Separation of antiferromagnetic and spin-liquid phases in degenerate antiferromagnetic semiconductors

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A degenerate antiferromagnetic semiconductor can separate into different phases in a manner different from that in the pioneering studies of this topic [É. L. Nagaev, JETP Lett. **16**, 394 (1972); V. A. Kashin and É. L. Nagaev, Sov. Phys. JETP **39**, 1036 (1974)]. The crystal can separate into alternating regions of a normally stable phase and a normally unstable one, both antiferromagnetic. All the charge carriers would be in the unstable phase. In the two-phase state, the crystal can be either an insulator or a good conductor, depending on whether the highly conducting phase is multiply or singly connected. A spin liquid can serve as an alternative highly conducting phase. There is a discussion of some specific phase transitions which occur in two-phase systems of this sort: the disappearance of order in the highly conducting phase, a temperature-induced magnetic-electronic percolation, and a melting of the two-phase state.

The problem of phase separation in degenerate semiconductors is by no means new: It was first formulated and studied back in the early 1970s in papers[1,2] by one of the present authors. The ground state of a homogeneous, degenerate antiferromagnetic semiconductor was studied in those papers. It was shown that with suitable parameter values the semiconductor should break up into alternating regions of a ferromagnetic phase and an antiferromagnetic one, with all the conduction electrons in the former. Ferromagnetic regions of this sort are essentially complexes of ferrons, which are quasiparticles typical of antiferromagnetic semiconductors: A conduction electron (or hole) gives rise to a microscopic region in the ferromagnetic crystal and stabilizes this region by localizing in it.[3] The authors of the present paper later studied the separation of antiferromagnetic and ferromagnetic phases at nonzero temperatures, in particular, the melting of a collective ferron state of this sort accompanied by a transition of the crystal to a single-phase magnetic state.[4,5]

The separation of ferromagnetic and antiferromagnetic phases predicted in Refs. 1 and 2 has been observed experimentally in the degenerate antiferromagnetic semiconductors EuTe and EuSe. It also occurs in several other materials.[6,7] Among the recent experimental studies in this field we should mention that by Osipov and Kochev,[8] who identified specific features of collective charge transport in very strong electric fields in highly degenerate EuTe below the transition to a two-phase ferromagnetic–antiferromagnetic state. A detailed analysis of that experiment was carried out in Ref. 9. It was soon recognized that the phase separation should be universal, not something peculiar to antiferromagnetic semiconductors.

Just recently, many investigators have taken up the problem of phase separation in degenerate antiferromagnetic semiconductors in connection with the problem of superconductivity (e.g., Refs. 10–13). These investigators have apparently been unaware of the earlier work on phase separation. For example, the Coulomb energy of highly charged regions of different phases and also their surface energy were totally ignored in Refs. 10–13. As was shown in Refs. 1 and 2, these energies are of fundamental importance for demonstrating that a phase separation is possible and also for determining the geometry of the two-phase state. For this reason, the results of such studies on phase separation cannot be regarded as having a solid basis. This is true in particular of Ref. 10, where, within the framework of the Hubbard model, the holes were assumed to be concentrated in regions which became nonmagnetic as a result.

In the present paper we discuss a different type of separation of a degenerate antiferromagnetic semiconductor into phases: a separation which is completely different in nature from that in the earlier papers on this topic. Specifically, we assume that there exist alternative antiferromagnetic phases in which the energy of the charge carriers is lower than in the antiferromagnetic phase which actually exists in their absence. (For definiteness, we will conduct the discussion in terms of conduction electrons, although everything below also applies to holes.) The crystal can then break up into alternating regions of a normally stable antiferromagnetic phase and a normally unstable one, with all the conduction electrons in the latter.

The regions of unstable phase are actually multielectron afmons. The concept of an "afmon" was recently introduced in Refs. 14–16: An afmon is a quasiparticle which arises in an antiferromagnetic semiconductor upon the self-localization of an electron in a region of an altered antiferromagnetic phase. A condition for the existence of such quasiparticles is that the Néel point be sufficiently high; this condition rules out the possible existence of ferrons in the same material. The afmon's "memory" of a ferron is a slight skew of the sublattice moments of the antiferromagnet in the region in which a conduction electron localizes. Afmons are most favored from the energy standpoint near

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the boundary between a normally stable antiferromagnetic phase and an antiferromagnetic phase which arises upon self-localization. However, afmons can also exist at a fair distance from this boundary at realistic parameter values. For example, the role of an antiferromagnetic structure which is stable in the absence of charge carriers might be played by a checkerboard Néel structure, while the role of a structure in which the carrier self-localizes might be played by a layered Landau structure.

In addition to the division of the crystal into two antiferromagnetic phases, we will discuss the possibility that the crystal breaks up into regions of a normally stable antiferromagnetic phase and a spin-liquid phase, with the latter having all the conduction electrons. By "spin liquid" we mean a system in which there is no magnetic order because of a competition between different exchange mechanisms. If a spin liquid exists at $T=0$, it should be regarded as a quantum system. However, it may also arise at a nonzero temperature, as the result of a disruption of magnetic order.

The possibility that a crystal will break up into antiferromagnetic and spin-liquid phases ultimately stems from the circumstance that the energy of an electron in a spin-liquid state is close to the energy of an electron in a Landau structure (at least in the nearest-neighbor approximation). The expenditure of magnetic energy on the conversion of a Néel order into a Landau order may be greater than the expenditure of energy on disordering through a disruption of a Néel structure. Accordingly, even at $T=0$, not to mention nonzero temperatures, the energy conditions or thermodynamic conditions will favor a breakup of the system into an antiferromagnetic phase and a spin-liquid phase, rather than a breakup into two antiferromagnetic phases. If the relations among the parameters are instead of such a nature that a two-phase antiferromagnetic state exists at $T=0$, then the Landau phase will melt when the temperature is raised, converting into a spin liquid which coexists with the Néel antiferromagnetic phase and which is electrically charged with respect to the latter.

The procedure developed in Refs. 1–5 for studying the geometry of the two-phase state and the melting of this state at nonzero temperatures, at which the antiferromagnetic phase coexists with a ferromagnetic phase, is generalized below to the case in which an antiferromagnetic phase coexists with another antiferromagnetic phase or with a spin liquid. The basic results of Refs. 1–4 remain valid for the case discussed here. In the two-phase state which we are discussing here, just as in the case of a collective ferron state, the part of the crystal occupied by electrons can have topologies of two types: (a) a multiply connected topology with highly conducting spheres with the Landau structure (or spin-liquid spheres) in an insulating matrix with a Néel structure (Fig. 1); and (b) a singly connected topology, with insulating spheres with the Néel structure in a highly conducting matrix with an Landau structure (Fig. 2).

If the topology of the first type is realized, then the system must be an insulator (a two-phase insulating state) since the conduction electrons are trapped inside each of their spheres. In the case of a topology of the second type, the crystal is in a two-phase conducting state. A change in topology may occur upon a change in the density of conduction electrons in the crystal. An increase in density converts the highly conducting phase from a multiply connected to a singly connected topology. This conversion corresponds to a percolation of conduction electrons and a layered antiferromagnetic order. An insulator–metal transition thus occurs.

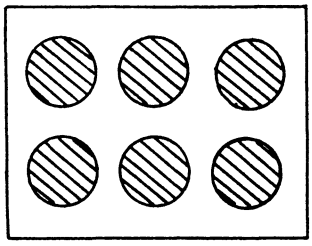

FIG. 1. Two-phase insulating state of a degenerate semiconductor. The highly conducting part of the crystal (a Landau antiferromagnetic phase or a spin liquid) is represented by the hatching; the insulating part of the crystal (a Néel antiferromagnetic phase) is represented by the unhatched region.

Percolation can also occur upon a change in temperature, because of an increase in the volume of the highly conducting phase. At high temperatures, the two-phase state melts, converting into a single-phase state.

Recent experiments[17] point to a separation of phases in certain high T_c superconductors; specifically, this is a separation into a spin-liquid conducting phase and an antiferromagnetic insulating phase. A separation of phases in a high T_c superconductor was also observed in Ref. 18. The experimental data of that study are evidence of a percolation in the superconducting phase.

METHOD FOR CALCULATIONS ON A TWO-PHASE ANTIFERROMAGNETIC STATE AT $T=0$

The calculation is carried out in the s–f model (or, equivalently, the s–d model) with the Hamiltonian

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

$$-\tfrac{1}{2}\sum J(\mathbf{f})\mathbf{S}_{\mathbf{g}}\mathbf{S}_{\mathbf{g}+\mathbf{f}}, \qquad (1)$$

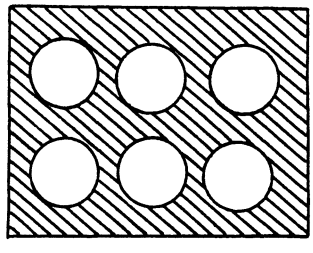

FIG. 2. Highly conducting two-phase state of a degenerate semiconductor. The hatched and unhatched regions have the same meaning as in Fig. 1.

where the operators $a^*_{\mathbf{g}\sigma}$ and $a_{\mathbf{g}\sigma}$ create and annihilate an s-wave electron with a spin projection σ at the magnetic atom of index $\mathbf{g}$, $\mathbf{S}_\mathbf{g}$ is the spin of the f-wave electrons of this atom, $\mathbf{s}_{\sigma\sigma'}$ are the Pauli matrices, and the vector $\mathbf{\Delta}$ numbers the nearest neighbors. In contrast with the kinetic-energy operator of the s-wave electrons [the first term in (1)], the Hamiltonian of the "direct" f–f exchange [the last term in (1)] incorporates the interaction between neighbors which are more distant than nearest neighbors. This point must be taken into account if we wish to study the vicinity of the boundary between antiferromagnetic phases: Layered antiferromagnetic structures can be stable only if neighbors out to at least the second-nearest are taken into account.

Like the problem[1–5] of the stratification of a crystal into antiferromagnetic and ferromagnetic phases, the problem of the stratification of a crystal into two distinct antiferromagnetic phases is solved by a variational method. In this case, however, the problem is far more complicated: Instead of two variational parameters it is necessary to introduce four.

It is assumed that one phase corresponds to a checkerboard antiferromagnetic order and does not contain conduction electrons, while the other corresponds to a layered antiferromagnetic order and contains all the conduction electrons. The first variational parameter is the ratio of the volumes of the first and second phases, X. Neglecting fluctuations of the electrostatic potential of an impurity, we also assume that the phase with the smaller volume forms a periodic structure of spherical inclusions of radius R inside the predominant phase. The quantity R is the second variational parameter of the problem.

The s–f exchange may distort the magnetic structure of the phase containing the electrons in such a way that the angle (2φ) between the spin directions in neighboring ferromagnetic layers becomes nonzero, and a magnetic moment arises in the corresponding part of the crystal. The skew angle of the moments of the sublattices, φ, serves as the third variational parameter.

For a crystal with a simple cubic lattice and a lattice constant a, as discussed below, the spin configuration of the magnetic structure with a vector $\mathbf{q}$ is specified by the following expressions:

$$S^x_\mathbf{q}=S\sin\varphi\cos(\mathbf{qg}),$$

$$S^y_\mathbf{q}=S\sin\varphi\sin(\mathbf{qg}), \tag{2}$$

$$S^z_\mathbf{q}=S\cos\varphi.$$

For a checkerboard order we would have $\mathbf{q}=\mathbf{q}_N=(\pi/a,\pi/a,\pi/a)$, for a chain order we would have $\mathbf{q}_C=(\pi/a,\pi/a,0)$, and for a layered order we would have $\mathbf{q}_L=(\pi/a,0,0)$.

The electron spectrum in a crystal with structure (2) consists of two subbands, whose bottoms have the energies

$$\tilde{E}_{bN}(\sigma)=-\tfrac{1}{2}\sqrt{(W+2\sigma AS\cos\varphi)^2+A^2S^2\sin^2\varphi},\quad \mathbf{q}=\mathbf{q}_N, \tag{3}$$

$$\tilde{E}_{bL}(\sigma)=-\frac{W}{3}-\frac{1}{2}\sqrt{\left(\frac{W}{3}+2\sigma AS\cos\varphi\right)^2+A^2S^2\sin^2\varphi},$$

$$\mathbf{q}=\mathbf{q}_L, \tag{4}$$

where W is the width of the seed conduction band. We will not discuss a chain structure, since according to Ref. 16 the energy of an electron in it is far higher than in a layered structure.

Expressions (3) and (4) follow from the general formula for the lower part of the electron spectrum in magnetic structure (2):

$$\tilde{E}_\mathbf{k}(\sigma)=\tfrac{1}{2}[E_\mathbf{k}+E_{\mathbf{k}+\mathbf{q}} - \sqrt{(E_\mathbf{k}-E_{\mathbf{k}+\mathbf{q}}+2\sigma AS\cos\varphi)^2+A^2S^2\sin^2\varphi}] \tag{5}$$

with

$$E_\mathbf{k}=2B[\cos(k_xa)+\cos(k_ya)+\cos(k_za)] \tag{6}$$

and $W=12|B|$. This formula is found through an exact diagonalization of Hamiltonian (1), if we assume that the spins $\mathbf{S}_\mathbf{g}$ in it are classical vectors described by Eqs. (2). Equation (5) holds under the condition $2S\gg1$ if $W\gg AS$, or $(2S)^{1/2}\gg1$ if $W\ll AS$ (Ref. 16). Under the condition $W\gg AS$ for the problem at hand, Eq. (5) actually also applies at small values of the spins of the f electrons, S, since the correction to it for quantum fluctuations of the spins is the same for all nonmagnetic structures.[16] This formula thus does not affect the energy benefit resulting from a phase separation of the type under consideration here (provided, of course, that there is no skew or only a slight skew of the sublattice moments).

It follows from (3) and (4) that the minimum electron energy in the case of a layered ordering is indeed lower than the corresponding energy in a checkerboard ordering. The difference can reach several tens of electron volts for typical values of the crystal parameters. This is why a two-phase antiferromagnetic state can be realized. Such a state could hardly arise in compounds of rare earth elements, in which the strong inequality $W\gg AS$ holds. It would be more likely in compounds of transition elements with $W\sim AS$.

The discussion below will be restricted to the case $W>AS$, and in general we will not assume that this inequality is very strong. Since the electron spectrum consists of many spin subbands, it is convenient to introduce a fourth variational parameter, y, which determines the distribution of electrons between the two subbands:

$$n(\sigma)=\frac{n}{2}(1+2\sigma Y), \tag{7}$$

where n is the total density of conduction electrons, and $n(\sigma)$ are the densities of electrons in the corresponding subbands. In principle, the parameter Y could be expressed in terms of φ by working from the condition that the Fermi energies in the two subbands are equal. However, the ex-

pression for the Fermi level in a sample of finite dimensions is complicated by the existence of a surface correction to its bulk part.[19] It is thus difficult to express Y in terms of φ analytically.

Correspondingly, the total energy of the electrons must also include a bulk part E_V and a surface part E_S (Ref. 19). This circumstance is of fundamental importance; the theory of Refs. 1–4 is constructed on the basis of this circumstance. In the case at hand, the expressions for E_V and E_S found from (3)–(7) by the methods of Ref. 19 are not of the same form as in Refs. 1–4:

$$E_V=-\frac{Wn}{3}-\frac{n}{4}(1+Y)\sqrt{\left(\frac{W}{3}+AS\cos\varphi\right)^2+A^2S^2\sin^2\varphi}$$
$$-\frac{n}{4}(1-Y)\sqrt{\left(\frac{W}{3}-AS\cos\varphi\right)^2+A^2S^2\sin^2\varphi}$$
$$+0.3n\mu(1+X)^{2/3}[(1+Y)^{5/3}+(1-Y)^{5/3}], \qquad (8)$$

$$E_S=\frac{3}{32}\left(\frac{\pi}{3}\right)^{1/3}\xi\mu n^{2/3}(1+X)^{1/3}$$
$$\times[(1+Y)^{4/3}+(1-Y)^{4/3}]\frac{1}{R}, \qquad (9)$$

where $\xi=3$ for the case of spheres filled by electrons, $\xi=3X$ for the case of insulating spheres, $\mu=(3\pi^2n)^{2/3}/2m^*$, and $1/m^*=Wa^2/6$.

The first term in (8) means the minimum energy of the conduction electrons corresponding to a motion of these electrons along the ferromagnetic layers. The second and third terms represent the minimum energy of the conduction electrons corresponding to their motion across the ferromagnetic layers, for the lower and upper spin subbands, respectively. The fourth term is the kinetic energy of the gas of degenerate electrons, reckoned from the bottom of the conduction band in the case $AS=0$ (μ is the Fermi energy in the case $AS=0$). In other words, this term is written in the zeroth approximation in AS/W, in contrast with the preceding terms. This approach is justified by the presence of the small parameter $\nu^{2/3}$ in this term; this parameter is not present in the preceding terms (the number of conduction electrons per atom, ν, is $\sim 10^{-4}$–10^{-2} in a degenerate semiconductor with an effective electron mass m^* on the order of the actual mass).

There is an especially noteworthy fact here: This small value of the kinetic energy, in accordance with ν, has the consequence that the lower position of the bottom of the conduction band in the case of a layered order than in the case of a checkerboard order guarantees that the electron energy for a layered order will be lower than that for a checkerboard order.

The term E_S, which gives the surface energy of the electrons, arises from a summation over the two spin subbands. It results from an expansion of the electron energy in $1/k_FR$ as a first-order correction to the bulk energy, (8) (k_F is the Fermi momentum).

The existence of negatively charged, highly conducting regions and positively charged insulating regions leads to a substantial increase in the energy of the two-phase state. To estimate the Coulomb energy we can use the method of Wigner cells as in Refs. 1–4. As a result we find

$$E_C=\frac{2\pi}{5\varepsilon}n^2e^2R^2f(X), \qquad (10)$$

where

$$f(X)=2X+3-3(1+X)^{2/3}$$

for spheres occupied by electrons (Fig. 1),

$$f(X)=X[3X+2-3X^{1/3}(1+X)^{2/3}]$$

for empty spheres (Fig. 2), and ε is the dielectric permittivity of the crystal.

We now consider the magnetic energy E_M of a two-phase antiferromagnetic state, i.e., the energy of direct interatomic exchange. It follows from (1) and (2) that the energy of the skewed magnetic structure with a vector $\mathbf{q}$ is given by the following expression, in which neighbors out to third-nearest are taken into account:

$$E_M(\mathbf{q})=-\frac{S^2N}{2}[J_\mathbf{q}+(J_0-J_\mathbf{q})\cos^2\varphi],$$

$$J_\mathbf{q}=2J_1[\cos(q_xa)+\cos(q_ya)+\cos(q_za)]$$
$$+4J_2[\cos(q_xa)\cos(q_ya)+\cos(q_ya)\cos(q_za)$$
$$+\cos(q_za)\cos(q_xa)]$$
$$+8J_3\cos(q_xa)\cos(q_ya)\cos(q_za). \qquad (11)$$

The corresponding exchange integrals are assumed to be negative ($J_1,J_2,J_3<0$). It is also assumed that the inequality $|J_3|>|J_1|/4$ holds. This inequality means that a layered Landau antiferromagnetic structure ($\mathbf{q}=\mathbf{q}_L$) is more favorable from the energy standpoint than a chain structure ($\mathbf{q}=\mathbf{q}_C$).

When surface effects at the interface are ignored, the magnetic energy of this system is the exchange energy of a sample of which a fraction $X/(1+X)$ has the Néel structure without a skew of moments ($\varphi=\pi/2$), while a fraction $1/(1+X)$ has a Landau structure with a skew. The density of this energy is correspondingly written

$$E_M=\frac{1}{a^3(1+X)}[D_1+(D_2-D_1)\cos^2\varphi],$$
$$D_1=S^2(4|J_1|-8|J_2|),\quad D_2=S^2(6|J_1|+8|J_3|). \qquad (12)$$

(This energy is reckoned from the exchange energy density in the case of an ideal collinear checkerboard order.) The quantity D_1 in (12) is the difference between the energies (per atom) of the unskewed layered and checkerboard structures. This difference vanishes at the interface; in general, it is small in comparison with D_2, which is another combination of exchange integrals.

The stationary state of the system is found by minimizing the total energy of the system:

$$E=E_V+E_S+E_C+E_M. \qquad (13)$$

Only the surface energy and the Coulomb energy of the system depend on the parameter R; i.e., the geometry of

the two-phase state is determined by these energies alone. A minimization of the sum of E_S and E_C leads to the following expressions:

$$Q=E_S+E_C$$
$$=0.42521n\left(\frac{\mu^2e^2n^{1/3}}{\varepsilon}\right)^{1/3}\xi\left[\frac{f(X)}{\xi}\right]^{1/3}$$
$$\times(1+X^{2/9})[(1+Y)^{4/3}+(1-Y)^{4/3}]^{2/3}, \tag{14}$$

$$nR^3=0.037880\frac{\mu\varepsilon}{e^2n^{1/3}}\xi\frac{(1+X)^{1/3}}{f(X)}$$
$$\times[(1+Y)^{4/3}+(1-Y)^{4/3}]. \tag{15}$$

After (14) is substituted into (13), the minimization in terms of the other variational parameters is carried out numerically. The minimized energy in (13) must be compared with the energies of the two-phase antiferromagnetic–ferromagnetic state and a uniform state of the crystal. In the calculation of the first of them it was assumed that the insulating phase has an ideal collinear Néel antiferromagnetic structure, while the conducting phase might be magnetized to a point less than saturation. If this is the case, then this phase, too, has a checkerboard structure, but one which is very skewed. The energy of such a state differs from (13) by the expressions for E_V and E_M:

$$E_V=-\frac{n}{4}(1+Y)\sqrt{(W+AS\cos\varphi)^2+A^2S^2\sin^2\varphi}$$
$$-\frac{n}{4}(1-Y)\sqrt{(W-AS\cos\varphi)^2+A^2S^2\sin^2\varphi}$$
$$+0.3n\mu(1+X)^{2/3}[(1+Y)^{5/3}+(1-Y)^{5/3}], \tag{15'}$$

$$E_M=\frac{1}{a^3(1+X)}D_2\cos^2\varphi. \tag{16}$$

Each of the first two terms in (15′) is the energy of the bottom of a corresponding spin subband. The third term gives the kinetic energy of the electrons in these subbands [in the leading approximation in AS/W, this third term is the same as the corresponding term in (8)].

As the energy of the uniform state we adopt the smaller of the energies of the following two states: 1) that in which the electrons uniformly fill the entire crystal, and the latter has a skewed Néel structure; 2) that in which the electrons uniformly fill the entire crystal, and the latter has a skewed Landau structure. The values of these energies are found by minimizing the two versions of expression (13) written above under the condition $X=0$.

CALCULATION FOR A PARTIALLY OR COMPLETELY DISORDERED TWO-PHASE STATE

The variational procedure described above for a two-phase antiferromagnetic structure can be generalized to nonzero temperatures. In the latter case, it must be formulated as a condition for a minimum of the free energy of the system. We restrict the discussion below to the case in which the skew of the sublattice moments can be ignored. A sufficient condition here is the inequality $\mu\gg AS$. The renormalization of the electron energy due to s–f exchange can thus be described in terms of the Hamiltonian for indirect exchange between f-wave spins. Since this Hamiltonian has a Heisenberg structure, a skew is not possible in this system in the absence of an external magnetic field.

In the absence of a skew, it is sufficient to use two variational parameters, R and X. Since the electron gas is degenerate at the temperatures under consideration here, the lowest energy of a nonuniform electron gas must be used in the expression for the total free energy of the system. As before, this energy is the sum of bulk, surface, and Coulomb parts; the latter two are the same as expression (9) for E_S and expression (10) for E_C. The energy of an electron in the one-site approximation, which does not depend on the configuration of the s-wave spins, should be used in the first of these components:

$$E_V=-\frac{Wn}{2}+\frac{3n\mu}{5}(1+X)^{2/3}$$
$$-\left(\frac{A}{2N}\right)^2\frac{S(S+1)}{a^3}\sum_{\mathbf{kq}}\frac{n_{\mathbf{k}}-n_{\mathbf{k+q}}}{E_{\mathbf{k+q}}-E_{\mathbf{k}}}, \tag{17}$$

where $n_{\mathbf{k}}$ is the electron Fermi function. The energy of the magnetic order in (13) should be replaced by the free energy of the magnetic subsystem, F_M. This free energy is expressed not only in terms of the direct-exchange integral $J_{\mathbf{q}}$ in (11) but also in terms of the indirect-exchange integral. The latter is directly related to the two-site correction to the electron energy.

In the molecular field approximation the free energy of the magnetic subsystem can be written

$$F_M=\frac{X}{1+X}f_N(T)+\frac{1}{1+X}f_L(T), \tag{18}$$

$$a^3f_N=\frac{1}{2}J(\mathbf{q}_N)S_N^2-T\ln\sum_{m=-S}^{S}\exp\left[\frac{mJ(\mathbf{q}_N)S_N}{T}\right], \tag{19}$$

$$a^3f_L=\frac{1}{2}\tilde{J}(\mathbf{q}_L)S_L^2-T\ln\sum_{m=-S}^{S}\exp\left[\frac{m\tilde{J}(\mathbf{q}_L)S_L}{T}\right], \tag{20}$$

$$\tilde{J}(\mathbf{q})=J(\mathbf{q})-\frac{A^2}{2N}\left(\sum_{\mathbf{k}}\frac{n_{\mathbf{k}}-n_{\mathbf{k+q}}}{E_{\mathbf{k}}-E_{\mathbf{k+q}}}-\frac{1}{N}\sum_{\mathbf{p}}\frac{n_{\mathbf{k}}-n_{\mathbf{k+p}}}{E_{\mathbf{k}}-E_{\mathbf{k+p}}}\right). \tag{21}$$

In place of expression (6) for $E_{\mathbf{k}}$ we can use the quadratic approximation in (17) and (21).

The expectation values of the spins are found from the self-consistency equations:

$$S_N=SB_S\left(\frac{SJ(\mathbf{q}_N)S_N}{T}\right),$$
$$S_L=SB_S\left(\frac{S\tilde{J}(\mathbf{q}_L)S_N}{T}\right), \tag{22}$$

where B_S is the Brillouin function, and $J(\mathbf{q}) \equiv \mathrm{J}_{\mathbf{q}}$.

We note that the indirect-exchange integral [the second term in (21)], which increases with decreasing $\mathbf{q}$, stabilizes the Landau structure even if $J(\mathbf{q}_N)$ is greater than $J(\mathbf{q}_L)$. However, the electron densities must be below the values at which $\tilde{J}(\mathbf{q})$ acquires a maximum at $\mathbf{q} < \mathbf{q}_L$.

We turn now to the case in which the self-localization of carriers occurs in regions in which there is absolutely no magnetic order. This situation appears to be possible in principle even at $T=0$, because the electron energy in the magnetically disordered regions, given by (17), is very close to the energy in a Landau antiferromagnetic structure, (4), under the condition $AS \ll W$ with dispersion relation (6) (it is assumed that there is no skew). To ensure that this assertion is valid not only for large spins S but also small ones, we should add to the expression found from (4) the energy of Coulomb fluctuations of spins:[16]

$$E^{(q)} = -\frac{A^2 P}{4N^2} \sum_{\mathbf{k},\mathbf{p}} \frac{1}{E_{\mathbf{k}+\mathbf{p}} - E_{\mathbf{p}}}, \tag{23}$$

where P means the principal value. It is simple to verify that the difference between the positions of the bottom of the conduction band in these two cases, divided by the energy of an s-wave electron, is only 1.5%.

On the other hand, the exchange energy expended on creating a spin-liquid region may be less than that expended on creating a region of Landau phase. With $J_3=0$, for example, the first of these energies, per atom, at the interface between checkerboard and chain phases is $3|J_1|S^2/2$ according to (11), and the second of these energies is larger by a factor of 4/3. For this reason, the idea that a semiconductor at $T=0$ might separate into regions of an antiferromagnetic phase and a spin-liquid phase, with all the carriers in the latter, does not look totally absurd. Nevertheless, this question can be unambiguously resolved only after we have verified that a two-phase antiferromagnetic–ferromagnetic state is not more favorable from the energy standpoint than a two-phase antiferromagnetic–spin-liquid state. The possible existence of such a state at nonzero temperatures is self-evident.

RESULTS OF THE CALCULATIONS AND DISCUSSION

In the numerical calculations we ignored spatial fluctuations of the potential of dopant atoms which make the semiconductor degenerate. These fluctuations have little effect on the equilibrium properties of the two-phase state, although they may be extremely important for the kinetics, e.g., causing a pinning of charged regions.

Calculations carried out for $T=0$ show that a two-phase antiferromagnetic state with alternating Néel and Landau phases can be more favorable from the energy standpoint than a uniform state at reasonable values of the parameters of the system. At low carrier densities, the Landau phase consists of distinct spherical droplets, which form a periodic structure within the Néel phase (Fig. 1). Since there are no conduction electrons in the latter, the presence of these electrons in the Landau-phase regions does not cause the crystal as a whole to become a conductor. The reason is that each electron is trapped in its own droplet and cannot carry its charge through the crystal.

With increasing electron density, the relative size of the Landau phase increases. At a certain density n_p, the regions of this phase make contact with each other. A highly conducting Landau phase thus undergoes a topological conversion at $n=n_p$ from a multiply connected phase to a singly connected one. Correspondingly, a percolation of the Landau order and the electron liquid also occurs at n_p; i.e., the crystal becomes highly conducting. At $n>n_p$, the insulating droplets of Néel phase form a periodic structure in a predominant Landau phase (Fig. 2). These droplets have only a slight effect on the conductivity of the crystal, since the conduction electrons circumvent these droplets as they move from the cathode toward the anode. With a further increase in carrier density, the crystal abruptly goes into a uniform state with a Landau order [if, at these values of n, no other value of $\mathbf{q}$ corresponds to a maximum of the total exchange integral $\tilde{J}(\mathbf{q})$; see (21)].

A two-phase antiferromagnetic state is stable at (for example) the following parameter values: $W=1.69$ eV (the effective electron mass is three times the actual mass), $a=3$ Å, $\varepsilon=20$, $AS=1$ eV, $D_1=5\cdot 10^{-4}$ eV, and $D_2=10^{-2}$ eV. A percolation of the Landau order occurs at $n_p=1.3\cdot 10^{20}$ cm^{-3}. The electron droplets are characterized by the values $\varphi=\pi/2$ and $Y=0$ of the variational parameters at the percolation point (the Landau phase is collinear, and the electrons are spin-depolarized). The number of electrons in a droplet at the percolation point is 7.8; the radius of a droplet is 19 Å.

A decrease in the Néel point of the crystal leads to a skew of the sublattice moments in the highly conducting phase and, correspondingly, to a polarization of the electrons. With the same parameter values for the system, except $D_2=5\cdot 10^{-3}$ eV, we thus have $n_p=1.305\cdot 10^{20}$ cm^{-3}, $\cos\varphi=0.097$, and $Y=0.51$ with the same droplet radius and with nearly the same average number of electrons in a droplet (8.2) at the percolation point. A further decrease in the Néel point has the consequence that a two-phase antiferromagnetic–ferromagnetic state is energetically more favorable than a two-phase antiferromagnetic state. With the same parameter values for the system, except $D_2=10^{-3}$ eV, there is thus a cooperative ferron state with a complete magnetization of the highly conducting part of the crystal ($\varphi=0$) and a complete spin polarization of the electrons. The percolation density is $1.715\cdot 10^{20}$ cm^{-3} here, the number of electrons in a droplet is 11, and the droplet radius at the percolation point is 19 Å.

The conditions are even more favorable for the realization of a two-phase antiferromagnetic state in a degenerate antiferromagnetic semiconductor if the effective mass of a carrier is reduced, by increasing the lattice constant, while there is essentially no change in the width of the conduction band, $W=1.83$ eV. If we use the same values of ε, AS, and D_1 and adopt the value $a=5$ Å (in which case the effective mass of an electron is equal to the actual mass), then with $D_2=10^{-2}$ eV there should be a collinear two-phase antiferromagnetic state with a percolation den-

sity $0.291 \cdot 10^{20}$ cm^{-3}, a droplet radius $R=38$ Å at the percolation point, and 14 electrons in a droplet.

A decrease in D_2 to $5 \cdot 10^{-3}$ eV leads to a two-phase antiferromagnetic state with a significant skew of the moments in the Landau phase: While the percolation density and the droplet radius remain essentially the same, with a slight increase in the number of electrons in a droplet, (17), we have $\cos\varphi=0.194$ and $y=0.92$. If we reduce D_2 to 10^{-3} eV, a two-phase antiferromagnetic–ferromagnetic state with a complete magnetization of the ferromagnetic part of the crystal and a complete spin polarization of the electrons becomes energetically favorable. However, the droplet radius and the number of electrons in a droplet change slightly: At the percolation point, $n_p=0.38 \cdot 10^{20}$ cm^{-3}, they are 20 and 39 Å, respectively.

It follows from these results that a two-phase antiferromagnetic state is indeed possible over a fairly broad range of completely realistic parameter values of the crystals. The relation between the parameters W and AS used above ($W \sim AS$) is completely realistic for compounds of transition metals, although it could hardly be possible in compounds of rare earth metals. The density interval 10^{19}–10^{20} cm^{-3}, in which the percolation density falls, is typical of degenerate semiconductors with an effective carrier mass on the order of the actual mass.

It was not possible to carry out numerical calculations for partially or completely disordered systems, including a two-phase antiferromagnet–spin-liquid system at $T=0$, since the procedure developed in the preceding section of this paper is not applicable in the most pertinent case, $W \sim AS$. At $W \gg AS$, the numerical results are not very informative for real systems. Nevertheless, it is possible to draw several qualitative conclusions from the results of the preceding section of the paper even without going through numerical calculations.

As we mentioned in the preceding section of this paper, the difference between the energies of an electron in a spin liquid and a Landau phase is small (only 1.5% under the condition $W \gg AS$). If the system is in a two-phase antiferromagnetic state at $T=0$, we would naturally expect, on this basis, that the Landau order would "melt" very rapidly as the temperature was raised, and a spin liquid would take its place. During heating, the system thus goes from a two-phase antiferromagnetic state into a two-phase antiferromagnet–spin-liquid state, with an insulating Néel antiferromagnetic phase, with all the charge carriers in the spin liquid.

As the temperature is raised further, the two-phase state should undergo a melting, with a phase transition to a uniform state. A detailed study of a two-phase antiferromagnetic–ferromagnetic state has shown[4] that this state converts into a single-phase state through a first-order phase transition. The volume of the highly conducting phase abruptly becomes equal to the volume of the entire crystal, and the ferromagnetism disappears. We would naturally expect that in the case under consideration here the melting of the two-phase state should occur through a first-order phase transition. If the crystal is in a two-phase insulating state below the point of the transition (Fig. 1), it would be in a highly conducting state above this point; i.e., there should be an abrupt insulator–metal transition. If the crystal is instead in a two-phase, highly conducting state below the point of the transition (Fig. 2), then the melting of the two-phase state should not be accompanied by a radical change in conductivity.

By analogy with Refs. 4 and 5, we could expect some interesting features of the phase transitions in two-phase systems near n_p. We might expect that the system could behave in two distinct ways.

1) The dimensions of the highly conducting droplets increase with the temperature. If n is just slightly smaller than n_p, then at some temperature there should be a thermally induced percolation of the Landau order (or of the spin-liquid state) and of the electron gas simultaneously. In other words, there should be a transition from the configuration in Fig. 1 to that in Fig. 2. Consequently, an insulator-metal transition occurs without a disruption of the two-phase nature of the state. At an even higher temperature, this state should melt, and the system should go into a single-phase state, but this melting would not be accompanied by a sharp change in the conductivity of the system. A calculation[4,5] for antiferromagnetic–ferromagnetic systems shows that this is precisely the situation which prevails in those cases.

2) Alternatively, there is a decrease in the dimensions of the highly conducting droplets with increasing temperature. If n is just slightly above n_p, then there might be a thermally induced phase transition from the highly conducting two-phase state in Fig. 2 into the insulating state in Fig. 1. As the temperature is raised further, however, the latter state should melt, and the crystals should revert to a highly conducting state. In other words, one would observe a reentrant insulator–metal transition.

In principle, there is yet a further possibility: phase transitions from a two-phase antiferromagnetic state to an antiferromagnetic–ferromagnetic state. However, at this stage of the research on the problem, that possibility is only hypothetical.

CONCLUSION

It can be seen from the discussion above that this separation of phases has nothing in common with the trivial separation of phases which occurs upon a first-order phase transition. This is essentially a specific nonuniform state of uniform crystals with alternating regions of oppositely charged phases, combined into a unified system by a Coulomb interaction. It is for this reason that models which ignore the Coulomb interaction (e.g., the t–J model in Ref. 10) are inadequate for describing this phenomenon. Furthermore, such models lead to results which are unstable with respect to the incorporation of a Coulomb interaction.

Really, if one assumes that the system is separated into two regions of different phases (and nothing else was stipulated in Refs. 10–13), then the Coulomb energy should be macroscopically large ($\sim N^{1/3}$ in the three-dimensional case and $\sim N^{1/2}$ in the two-dimensional case where N is the number of charge carriers in a phase). For this reason the Coulomb explosion would be inevitable. In principle, the

Coulomb interaction might be suppressed if in the region where all the carriers are concentrated, donors or acceptors compensating their charge are concentrated, donors or acceptors compensating their charge are concentrated, too.[10] But since the diffusion of impurity atoms is very weak their distribution is frozen in the crystal virtually in all the cases of interest. Thus, apparently, the only way to weaken the Coulomb interaction consists in mixing up the phases in the fashion described above.

It should be mentioned then the ferron state of a single charge discovered in Ref. 3 and rediscovered recently in Ref. 10 is in the t–J-model less energetically favored than the magnetic string.[6] On the other hand, separation of the crystal into a magnetic and nonmagnetic phase is impossible in real systems as in them the inequality $t \gg J$ should hold instead of the opposite inequality assumed in Ref. 10.

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