

# Temperature-induced percolation and melting in inhomogeneous electronic-magnetic systems

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We carry out a finite-temperature investigation of the heterophase state of a uniform magnetic conductor in which the conduction electrons interact strongly with the magnetic subsystem. Our calculations apply to a strongly degenerate antiferromagnetic semiconductor which spontaneously decays into alternating ferro- and antiferromagnetic regions at  $T = 0$ , with the conduction electrons concentrated in the former regions. At finite temperatures, a possible state of this system is one with alternating high-conductivity ferromagnetic and low-conductivity paramagnetic regions. As  $T$  increases this heterophase structure melts, so that a bulk sample will pass discontinuously to the uniform state. In a number of cases this melting is preceded by a change in the topology of the ferromagnetic high-conductivity portion of the crystal: as the temperature increases, the crystal topology changes from multiply connected to simply connected, which corresponds to percolation of the conduction electrons and to ferromagnetic order. In other words, an insulator-metal transition occurs. In thin films the heterophase state, which is initially layered, passes continuously into the uniform state with increasing temperature. In these systems, re-entrant transitions are also possible when the heterophase state is unstable at  $T = 0$ : first from the uniform to the heterophase state, and then conversely.

A conducting crystal in which the conduction electrons interact strongly with some other subsystem (i.e., with phonons or magnons) can have states in which the current-carrier distribution is spatially inhomogeneous. In nonmagnetic metals these states are associated with charge-density waves, while in magnetic conductors the states with spatially inhomogeneous charge distributions can have a much more complex structure.

It was shown in Refs. 1–3 that a strongly degenerate antiferromagnetic semiconductor can break up into alternating antiferro- and ferromagnetic regions, with all the conduction electrons concentrated in the latter regions at  $T = 0$ . This heterophase state of the uniform crystal can be viewed as a superposition of nonlinear spin- and charge-density waves coupled to each other. However, it is more natural to regard these inhomogeneous electronic-magnetic states as arising from “ferron” effects in a system of ferromagnetic regions. Ferrons are the quasiparticles of a nonferromagnetic or not-entirely-ferromagnetic crystal; their properties derive from the fact that a conduction electron in such a crystal can create a microregion of increased magnetization around it and thereby “self-localize” within this microregion.<sup>4</sup>

At the present time the concept of ferrons is an attractive one for explaining high-temperature superconductivity in magnets whose magnetic atoms have low spin (see, e.g., Ref. 5). However, for magnetic ions with higher spin the mobility of ferrons is extremely low, and is even lower for ferron complexes which occur in degenerate semiconductors. In this latter case it should also be kept in mind that the electrostatic potential of the ionized donors, which fluctuates in space, causes pinning of the ferron complexes; because they are in weak electric fields, these complexes are actually immovable. In these systems only insulating or normally conducting states are possible; as the magnetic field or temperature changes, phase transitions occur between these states.<sup>1)</sup> The heterophase state under discussion here is observed experimentally in strongly doped antiferromagnetic semiconductors such as EuSe and EuTe with large values of

the spin  $S = 7/2$  (an analysis of an experiment which demonstrates this is given in Ref. 3).

Heterophase states of a strongly degenerate magnetic semiconductor were investigated for  $T = 0$  in Refs. 1–3. In this paper we will discuss these states at finite temperatures, along with a number of qualitatively new results that we have succeeded in obtaining for this case. First of all, we will show that the ferro-antiferromagnetic heterophase state is not the only state possible: a ferro-paramagnetic heterophase state can also occur in degenerate semiconductors, for which the number of conduction electrons in the paramagnetic regions is exponentially small.

Secondly, we will investigate phase transitions which take bulk samples from the heterophase to the uniform state; in essence, this amounts to melting of the nonuniform electronic-magnetic state. In the self-consistent field approximation we find that this is a first-order transition. This result is by no means as self-evident as might be supposed: arguments based on a simple phenomenological model might lead us to conclude that within the self-consistent field approximation this melting ought to be a continuous phase transition, and that it becomes discontinuous because of critical fluctuations.<sup>6</sup> The fact that the model discussed here admits a first-order phase transition even in the absence of critical fluctuations suggests that large jumps in the system parameters are possible at the transition point.

Thirdly, we establish the possibility that the topology of the heterophase state may change as the temperature increases: specifically, the ferromagnetic portion of the bulk crystal can change from multiply connected to simply connected. This is equivalent to simultaneous percolation of the electron liquid and the ferromagnetic order.

Fourthly, within the approximations used here, we find that the disappearance of the heterophase state in a thin film takes place by way of a second-order phase transition; however, the geometry of the heterophase state is such that this transition does not have the characteristics of a melting transition. Re-entrant phase transitions from the uniform to the

heterophase state are possible, in which the crystal is uniform in its ground state, but becomes a two-phase material as the temperature increases, followed by the disappearance of the inhomogeneous electronic-magnetic structure at some higher  $T$ .

## 1. MICROSCOPIC MODEL

The systems under discussion in this paper are degenerate antiferromagnetic semiconductors in the heterophase state. The existence of this state is a consequence of the following physics: when the energy of the conduction electrons is smaller in ferromagnetic order than in antiferromagnetic order, these electrons tend to drive the crystal in the direction of ferromagnetic order. If their concentration  $n$  is not high enough, they cannot make the entire crystal ferromagnetic; however, if these electrons can "self-localize" in certain regions of the crystal, they will make these regions ferromagnetic.

Let us first discuss the case of a bulk sample. By calculating the energy of the ground state in the approximation in which fluctuations in the potential of the ionized donors are neglected, Kashin and Nagaev<sup>2</sup> showed that for sufficiently small  $n$  the ferromagnetic regions are spheres that are isolated from each other within an antiferromagnetic matrix (Fig. 1a). As  $n$  increases the volume of the ferromagnetic region of the crystal increases. At a certain concentration  $n_p$  the topology changes: the ferromagnetic region converts from multiply connected to simply connected. This implies that for  $n > n_p$  the antiferromagnetic regions form spheres that are isolated from one another within a ferromagnetic matrix (Fig. 1b).

Since at  $T = 0$  only the ferromagnetic regions contain electrons, for  $n < n_p$  the crystal behaves as an insulator, while for  $n > n_p$  it behaves as a good conductor. From this we see that for  $T = 0$  the concentration  $n_p$  at which the percolation of electrons and ferromagnetic order begins corresponds to a transition of the crystal from a two-phase insulating to a two-phase conducting state. For  $n = n_p$  the volumes of the ferromagnetic and antiferromagnetic regions of the crystal in the model we will discuss below are equal to one another. It is obvious that at this concentration the models based on spheres (Figs. 1a, 1b) must be inadequate, and that the structure of the heterophase state must be considerably more complicated.

In the system we will discuss here, the symmetry of the system is purely translational. For  $n < n_p$  the period of its structure is determined by the position of the ferromagnetic spheres in the antiferromagnetic matrix; as  $n$  approaches  $n_p$

this period decreases. For  $n > n_p$  the period of the structure is determined by the position of the antiferromagnetic spheres within the ferromagnetic matrix, and increases as  $n$  departs from  $n_p$ . From this we see that at the percolation concentration  $n_p$  the period of the structure is a minimum. It is quite evident that the symmetry group of the system in the insulating heterophase state is not a subgroup of the symmetry group in the conducting heterophase state.

At finite temperatures, the ratio  $x$  of the volumes of the antiferromagnetic and ferromagnetic regions of the crystal must vary with  $T$ , in view of the different temperature dependence of the free energies of the ferro- and antiferromagnetic portions of the crystal (here we will assume that the concentration  $n$  is fixed). Therefore, if the concentration  $n$  differs from  $n_p$  at  $T = 0$ , while  $x$  differs from unity, as the temperature rises the parameter  $x$  can reach its critical value  $x = 1$ . When this happens a phase transition occurs, accompanied by a change in the type of conductivity. As we will show below, this phenomenon, i.e., in which the crystal passes from an insulating to a highly-conducting state as the temperature rises, is possible only for  $n < n_p$ . It is this transition which corresponds to thermally-induced percolation of the electron liquid mentioned above, accompanied simultaneously by the percolation of ferromagnetic order (i.e., the unification of ferromagnetic microregions into a simply-connected region).

Independent of whether or not a percolation phase transition occurs in the crystal, at high temperatures a first-order phase transition should occur from the nonuniform to the uniform state. If this transition occurs from the insulating heterophase state, then the crystal is converted discontinuously to a highly conducting state.

In this paper we will discuss thin films as well as bulk samples. In such films the condition that the surface energy be a minimum gives rise to a tendency towards a heterophase state with layered structure (Fig. 1c). In this case the phase transition from the inhomogeneous to the uniform state is second order. Our calculation shows that it is possible to have a re-entrant transition in this case, for which, as the temperature increases, the system passes from a uniform conducting state to a nonuniform insulating state and then back again to the uniform state.

This transition is to some extent similar to self-localization of individual current carriers in a bulk sample of ferromagnetic semiconductor, whose possibility in principle was proven in Ref. 7: like the transition discussed in this paper, self-localization can occur only at a rather high temperature, and above a certain still higher temperature it becomes impossible once more. However, in contrast to the results pre-

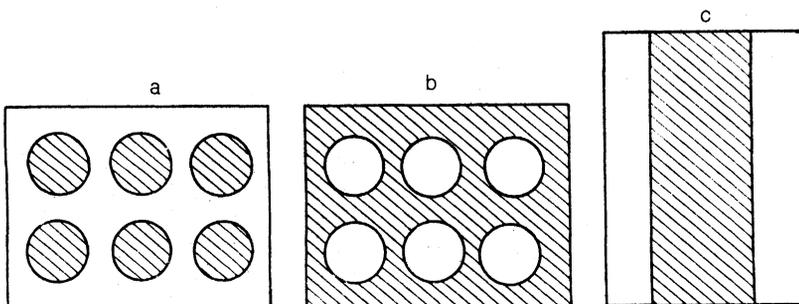


FIG. 1. Two-phase states of a degenerate antiferromagnetic semiconductor: insulating (a) and conducting (b) states in a bulk sample; insulating state in a thin film (c). The ferromagnetic regions of the crystal are crosshatched; the antiferromagnetic regions are not crosshatched.

sented here, both the transition to self-localization and to its disruption take place discontinuously.

The authors of Ref. 3 concluded that for realistic system parameters such processes are impossible. Clearly this also explains the following feature of our model as well: for the case of bulk samples, where the phase transition from uniform to nonuniform state is first order, we also do not observe re-entrant transitions for realistic values of the parameters. However, the situation for thin films turns out to be considerably different, since the conditions for existence of heterophase states in these films are strongly relaxed compared to those of bulk samples. In the final analysis, this reflects the overall energetic advantage of self-localization in two-dimensional as compared to three-dimensional systems.<sup>8</sup>

In addition to characterizing the phase transitions that are possible in these systems, our calculations reveal the strong temperature and external magnetic field dependence of the parameters of a degenerate antiferromagnetic semiconductor in the uniform state. This implies that it is possible to create inhomogeneous systems with parameters whose variation is directional by means of gradients in the temperature or field. For example, in an inhomogeneous magnetic field, if the configuration shown in Fig. 1a is created, then in high-field regions the size of the ferromagnetic spheres and the number of electrons in each of them will be larger than in the low-field regions. Systems of this kind should exhibit nonlinear properties similar to the diode effects observed in island films with directed variation of the island parameters.<sup>9</sup>

## 2. CALCULATIONS FOR A BULK SAMPLE

Let us describe the antiferromagnetic semiconductor using the  $s$ - $f$  model. Our calculations are based on a variational principle for the free energy of the system, which generalizes the approach used in Refs. 1-3 to finite temperatures. In this generalized variational principle we can build in the fact that the magnetization of a region where electrons are concentrated is different from its maximum value. Accordingly, in regions where there are no electrons, the antiferromagnetic order at finite temperatures is either complete or partially destroyed (strictly speaking, at finite temperatures conduction electrons should also be present in these regions; however, their number is exponentially small). These regions can be magnetized by an external magnetic field, whose presence is also included in the calculation. The ratio  $x$  of the volumes of these regions to the volume of the ferromagnetic regions is a variational parameter. The geometry of the inhomogeneous state is given by Figs. 1a and 1b, as before. The radius  $R$  of a spherical inclusion of a different phase is a second variational parameter.

In view of the degeneracy of the electrons, the contribution of thermal excitations to their free energy can be neglected. In complete correspondence with Refs. 1-3, the volume portion  $E_V$  and the surface portion  $E_S$  of the electron energy, and also the Coulomb energy  $E_Q$  (all these quantities are per unit volume in the calculation) are written in the form

$$E_V = \frac{3}{5} \mu_p(n) n (1+x)^{3/2}, \quad \mu_p(n) = \frac{(6\pi^2 n)^{2/3}}{2m^*}, \quad (1)$$

$$E_S = \frac{5}{16} \left( \frac{\pi}{6} \right)^{1/2} \beta \frac{E_V}{n^{1/2} (1+x)^{1/2} R}, \quad (2)$$

$$E_Q = \frac{2\pi}{5\epsilon_0} n^2 e^2 R^2 f(x), \quad (3)$$

where

$$f(x) = \begin{cases} 2x + 3 - 3(1+x)^{2/3} & \text{for a sphere occupied} \\ & \text{by electrons,} \\ x[3x + 2 - 3x^{1/3}(1+x)^{2/3}] & \text{for empty spheres.} \end{cases}$$

Here  $n$  is the average concentration of conduction electrons in the crystal, which we assume is given;  $m^*$  is the effective mass of the electrons;  $\epsilon_0$  is the dielectric permittivity of the crystal; and we have  $\beta = 3$  for the case of spheres occupied by electrons and  $\beta = 3x$  for the case of empty spheres. Equation (3) allows us to establish that for  $x < 1$  a minimum in the energy is ensured by a geometry in which the phase occupied by electrons is the dominant one (i.e., empty spheres), while for  $x > 1$  spheres occupied by electrons are found within an insulating matrix.

The free energy of the magnetic subsystem  $F_M$  (taken per unit volume in this calculation) is calculated at a given temperature  $T$  and external magnetic field  $H$  within the self-consistent field approximation, which takes into account the fact that in the phase occupied by electrons an additional "electronic" magnetic field  $H_e = Ana^3(1+x)/2$  acts on the  $f$  spins, whose origin is the  $s$ - $f$  exchange interaction. In this case it is assumed that electrons in the strongly magnetized phase are completely polarized with respect to spin. Then  $F_M$  is given by a sum of contributions from the weakly- and strongly-magnetized phases<sup>2)</sup>:

$$F_M = \frac{x}{1+x} F_M(H, T) + \frac{1}{1+x} F_M(H+H_e, T), \quad (4)$$

$$a^3 F_M(H, T)$$

$$= \begin{cases} -\frac{H^2}{4|J_0|} + \frac{1}{2} |J_0| S_1^2 - T \ln \left[ \sum_{m=-S}^S \exp\left(\frac{m|J_0|S_1}{T}\right) \right] \\ \text{for } H \leq 2|J_0|S_1, \\ -\frac{1}{2} |J_0| S_2^2 - T \ln \left\{ \sum_{m=-S}^S \exp\left[\frac{m}{T}(H - |J_0|S_2)\right] \right\} \\ \text{for } H \geq 2|J_0|S_1, \end{cases}$$

where  $A$  is the  $s$ - $f$  exchange integral,  $a$  is the lattice constant,  $S$  is the value of the spin of the magnetic atoms,  $J_0$  is the first exchange integral, and  $S_1$  and  $S_2$  satisfy the following self-consistent equations ( $B_S$  is the Brillouin function):

$$S_1 = SB_S \left( \frac{S|J_0|S_1}{T} \right)$$

$$S_2 = SB_S \left[ \frac{S}{T} (H - |J_0|S_2) \right].$$

The expression for  $F_M(H, T)$  in (4) is obtained by the standard self-consistency procedure applied to a two-sublattice antiferromagnet with exchange between nearest neighbors. The case of small fields  $H \leq 2|J_0|S_1$  corresponds to the noncollinear-antiferromagnet situation, where  $S_1$  is the value of the sublattice atomic moment, while the angle between the sublattice moments  $2\theta$  is such that  $\cos \theta = H/2|J_0|S_1$ . In the high field case  $H \geq 2|J_0|S_1$  (which is realized for any

field at temperatures larger than the Néel temperature  $T_N = |J_0|S(S+1)/3$ , since in this case  $S_1 = 0$ ) the crystal is an unsaturated ferromagnet with  $S_2$  being the value of the atomic moments.

The stationary state of the system is determined from the condition that the total free energy of the system be a minimum

$$F = E_V + E_S + E_Q + F_M. \quad (5)$$

It is clear from Eqs. (1)–(4) that only the quantities  $E_S$  and  $E_Q$  depend on the parameter  $R$ . Minimizing the sum  $Q = E_S + E_Q$  with respect to  $R$  for fixed  $x$  leads to the following expressions:

$$Q = 0.57930 \cdot n \gamma \beta \left[ \frac{f(x)}{\beta} \right]^{1/2} (1+x)^{3/2}, \quad \gamma = [\mu_p^2 e^2 n^{1/2} / \epsilon_0]^{1/2},$$

$$nR^3 = 0.06013 \frac{(1+x)^{1/2} \mu_p \epsilon_0}{f(x) e^2 n^{1/2}} \quad (6)$$

After substituting (6) into (5), we carry out the minimization with respect to  $x$  numerically for the same values of the system parameters as were chosen in Refs. 1–3 (they correspond to rare-earth compounds of EuTe type<sup>3</sup>):  $S = 7/2$ ;  $|J_0|S^2 = 10^{-3}$  eV ( $T_N = 5$  K, while the field at which the sublattices collapse is 98.7 kOe);  $AS = 1$  eV;  $\epsilon_0 = 20$ ;  $a^{-3} = 4 \cdot 10^{22}$  cm<sup>-3</sup>; and the effective mass equals the free electron mass. The electron concentration that corresponds to these values of the parameters is  $n_p = 1.05 \cdot 10^{20}$  cm<sup>-3</sup>; at  $T = 0$ , a transition occurs in the crystal from the two-phase insulating state to a two-phase conducting state when  $n$  equals this value. The corresponding values of the ferromagnetic droplet radius  $R$  and the number of electrons  $N$  in a droplet, which are computed more precisely here than in Refs. 1–3, turn out to be the following:  $R = 31.4$  Å,  $N = 28$ . In this work we will consider three cases corresponding to the following values of the electron concentration, all of which are smaller than  $n_p$ :  $10^{19}$ ,  $5 \cdot 10^{19}$ , and  $10^{20}$  cm<sup>-3</sup>.

The results of our numerical calculation for these three values of electron concentration are shown in Figs. 2a, 2b, and 2c in the form of phase diagrams in the  $T$ - $H$  plane. Here Figs. 1, 2, 3 denote regions of absolute stability for the uniform state, the two-phase conducting state, and the two-phase insulating states, respectively.

It is clear from Fig. 2a that for sufficiently small electron concentrations only a transition from the two-phase insulating state to the uniform conducting state is possible. In contrast to the crystal-to-liquid transition, this transition, even in the self-consistent field approximation, is found to be discontinuous with regard to the magnetic structure of the crystal and the spatial distribution of electrons (i.e., the quantity  $x$  is discontinuous).

In the region where the two-phase insulating state is stable, the parameters of this state change in the following way as  $T$  and  $H$  increase (these trends are also preserved for the two other electron concentrations treated here): the radius of the electron spheres and the number of electrons within a sphere increase, while the concentration of these spheres and the value of  $x$  decrease.

At sufficiently high electron concentration, three states can be stable within the framework of the approximations used here: the uniform phase, the conducting heterophase, and the insulating heterophase (see Figs. 2b, 2c). Accordingly, as the temperature changes the class of phase transitions that are possible in such a system is enlarged: along with transitions from the heterophase to the uniform state, it now becomes possible to have phase transitions between the heterophase conducting and heterophase insulating states. At an electron concentration of  $5 \cdot 10^{19}$  cm<sup>-3</sup> such transitions occur only in a magnetic field, while for concentration  $10^{20}$  cm<sup>-3</sup> they can occur in its absence.

Physically, these transitions occur because the magnetic portion of the free energy  $F_M$  in the antiferromagnetic (or paramagnetic) regions of the crystal decreases more slowly with temperature than in the ferromagnetic regions. Therefore, as the temperature increases the volume of the ferromagnetic portion grows. This implies that if the crystal is found in the heterophase insulating state at  $T = 0$ , then as the temperature increases it enters the heterophase conducting state, i.e., an insulator-metal transition occurs.

Note also that the transition of the system to the uniform state both from the insulating and from the conducting heterophase states is a first-order transition for all the electron concentrations investigated above. In this case, as is usual for first-order transitions, within a certain neighborhood of the transition point the corresponding states (uniform from the side of low temperatures and low fields, non-uniform from the side of high temperatures and high fields) continue to exist as metastable states.

As for transitions between insulating and conducting heterophase states (the dashed curves in Fig. 2 correspond to these transitions), it is unfortunate that the geometries adopted here (spheres of one phase within a matrix of the other phase), although completely satisfactory far from the transition point, do not allow us to describe the heterophase state exactly near the transition point: there its geometry is much more complicated. Meanwhile, the geometries of Fig. 1a and Fig. 1b correspond formally to two different branches of the free energy  $F_a$  and  $F_b$ , since for these branches the expressions for  $E_S$  (2) and  $E_Q$  (3) are different. As a consequence of this, the transition from insulating to conducting heterophase states, while formally of first order, is in fact close to second order. Actually, the jumps in the parameters are not large: for  $n = 10^{20}$  cm<sup>-3</sup> the parameter  $x = 1.1$  in the insulating phase and  $x = 0.9$  in the conducting phase.

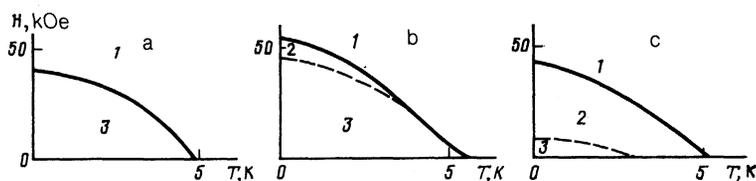


FIG. 2. Phase diagrams of a bulk sample for different values of the electron concentration; a)  $n = 10^{19}$ ; b)  $5 \cdot 10^{19}$ ; c)  $10^{20}$  cm<sup>-3</sup>.

It is natural to assume that the geometry of Fig. 1a in fact evolves continuously into the geometry of Fig. 1b. Then the heterophase state would correspond to a single branch of the free energy, and  $F_a$  would continuously evolve into  $F_b$  near the transition point. Because the structure of the inhomogeneous state can vary continuously with  $T$  or  $n$ , the problem of ordering in the heterophase state differs from the analogous problem for a system of spheres of fixed radius, where it is well known that minima in the energy correspond directly to certain types of packing (we should also add that in this problem the heterophase state with the closest packing is much more energetically disadvantageous because of the Coulomb repulsion).

A further indication that the phase transition is continuous is the fact that on both sides of the transition temperature  $T_0$  the derivative  $-\partial^2 F / \partial T^2$  increases as the temperature approaches  $T_0$ , although not by much (on the order of  $\sim 1\%$  of its value far from  $T_0$ ). Possibly this implies the existence of a peak in the heat capacity at the point  $T_0$ . If the phase transition is in fact a second-order transition, then it differs from the usual type of transition in the absence of an increase in the symmetry of the system at the transition point. A phenomenological theory of such phase transitions was given in Ref. 10.

Although the calculations presented above do not allow us to rigorously confirm these considerations regarding continuity of the phase transition, we may consider the fact of thermally induced percolation to be firmly established. Usually percolation is associated with an increase in the concentration of percolating "substations" and the temperature plays no role. In our case, however, percolation comes about by thermal expansion of the electron-ferromagnetic droplets (naturally this has nothing in common with the expansion of the crystal lattice).

If the concentration of conduction electrons is larger than  $n_p$  but smaller than  $n_0 = 1.9 \cdot 10^{20} \text{ cm}^{-3}$ , then at zero temperature and external magnetic field the system is in a conducting heterophase state. As the temperature or field increases this state passes into the uniform state by way of a first-order phase transition. If  $n > n_0$ , then the system is found in the uniform state for any temperature and field.

### 3. CALCULATIONS FOR A THIN FILM

Let us now consider a situation where the degenerate antiferromagnetic semiconductor is a thin film whose thickness is comparable to the size of inhomogeneities in a bulk sample made of the same material. In this case the geometry of a state that is inhomogeneous with respect to magnetization and electron density changes radically and will apparently be layerlike. As was shown in Ref. 2, the difference in energy between inhomogeneous states with geometries like those shown in Figs. 1a and 1b, and states shown in Fig. 1c, are not large even for bulk samples. Therefore rather small surface interactions are sufficient to fix the layered structure in the film. In what follows we will show that in fact the region of concentrations in which it is possible to have a layered heterophase state is wider for the film than for a bulk sample with the structures shown in Figs. 1a and 1b.

Let us consider an inhomogeneous state of a film of thickness  $L$  for which all the conduction electrons are concentrated in a layer of increased magnetization at the center of the film (Fig. 1c). An alternating layered structure whose

center is in the antiferromagnetic phase is less advantageous energetically, since for an equal volume of the ferromagnetic phase spatial quantization raises the energy of the electrons of a simply connected ferromagnetic region less than for a multiply connected region. Fixing of the electron layer in the center of the film can be facilitated by the small repulsive potential of the film surfaces which causes a kink at the top of the conduction band; however, this will not be taken into account explicitly here. Because the electron layer is isolated from the surface of the film by layers of the insulating phase, this inhomogeneous state will be insulating for currents flowing in the direction perpendicular to the surface of the film.

The ratio  $x$  of the volume of the insulating phase to the volume of the conducting phase (implying that the thickness of the electron layer equals  $L / (1 + x)$ ) is a variational parameter as before. Because the bulk part of the electronic energy  $E_V$  and the free energy of the magnetic subsystem  $F_M$  do not depend on the geometry of the inhomogeneous state, Eqs. (1) and (4) are also valid for the case of a layered structure. However, the expressions for the surface part of the electronic energy  $E_S$  and the Coulomb energy  $E_Q$  in this case have the form

$$E_S = \frac{5}{8} \left( \frac{\pi}{6} \right)^{1/2} \frac{(1+x)^{3/2}}{Ln^{1/2}} E_V, \quad (7)$$

$$E_Q = \frac{\pi}{6\epsilon_0} n^2 e^2 L^2 \left( \frac{x}{1+x} \right)^2. \quad (8)$$

In the general case, it is necessary to consider yet another variational parameter, the number of electronic layers. As this number increases the Coulomb energy decreases while the surface energy increases. In particular, in order that the conducting phase be a single electronic layer, it is necessary that the inequality

$$\frac{3}{4} E_Q < E_S$$

be fulfilled, where  $E_S$  and  $E_Q$  are determined by Eqs. (7) and (8). For the system parameters used here for our numerical calculations, this inequality is fulfilled for any values of  $x$ .

A stationary state of the system is found from the condition that the total free energy (5) be a minimum, where the terms of this free energy are defined by Eqs. (1), (7), (8) and (4). Numerical minimization with respect to the single variational parameter  $x$  was carried out for  $L = 50 \text{ \AA}$  and the same remaining parameters as for the previous case of a bulk sample. The results of the calculation are shown in Fig. 3; the region of stability of the heterophase insulating state lies below the corresponding curve, while that of the uniform conducting phase lies above it. In contrast to a bulk sample, in

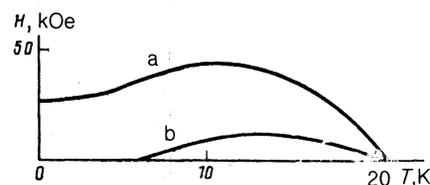


FIG. 3. Phase diagram for thin film for different values of electron concentration: a)  $n = 10^{19}$ ; b)  $5 \cdot 10^{19} \text{ cm}^{-3}$ .

the self-consistent field approximation the transition between these two states is always second-order: the thickness of the near-surface antiferromagnetic regions drops to zero gradually. In principle the possibility exists that this transition will become first-order due to critical fluctuations; however, here again the transition will remain close to second-order.

Note that, in contrast to the bulk case, the heterophase state is always insulating in the situation discussed here, leading to a smaller variety of possible phase transitions. The characteristic feature of these results is the presence of a temperature region where the boundary magnetic field grows with temperature. This leads to the possibility of a re-entrant transition: within a certain range of values of the magnetic field, a transition can occur from a uniform to a nonuniform state as the temperature increases. As the temperature increases further, the system always evolves into the uniform state. For  $n > 4 \cdot 10^{20} \text{ cm}^{-3}$  the system is in the uniform state for any temperature and field. Note that for a bulk sample with the same parameters, the uniform state is established even for  $n_0 = 1.9 \cdot 10^{20} \text{ cm}^{-3}$ , i.e., the region of stability of the nonuniform state is wider in the film than in the bulk.

<sup>1</sup>) A similar insulator-conductor transition is possible at intermediate doping, where the potential of the defects plays a decisive role. These transitions are caused by the dependence of the critical donor concentration at which delocalization of the donor electrons occurs on the local and average magnetizations of the crystal.<sup>3</sup>

<sup>2</sup>) For  $T = 0$  and  $H = 0$  in the nearest-neighbor approximation, or for

$H > 2|J_0|S$ , the magnetic free energy reduces to a purely bulk term determined by Eq. (4) for any exchange radius, while the surface contribution to this energy reduces to zero. At finite temperatures this surface contribution is different from zero; however, we can assume that it is small compared to the electronic energy.

<sup>3</sup>) Conditions for the appearance of the heterophase state in EuSe are considerably more favorable; however, theoretical investigation of this state is hindered by the fact that EuSe is a metamagnet with an extremely complicated phase diagram.

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