Phase transitions stimulated by impurities in superconductors with a multicomponent order parameter

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An investigation is reported of the influence of an anisotropic interaction with impurities on the superconducting states with a complex vector order parameter. The Ginzburg–Landau theory is used to show that when the impurity concentration n exceeds a certain critical value n_{c1}, the phase transition from the normal to the superconducting state splits into two transitions: the first, with a real vector order parameter, occurs at $T = T_{c1}$, and at lower temperatures by a second-order phase transition to a state with a complex vector order parameter. This state is separated from the region $n < n_{c1}$ by a second-order phase transition. It is pointed out that this situation may not occur in a real superconductor if the superconductivity appears before the point $n = n_{c1}$ is reached on the concentration axis. The physical conditions under which the above splitting into two transitions nevertheless takes place are given. Moreover, at an impurity concentration $n = n_{c2} > n_{c1}$ the T_{c1} line exhibits a tricritical point: the superconducting transition in the range $n > n_{c2}$ becomes first-order if the superconductivity does not appear at lower concentrations.

1. INTRODUCTION

Replacement of uranium with thorium atoms in the heavy-fermion compound UBe_{13} splits the phase transition to the superconducting state into two at thorium concentrations exceeding a certain critical value x_c (Refs. 1 and 2). Such phase transition behavior is possible when the superconducting state represents a mixture of two superconducting phases a and b, which have different symmetry groups and whose critical temperatures depend in different ways on the impurity concentration: $T_{cb} > T_{ca}$ when $x < x_c$ and $T_{cb} < T_{ca}$ when $x > x_c$. A theory of phase transitions of cubic semiconductors based on this hypothesis is developed in Ref. 3 and other possible explanations of the phase diagram of the system $U_{1-x}Th_xBe_{13}$ are considered there.

Phase transitions between superconducting phases of different symmetry are possible also within the framework of one representation. We shall show that such a situation does occur in a superconductor containing impurities and characterized by a complex vector order parameter ψ, which transforms in accordance with one of the two- or three-dimensional representations of the symmetry group.[4]

The interaction with impurities in superconductors with a multicomponent order parameter is anisotropic, i.e., it depends on the position (relative to the center of a unit cell) of a given impurity. Averaging over the random orientations of the anisotropy axes gives rise to an effective functional of the free energy and minimization of this functional determines the phase diagram of the superconducting states.

In the range of impurity concentrations n exceeding a certain critical value n_{c1} the phase transition from the normal to superconducting state splits into two. First, a second-order transition takes place at $T = T_{c1}$, producing a superconducting phase *1* with a real order parameter ($|\psi^2| = |\psi|^2$), and this is followed by a second-order transition to a phase *2* with a complex order parameter ($0 < |\psi^2| < |\psi|^2$), separated from a region *3* with a complex order parameter ($|\psi^2| = 0$) by a second-order phase transition. When the impurity concentration is $n = n_{c2} > n_{c1}$, the T_{c1} line may exhibit a tricritical point the phase transition to the superconducting state with $n > n_{c2}$ becomes first-order. However, in the case of physically meaningful parameters of the theory the superconductivity is clearly suppressed at impurity concentrations less than $n > n_{c2}$. This qualitative behavior is the same for two- and three-dimensional representations of the symmetry group of the particular crystal.

We shall identify the superconducting classes to which the new phases belong and establish inequalities governing the parameters of the proposed model and ensuring the phase transitions just described.

2. SELECTION OF THE MODEL AND AVERAGING OVER IMPURITIES

We shall consider a superconductor described by the Ginzburg-Landau functional (see Refs. 4 and 5):

$$F=\int_V dV\left[-\alpha\tau|\psi|^2+\beta_1|\psi|^4+\beta_2|\psi^2|^2+\beta_3\sum_i|\psi_i|^4\right]+F_{im}, \tag{1}$$

$$F_{im}=\sum_a \psi_i(\mathbf{r}_a)\psi_k^*(\mathbf{r}_a)U_{ik}^a,$$

$$\tau=(T_c-T)/T_c, \tag{2}$$

where α, β_1, β_2, and β_3 are numbers; V is the volume of the superconductor; and the index a labels the various impurities. In this model U^a_{ik} is a symmetric matrix which corresponds to an ellipsoid of revolution with the eigenvalues λ_1, λ_2, and λ_3 whereas in the expression (2) for F_{im} we have to sum over the impurities present, whose concentration is n. The problem of the appearance of a term $\beta_2|\psi^2|^2$ in Eq. (1) and of those superconducting classes which admit the existence of this term will be discussed in Sec. 4.

The subsequent procedure is as follows: Averaging is carried out over the impurity orientations and their positions in space. Then, minimization of the Ginzburg-Landau functional establishes the nature of the superconducting state.

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One important point should be noted. In the calculation of the main physical quantities we have to average the logarithm of the partition function, i.e., the free energy and not the partition function itself. Nevertheless, we shall average the partition function. However, it can easily be shown (see the Appendix) that to fourth order in ψ, the results of calculations made by these two methods are identical. It follows from the above comments that the functional form of the true free energy will differ from that given above, but this does not affect the final result, because in this result we need to know the free energy only to fourth order in ψ.

We now carry out averaging over the impurity-orientations:

$$U_{ik}{}^{a}=(S^{a})^{t}\Lambda S^{a},$$
$$\Lambda_{ij}=0,\quad i\neq j;\quad \Lambda_{11}=\lambda_1;\quad \Lambda_{ii}=\lambda_2,\quad i\neq 1, \tag{3}$$

where S^a is an orthogonal matrix. The dimensionality of the matrices Λ and S^a is equal to the dimensionality of the order parameter.

The partition function can be described by the following readily derived expression:

$$\langle\exp(-\rho F_{im})\rangle_{im}=\left\{\int\exp[-\beta f_{im}(\psi(\mathbf{r}),\psi^*(\mathbf{r}))]\frac{d\mathbf{r}}{V}\right\}^{nV}, \tag{4}$$

$${}_{m}(\psi(\mathbf{r}_a),\psi^*(\mathbf{r}_a))=-\frac{1}{\beta}\ln\left[\int DS^{a}\exp(-\beta F_{im}{}^{a})\right],\quad \beta=\frac{1}{T}. \tag{5}$$

Complete integration of Eq. (5) is possible only if the order parameter has the dimensionality $d_s=2$. In this case the calculation gives

$$f_{im}(\psi,\psi^*)=\frac{\lambda_1+\lambda_2}{2}|\psi|^2-\frac{1}{\beta}\ln\left[I_0\left(\frac{|\lambda_1-\lambda_2|}{2}\beta|\psi^2|\right)\right]. \tag{6}$$

If $d_s=3$, then

$$f_{im}(\psi,\psi^*)=\frac{\lambda_1+\lambda_2}{2}|\psi|^2$$
$$-\frac{1}{\beta}\ln\left\{\int_{-1}^{1}dx\exp\left[-\frac{x^2}{2}(\lambda_1-\lambda_2)\beta|\psi|^2\right]\right.$$
$$\left.\times I_0\left(\frac{1-x^2}{2}\beta|\psi^2|(\lambda_1-\lambda_2)\right)\right\}, \tag{7}$$

where $I_0(t)$ is a modified Bessel function of the zeroth order. We shall consider only the case $d_s=2$.

3. PHASE DIAGRAM

We thus find that the density of the average Ginzburg–Landau functional is

$$f=-\alpha\tau|\psi|^2+n\frac{\lambda_1+\lambda_2}{2}|\psi|^2+\beta_1|\psi|^4+\beta_2|\psi^2|^2$$
$$+\beta_3\sum_i|\psi_i|^4-\frac{n}{\beta}\ln\left[I_0\left(\frac{\beta}{2}(\lambda_1-\lambda_2)|\psi^2|\right)\right]. \tag{8}$$

For $\beta_3>0$, then for arbitrary values of $|\psi|^2$ and $|\psi^2|$ we can assume $|\psi_x|^2=|\psi_y|^2$. Then, introducing

$$-\alpha\tau+n(\lambda_1+\lambda_2)/2=-a(T,n),$$
$$\beta=\beta_1+\beta_3/2, \tag{9}$$

we can reduce f to

$$f=f_1(|\psi|^2)+f_2(|\psi^2|), \tag{10}$$

where

$$f_1(t)=-at+\beta t^2,$$
$$f_2(t)=\beta_2t^2-(n/\beta)\ln[I_0(^1/_2\beta t|\lambda_1-\lambda_2|)].$$

For $\beta_2\leqslant 0$, we have $f_2\leqslant 0$ and the state is always described by the relationship $|\psi|^2=|\psi|^2|$. However, for $\beta_2>0$, there is a critical concentration $n_{c1}=16\beta_2/\beta|\lambda_1-\lambda_2|^2$, such that for $0\leqslant n<n_{c1}$ the graph has the form shown in Fig. 1a, whereas for $n>n_{c1}$ the graph is different (Fig. 1b).

Just this qualitative discussion is sufficient to show that in the former case the superconducting state is characterized by $|\psi^2|=0$, whereas in the latter case there is a transition from the state 1 ($|\psi|^2=|\psi^2|$) to the state 2 ($0<|\psi^2|<|\psi|^2$) at a temperature $T_{c2}(n)$. We find this dependence and show that this is a second-order phase transition.

We can easily see that a minimum of f corresponds to states with the following order parameters:

$$\begin{aligned}&1)\ |\psi|^2=|\psi^2|,\quad \text{if}\quad |\psi|^2\leqslant\xi_0;\\&2)\ |\psi|^2>|\psi^2|=\xi_0,\quad \text{if}\quad |\psi|^2>\xi_0,\end{aligned} \tag{11}$$

where $\xi_0>0$ is such that $\min f_2(\xi)=f_2(\xi_0)$. It should be noted that ξ_0 is independent of temperature and is affected only by the impurity concentration and other parameters. We thus find

$$f(\psi,\psi^*)=\begin{cases}f_1(|\psi|^2)+f_2(|\psi|^2), & |\psi|^2\leqslant\xi_0,\\ f_1(|\psi|^2)+f_2(\xi_0), & |\psi|^2>\xi_0.\end{cases} \tag{12}$$

We can also readily determine the dependence of $|\psi|^2$ on T by representing f_2 in the vicinity of ξ_0 by the expression

$$f_2(\xi)=f_2(\xi_0)+\gamma(\xi-\xi_0)^2. \tag{13}$$

Then the relationship

$$a=2\xi_0\beta \tag{14}$$

governs the temperature of the transition from the phase *1* to the phase *2*.

We can also calculate easily the specific heat discontinuity as a result of this phase transition:

$$\Delta c=T^2(\xi_0)\left(\frac{\partial^2f_2(\xi)}{\partial\xi^2}\right)_{\xi=\xi_0}\left(\frac{\partial\xi}{\partial T}\right)^2. \tag{15}$$

In view of the continuity of the first derivative with respect to f, it follows that the $1\to 2$ transition is second-order.

We now consider the line separating the phases labeled *2* and *3*. It is a segment joining the points $[T_c(n_{c1}),n_{c1}]$ and $(0,n_{c1})$ because its position is governed by the existence of a nonzero minimum of the function f_2, the appearance of

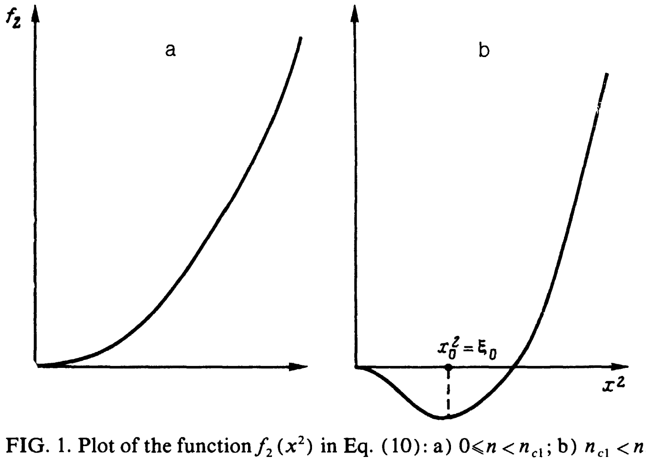

FIG. 1. Plot of the function $f_2(x^2)$ in Eq. (10): a) $0\leqslant n<n_{c1}$; b) $n_{c1}<n$.

which below $T_c(n_{c1})$ is independent of temperature. We show that this line represents second-order phase transitions. The quantity ξ_0 can be approximated satisfactorily by the expression

$$\xi_0 = \frac{4}{|\lambda_1-\lambda_2|}\left[\left(\frac{n}{n_{c1}}\right)^2 - 1\right]^{1/2}. \tag{16}$$

We can then readily find the change in the free energy as a result of the $2\to 3$ transition:

$$\Delta f_{2\to 3} = f_2(\xi_0(n)) \approx \mathrm{const}\cdot\xi_0^4. \tag{17}$$

It readily follows from the above expression that the $2\to 3$ transition is second-order, because the free energy in the dependence on the impurity concentration is a continuously differentiable function, but its second derivative has a discontinuity along the n_{c1} line.

If the impurity concentration n exceeds $n_{c2} = 16T(\bar{\beta}+\beta_2)/(\lambda_1-\lambda_2)^2$, the dependence of the free energy on the order parameter has two different minima indicating that the line separating the normal and superconducting (1) phases is a line of first-order phase transitions and that the minimum of $|\psi|\neq 0$ is lost when the superconducting phase is absolutely unstable, whereas the minimum $|\psi|=0$ disappears when the normal phase becomes absolutely unstable. This is illustrated in Figs. 2 and 3.

In the next section we shall identify the superconducting classes to which this model can be applied, as well as the correspondence between these classes and the phases predicted by the model.

4. SYMMETRY OF SUPERCONDUCTING PHASES

The various classes can be made to correspond to the different phases of the model by first identifying the symmetry groups of the normal phase which admit the existence of a term $\beta_2|\psi^2|^2$ in the functional (1). We can readily show that if the order parameter transforms in accordance with the two-dimensional representation of the corresponding group, i.e., if the vector ψ is two-dimensional, the symmetry group of the superconductor should be axial (it should have a preferred symmetry axis). However, if the vector ψ is three-dimensional, then in general any point group allows the existence of the term in question.

Since we shall concentrate on the two-dimensional order parameter and allow for the influence of the term $\beta_3\Sigma|\psi_i|^4$ in the functional (1), it is clear that the maximum combined group corresponding to the normal state consistent with a model described by the functional (1) is the group $D_4\times U(1)\times R$.

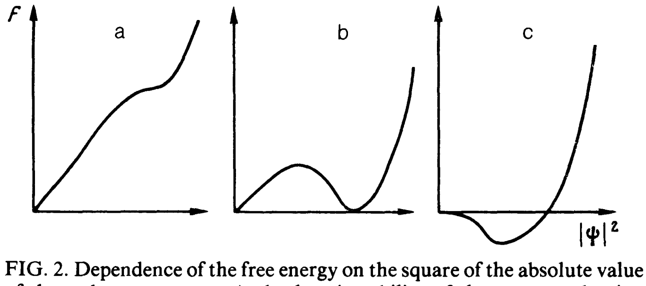

FIG. 2. Dependence of the free energy on the square of the absolute value of the order parameter: a) absolute instability of the superconducting phase; b) first-order phase transition; c) absolute instability of the normal phase.

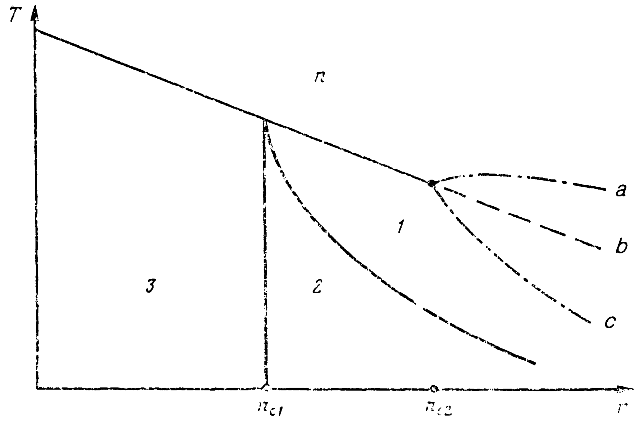

FIG. 3. Phase diagram of a superconductor described by the model of a functional of Eq. (8): here, n is the normal phase ($|\psi|^2=0$); *1*) superconducting phase ($|\psi|^2=|\psi^2|\neq 0$); *2*) magnetic superconducting phase ($0<|\psi^2|<|\psi|^2$); *3*) magnetic superconducting phase ($|\psi^2|=0, |\psi|^2\neq 0$). The continuous curves represent second-order phase transitions; the dashed line represents first-order phase transitions (line b); the chain curves represent the lines of absolute instability of the superconducting (a) and normal (c) phases.

The superconducting classes were found in Ref. 4. We use the results given there and establish the necessary correspondence between the superconducting phases and the relative classes. Using the condition $|\psi_x|^2=|\psi_y|^2$, we select the order parameter for the phase *1* (Fig. 2) in the form $\psi=(\exp[i\pi/4], \exp[i\pi/4])$; the corresponding classes $D_2'(C_2)$ (Ref. 4) which must be transformed to

$$\tilde{D}_2'(C_2) = e^{i\pi/4}D_2'(C_2)e^{-i\pi/4},$$

since the order parameter of the phase *1* is selected—for the sake of convenience—in the complex form. In phase *3* the corresponding class is $D_4(E)$ with the order parameter $\psi=(1,i)$. However, in the case of phase *2* there is a spontaneous breaking of the $D_4(E)$ and $\tilde{D}_2'(C_2)$ symmetry, yielding a certain class X which is a subgroup of these two groups. We shall write down explicitly the elements of the class X:

$$X=\{E, C_2e^{i\pi}, e^{i\pi/2}RU_2'^{(1)}, e^{-i\pi/2}RU_2'^{(2)}\}, \tag{18}$$

as well as the expression for the order parameters of the phases *1*, *2*, and *3*:

$$\psi^2=(e^{i\varphi}, ie^{-i\varphi}),$$

transforming to ψ_3 when $\varphi=0$ and to φ_1 when $\varphi=\pi/4$. The $U_2'^{(1)}$ and $U_2'^{(2)}$ axes are bisectors of the angles XOY and $-XOY$, respectively. Therefore, we obtain the phase diagram shown in Fig. 4. We now give the expressions for the corresponding order parameters:

$$\begin{aligned}\psi_1&=e^{i\pi/4}(1,1),\\ \psi_2&=(e^{i\varphi}, ie^{-i\varphi}),\\ \psi_3&=(1,i).\end{aligned} \tag{19}$$

Naturally the groups $D_4(E)$ and $\tilde{D}_2'(C_2)$ are subgroups of the complete combined group $D_4\times U(1)\times R$ and the group X is a subgroup of all three groups.

The order parameter ψ is truly complex, because the vectors Re ψ_2 and Im ψ_2 are noncollinear, which means that the phase *2* is magnetic. Electrons in this phase are paired in a nontrivial manner: The pairs now have a nonzero orbital momentum, but this is not the state with a fixed momentum

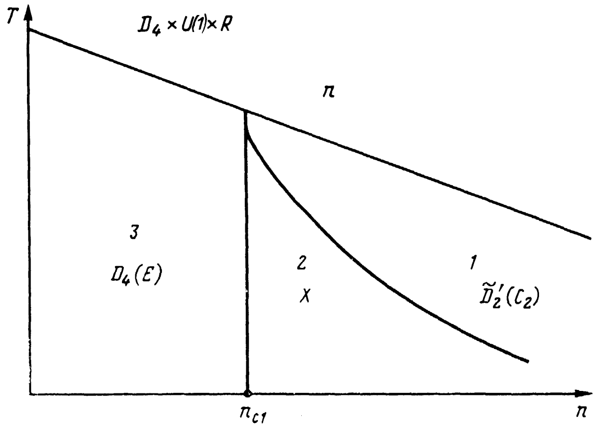

FIG. 4. Correspondence between the phases and the superconducting classes of the system.

since the symmetry group of the crystal does not have a complete rotational symmetry. This state is a sum of different spherical harmonics corresponding to different values of the orbital momentum.

5. CONCLUSIONS

The situation considered above is one in which nontrivial pairing occurs in a superconductor and the order parameter transforms in accordance with the two-dimensional representation of the symmetry group. The anisotropic interaction with impurities stimulates a second-order phase transition from a nonmagnetic superconducting phase to a magnetic one when the impurity concentration exceeds a certain critical value n_{c1}.

The question now arises about the validity of this model associated mainly with the selection of the impurity term in the functional (1) in the form given by Eq. (2), and relating to the procedure of averaging over the impurities. Moreover, it may happen that the constants of the interaction with the impurities λ_1 and λ_2 are equal and then the model is invalid.

We consider these problems now and identify a physical situation in which the values of λ_1 and λ_2 are not only not equal to one another, but also one of them vanishes identically.

The most general form of the impurity term in the Ginzburg–Landau theory was obtained in Ref. 6. When applied to the present model, this term becomes

$$F_{\rm imp} = C\int\int \Delta(\mathbf{r})[\operatorname{Im}\hat{T}(\mathbf{r},\mathbf{r}')]\Delta^*(\mathbf{r}')\,d\mathbf{r}\,d\mathbf{r}', \tag{20}$$

where C is a constant and the expression $\operatorname{Im}\hat{T}(\mathbf{r},\mathbf{r}')$ represents the imaginary part of the exact scattering matrix considered in the coordinate representation.

We assume that the impurity is located at some point $\mathbf{r}_0$. Then, calculation of $\operatorname{Im}\hat{T}(\mathbf{r},\mathbf{r}')$ to second order in the interaction with the impurity $[V(\mathbf{r}) = V\delta(\mathbf{r}-\mathbf{r}_0)]$ yields the expression

$$\operatorname{Im}\hat{T}(\mathbf{r},\mathbf{r}') = V\delta(\mathbf{r}-\mathbf{r}_0)\delta(\mathbf{r}-\mathbf{r}'). \tag{21}$$

Substituting Eq. (21) into (20), we obtain an impurity term in the form

$$F_{\rm imp} = \psi_i^* U_{ik}\psi_k,$$
$$U_{ik} = CV\Phi_i^*(\mathbf{r}_0)\Phi_k(\mathbf{r}_0). \tag{22}$$

Here Φ_1 and Φ_2 are the basis functions of the irreducible representation which governs the transformation of the order parameter ψ. It should be pointed out that the matrix U_{ik} is degenerate, so that

$$\lambda_1=0,\quad \lambda_2=\operatorname{Tr} U_{ik}=CV[\Phi_1^*(\mathbf{r}_0)\Phi_1(\mathbf{r}_0)+\Phi_2^*(\mathbf{r}_0)\Phi_2(\mathbf{r}_0)]. \tag{23}$$

The value of λ_2 is generally nonzero.

We can easily understand also the meaning of the averaging over the "orientations" of an impurity for fixed values of λ_1 and λ_2. It simply means averaging over the position of an impurity in the unit cell on condition that the impurity is located on an equipotential surface inside the well created by the crystal field.

The symmetry analysis in Sec. 4 shows that this model applies to real superconductors with a symmetry group that allows nontrivial pairing in a superconductor with a multicomponent order parameter and the appearance of terms of the $\beta_2|\psi^2|^2$ type in the Ginzburg–Landau functional. However, we have to determine the physical conditions under which this model is acceptable and identify those relationships between the constants of the theory which ensure that the splitting of the phase transition occurs before suppression of the superconductivity. This happens if the following inequality is satisfied:

$$2\alpha/(\lambda_1+\lambda_2) > 16\beta_2 T/(\lambda_1-\lambda_2)^2. \tag{24}$$

On the left we have the expression for the impurity concentration n_{c3} at which the superconductivity is suppressed and on the right we have the impurity concentration n_{c1} at which the transition splits.

If we use the expression for the constants given by $\lambda_i \sim \sigma_{\rm tr} k_F^2/16\pi$, for example that derived in Ref. 7, we obtain the following expression for the quantities n_{c1} and n_{c3}:

$$n_{c1} \sim (16\pi/\sigma_{tr}k_F^2)^2 N_0 T_c \varepsilon^2,$$
$$n_{c3} \sim (16\pi/\sigma_{tr}k_F^2) N_0 T_c. \tag{25}$$

Here, N_0 is the density of states at the Fermi level; $\sigma_{\rm tr}$ is the transport cross section of an impurity, and the coefficient ε represents the smallness of the coefficient β_2 associated with, for example, the fact that the crystal symmetry is tetragonal but differs slightly from the cubic (the degree of compression of the cube is proportional to ε). It then follows from the expressions in Eq. (5) that this phase transition behavior occurs in two important cases: for $\sigma_{\rm tr} k_F^2 \sim 16\pi \gg 1$, i.e., when the impurity has "giant" dimensions, and for $\varepsilon \ll 1$ (in the slightly noncubic case, which is more realistic than the former condition).

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APPENDIX

We shall show that the coefficient in front of $|\psi^2|^2$ in the expression for the effective free energy is the same irrespective of whether it is calculated by averaging the partition function and then taking the logarithm of the average or by averaging the logarithm of the partition function. Moreover, the free energy is an increasing function in the limit $|\psi^2|\to\infty$. Therefore, if the free energy falls at low values of $|\psi^2|$, then at some value $|\psi^2|\neq 0$, it has a minimum when considered as a function of $|\psi^2|$, which means that the phase diagram obtained above applies in this case.

We now calculate the impurity term in the effective free energy by the two methods mentioned above.

1. We assume that $f_1 = -T\ln\langle Z\rangle_v$, where Z is the partition function and the angular brackets denote averaging over the impurities. We then have

$$Z=\left\langle \exp\left(\sum_a \psi_a^{i*}V_a^{ik}\psi_a^k\right)\right\rangle_{F_0}.$$

Here and below the index a labels the impurities over which the summation is carried out and the expression $\langle A\rangle_{F_0}$ means that

$$\langle A(\psi,\psi^*)\rangle_{F_0}=\int D\psi^* D\psi\exp(-F_0)A(\psi,\psi^*),$$

i.e., it implies a functional integral of $A(\psi,\psi^*)$ with respect to ψ and the use of the function F_0 which is the Ginzburg–Landau functional of a pure superconductor. In the exponential function and the logarithm to within fourth-order terms in ψ, we obtain

$$f_1=-T\left\langle\left\langle \sum_a(\psi_a^* V_a\psi_a)\right\rangle_{F_0}\right\rangle_V -\frac{T}{2}\left\langle\left\langle\sum_{a,b}(\psi_a^*V_a\psi_a)(\psi_b^*V_b\psi_b)\right\rangle_{F_0}\right\rangle_V +\frac{T}{2}\left(\left\langle\left\langle\sum_a(\psi_a^*V_a\psi_a)\right\rangle_{F_0}\right\rangle_V\right)^2+o(|\psi|^4).$$

Here,

$$(\psi_a^*V_a\psi_a)=\sum_{ik}\psi_a^{i*}V_a^{ik}\psi_a^k,$$

$$V^{ik}=U^{ik}-\frac{\lambda_1+\lambda_2}{2}\delta^{ik},$$

and the matrix U is defined above. Therefore, we have

$$f_1=-\frac{T}{2}\left\langle\left\langle\sum_{a,b}(\psi_a^*V_a\psi_a)(\psi_b^*V_b\psi_b)\right\rangle_{F_0}\right\rangle_V+o(|\psi|^4).$$

2. In the second case, we obtain

$$f_2=-T\langle\ln Z\rangle_V=-T\left\langle\left\langle\sum_a(\psi_a^*V_a\psi_a)\right\rangle_{F_0}\right\rangle_V -\frac{T}{2}\left\langle\left\langle\sum_{a,b}(\psi_a^*V_a\psi_a)(\psi_b^*V_b\psi_b)\right\rangle_{F_0}\right\rangle_V +\frac{T}{2}\left\langle\left\langle\sum_a(\psi_a^*V_a\psi_a)\right\rangle_{F_0}\times\left\langle\sum_b(\psi_b^*V_b\psi_b)\right\rangle_{F_0}\right\rangle_V+o(|\psi|^4).$$

The last term in the above expressions depends only on $|\psi|^2$ and not on $|\psi^2|$, whereas the first term is identical with f_1 apart from the correction $o(|\psi|^4)$, which means that the coefficients in front of $|\psi^2|^2$ are identical in the expressions for f_1 and f_2.

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