin^2\varphi\exp[i\mathbf{Q}(\mathbf{g}-\mathbf{f})]\}+S\delta_{\mathbf{g}\mathbf{f}}. \tag{10b}$$

The first term in (10b) corresponds to treatment of the spins as quasiclassical vectors, and the second describes the quantum corrections to them.

In the three-dimensional case, putting $E_{\mathbf{k}\sigma} = E^0_\mathbf{k}$ in the second term of (9) and using (8), we obtain the following expression for the carrier energy near the bottom of the band:

$$E_{\mathbf{k}\sigma}=E_{\mathbf{k}\sigma}{}^c+E_{\mathbf{k}\sigma}{}^q, \tag{11}$$

$$E_{\mathbf{k}\sigma}{}^c=E_\mathbf{k}{}^0-AS\sigma\cos\varphi+A^2S^2\sin^2\varphi/4(E_\mathbf{k}{}^0-E^0_{\mathbf{k}+\mathbf{Q}}), \tag{12}$$

$$E_{\mathbf{k}\sigma}{}^q=A^2S(1-2\sigma\cos\varphi)G(E_\mathbf{k}{}^0)/4;\quad G(E_\mathbf{k}{}^0)\approx 3/W, \tag{13}$$

where

$$G(E)=\frac{1}{N}\sum_\mathbf{p}\frac{1}{E-E_\mathbf{p}{}^0-i0},$$

the term $E^c_{\mathbf{k}\sigma}$ corresponds to spins as classical vectors and is obtained from (7) by expansion in terms of AS/W. The second term $E^q_{\mathbf{k}\sigma}$, which gives the quantum correction, was calculated using Eq. (8). It is obviously of order $1/S$ compared with $E^c_{\mathbf{k}\sigma}$.

In fact, however, expression (7) is not always accurate to $1/S$. The described procedure leads in the two-dimensional case to a logarithmic divergence of G. An accurate calculation of G for $D = 2$ is in itself a rather serious problem. It need not be considered at all, however, if only the energy difference of unequal AFM phases is of interest, since $E^q_{\mathbf{k}\sigma}$ is independent of the type of the AFM structure and therefore does not enter into this difference. The quantum correction can be neglected also when the skews of the moments are small. On the other hand, however, the quantity G enters in the energy difference between an AFM phase and a spin fluid in which the electron energy is given, according to (9) and (10b), by the expression

$$E_\mathbf{k}=E_\mathbf{k}{}^0-A^2S(S+1)G(E_\mathbf{k})/4. \tag{14}$$

In the three-dimensional case it is possible, as before to replace $E_\mathbf{k}$ in the expression for G by $E^0_\mathbf{k}$. In the two-dimensional case, however, one obtains for G the estimate

$$G\approx(3/\pi W)\ln(3A^2S/4\pi W^2). \tag{15}$$

The value of G for small atom numbers N is proportional to $\ln N$. It follows from (11) and (15) that in the three-dimensional case the classical-spin picture (7) is valid if

$$\frac{1}{S}(2\ln|W/AS|+\ln S)\ll 1. \tag{16}$$

The validity of (7) is even more restricted in the limit of narrow bands, $W \ll AS$, regardless of the dimensionality of space. When the quantum character of the spins is taken into account, the result for small S depends substantially on the sign of A. In particular, a magnetic string (quasioscillator) should be realized for $S = 1/2$ and $A < 0$ (Ref. 4). We therefore consider below the case $A > 0$. At sufficiently large S, the results that follow can be easily generalized to include negative A.

According to (7), for chessboard ordering and as $A \to \infty$, the width of the s-electron energy band should vanish in the nearest-neighbor approximation (8) regardless of the value of S; this is most doubtful from the physical point of view. To obtain a correct result one must take into account the possibility of a d-spin direction change correlated with the s-electron spin. The d-spin direction change should be such as to restore the skewed AFM ordering in the crystal after the departure of the s-electron from the atom (otherwise a magnetic string would be produced in the crystal, which is incompatible with the assumption that A is positive).

Beside the inequality $W \ll AS$, account is taken in the calculation of the inequality $W \gg T_N$ mentioned in the Introduction. The zeroth-approximation Hamiltonian is therefore chosen to be the second term of (1). Its eigenfunctions corresponding to states with lowest s-d exchange energy, are characterized by the total spin $S + 1/2$ of a magnetic atom carrying an electron:

$$\begin{aligned}\psi_{\mathbf{g}}(M) = (2S+1)^{-1/2}\{&(S+M+{}^1\!/_2)^{1/2}\delta(S_{\mathbf{g}}^z, M-{}^1\!/_2)\,a_{\mathbf{g},1/2}^{*}\\ &+(S-M+{}^1\!/_2)^{1/2}\\ &\times\delta(S_{\mathbf{g}}^z, M+{}^1\!/_2)\,a_{\mathbf{g},-1/2}^{*}\}\,|0\rangle,\end{aligned} \qquad (17)$$

where $\delta(n,m)$ is a delta function with discrete argument; $M - z$ is the projection of the total spin; all the spin projections in (17) are taken in a local coordinate frame with z axis directed along the moment of the sublattice to which the atom belongs; $|0\rangle$ is the vacuum wave function for the s-electron

It will be assumed below that the s-electron does not perturb the ordering in the sublattice 1, but when it goes over to sublattice 2 it deflects the spin of its target atom from the normal direction of the latter. When the s-electron returns to sublattice 1, the direction of the deflected spin is restored. Neglecting the zero-point oscillations of the spins, the wave function of the system takes the form

$$\begin{aligned}\psi = X\sum_{\mathbf{g}}\prod_{i=1}^{D}&\sin[k_i a(g_i+1)]\,\psi_{\mathbf{g}}(S+{}^1\!/_2)\,\Phi(\mathbf{g})\\ +\sum_{\mathbf{h}}\prod_{i=1}^{D}&\sin[k_i a(h_i+1)]\\ \times[&Y\psi_{\mathbf{h}}(S+{}^1\!/_2)+Z\psi_{\mathbf{h}}(S-{}^1\!/_2)]\,\Phi(\mathbf{h}),\\ \Phi(\mathbf{j}) = &\prod_{\mathbf{f}\neq\mathbf{j}}\delta(S_{\mathbf{f}}^z, S),\end{aligned} \qquad (18)$$

where the subscripts **g** and **h** number the atoms of the first and second sublattice, respectively. The coefficients X, Y, and Z should be obtained from the minimum-energy condition.

Using (1), (17), and (18) we obtain the following expression for the s-electron energy in a Néel structure:

$$E_{\mathbf{k}} = 2B\gamma_{\mathbf{k}}\left(\cos^2\varphi + \frac{\sin^2\varphi}{2S+1}\right)^{1/2} - \frac{AS}{2},\qquad \gamma_{\mathbf{k}} = \frac{1}{D}\sum_{i=1}^{D}\cos k_i. \qquad (19)$$

The solution for layered ordering with $\mathbf{Q}_1 = (\pi, 0, 0)$ is in general too unwieldy. For $2S \gg 1$ and arbitrary φ it can be written in the form

$$E_{\mathbf{k}} = 2B\left[\Gamma_{\mathbf{k}} + \cos k_1\left(\frac{\sin^2\varphi}{2S+1} + \cos^2\varphi\right)^{1/2}\right] - \frac{AS}{2},\qquad \Gamma_{\mathbf{k}} = \sum_{i=2}^{D}\cos k_i. \qquad (20)$$

The corresponding equation for $\varphi = \pi/2$ and arbitrary spins is

$$E_{\mathbf{k}} = B\left[\frac{4S+1}{2S+1}\Gamma_{\mathbf{k}} + \frac{\Gamma_{\mathbf{k}}^2}{(2S+1)^2} + \frac{4\cos^2 k_1}{2S+1}\right]^{1/2} - \frac{AS}{2}. \qquad (21)$$

If $\varphi = \pi/2$ the result (19) coincides with that of Ref. 2. Obviously, Eqs. (19)–(21) yield a ground-state energy much lower than (7), thus proving that they are more accurate. As seen from (19), at angles φ close to $\pi/2$ the carrier energy bandwidth remains finite as $A \to \infty$. With increase of spin, however, it decreases like $(2S)^{-1/2}$. Thus, in the limit $W \ll AS$ Eq. (7) is valid only if $(2S)^{1/2} \gg 1$, i.e., for utterly fantastic spins exceeding 10. Equation (19), however, becomes equivalent to (7) if $\cot\varphi \gg (2S+1)^{-1/2}$.

3. CRITERIA FOR THE EXISTENCE OF AFMONS

The results of the preceding section yield estimates of the potential-well depth U for an s-electron, such as the region of a layered AFM phase inside a chessboard one. These estimates will be obtained for non-skewed moments ($\varphi = \pi/2$), and their skew increases even more the depth of the potential well. In the region where Eq. (7) or (11) is valid, assuming $W = AS = 3$ eV (this corresponds to an electron effective mass $m = (2|B|a^2)^{-1}$ close to the true one) we obtain in the three-dimensional case $U_1 = 0.4$ eV and $U_2 = 0.1$ eV respectively for layered phases 1 and 2 (see Sec. 1). In the two-dimensional case, at the same parameter values, the depth of the potential well corresponding to the layered phase L turns out to be 0.3 eV. In the opposite limit $W \ll AS$, the potential well depth turns out, according to (19) and (21), to be practically of the same order: for the AFM phase 1 it is close to $(D-1)W/6$, i.e., it can reach even 1 eV at the indicated values of the parameters. With decrease of spin, however, the potential-well depth decreases drastically in the case of narrow s-bands.

The presented values of U are comparable in practice with the potential-well depth U_F for the ferron. They are most likely to be reached in transition-metal compounds, while for rare-earth compounds the inequality $AS \ll W$ is usually so strong that the potential well is only hundredths of an eV deep. When it comes to HTSC, at the present level of our knowledge nothing definite can be said concerning the possible presence of afmons in them. On the one hand, although HTSC are frequently described by the Hubbard model, it is by far not evident that the appearance of a hole on a Cu^{2+} will cause the spin of the latter to vanish, i.e., that a Hubbard model which excludes afmons is adequate for such HTSC.[5)] On the other hand, however, the parameters of the hole are unknown, and it not clear with which virtual AFM structures the actually realized ones are to be compared.

Disregarding the skew of the moments, the afmon energy is calculated using exactly the same variational scheme as for ferrons.[2,5] It is assumed a layered-phase region inside a chessboard phase has a spherical ($D = 3$) or circular ($D = 2$) form with radius R. The total energy of the system consists of the energy of the s-electron inside a potential well of depth U and the d-d exchange energy spent to produce the layered-phase region. The first of them decreases and the second increases with increase of R, so that R is regarded as a variational parameter obtainable from the condition that the total system energy be a minimum. This, together with the requirement that the energy of the self-trapped state of the carrier be lower than the energy of the free carrier in an ideally ordered AFM crystal, leads to the following criterion for existence of a self-trapped state:[2,11]

$$K/W \leqslant \tilde{K}/W = 0{,}2(4-D)(U/W)^{1+D/2}, \tag{22}$$

where K is the energy per atom needed to produce the new phase, and $\tilde{K}$ is its critical value.

As indicated above, at $D = 3$ the most favorable for the onset of an afmon is phase with $\mathbf{Q}_1 = (\pi, 0, 0)$, if the energy needed to produce it is not high, e.g., near a triple point.[5] The sufficient condition for the existence of an afmon is that the inequality (22) hold for the afmon but not for a ferron. If $W \gg AS$, the potential-well depth U_F for a ferron, to first-order in AS/W, is $AS/2$. Taking this and (11) into account we obtain the following expression for the ratio of the critical energies $\tilde{K}_F$ and $\tilde{K}_A$ for ferron and afmon production is

$$r = \tilde{K}_A/\tilde{K}_F = [(D+1)AS/4W]^{1+D/2}.$$

If $AS/W = 2/3$, this ratio exceeds 0.25 for $D = 2$ and 0.35 for $D = 3$. Similar estimates are obtained for large spins and when (19) and (21) are employed. Using the known expression $J(\mathbf{Q})S(S+1)/3$, for the Néel point T_N we find from (3) and (5) that at the triple point

$$K_F = E^M(\mathbf{Q}_F) - E^M(\mathbf{Q}_N) = 12(1+1/S)^{-1}T_N.$$

If, however, $J_3 = 0$, we have $J_1 = 4J_2$ a value of K_F as large on the boundary between phases N and 2. Thus, an afmon can exist at the values indicated for the parameters or close to them even if $K_A = E^M(\mathbf{Q}_1) - E^M(\mathbf{Q}_N)$ amounts to several times T_N, i.e., the antiferromagnet parameters are far from the Néel-phase boundary.

It is interesting to note that at $D = 3$, according to (13) and (14), the paramagnetic region inside the Néel phase is for the s-electron a potential well of practically the same depth as the region of the layered phase 1. One can expect that the carriers are subject to dispersion laws, other than (8), for which the carrier energy in a spin fluid is even lower than in a layered AFM phase. On the other hand, there is also an exchange-integral region in which the energy K_{sl} needed to disorder the Néel phase is lower than that needed to produce phase 1. For example, if $J_3 = 0$ on the boundary of phases N and 2, the first of these quantities is equal to $3|J_1|S^2/2$ and the second is 4/3 times larger. This raises the question whether there exists a parameter region in which self-trapping at $T = 0$ occurs not in a microregion of an altered AFM phase, but in a region with loss or order (spin fluid).

Self-trapping of a carrier in a spin fluid, however, requires that it be energywise favored also over self-trapping in a FM region. In the case considered here $K_F/K_{sl} = 4$, and according to (22) self-trapping is possible in the spin-fluid region but forbidden in the FM region if $AS/W > 0.7$. Since, however, the equations used above were derived for the case $AS/W \ll 1$, the inequality obtained above, being marginal, does not guarantee feasibility of self-trapping in a spin fluid. Possibly the conditions of cooperative self-trapping are more favorable for a spin fluid than for a ferron, for owing to s-electron polarization in spin the electron energy in the FM region increases with carrier density more rapidly than in a spin fluid (see Sec. 5). On the other hand, the structure of expressions (14) and (15) points to a possibility that the self-trapping conditions will be more favorable in the two-dimensional than in the three-dimensional case.

4. LARGE-RADIUS AFMONS

Explicit expressions for the afmon parameters can be obtained if the carrier self-trapping region is large. The calculation that follows corresponds to the limit $W \gg AS$ and is based on expressions (11) and (12), although it can equally well be carried out in the limit $W \ll AS$ using (19) and (20). It is more convenient here to assume that the region of layered phase I inside a Néel AFM phase has the form of a cube ($D = 3$) or square ($D = 2$) with side $L \gg a$. The quasiparticle energy is then obtained by minimizing with respect to L and φ the expression ($\hbar = 1$)

$$E_A = \frac{D\pi^2}{2mL^2} - \frac{AS\cos\varphi}{2} - \frac{3A^2S^2}{4W}\left(\sin^2\varphi - \frac{1}{D}\right) + \left(\frac{L}{a}\right)^D K_1(\varphi), \tag{23}$$

where K_1 is written for $D = 3$ and $D = 2$ with allowance for the third-next and second-next nearest neighbor, respectively [see (3) and (4)]:

$$K_1 = \{-(2D-1)J_1 + 2J_2(D-1) - [J_1 + 2(D-1)J_2]\cos 2\varphi - 4(D-2)J_3(1+\cos 2\varphi)\}S^2. \tag{24}$$

According to the condition that the Néel ordering be stable in the absence of carriers, it is assumed that the inequality $J_1 < 2J_2 < 0$ is satisfied, as well as $J_1 > 4J_3$ in the three-dimensional case.

Putting $l = (J_2 - J_1)S^2$ and confining ourselves to small values of l, we obtain from (24) in the two-dimensional case

$$\cos\varphi = C(1-4I_1C^2)^{-1/2}(2l)^{1/2}, \tag{25}$$

where

$$C = AS(ma^2)^{1/2}/16\pi I_1, \quad I_1 = |J_1|S^2. \tag{26}$$

The optimal dimension L and the afmon energy E_A are given by

$$L = L_0(1-4I_1C^2)^{1/4}, \quad L_0 = (\pi a^2/2lm)^{1/4}. \tag{27}$$

$$E_A = -3A^2S^2/8W + E_0(1-4C^2I_1), \tag{28}$$

where

$$E_0 = (8\pi^2 l/ma^2)^{1/2},$$

L_0 and E_0 are the corresponding quantities without allowance for the skew.

As $l \to 0$ in the three-dimensional case, we have for the corresponding quantities: the skew angle

$$\cos\varphi=|AS/4I|(4ma^2/\pi^2)^{3/5}l^{3/5}, \qquad (29)$$

$$I=(6J_1+8J_3)S^2,$$

the dimension of the region

$$L=(\pi^2a^3/4ml)^{1/5} \qquad (30)$$

and the afmon energy

$$E_A=-A^2S^2/2W+\frac{5\cdot2^{-1/5}\pi^{6/5}}{(ma^2)^{3/5}}l^{2/5}. \qquad (31)$$

It follows from (25)–(27), (29), and (30) that as $l\rightarrow0$, i.e., as the phase boundary is approached, the size of the region tends to infinity, and the skew angle tends to zero. This result is physically clear: as $L\rightarrow\infty$ the probability of an electron staying on a magnetic atom, meaning also the molecular field applied to the electron by the atom, tends to zero. The total moment of the afmon, however, remains finite a $l\rightarrow0$ in both the two- and three-dimensional cases.

Next, the influence of the skew on the energy and size of the afmon as $l\rightarrow0$ is much stronger in the two-dimensional than in the three-dimensional case: at $D=3$ the skew contribution to E_A and L is of the order of $l^{1/5}$ and is therefore disregarded in Eqs. (30) and (31). In the two-dimensional case, with decrease of $|J_1|$, meaning T_N, the afmon energy for a given l increases, and its size decreases. It is interesting to note that when the condition (22) for the existence of a ferron is satisfied, expressions (25)–(28) become meaningless, i.e., the afmon and ferron are mutually exclusive states of a carrier (strictly speaking, the critical value obtained for I_1 from (25)–(28) differs from $\widetilde{K}_F$ in (22) by a factor ~1. This is quite natural, bearing in mind that the calculation is approximate and that furthermore the geometries of the self-trapping regions are somewhat different).

It is interesting to note that the dependence of the afmon dimension (27), (30) on the energy needed to produce the new phase and on the width of the carrier energy band turns out here to be exactly the same as for the ferron.[5,11] Just as in the case of ferrons,[2] the polarization of a lattice by the afmons lowers their energy additionally.

5. AFMON PAIRING AND COOPERATIVE AFMON EFFECTS

If the carrier density in the crystal is high enough, afmons can become paired to form charged particles with zero spin. The reason is that placing a second carrier in an AFM microscopic region of containing the first requires less energy than the production of a second region for the second carrier. Then, however, the Coulomb energy of the system is increased, and furthermore if the spins of both s-electrons are parallel the skew of the sublattice moments and the ensuing energy gain vanish. If, however, the electrons are placed in states with antiparallel spins, the second electron should be in a state with higher orbital energy than the first, so that pairing increases the kinetic energy of the trapped electrons. The number of carriers in the potential well should therefore be obtained from the condition that the system total energy be a minimum, and biafmon can be stable only in a definite interval of the system parameters and at definite doping levels. Owing to the dependence on the carrier density, afmon pairing in degenerate semiconductors is a cooperative effect.

We shall consider below only the three-dimensional case. The first two orbital states of a free electron in a spherical potential well of radius R with Dirichlet boundary conditions on its surface are[12]

$$\psi_1=C_1\sin(k_1r)/r,\quad C_1^2=(2\pi R)^{-3},\quad k_1=\pi/R \qquad (32)$$

and

$$\psi_2=C_2[\sin(k_2r)/k_2^2r^2-\cos(k_2r)/k_2r]Y_{1m},$$

$$C_2^2=k_2^3[\cos(2k_2R)/2k_2R$$

$$+\sin(2k_2R)/4+{}^1/_2(k_2R-1/k_2R)]^{-1}\approx40/R^3, \qquad (33)$$

where Y_{lm} is a spherical harmonic and $k_2=4.4934/R$.

It is easy to verify that as $l\rightarrow0$ both s electrons in a biafmon should be in the state ψ_1 with antiparallel spins. The state ψ_2 will be needed below only to investigate the stability of the biafmon to coalescence with a third afmon. The preceding statement concerning the electron configuration of the biafmon is justified by the following considerations. It follows from (29) that the skew of the sublattice moments lowers the afmon energy by an amount $l^{3/5}$. Recognizing that from (30) we have $R\propto l^{-1/5}$, the orbital energy E_2 exceeds E_1 by an amount $\propto l^{2/5}$.

If a degenerate semiconductor is considered, the system energy with all the carriers paired should be compared with the case when they are not paired. It should be recognized here that a carrier undergoes Coulomb interaction not only with the carrier with which it shared a potential well, but also with the remaining carriers and ionized defects. Coulomb interaction in such a system can be approximately taken into account by the method of Wigner cells, when the crystal is subdivided into spheres centered about biafmons or monoafmons, while the remainder of the sphere is occupied by the compensating charge. The radius R_W of the Wigner sphere is obtained from the condition that it be electrically neutral:

$${}^4/_3\pi nR_W^3=z, \qquad (34)$$

where n is the density of carriers with oppositely charged defects and z is the quasiparticle charge. Taking (34) into account under the condition $R_W\gg R$, the energy of interaction between the quasiparticle and the compensating charge is given by the expression

$$Q_z=-2\pi e^2z^{5/3}n^{1/3}/\varepsilon, \qquad (35)$$

where ε is the dielectric constant of the crystal.

In accordance with the foregoing, the biafmon energy is

$$E_B=\frac{\alpha}{R^2}+\frac{\beta}{R}+\gamma R^3-2U_1+Q_2, \qquad (36)$$

where

$$\alpha=\pi^2/m,\ \beta=3e^2/2\varepsilon,\quad \gamma=4\pi K_1/3a^3,\ U_1=A^2S^2/2W.$$

The constant β is obtained by calculating the Coulomb integral

$$q_{ik}=\int d^3r_1\,d^3r_2|\psi_i(\mathbf{r}_1)|^2|\psi_k(\mathbf{r}_2)|^2|\mathbf{r}_1-\mathbf{r}_2|^{-1} \qquad (37)$$

and using (32) as well as the expansion[13]

$$\frac{1}{|\mathbf{r}_1-\mathbf{r}_2|}=\sum_{n=0}^{\infty}\left\{\frac{r_1{}^n}{r_2{}^{n+1}}\theta(r_2-r_1)+\frac{r_2{}^n}{r_1{}^{n+1}}\theta(r_1-r_2)\right\}\frac{4\pi}{2n+1}$$

$$\times\sum_m Y_{nm}{}^*(\theta_1,\varphi_1)\,Y_{nm}(\theta_2,r_2), \qquad (38)$$

where $\theta(x)$ is the Heaviside unit step function. According to (37) and (38), $q_{11}=3/2R$.

It is easy to verify that the biafmon is not energywise favored at extremely small values of l. In fact, if the first term in the right-hand side of (36) is neglected, the biafmon energy E_B+2U_1 reckoned from the bottom of the potential well is of order $l^{1/4}$ whereas the corresponding energy for the mono-afmon is of the order of $l^{2/5}$. At somewhat higher value of l one can used perturbation theory in terms of the additional small parameter $e^2/\varepsilon aW$. The energy difference between two monoafmons and a biafmon is then

$$\delta_2=2E_A-E_B=(2-2^{3/5})\tilde{E}_A-\beta/R_2+\frac{2\pi e^2n^{1/3}}{\varepsilon}\left(\frac{3}{4\pi}\right)^{2/3}(2^{5/3}-2), \qquad (39)$$

where

$$\tilde{E}_A={}^5/_6(9\alpha^3\gamma^2)^{1/5},\ R_2=2^{1/5}R_1,\ R_1=(\alpha/3\gamma)^{1/5}.$$

It follows from (39) that if the carrier density is low enough to be able to neglect the last term of the last expression, a biafmon can be realized only if the energy K_1 needed to form a new phase satisfies the inequality

$$K_1/W>(e^2/\varepsilon aW)^5. \qquad (40)$$

At $\varepsilon=10$, $a=3$ Å, $W=3$ eV, the critical value K_1^c starting with which the inequality (40) is valid for K_1 is $4\cdot10^{-4}$ eV, while at $W=2$ eV we have $K_1^c=2\cdot10^{-3}$ eV.

It is necessary, however, to verify that the triafmon state does not turn out to be energywise even more favored than the biafmon state. In the calculation of the triafmon energy E_T it is assumed that two electrons are in a state ψ_1 and the third in a state ψ_2 [Eqs. (32) and (33)]. The Coulomb energy is calculated with the aid of (37) and (38). The resultant expression is unwieldy, and we cite only the numerical value $q_{12}\approx1/R$. We have then

$$\delta_3=E_A+E_B-E_T=(2^{3/5}+1-4^{3/5})\tilde{E}_A-\frac{e^2}{\varepsilon R_2}[2^{4/5}+{}^3/_2(2^{-1/5}-1)]$$

$$-\frac{2\pi e^2n^{1/3}}{\varepsilon}\left(\frac{3}{4\pi}\right)^{2/3}(2^{5/3}+1-3^{5/3}). \qquad (41)$$

It follows from (41) that if n is small triafmons are impossible at all values of the parameters. They do become possible, however, for larger n. There exists thus a density interval in which only carrier pairing is favored. If the densities are higher, multi-afmon complexes should be formed and lead to phase separation in the crystal, into an old and new AMF phase with all the carriers concentrated in the new one.

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1) This must not be taken to mean that carrier self-trapping is the only possible explanation of HTSC. Other mechanisms are also possible (see, e.g., the survey in Ref. 14).

2) The term "afmon" stems from an abbreviation for antiferromagnet. The more convenient and better sounding "antiferron" cannot be used for the quasiparticle considered here, since it was previously introduced to denote quasiparticles of an entirely different type.[2]

3) The possible skewing of the moments of the sublattices of a collinear antiferromagnet by the action of carriers was proved and investigated in detail in Ref. 2. It is shown there, in particular, that for small skews the magnon spectrum of such a system is positive-definite.

4) Several exchange integrals must be taken into account because the afmon is energywise most favored near a boundary between different AFM phases. A phase boundary, however, exists only if several exchange integrals differ from zero, since the competition between them determines the stability of one phase or another [see Eqs. (5) and (6)].

5) The appearance of a hole on the Cu^{2+} ion means a transition into the Cu^{3+} state. An isolated Cu^{3+} ion certainly satisfies Hund's rule, and its spin should therefore be not zero but unity. The levels of the partially filled *d*-shell are split in the crystal, so that Hund's rule may be violated. The Cu^{3+} spin can then indeed be zero. It would hardly be possible, however, to estimate by using purely theoretical calculations whether the crystal field suffices for this purpose.

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