On the structure of the superdiamagnetic state

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The general form of gauge-invariant functional of free energy, valid for the ferromagnetic and superdiamagnetic cases, is shown. Using an example of the systems with electron-hole pairing, it is shown that the superdiamagnetic state can occur only as an inhomogeneous state. In particular, it can occur in the electron-type ferroelectrics in the region of domain walls.

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1. In this paper we show that a systematic account of the interband interactions in systems with electron-hole pairing reduces the homogeneous current[1] to zero. On the other hand, as was shown in Ref. 2, the response of the system $\chi' = M/B$ to the total magnetic field "B" (M is magnetization) goes to $-\infty$ at a temperature $T_{\rm Im}$, which corresponds to the onset of an imaginary parameter of order $\Delta_{\rm Im}$. Therefore, according to the expression $H = B/\mu$ the magnetic field H is produced spontaneously at $T = T_{\rm Im}$ even in the absence of magnetic induction B ($\mu = 0$ at $T = T_{\rm Im}$).

The ferromagnetic state (spontaneous B), however, occurs when the response of χ to H ($\chi = M/H$) goes to $+\infty$, i.e., $\mu = +\infty$, and hence $B = \mu H$ is finite even at $H = 0$. To show in the general form that orbital ordering is possible with respect to H (superdiamagnetism) and with respect to B (ferromagnetism), we shall use here the free-energy functional without solving it, in which the nonequilibrium generalized momentum $\mathbf{p}$ plays the part of the order parameter:

$$F(p) = -\frac{1}{2\pi}\mathbf{p}^2 - \mathbf{p}\mathbf{A} + n\frac{\mathbf{A}^2}{2}, \qquad \mathrm{rot}\,\mathbf{A} = \mathbf{B}. \tag{1}$$

Here π is the total polarization operator of the system, whose Fourier components have generalized-momentum operators at the vertices

$$\hat{p}_q = \frac{1}{2i}\left(\nabla e^{i\mathbf{q}\mathbf{r}} + e^{i\mathbf{q}\mathbf{r}}\nabla\right), \qquad e = \hbar = c = m = 1,$$

where h is the particle density of the system. Variation of functional (1) with respect to $\mathbf{A}$ gives the current, and the equilibrium value $\mathbf{p} = \mathbf{p}_0$ is determined by minimizing (1) with respect to $\mathbf{p}$.

Taking this into account, we can easily see that the response of the system to the field $\mathbf{A}$ is

$$\mathbf{j}_\mathbf{q} = -(\pi_\mathbf{q} + n)\mathbf{A}_\mathbf{q}. \tag{2}$$

In accordance with the condition for gauge invariance, $\pi_\mathbf{q} = -n$ for all longitudinal $\mathbf{q}(\mathbf{q}\|\mathbf{A})$. Therefore, when the transverse momenta $\mathbf{q}_\perp$ are small, it appears that $\mathbf{j}_\mathbf{q}$

$\sim\chi' \mathbf{q}_\perp^2 \mathbf{A}_\mathbf{q}$ and evidently χ' may diverge in the paramagnetic and diamagnetic directions, depending on the sign of the sum $(\pi_\mathbf{q} + n)$. It is important to note that both types of divergence are described by the same gauge-invariant functional, and the standard functional of the ferromagnet $F = \alpha \mathbf{M}^2 - \mathbf{MH}$ is a special case of functional (1) in which the third (diamagnetic) term $n(\mathbf{A}^2/2)$ was dropped.

2. The electron-hole pairing with an imaginary order parameter is unique in that the Bloch representation must take into account the interband-interaction terms of the type $g_3 a_1^+ a_1 a_1^+ a_2$, which without leading to a logarithmic singularity, are the sources of $\Delta_{\rm Im}$ when the interband matrix element P_{12} is nonvanishing. Although the indicated terms are missing in the Cohn–Lattinger representation, it has a hybridization of the type $\Sigma_\mathbf{k} (\mathbf{p}_{12} \times \mathbf{k}) a_{1\mathbf{k}}^+ a_{2\mathbf{k}}$, which we take into account simultaneously with $\Delta_{\rm Im}$. As a result, the equations for the Green's function G_{ij} $(\mathbf{r},\mathbf{r}')$ $(i,j = 1,2$ are the band indices) have the form

$$(\omega - \hat{\epsilon}_1) G_{11}(\mathbf{r}, \mathbf{r}') + \left(\mathbf{P}\frac{\nabla}{i}\right) G_{21}(\mathbf{r}, \mathbf{r}') + \int d\mathbf{r}'' \Delta_{\rm Im}(\mathbf{r}, \mathbf{r}'') G_{21}(\mathbf{r}'', \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'), \tag{3}$$

$$(\omega - \hat{\epsilon}_2) G_{21}(\mathbf{r}, \mathbf{r}') + \left(\mathbf{P}^* \frac{\vec{\nabla}}{i}\right) G_{11}(\mathbf{r}, \mathbf{r}') + \int d\mathbf{r}'' \Delta_{\rm Im}^*(\mathbf{r}, \mathbf{r}'') G_{11}(\mathbf{r}'', \mathbf{r}') = 0.$$

If we assume that the parameter $\Delta_{\rm Im}$ (r,r') in Eq. (3) depends only on the difference $(r - r')$, then the spectrum for single-particle excitations $\tilde{\epsilon}_{1,2}(k)$ $= \pm \sqrt{(\mathbf{v}_F \mathbf{k})^2 + (\Delta + \mathbf{P} \times \mathbf{k})^2}$, i.e., it is asymmetric with respect to $\mathbf{k}$ when $\Delta_{\rm Im}(\mathbf{k})$ is even.

Determining in this case the G_{ij} functions from Eq. (3) and substituting them in the usual expression for the current

$$\mathbf{j}(\mathbf{r}) = T \sum_n \left\{ \frac{ie}{m} (\vec{\nabla}_{\mathbf{r}'} - \vec{\nabla}_\mathbf{r}) [G_{11}(\mathbf{r}, \mathbf{r}') - G_{22}(\mathbf{r}, \mathbf{r}')] + 2e[\mathbf{P}_{12} G_{21}(\mathbf{r}, \mathbf{r}') + \mathbf{P}_{21} G_{12}(\mathbf{r}, \mathbf{r}')] \right\}_{\mathbf{r}' \to \mathbf{r}} \tag{4}$$

we can see that the homogeneous current vanishes, since the interband current[1] is exactly compensated for by the intraband current, which is produced because of the aforementioned asymmetry of the single-particle spectrum.

Note that Batyev[3] attempted to eliminate the homogeneous current by redefining the current operator without allowance for the interaction of the type $g_3 a_1^+ a_1 a_1^+ a_2$ or for the hybridization. In fact, he used the time-dependent, first-order equation in the problem involving the exchange potential, which is incorrect.[4,5] Moreover, even if such an assumption were valid, the additional term in the current[3] remains finite only in the case of an infinite-range potential. We emphasize that an incorrect determination of the interband current[3] precludes its compensation by the intraband current, i.e., the homogeneous current is finite, which contradicts the Bloch theorem.

3. If the dependence of $\Delta_{\rm Im}$ on the center-of-mass coordinate $\mathbf{R} = (\mathbf{r} + \mathbf{r}')/2$ of the electron-hole pair is retained in Eqs. (3) and they are expanded in Δ ($\mathbf{R}$) and $\mathbf{P}_{12} \times \overset{\Delta}{\mathbf{k}}$ near $T_{\rm Im}$,[6] then we obtain the following expression for the current after averaging over the unit cell:

$$\mathbf{j}(\mathbf{R}) = \frac{7\xi(3)n}{8(\pi T_{\rm Im})^2} [P_{12}\vec{\nabla}_{\mathbf{R}}^2 - \vec{\nabla}_{\mathbf{R}}(\mathbf{P}\nabla_{\mathbf{R}})]\,\Delta(\mathbf{R}). \tag{5}$$

Note that expression (5) satisfies the condition for the transverse current $\operatorname{div}\mathbf{j} = 0$. In the two-band scheme $\operatorname{div}\mathbf{j} \sim u_1^* u_2$ vanishes after integration over the unit cell because of the orthogonality condition (u_1 and u_2 are Bloch modulation factors of bands 1 and 2).

Without restricting ourselves to the finite number of bands, we can obtain the following expressions for div j:

$$\operatorname{div}\mathbf{j} = \frac{e}{i}\int V(\mathbf{r}, \mathbf{r}'')\,\rho(\mathbf{r}'', \mathbf{r})\,d\mathbf{r}'' + \text{c.c.} \tag{6}$$

where

$$V(\mathbf{r}, \mathbf{r}'') = V(|\mathbf{r} - \mathbf{r}''|)\,\rho(\mathbf{r}, \mathbf{r}'')$$

$\rho(\mathbf{r},\mathbf{r}'')$ is the complete density matrix, which takes into account all the bands and $V(|\mathbf{r} - \mathbf{r}''|)$ is the exchange-interaction potential. Using the Hermitian density matrix, we can see that the condition $\operatorname{div}\mathbf{j} = 0$, allowing for all the bands, is fulfilled locally.

4. As seen in Eq. (5), the current in the ground (superdiamagnetic) state is nonvanishing if the minimum of the free energy corresponds to the inhomogeneous parameter $\Delta_{\rm Im}$. As is well known, the inhomogeneous state occurs when the Fermi surfaces of bands 1 and 2 are not completely congruent, regardless of the phase of the parameter Δ.[7] The coefficient γ_1 of expansion of the free energy F with respect to the order parameter in the $\gamma_1(\partial\Delta/\partial\mathbf{R})^2$ term or the $\gamma_1 q^2\Delta^2$ term changes its sign at a higher temperature than the coefficient α in the $\alpha\Delta^2$ term. A peculiarity of states with $\Delta_{\rm Im}$ is the anisotropy of the coefficient γ_1. Note that the true order parameter in this case is $\mathbf{q}\Delta$, rather than Δ, which, if we take into account expression (5) and the Maxwell equation $\operatorname{curl}\mathbf{H} = 4\pi\mathbf{j}$, corresponds to ordering in the magnetic field $\mathbf{H}$.

The asymmetry of the single-particle spectrum indicated above, after substituting $-\mathbf{k}$ for $\mathbf{k}$ in the case of homogeneous $\Delta_{\rm Im}$, leads to an inhomogeneous state even when the Fermi surfaces are totally congruent. In this case $q_{\rm opt}^2 \sim \Delta_{\rm Im}$, i.e., $j \sim \Delta_{\rm Im}^2$.

5. The assertion that the interband interactions specify the phase of the order parameter is valid only in the homogeneous case. Because an invariant of the type

$$\mathbf{P}_{12}\Delta_{\rm Im}(\Delta_{\rm Re}\operatorname{grad}\Delta_{\rm Im} - \Delta_{\rm Im}\operatorname{grad}\Delta_{\rm Re}) \tag{7}$$

exists in the free energy, the phase of the parameter Δ is a function of the coordinates. Such a state can be the ground state in the n-type ferroelectric materials[8] in which the polarization is proportional to $\Delta_{\rm Re}$. The maximum current occurs in the region of the domain wall in which the phase passes smoothly through $\pi/2$.

It follows from the above discussion that the structure of the superdiamagnetic state is highly sensitive to the high-temperature symmetric phase.

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