Curie law in diamagnetic susceptibility

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(Submitted 8 April 1978)
Pis'ma Zh. Eksp. Teor. Fiz. **27**, No. 11, 615–618 (5 June 1978)

The magnetic susceptibility of systems that are unstable to electron-hole pairing is investigated in the self-consistent-field approximation. It is shown that the response of the system is diamagnetic, and that its temperature dependence obeys the Curie law in the case of an imaginary order parameter.

PACS numbers: 75.20.−g, 75.30.Cr

1. The purpose of this paper is to determine the directions in which searches can be made for systems with superdiamagnetic properties. It will be shown that for substances whose temperature of the transition to the current state $T_{\rm Im}$ is higher than the temperature of the transition to the ferroelectric state $T_{\rm Re}$, the diamagnetic susceptibility χ' obeys the Curie-Weiss law. For substances having $T_{\rm Re}$ larger than $T_{\rm Im}$, the diamagnetic susceptibility has a peak near $T_{\rm Re}$. This peak should be observable if $T_{\rm Re}$ and $T_{\rm Im}$ have close values, and this is possible for ferroelectrics in which the ion contribution to the polarizability is not too large compared with the electron contribu-

tion. Introduction of impurities in such ferroelectrics should[1] change the ratio of the temperatures T_{Re} and T_{Im} in favor of the latter. (Of course, the phase transitions in "pure" ferroelectrics should be close to second-order transition.)

It is important to note that the anomalous diamagnetic response in our system takes place in the self-consistent approximation. That is to say, the magnetic field in a two-band mode with coinciding band extrema and with allowed interband dipole transitions induces a current-dependent order parameter. These distinguish our anomalous-diamagnetism mechanism in principle from the fluctuation mechanism, with which is connected the Curie law in χ' of a superconductor or of a Peierls dielectric at $T \geqslant T_c$. In particular, the temperature interval in which the anomalies of χ' should be observed turns out to be much wider.

2. Assume that in the non-reconstructed phase the system is described by the two-band model of an isotropic "semimetal" and $\epsilon_1(\mathbf{p}) = -\epsilon_2(\mathbf{p})$ (1 and 2 are the band indices). We examine the response of such a system, in which interband dipole transitions are allowed, to stationary electric and magnetic fields that vary slowly in space. We carry out the investigation with the aid of temperature Green's functions, for which we can obtain the following system of equations:

$$\left[i\omega - \frac{1}{2m}\left(\frac{\vec{\nabla}}{i} - eA\right)^2 + \epsilon_F\right] G_{11}(\mathbf{r},\mathbf{r}')$$

$$+ \left[\frac{\mathbf{P}^*}{m}\left(\frac{\vec{\nabla}}{i} - e\mathbf{A}\right) + \mathbf{dE} + \lambda(\mathbf{r})\right] G_{21}(\mathbf{r};\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}'),$$

$$\left[i\omega + \frac{1}{2m}\left(\frac{\vec{\nabla}}{i} - e\mathbf{A}\right)^2 - \epsilon_F\right] G_{21}(\mathbf{r},\mathbf{r}') + \left[\frac{\mathbf{P}}{m}\left(\frac{\vec{\nabla}}{i} - e\mathbf{A}\right) + \mathbf{dE}\right.$$

$$\left. + \lambda^*(\mathbf{r})\right] G_{11}(\mathbf{r},\mathbf{r}') = 0. \tag{1}$$

Here $\mathbf{A}$ is the vector potential describing the magnetic field, $\mathbf{E}$ is the electric-field intensity vector, $\mathbf{d}$ is the matrix element of the dipole transition between bands 1 and 2, $\mathbf{P}$ is the interband matrix element of the momentum operator, $\epsilon_F = P_F^2/2m$, P_F is the Fermi momentum, $\lambda(\mathbf{r})$ is the order parameter expressed in terms of the anomalous mean values of $\Delta(\mathbf{r})$ introduced in[1] and characterizing singlet electron-hole pairing:

$$\lambda(r) = g_{\mathrm{Re}}\Delta_{\mathrm{Re}}(\mathbf{r}) + i g_{\mathrm{Im}}\Delta_{\mathrm{Im}}(\mathbf{r}), \tag{2}$$

g_{Re} and g_{Im} are the coupling constants corresponding to states with real and imaginary order parameters, respectively.

We discuss first a situation wherein there is no magnetic field in a system, and the electric field differs from zero and obviously plays the role of the source of Bose condensates of electron-hole pairs. We assume that the situation $T_{\mathrm{Re}} > T_{\mathrm{Im}}$ is realized, i.e., the system is ferroelectric at $T < T_{\mathrm{Re}}$ (T_{Re} and T_{Im} are respectively the temperatures of the transition to states with real and imaginary order parameters and are connected with the corresponding coupling constants g_{Re} and g_{Im} by known relations.) The electric field **E**, in the approximation linear in **E**, induces in the system only a real order parameter, as follows from (1). By the method used below to derive expression (5) for the diamagnetic susceptibility, we can show that the dielectric constant of the system satisfies the Curie-Weiss law. Near T_{Re}, ϵ diverges like $(T-T_{\mathrm{Re}})^{-1}$, and below T_{Re} the system goes over into the ferroelectric state. In the absence of a magnetic field the state with imaginary order parameter cannot be realized at $T_{\mathrm{Re}}>T_{\mathrm{Im}}$ ($\Delta_{\mathrm{Im}}=0$).

3. We consider now the case when there is no electric field in the system and the magnetic field differs from zero. At $T>\max(T_{\mathrm{Re}},T_{\mathrm{Im}})$ no real order parameter can be realized in the system. Since the vector **P** in (1) is imaginary, we can easily show that the magnetic field serves as a source of a Bose condensate with imaginary order parameter. At $T>\max(T_{\mathrm{Re}},T_{\mathrm{Im}})$ we can confine ourselves to linearization of the system (1) with respect to **A** and $\lambda(\mathbf{r})$. Let **P** be small enough, so that only the first-order correction of the corresponding hybridization term need be taken into account. In this approximation, it follows from (1) and (2) that:

$$g_{\mathrm{Im}}\Delta_{\mathrm{Im}} = -\frac{q^2 v_F^2}{6(\pi T_{\mathrm{Im}})^2}\frac{e}{m}A_q|\mathbf{P}|\frac{T_{\mathrm{Im}}}{T-T_{\mathrm{Im}}}, \tag{3}$$

where v_F is the velocity on the Fermi surface. Thus, the magnetic field **A** actually induces Δ_{Im} in an approximation linear in **A**. The response of the system to the total magnetic field can be obtained with the aid of the expression for the interband component of the current density[1]:

$$\mathbf{j} = \frac{2e}{m}|\mathbf{P}|\Delta_{\mathrm{Im}}. \tag{4}$$

In the determination of the susceptibility we shall not take into account the usual diamagnetic current due to the intraband processes,[2] since it contains no singularities whatever in the region of the transition temperature.

We consider the case $T_{\mathrm{Im}}>T_{\mathrm{Re}}$. It can be realized, first, at $g_{\mathrm{Im}}<g_{\mathrm{Re}}$. It is, however, quite difficult to obtain this relation between the coupling constants. A more realistic situation is $g_{\mathrm{Im}}>g_{\mathrm{Re}}$, but since the scattering by the impurities suppresses the temperature of the ferroelectric transition more strongly than the temperature of the transition to the current state, T_{Im} may turn out to be larger than T_{Re}.[1] Our description then pertains to a system with spontaneous currents at $T>T_{\mathrm{Im}}$. The function of the response of χ' to the total field is anisotropic, as follows from (3) and (4). Substituting Δ_{Im} from (3) and (4), we obtain at $\mathbf{A}\|\mathbf{P}$:

$$\chi' = \frac{2}{g_{\mathrm{Im}}N(0)}\frac{T_{\mathrm{Im}}}{T-T_{\mathrm{Im}}}\frac{|\mathbf{P}|^2 v_F^2}{(\pi T_{\mathrm{Im}})^2}\chi_L, \tag{5}$$

where χ_L is the Landau diamagnetic susceptibility. The response χ of the system to the external field is[3] $\chi=\chi'/(1-4\pi\chi')$. It is seen that as $T\to T_{\mathrm{Im}}$ the susceptibility tends to the ideal value $1/4\pi$, thus confirming the presence of "superdiamagnetism" in a state with spontaneous current $(T<T_{\rm Im})$.[3]

In the case $T_{\rm Im}<T_{\rm Re}$ the quantity $|\chi'|$ does not become infinite anywhere. Expression (5) is valid at $T\geqslant T_{\rm Re}$. At $T<T_{\rm Re}$ there appears in the system, besides the imaginary order parameter induced by the magnetic field, a real parameter connected with the transition to the ferroelectric state. From the system (1) we can deduce that $|\chi'|$ has in this case a maximum near $T_{\rm Re}$ and falls off with decreasing T, owing to the increase of $\Delta_{\rm Re}$.

It is of interest to note in this connection the results of[4], where an increase of the diamagnetic susceptibility of SnTe is observed near the ferroelectric-transition temperature.

It can be shown that at $T<T_{\rm Im}$ (in the current state) the modulus of the differential susceptibility χ' decreases with changing $|T-T_{\rm Im}|$ twice as fast as at $T>T_{\rm Im}$ (the "rule of two").

The authors thank V.L. Ginzburg for a useful discussion.

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