Phase separation in correlated systems with a short-range order

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A model describing the onset of phase separation in systems with a fluctuation short-range order is proposed. Over a wide region of the phase diagram, an insulating short-range order leads to an absolute instability of a homogeneous state of the system.

Phase separation is widely observed in various high-T_c superconductors in very diverse experiments. Interest in this effect continues to grow; in fact it is fair to say that this topic has now emerged as an independent direction in research on the properties of high-T_c superconductors. A band theory for phase separation, which has been worked out by the present authors, has made it possible to describe, from a common standpoint, a wide range of experimental results which seem, at first glance, to be unrelated.[1] The condition for phase separation is that the derivative of the chemical potential μ with respect to the number of particles n be negative ($\partial\mu/\partial n<0$). This negativity expresses the general thermodynamic condition that a homogeneous state of the system is absolutely unstable. According to band theory and in the mean-field approximation,[1] this condition is realized because of a rapid suppression of the insulating order parameter by doping. This order parameter determines the gap in the carrier spectrum in a phase with magnetic or charge order. Convincing experimental evidence has recently been found for the existence of strong insulating correlations throughout the region in which a superconducting state exists.[2,3] However, there is the problem that the long-range order in this region is suppressed by doping and large fluctuations, while the insulating correlations are manifested in the form of a short-range order.[3] In order to describe effects of a short-range order, it is necessary in principle to abandon the mean-field approximation.

In the present letter we construct a description of this sort for 2D systems with an interband exciton instability of the type discussed in Ref. 4. That an instability of this sort can exist in the CuO_2 plane of a high-T_c superconductor was demonstrated in Ref. 5. We thus obtain a formulation of the problem of the 2D doping of a semiconductor whose valence and conduction bands are separated by a gap ϵ_g and whose upper band has a particle density n. A phase separation is possible in such a system because the free electrons, while suppressing the interband instability (the effect is analogous to a Burshteĭn shift), cause an effective lowering of the edge of the conduction band and thus a possible lowering of the level of μ with increasing n. As we show below, this mechanism for phase separation does not depend on whether there is a long-range insulating order (an exciton condensate), but it is realized in the fluctuation region in which there is only a short-range order.

The partition function of the system can be written as the functional integral

$$Z=\int D\bar{\Psi}D\Psi\exp[-S\{\bar{\Psi},\Psi\}, \quad \bar{\Psi}=(\psi_c^*,\psi_v^*),$$

$$S=\int_0^{1/T}d\tau\int d^2r\left\{\bar{\Psi}\left[\partial_\tau-\mu+\sigma_z\left(-\frac{\nabla^2}{2m}+\frac{1}{2}\epsilon_g\right)\right]\bar{\Psi}+g\psi_v^*\psi_c^*\psi_c\psi_v\right\}, \tag{1}$$

where g is the interaction potential in the interband channel, and $\psi_{c(v)}$ are Grassmann fields corresponding to the conduction band (to the valence band). When intraband interactions are taken into account, the vertex g becomes dressed, and an instability may also result in the superconducting channel. We will not discuss the latter possibility in the present letter (for example, a temperature above the superconducting transition temperature). We accordingly restrict the discussion to the interband interaction, which is of importance to the mechanism for the phase separation, taking g to be the dressed vertex.

Using a Hubbard–Stratonovich transformation, we can put the fermion part of the action in Gaussian form:

$$S=\int d\tau d^2r\left\{\bar{\Psi}M\Psi+\frac{(\Delta^*\Delta)}{g}\right\},$$

$$M=G^{-1}+\hat{\Delta}, \quad \hat{\Delta}=\sigma_+\Delta+\sigma_-\Delta^*. \tag{2}$$

The operator

$$G=\left[\partial_\tau-\mu+\sigma_z\left(-\frac{\nabla^2}{2m}+\frac{1}{2}\epsilon_g\right)\right]^{-1}$$

corresponds to the Green's function of the free fields. Let us consider the case in which the exciton energy ϵ_{ex}, the doping, and thus the temperature interval of interest here are such that the following condition holds:

$$\nu\equiv\max\left\{\mu-\frac{1}{2}\epsilon_g,\ T,\ \epsilon_{ex}-\epsilon_g\right\}\ll\epsilon_g.$$

In the fields ψ we distinguish parts which vary rapidly and slowly:

$$\psi=\psi^{(0)}+\psi^{(1)}, \quad \psi^{(0)}=\sum_{k>\tilde{k}}\psi(k)e^{ikx}, \quad \psi^{(1)}=\sum_{k<\tilde{k}}\psi(k)e^{ikz}.$$

The momentum $\tilde{k}$ is chosen to satisfy

$$\nu\ll\tilde{k}^2/2m\ll\epsilon_g. \tag{3}$$

We integrate the expression for the partition function with action (2) over the Fermi fields. We then multiply and divide the result by $\exp\{\mathrm{Tr}^{(1)}\ln\tilde{M}\}$, where $\tilde{M}$ corresponds to the operator M in (2) at a zero chemical potential $[\tilde{M}=M(\mu=0)]$, and the superscript 1 on the Tr operation means that the trace is taken over only the slow variables $(k<\tilde{k})$. As a result we find

$$Z\sim\int D\Delta D\psi^{(1)}\exp[-\{S_B+S_{FB}\}]. \tag{4}$$

The boson part of the action, S_B, is

$$S_B = -\mathrm{Tr}^{(0)}\ln(1+G\hat{\Delta}) - \mathrm{Tr}^{(1)}\ln(1+\tilde{G}\hat{\Delta}) + \int d\tau d^2r\,\frac{|\Delta|^2}{g}. \qquad (5)$$

Here $\tilde{G}\equiv G(\mu=0)$ is the Green's function of the undoped system. The part of the action which describes fermions and the fermion–exciton interaction is

$$S_{FB} = \int d\tau d^2r \bar{\psi}^{(1)} M\tilde{M}^{-1}\psi^{(1)}. \qquad (6)$$

Using condition (3), we can calculate the product $M\tilde{M}^{-1}$ and eliminate states of the valence band. In the leading order in ϵ_g^{-1}, we thus have the following expression for S_{FB}:

$$S_{FB} = \int d\tau d^2r\left\{\psi^*\left(\partial_\tau - \mu - \frac{\nabla^2}{2m}\right)\psi + \frac{1}{\epsilon_g}\Delta^*\Delta\psi^*\psi\right\}. \qquad (7)$$

The physical nature of the fermion–boson repulsion which has appeared in (7) is determined by the circumstance that the electrons, by filling band states, reduce the phase volume involved in forming excitons and thereby suppress the tendency toward an exciton instability.

We wish now to transform the boson part of the action, (5). Since conditions (3) are satisfied, the result of taking the trace over the fast variables (the region $k>\tilde{k}$) does not depend on the presence of $\mu\neq 0$ in the first term in (5). Consequently, we find the following result for S_B:

$$S_B = -\mathrm{Tr}\ln(1+\tilde{G}\hat{\Delta}) + \int d\tau d^2r\,\frac{|\Delta|^2}{g}. \qquad (8)$$

In the case under consideration here ($|\epsilon_g - \epsilon_{\mathrm{ex}}| \ll \epsilon_g$), an expansion of (8) in powers of $\Delta(r,\tau)$ is an expansion in terms of a "gas" parameter, which is a measure of the extent to which the excitons can be considered to constitute a gas. It can be shown[6] that it is sufficient to restrict the discussion to terms of up to fourth order, regardless of the dimensionality. In $D=2$ we obtain the following effective action, which describes electrons in the upper band which are interacting with the charge fluctuation field $\varphi=(m/2\pi\epsilon_g)^{1/2}\Delta$:

$$S_{\mathrm{eff}} = \int d\tau d^2r\varphi^*\left\{\left(\partial_\tau - \frac{\nabla^2}{4m} + \alpha\right)\varphi + \frac{1}{2}\lambda(\varphi^*\varphi)^2 + \psi^*\left(\partial_\tau - \frac{\nabla^2}{2m} - \mu\right)\psi + \lambda\varphi^*\varphi\psi^*\psi\right\}, \qquad (9)$$

where the interaction constant is $\lambda=2\pi/m$.

In 2D systems, for any $T\neq 0$, the long-range order, in the sense that there is a Bose condensate, is disrupted (we have $\langle\varphi\rangle\equiv 0$, and the binary correlation function vanishes at infinity). However, it follows directly from the form of the fermion–boson interaction in (9) that the existence of a short-range order (i.e., $\langle\varphi^2\rangle\not\equiv 0$) is sufficient for a lowering of the edge of the electron band, which leads to the possibility of a phase separation. The specific form of the short-range order which results is not of fundamental importance. [For example, in the model which we are using here, (1), the appearance of a topological "long-range order" with a power-law decay of the correlation function as the result of a (Berezinskiĭ–) Kosterlitz–Thouless phase transition is quite probable.]

The possibility that the condition $\partial\mu/\partial n<0$ would hold, and that a phase separation would occur, can be demonstrated even in the very simple self-consistent approximation for the eigenenergy parts of the bosons and the fermions, which are, respectively,

$$\sum_B=\lambda\langle\varphi^2\rangle+\lambda n,\quad \sum_F=\lambda\langle\varphi^2\rangle. \tag{10}$$

The role played by the self-consistency equations is played in this case by equations which determine the electron density n and the mean-square value of the field φ, which describes the presence of an insulating short-range order in the system:

$$n=2\int f_F\left(\frac{k^2/2m-\mu_F}{T}\right)\frac{d^2k}{(2\pi)^2},\quad \mu_F=\mu-\lambda\langle\varphi^2\rangle,$$

$$\langle\varphi^2\rangle=\int f_B\left(\frac{k^2/4m-\mu_B}{T}\right)\frac{d^2k}{(2\pi)^2},\quad -\mu_B=\alpha+\lambda n+\lambda\langle\varphi^2\rangle, \tag{11}$$

where f_F and f_B are respectively Fermi and Bose distributions. Integrating Eqs. (11), we find the explicit dependence of the quantities $\langle\varphi^2\rangle$ and $\partial\mu/\partial n$ on the number of particles n and on the temperature T:

$$\langle\varphi^2\rangle=\frac{2T}{\lambda}\ln\frac{x}{\sqrt{1+2x}-1}\equiv\frac{2T}{\lambda}\ln\beta,\quad x=2\exp\left\{-\frac{\alpha+\lambda n}{T}\right\}, \tag{12}$$

$$\frac{\partial\mu}{\partial n}=-\frac{1}{2}\lambda\left\{4\frac{2\beta-1}{\beta-1}-[1-\exp(-\lambda n/2T)]^{-1}\right\}. \tag{13}$$

In the case $T\equiv0$, the boson chemical potential is $\mu_B=0$, and $\langle\varphi^2\rangle$ is equal to the square of the mean-field order parameter $(\langle\varphi^2\rangle=\langle\varphi_0\rangle^2_{HF})$. In other words, there is a long-range order (with $\alpha+\lambda n<0$). In the case $T\not\equiv0$, however, this long-range order is completely disrupted, and we are left with only a short-range order $(\langle\varphi\rangle\equiv0,$ $\langle\varphi^2\rangle\not\equiv0)$. Analysis of expression (13) shows that there exists a region of short-range order in which the condition $\partial\mu/\partial n<0$ holds. In particular, under the condition $\lambda n/2T\ll1$, the critical density n_c, below which the system is absolutely unstable with respect to separation, has the following T dependence:

$$n_c=m/2\pi\,[\epsilon_{ex}-\epsilon_g+T\ln(4/3)].$$

In this case the nature of the phase separation is governed by a renormalization of the spectrum of charge carriers because of the condition $\langle\varphi^2\rangle\not\equiv0$, which leads to a decrease in the band gap with the doping and to a resultant decrease in the level of μ with increasing n. The phase-separation mechanism discussed here is not directly related to an attraction of charge carriers in real space, which is also responsible for the onset of superconductivity, as in models based on the strong-coupling approximation.[7,8] In the case at hand, the superconductivity develops in an intraband channel in parallel with the phase separation, without competing with the latter, in contrast with the case in Refs. 7 and 8.

In the phase-separation models[1,7,8] which have been proposed previously, phases characterized by a long-range order of some type or other have coexisted. As has been shown in the present letter, however, the existence of well-developed local correlations, which form regions of short-range order, is sufficient for the onset of a phase separation. As in systems with a long-range order, a phase separation may be induced by a superconductivity.[1] A specific interaction of this type, of internal degrees of freedom responsible for the onset of a dielectric state and for superconductivity, is apparently of fairly general applicability. In particular, it should occur in disordered superconductors near a metal–insulator transition.[9] Whether it is possible to construct a model for phase separation in such systems on the basis of ideas concerning an insulating order parameter requires a special study.

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