Macroscopic current states in crystals

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The conditions for the appearance of a spontaneous current in a crystal are investigated. It is shown that there is no uniform current under equilibrium conditions. In the two-band model unstable to dielectric pairing, with allowed dipolar interband transitions, an expression is obtained for an inhomogeneous spontaneous current and satisfies the transversality condition. The current arises in proportion to the symmetric imaginary and antisymmetric real order parameters. The current-state structure in the region of the phase soliton is obtained. It turns out that the magnetic moment of this state is strictly zero.

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INTRODUCTION

The magnetic properties of solids are determined by the spin or by the orbit degrees of freedom. The characteristic scale of the loops of the current connected with the orbital motion is usually of the order of the atomic scale. In principle, however, macroscopic current structures can occur. A model description of these structures was proposed in Refs. 1 and 2 in the form of a phase transition into a state with a spontaneous current. The possibility of a current state with a large-size inhomogeneity was discussed in Ref. 3 in connection with the published reports[4] of the anomalous diamagnetism of CuCl.

The excitonic insulator model proposed in Ref. 5 was used in Refs. 1 and 2 to describe a phase transition into the current state. This model made it possible previously to obtain various phase transitions: structural transformations, metal–insulator transitions,[6] and transitions into a ferroelectric,[7] antiferromagnetic,[8] and ferromagnetic[8] phase. This variety of physical properties that appear in the excitonic-insulator model of the phase transitions is that the electron-hole system has a large number of "degrees of freedom." First is the spin degree of freedom due to the fact that different particles are paired in the exciton condensate, namely electron with a hole (and not an electron with an electron as in the BCS theory). As a result, the spin and coordinate structures of the order parameters are not linked, and this leads, for example, to the possible appearance of a charge density wave (CDW) or a spin density wave (SDW) with a coordinate structure of the order parameter unchanged.

Second, since the wave functions of the particles in the electron and hole bands can have different symmetry, there exists a "symmetry" degree of freedom. The relative symmetry of these bands specifies the form of the single-particle operators for which the interband matrix elements differ from zero. Therefore, if an order parameter connected with the Bose condensation of the excitons appears in the system, the mean values of the physical quantities determined by these operators turn out to differ from zero, i.e., corresponding physical properties appear. It must also be noted that the relative phase difference of the interband matrix elements and of the order parameter determines which of the possible mean values appears. For example, if the extrema of the electron and hole bands do not coincide in momentum space when a singlet order parameter is produced then, as noted by Halperin and Rice,[10] a charge-density wave appears in the system if the parameter is real, and a current-density wave if the parameter is imaginary.

More curious is the situation when the band extrema coincide and dipolar transitions are allowed between them. Then, at a real order parameter, ferroelectric properties appear,[1,7] and in the case of an imaginary one, owing to the relation $v_{12}=i\omega_{12}d_{12}$ (v_{12} and d_{12} are the interband matrix elements of the velocity and of the coordinate, ω_{12} is the frequency of the interband transitions in the unrestructured phase), there should apparently be observed a uniform current.[1] This would be in fact the case if the self-consistency equations were not to impose a strictly defined coordinate dependence of the order parameter. This dependence turns out to be such that the uniform current vanishes,[2] but the existence of inhomogeneous currents remains possible in the presence of a spatially inhomogeneous imaginary order parameter. This question will be studied in more detail in §§1 and 2 of the present article. The question of the onset of ferroelectric properties will also be discussed there with greater rigor than before. In §3 is considered the Landau functional corresponding to transitions into a state with spontaneous current, and the structure of the current state is obtained.

§1. DETERMINATION OF THE CURRENT OPERATOR

There are three known methods[11] of determining the form of the current operator $\hat{\mathbf{j}}$: from the equation of motion for the coordinate operator $\hat{\mathbf{r}}$

$$\dot{\hat{\mathbf{r}}}=i[\hat{H},\hat{\mathbf{r}}],\tag{1}$$

in terms of the continuity equation for the charge-density operator $e\hat{n}(r)$:

$$\partial e\hat{n}(\mathbf{r})/\partial t=i[\hat{H},e\hat{n}(\mathbf{r})]\equiv-\operatorname{div}\hat{\mathbf{j}},\tag{2}$$

and as the variation of the Hamiltonian $\hat{H}$ with respect to the vector potential $\hat{\mathbf{A}}$:

$$\hat{\mathbf{j}}=-c\delta\hat{H}/\delta\hat{\mathbf{A}}.\tag{3}$$

For the usual gauge-invariant local Hamiltonian with an interaction that depends only on the difference of the

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coordinates, and for a quadratic form of the kinetic-energy operator, all three methods lead to the same result:

$$\hat{\mathbf{j}}=\frac{e}{m}\sum_i\hat{\mathbf{p}}_i, \tag{4}$$

where e is the particle charge, m is its mass, $\hat{\mathbf{p}}_i=-i\nabla_i$ is the momentum of the i-th particle. However, if approximate methods are used in the calculation, such as the Hartree-Fock equations or inclusion of only a finite number of bands in the crystal, certain caution must be exercised, even though formula (4) undoubtedly remains in force.

If, for example, a two-band model of the crystal is investigated, then its Hamiltonian $\hat{H}_0$ for noninteracting electron is patently gauge-invariant in the Luttinger-Kohn basis (see, e.g., Ref. 12):

$$\hat{H}_0=\begin{pmatrix} \varepsilon_1\left(\frac{\nabla}{i}-\frac{e}{c}\mathbf{A}\right) & \frac{1}{m}\mathbf{P}\left(\frac{\nabla}{i}-\frac{e}{c}\mathbf{A}\right) \\ \frac{1}{m}\mathbf{P}^*\left(\frac{\nabla}{i}-\frac{e}{c}\mathbf{A}\right) & \varepsilon_2\left(\frac{\nabla}{i}-\frac{e}{c}\mathbf{A}\right) \end{pmatrix}. \tag{5}$$

Here ε_1 and ε_2 are the spectra of the first and second bands, $\mathbf{P}=i\langle\varphi_{2k}|\nabla|\varphi_{ik}\rangle$ is the interband matrix element of the momentum operator, and $\varphi_{ik}=u_i(\mathbf{r})\exp(i\mathbf{k}\cdot\mathbf{r})$ are the wave functions of the Kohn-Luttinger basis, in which the Bloch factors $u_i(\mathbf{r})$ do not depend on the quasimomentum $\mathbf{k}$.

The matrix element $\mathbf{P}$ differs from zero only when the functions u_1 and u_2 are of unlike parity. In this case, however, it might seem that the interband matrix element of the coordinate should also differ from zero, if this element is calculated from the formula $\mathbf{d}=\langle\varphi_{2k}|\mathbf{r}|\varphi_{im}\rangle$. This, however, is not so, and in the Kohn-Luttinger representation the coordinate operator $\hat{\mathbf{r}}$ is always diagonal in the band indices.[1] Indeed, the current operator for the Hamiltonian $\hat{H}_0$ takes in accord with the definition (3) the form

$$\hat{\mathbf{j}}=\begin{pmatrix} -c\frac{\delta\varepsilon_1}{\delta\mathbf{A}} & \frac{e}{m}\mathbf{P} \\ \frac{e}{m}\mathbf{P}^* & -c\frac{\delta\varepsilon_2}{\delta\mathbf{A}} \end{pmatrix}. \tag{6}$$

It is easy to verify by direct calculation that to reconcile this definition of the current with its definition by the equation of motion (1) for the coordinate operator, the latter must be regarded as diagonal:

$$\hat{\mathbf{r}}=\begin{pmatrix} \mathbf{r} & 0 \\ 0 & \mathbf{r} \end{pmatrix}. \tag{7}$$

Moreover, if it is assumed that the matrix $\hat{\mathbf{r}}$ has off-diagonal elements in the Kohn-Luttinger representation, the usual commutation relation between the momentum and coordinate operators is also violated, and the current operator turns out to be non-Hermitian. A similar difficulty arises also when the current is determined with the aid of the continuity equation (2) unless one postulates beforehand that the local-density operator $\hat{n}(\mathbf{r})$ is diagonal in the band indices.

Thus, the gauge-invariant procedure (3) for determining the current operator, as applied to the Hamiltonian (5), makes it possible to determine correctly the forms of the coordinate and density operators. In addition, the Kohn-Luttinger basis turns out to be more convenient than the Bloch diagonal representation if account must be taken of interelectron interaction effects. In this basis (owing to the diagonality of the density operator), the interaction operator is simplest in form. It depends only on the coordinate difference and does not contain interband-scattering processes. This is precisely why the Hamiltonian $\hat{H}_0$ (5), supplemented by an interband interaction Hamiltonian $\hat{H}_{\text{int}}$ of the density-density type:

$$\hat{H}_{int}=\int d\mathbf{r}_1 d\mathbf{r}_2 V(\mathbf{r}_1-\mathbf{r}_2)\psi_1^+(\mathbf{r}_1)\psi_2^+(\mathbf{r}_2)\psi_2(\mathbf{r}_2)\psi_1(\mathbf{r}_1), \tag{8}$$

[$\psi_i(\mathbf{r})$ are the electron annihilation operators in the i-th band at the point $\mathbf{r}$ in the Kohn-Luttinger basis] will be used here to describe the current states. It is convenient to assume for simplicity that the electron dispersion laws are isotropic in both bands and are described by effective masses m^* that are equal in absolute value, i.e., $\varepsilon=\varepsilon_1=-\varepsilon_2=k^2/2m^*-\varepsilon_F$ ($\varepsilon_F>0$). The matrix element P will be assumed small enough ($m^{-1}|\mathbf{P}|k_F<\varepsilon_F$). This allows us to describe the transition into the current state as a transition into the excitonic-insulator phase in a semimetal.[5] This transition is described in standard fashion by introducing a nondiagonal Green's function $G_{12}=-i\langle T\psi_1^+\psi_2\rangle$ that depends on the anomalous mean value of $\Delta(\mathbf{r}_1,\mathbf{r}_2)$ determined from the self-consistency equation:

$$\Delta(\mathbf{r}_1,\mathbf{r}_2)=\int\frac{d\omega}{2\pi i}G_{12}(\mathbf{r}_1,\mathbf{r}_2,\omega)V(\mathbf{r}_1-\mathbf{r}_2). \tag{9}$$

The Green's function G_{12} itself is determined here from the solution of a system of Hartree-Fock equations with a nonlocal potential $\hat{\Delta}$ (9)

$$\begin{aligned}(\omega-\varepsilon)G_{11}&=\left[\frac{1}{im}\mathbf{P}\nabla+\hat{\Delta}\right]G_{21}+\delta(\mathbf{r}_1-\mathbf{r}_2),\\ (\omega+\varepsilon)G_{21}&=\left[\frac{1}{im}\mathbf{P}^*\nabla+\hat{\Delta}^+\right]G_{11}.\end{aligned} \tag{10}$$

In the spatially homogeneous case, the order parameter $\Delta(\mathbf{r}_1,\mathbf{r}_2)$ depends only on the difference between the coordinates. Then, changing to the momentum representation, we easily obtain from (9) and (10)

$$\begin{aligned}G_{11(22)}&=\frac{1}{2}\left\{1(\mp)\frac{\varepsilon(\mathbf{k})}{[\varepsilon^2(\mathbf{k})+|m^{-1}\mathbf{Pk}-\Delta(\mathbf{k})|^2]^{1/2}}\right\}\\ G_{12}=G_{21}^*&=\frac{\Delta(k)-m^{-1}\mathbf{Pk}}{[\varepsilon^2(\mathbf{k})+|m^{-1}\mathbf{Pk}-\Delta(\mathbf{k})|^2]^{1/2}},\\ \Delta(\mathbf{k})&=\int\frac{d^3\mathbf{k}'}{(2\pi)^3}V(\mathbf{k}-\mathbf{k}')\frac{\Delta(\mathbf{k}')-m^{-1}\mathbf{Pk}'}{[\varepsilon^2(\mathbf{k}')+|m^{-1}\mathbf{Pk}'-\Delta(\mathbf{k}')|^2]^{1/2}}.\end{aligned} \tag{11}$$

The uniform spontaneous current obtained in Ref. 1 is cancelled out in the spatially homogeneous case in the following manner. Taking into account the momentum-representation definition of the current operator (6), namely,

$$\hat{\mathbf{j}}=\begin{pmatrix} e\frac{\partial\varepsilon}{\partial\mathbf{k}} & \frac{e}{m}\mathbf{P} \\ \frac{e}{m}\mathbf{P}^* & -e\frac{\partial\varepsilon}{\partial\mathbf{k}} \end{pmatrix}, \tag{12}$$

we can obtain from (10) an expression for the average interband component (connected with $\mathbf{P}$) of the current density:

$$\mathbf{j}_{12}+\mathbf{j}_{21}=\frac{e}{m}\sum_{\mathbf{k}}\mathbf{P}G_{12}+\text{c.c.}=\frac{e}{m}\sum_{\mathbf{k}}\frac{\mathbf{P}\Delta(\mathbf{k})+\mathbf{P}^{*}\Delta^{*}(\mathbf{k})}{[\varepsilon^{2}(\mathbf{k})+|m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})|^{2}]^{1/2}}. \tag{13}$$

Noticing that the interband matrix element of the momentum **P** is pure imaginary, $\mathbf{P}=-\mathbf{P}^{*}$, it follows from (14) that a finite interband current appears when $\Delta(\mathbf{k})$ acquires an imaginary symmetrical component, i.e., when a nontrivial $[\Delta(\mathbf{k})=\Delta(-\mathbf{k})\neq 0]$ solution of the consistency equation appears in (11). This is indeed the result obtained in Ref. 1. Owing to the inconvenience of the diagonal Bloch representation, in which it is difficult to satisfy gauge invariance, the intraband current that cancels out (13) was not obtained in Ref. 1; this current is now

$$\mathbf{j}_{11}+\mathbf{j}_{22}=e\sum_{\mathbf{k}}\frac{\partial\varepsilon}{\partial\mathbf{k}}(G_{11}+G_{22})=e\sum_{\mathbf{k}}\frac{\partial\varepsilon(\mathbf{k})}{\partial\mathbf{k}}\frac{\varepsilon(\mathbf{k})}{[\varepsilon^{2}(\mathbf{k})+|m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})|^{2}]^{1/2}}. \tag{14}$$

Addition of (13) to (14), using the identity

$$\frac{\partial}{\partial\mathbf{k}}\left[\varepsilon^{2}(\mathbf{k})+\left|\frac{1}{m}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})\right|^{2}\right]^{1/2}=\left\{\frac{\partial\varepsilon(\mathbf{k})}{\partial\mathbf{k}}\varepsilon(\mathbf{k})+\frac{1}{m}\mathbf{P}(\Delta(\mathbf{k})-m^{-1}\mathbf{P}\mathbf{k})\right.$$
$$\left.-\frac{\partial\Delta(\mathbf{k})}{\partial\mathbf{k}}\left(\Delta(\mathbf{k})-\frac{1}{m}\mathbf{P}\mathbf{k}\right)\right\}\left[\varepsilon^{2}(\mathbf{k})+\left|\frac{1}{m}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})\right|^{2}\right]^{1/2},$$

which is valid for imaginary Δ, yields the following expression for the total current:

$$\mathbf{j}_{11}+\mathbf{j}_{22}+\mathbf{j}_{12}+\mathbf{j}_{21}=e\sum_{\mathbf{k}}\left\{\frac{\partial}{\partial\mathbf{k}}\left[\varepsilon^{2}(\mathbf{k})+\left|\frac{1}{m}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})\right|^{2}\right]^{1/2}\right.$$
$$\left.+\frac{\partial\Delta(\mathbf{k})}{\partial\mathbf{k}}\frac{m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})}{[\varepsilon^{2}(\mathbf{k})+|m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})|^{2}]^{1/2}}\right\}. \tag{15}$$

The first term of (15) vanishes upon integration with any distribution function that depends only on the energy. To calculate the second we must resort to the self-consistency equation (11). Differentiating it and substituting in (15) in place of $\partial\Delta(\mathbf{k})/\partial\mathbf{k}$, we can verify that

$$\sum_{\mathbf{k}}\frac{\partial\Delta(\mathbf{k})}{\partial\mathbf{k}}\frac{m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})}{[\varepsilon^{2}(\mathbf{k})+|m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})|^{2}]^{1/2}}=-\sum_{\mathbf{k},\mathbf{k}'}\frac{\partial V(\mathbf{k}-\mathbf{k}')}{\partial\mathbf{k}}$$
$$\times\frac{\Delta(\mathbf{k})-m^{-1}\mathbf{P}\mathbf{k}}{[\varepsilon^{2}(\mathbf{k})+|m^{-1}\mathbf{P}\mathbf{k}-\Delta(\mathbf{k})|^{2}]^{1/2}}\frac{\Delta(\mathbf{k}')-m^{-1}\mathbf{P}\mathbf{k}'}{[\varepsilon^{2}(\mathbf{k}')+|m^{-1}\mathbf{P}\mathbf{k}'-\Delta(\mathbf{k}')|^{2}]^{1/2}}\equiv 0 \tag{16}$$

by virtue of the fact that the potential $V(\mathbf{k}-\mathbf{k}')$ is even in $\mathbf{k}-\mathbf{k}'$ [in (16) it suffices to make the substitution $k\rightleftarrows k'$].

It has thus been shown that, first, when the current operator is correctly defined in gauge-invariant form, there is no uniform spontaneous current in the equilibrium system and second, the exchange correction makes no contribution to the average current. The procedure of redefining the current operator via an exchange correction, proposed by Batyev[13] to prove the absence of a uniform spontaneous current, seems therefore doubtful.

One more attempt to annihilate the homogeneous spontaneous current was made by Nozieres and Saint-James.[14] Their model differs from our (5) in that the interband matrix element of the momentum operator in Ref. 14 reverses sign on moving through the Brillouin zone, so that its integral is zero over the entire zone, whereas in (5) it is constant. Thus, the result obtained in Ref. 14 does not hold for the model considered here (and in Ref. 1).

It is of interest to trace how the ferroelectric properties discussed earlier in Refs. 1 and 7 appear in the model with the $\mathbf{k}\cdot\mathbf{P}$ interaction (5) for a real order parameter Δ. The impression at first glance is that since the Hamiltonian has no dipolar matrix elements **d** of the coordinate operator (7), neither spontaneous polarization nor a uniform current appear in the case of a spatially homogeneous order parameter $\Delta=\mathrm{Re}\Delta$. Indeed, the appearance of the anomalous Green's function G_{12} does not lead automatically to the appearance of a mean value of the operator $\hat{\mathbf{r}}$ (7), since the latter is diagonal, and the function G_{11} (11) at $\Delta=\mathrm{Re}\Delta$ is quadratic in Δ, so that it seems that there should be no spontaneous polarization linear in Δ. It will be shown in §2 that a spontaneous polarization linear in $\Delta=\mathrm{Re}\Delta$ does indeed appear, and the seeming paradox is due to an incorrect transition to the limit of homogeneous Δ.

We have considered here a two-band model. A general proof of the absence of uniform spontaneous current in equilibrium systems, is given in the Appendix in the Hartree-Fock approximation. The first method used for the proof is based on the definition (4) of the current and is due to L. V. Keldysh. The second is based on considerations of gauge invariance for Hamiltonians with nonlocal Hartree-Fock potentials. Both require that the density matrix be Hermitian. In the first case the current calculated is that of the particles, and in the second that of the quasiparticles.

§2. DENSITY OF INHOMOGENEOUS CURRENT

The absence of a homogeneous current does not prevent the existence of macroscopic inhomogeneous transverse current, if the parameter Δ (9) itself is macroscopically inhomogeneous. To demonstrate this, it suffices to calculate the currents in an approximation linear in Δ and **P**, assuming that the order parameter Δ is imaginary and varies slowly in space. It is convenient to represent the paraemeter Δ in the form

$$\Delta(\mathbf{r}_1,\mathbf{r}_2)=\sum_{\mathbf{k}}\Delta(\mathbf{k},\mathbf{R})\exp i\mathbf{k}(\mathbf{r}_1-\mathbf{r}_2), \tag{17}$$

where the relative and average coordinates $\mathbf{r}_1-\mathbf{r}_2$ and $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$, respectively, have been introduced. The slow variation of $\Delta(\mathbf{k},\mathbf{R})$ with **R** will in fact be used. It is possible to separate in $\Delta(\mathbf{k},\mathbf{R})$ the symmetrical and antisymmetrical parts Δ^{s} and Δ^{a}, respectively:

$$\Delta(\mathbf{k},\mathbf{R})=\Delta^{s}(\mathbf{k},\mathbf{R})+\frac{\mathbf{k}}{|\mathbf{k}|}\Delta^{a}(\mathbf{k},\mathbf{R}),$$
$$\Delta^{s}(\mathbf{k},\mathbf{R})=\Delta^{s}(-\mathbf{k},\mathbf{R}),\qquad \Delta^{a}(\mathbf{k},\mathbf{R})=-\Delta^{a}(-\mathbf{k},\mathbf{R}). \tag{18}$$

In the spatially inhomogeneous case, $\Delta(\mathbf{k},\mathbf{R})$ is independent of **R** and, as shown in §1, there is likewise no homogeneous spontaneous current. In the inhomogeneous case, the current can be calculated in the approximation linear in Δ and **P**, provided the corresponding Green's functions are known:

$$G_{11}(\mathbf{r}_1,\mathbf{r}_2)=G_{11}^{0}(\mathbf{r}_1-\mathbf{r}_2)-\iiint G_{11}^{0}(\mathbf{r}_1-\mathbf{r}')\frac{1}{im}\mathbf{P}\nabla_{\mathbf{r}'}G_{22}^{0}$$
$$\times(\mathbf{r}'-\mathbf{r}'')\Delta^{*}(\mathbf{r}'',\mathbf{r}''')G_{11}^{0}(\mathbf{r}'''-\mathbf{r}_2)\,d\mathbf{r}'\,d\mathbf{r}''\,d\mathbf{r}'''-\iiint G_{11}^{0}(\mathbf{r}_1-\mathbf{r}')\Delta(\mathbf{r}',\mathbf{r}'')$$
$$\times G_{22}^{0}(\mathbf{r}''-\mathbf{r}''')\frac{1}{im}\mathbf{P}\nabla_{\mathbf{r}'''}G_{11}^{0}(\mathbf{r}'''-\mathbf{r}_2)\,d\mathbf{r}'\,d\mathbf{r}''\,d\mathbf{r}''',$$

$$G_{12}(\mathbf{r}_1,\mathbf{r}_2)=\int G_{11}{}^0(\mathbf{r}_1-\mathbf{r}')\frac{1}{im}\mathbf{P}\nabla_{\mathbf{r}'}G_{22}{}^0(\mathbf{r}'-\mathbf{r}_2)\,d\mathbf{r}'$$
$$-\iint G_{11}{}^0(\mathbf{r}_1-\mathbf{r}')\Delta(\mathbf{r}',\mathbf{r}'')G_{22}{}^0(\mathbf{r}''-\mathbf{r}_2)\,d\mathbf{r}'\,d\mathbf{r}''. \qquad (19)$$

Substituting in (19) the expression for the order parameter $\Delta(\mathbf{k},\mathbf{R})$ (17) and calculating with the aid of (19) the average value j of the current operators (6), we can obtain

$$\mathbf{j}=\mathbf{j}_a+\mathbf{j}_s,$$
$$\mathbf{j}_s=\frac{e}{m}L\operatorname{Re}\operatorname{rot}[\mathbf{P}\operatorname{grad}\Delta^s(\mathbf{R})], \qquad (20)$$
$$\mathbf{j}_a=\frac{e}{m}Q\operatorname{Im}\operatorname{rot}[\mathbf{P}\Delta^a(\mathbf{R})].$$

Here

$$L=-\frac{1}{6}\frac{k_F{}^3}{m^*}\frac{T}{\pi}\sum_{n\geqslant0}\frac{\omega_n(\omega_n{}^2-3\mu^2)}{(\mu^2+\omega_n{}^2)^3},$$
$$Q=\frac{2}{3}k_F{}^2\frac{T}{\pi}\sum_{n\geqslant0}\mu\frac{\omega_n}{(\mu^2+\omega_n{}^2)^2},\quad \omega_n=(2n+1)\pi T. \qquad (21)$$

These formulas were obtained assuming a weak dependence of $\Delta(\mathbf{k},\mathbf{R})$ on k. This is justified for the case of a short-range potential $V(\mathbf{r}_1-\mathbf{r}_2)$ (8), and account was taken here only of the lower derivatives of $\Delta(\mathbf{k},\mathbf{R})$ with respect to the coordinate $\mathbf{R}$ (large inhomogeneity scale). It follows from (20) that the spontaneous current is patently transverse, so that the continuity equation is satisfied explicitly. Furthermore, this current appears only in the case of an order parameter that is inhomogeneous in space, its amplitude increases with increasing degree of inhomogeneity, and the scale of the current turns out to be macroscopic and is determined by the inhomogeneity scale, and the inhomogeneity must have a transverse character if a spontaneous current is to exist. In other words, the change of the amplitude $\Delta(\mathbf{R})$ should be in a direction perpendicular to $\mathbf{P}$, in which case the current is directed along the $\mathbf{P}$ anisotropy axis. The expressions obtained here for the spontaneous current (20) show that it consists of two components, $\mathbf{j}_s$ and $\mathbf{j}_a$. The first is connected with the appearance of an imaginary symmetrical order parameter Δ_s and exists at all doping levels. The second appears only in accord with the degree of doping μ (the noncongruence of the electron and hole Fermi surface) in the presence of an antisymmetrical real part $\Delta^a(\mathbf{R})$ of the order parameter (18). Principal attention will therefore be paid hereafter to the symmetrical current component $\mathbf{j}_s$:

It seems useful to obtain results similar to (20) in the semiconductor model of an excitonic insulator. This makes it possible to make these results more lucid, since the calculations are technically simpler. In addition, the semiconductor model will be used here to explain the paradox described in §1 concerning the appearance of ferroelectric properties in the system at a real value of the symmetrical part of the order parameter Δ. The Hamiltonian of the semiconductor model does not differ in any way from the Hamiltonian (5), (8) of the semimetal model, and it is assumed only that the electron (ε_1) and hole (ε_2) bands do not overlap and are separated by an energy gap ε_g. The equations for the Green's function (19) and the self-consistency equations (9) retain their form in the low-density limit $(\varepsilon_{ex}-\varepsilon_g)/\varepsilon_g\ll1$ (ε_{ex} is the exciton binding energy), and expressions (19) likewise remain in force. In the momentum representation with respect to the relative and average coordinates $(\mathbf{r}_1-\mathbf{r}_2)$ and $\mathbf{R}$ it is necessary to replace (17) by

$$\Delta(\mathbf{k},\mathbf{q})=\int\Delta(\mathbf{k},\mathbf{R})e^{-i\mathbf{q}\mathbf{R}}\,d^3\mathbf{R}. \qquad (22)$$

Relations (19) at the temperature $T=0$ then take the form

$$G_{11}(\mathbf{k},\mathbf{k}+\mathbf{q})=G_1{}^0(\mathbf{k})\mathbf{P}\mathbf{k}G_2{}^0(\mathbf{k})\Delta_q{}^*(\mathbf{k})G_1{}^0(\mathbf{k}+\mathbf{q})$$
$$+G_1{}^0(\mathbf{k})\Delta_{\mathbf{q}}(\mathbf{k})G_2{}^0(\mathbf{k}+\mathbf{q})\mathbf{P}(\mathbf{k}+\mathbf{q})G_1{}^0(\mathbf{k}+\mathbf{q}),$$
$$G_{12}(\mathbf{k},\mathbf{k}+\mathbf{q})=G_1{}^0(\mathbf{k})\Delta_{\mathbf{q}}(\mathbf{k})G_2{}^0(\mathbf{k}+\mathbf{q})+\delta(\mathbf{q})G_1{}^0(\mathbf{k})\mathbf{P}\mathbf{k}G_2{}^0(\mathbf{k}), \qquad (23)$$
$$G_1{}^0(\mathbf{k})=(\omega-\varepsilon_1(\mathbf{k}))^{-1},\qquad G_2{}^0(\mathbf{k})=(\omega-\varepsilon_2(\mathbf{k}))^{-1}.$$

With account taken of G_{21} and G_{22} and of the substitution q → –q, there are altogether eight such expressions. The matrix of the Fourier component of the current operator (6) takes in the momentum representation the form

$$\mathbf{j}_{\mathbf{q}}=e\begin{pmatrix}\frac{1}{m^*}\left(\mathbf{k}+\frac{\mathbf{q}}{2}\right) & \frac{1}{m}\mathbf{P}\\ \frac{1}{m}\mathbf{P}^* & -\frac{1}{m^*}\left(\mathbf{k}+\frac{\mathbf{q}}{2}\right)\end{pmatrix}, \qquad (24)$$

since the operator of the intraband current density equals in the coordinate representation

$$\hat{\mathbf{j}}_{11}(\mathbf{r})=\frac{e}{2m^*}\sum_{\mathbf{k}}(\hat{\mathbf{p}}e^{i\mathbf{q}\mathbf{r}}+e^{i\mathbf{q}\mathbf{r}}\hat{\mathbf{p}}). \qquad (25)$$

It follows then from (23) and (24) that in the approximation linear in Δ and $\mathbf{P}$ the mean value of the q-th component of the current is

$$\mathbf{j}_{\mathbf{q}}=e\int\frac{d\omega\,d^3\mathbf{k}}{i(2\pi)^4}\left\{\frac{1}{m^*}\left(\mathbf{k}+\frac{\mathbf{q}}{2}\right)G_{11}(\mathbf{k},\mathbf{k}+\mathbf{q})+\frac{1}{m}\mathbf{P}G_{12}(\mathbf{k},\mathbf{k}+\mathbf{q})\right.$$
$$\left.-\frac{1}{m^*}\left(\mathbf{k}+\frac{\mathbf{q}}{2}\right)G_{22}(\mathbf{k},\mathbf{k}+q)+\frac{1}{m}\mathbf{P}^*G_{21}(\mathbf{k},\mathbf{k}+\mathbf{q})\right\}. \qquad (26)$$

After integrating in (26) with respect to the frequencies, it turns out that, accurate to the q^2 terms,

$$\mathbf{j}_{\mathbf{q}}=\frac{e}{m}\operatorname{Re}[\mathbf{q}[\mathbf{q}\mathbf{P}]]\Delta_{\mathbf{q}}{}^*\int\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{m^*(\varepsilon_1(\mathbf{k})+\varepsilon_2(\mathbf{k}))^2}. \qquad (27)$$

This expression was obtained by expanding in (26) in powers of q, followed by the use of the formula for the integration by parts:

$$\int d^3\mathbf{k}\,y\operatorname{grad}z=-\int d^3\mathbf{k}\,z\operatorname{grad}y.$$

It is seen that expression (27) duplicates exactly the structure of the formula for the symmetrical part $\mathbf{j}_s$ of the spontaneous current of a semimetal, if (20) is recast in the momentum representation.

It was stated in Refs. 1 and 7 that a system with allowed interband transitions acquires ferroelectric properties if a real order parameter sets in. It is easy to show that for the model considered here this is indeed the case. It is necessary first to determine the charge-density operator $\hat{\rho}_k$. In accord with the statements made in §1, that the operator of the coordinate $\hat{\mathbf{r}}$ and the electron density $\hat{n}(\mathbf{r})$ are diagonal in a system with Hamiltonian (5), it must be assumed that the operator of the Fourier component of the charge density $\hat{\rho}_k$ is also diagonal in the band indices. Therefore the electric-field intensity operator $\hat{E}_q$ takes the form

$$\hat{E}_{\mathbf{q}}=\begin{pmatrix} i\dfrac{4\pi e\mathbf{q}}{q^2} & 0 \\ 0 & i\dfrac{4\pi e\mathbf{q}}{q^2} \end{pmatrix}. \tag{28}$$

Now, to find the polarization $\mathbf{E}$ in a system with a spatially homogeneous symmetrical order parameter, we must use expressions (23) for the Green's functions in the inhomogeneous case, and then find the limit of the corresponding expression as $\mathbf{q}\to 0$. We then obtain from (23) and (28)

$$\mathbf{E}_{\mathbf{q}}=e\int\frac{d\omega\, d^3\mathbf{k}}{i(2\pi)^4}\, i\frac{4\pi\mathbf{q}}{q^2}\{G_{11}(\mathbf{k},\mathbf{k}+\mathbf{q})+G_{22}(\mathbf{k},\mathbf{k}+\mathbf{q})\}$$

$$\approx\frac{4\pi e\mathbf{q}}{q^2}\,\mathrm{Im}(\mathbf{qP})\Delta_{\mathbf{q}}\int\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{1}{(\varepsilon_1(\mathbf{k})+\varepsilon_2(\mathbf{k}))^2}. \tag{29}$$

In the limit as $\mathbf{q}\to 0$, this quantity is finite if $\mathbf{q}\parallel\mathbf{P}$, this being evidence of the transition of the system into a state with homogeneous spontaneous polarization when a real order parameter appears in it, as was indeed stated in Refs. 1 and 7. In a recent paper[15] Batyev considered likewise the possibility of a ferroelectric state in a model with the Hamiltonian (5). The calculation in his paper is incorrect, since the ferroelectric properties appeared as a result the presence of nondiagonal matrix elements in the coordinate operator $\hat{\mathbf{r}}$, which was shown in §1 to be incorrect.

§3. STRUCTURE OF THE CURRENT STATE

In this section we consider the possible spatial structure of a state with spontaneous current on the basis of the free-energy functional $\mathscr{F}$. Such a functional was obtained macroscopically for a semimetal model earlier in Ref. 16, where the corresponding phase diagram was also constructed. In that reference, only a symmetric complex order parameter $\Delta^s(\mathbf{R})$ was considered. We have shown here in (20), however, that when a real antisymmetrical parameter $\Delta^a(\mathbf{R})$ appears a spontaneous current can also be produced. We shall threfore discuss briefly in the conclusion of this section the changes brought about by the existence of $\Delta^a(\mathbf{R})$.

The simplest structure of a functional for a symmetrical complex order parameter was macroscopically determined in Ref. 16. For a system with a symmetry axis ($0z\parallel\mathbf{P}$) it can be obtained also purely phenomenologically:

$$\mathscr{F}[\Delta^s(\mathbf{R})]=\int d\mathbf{R}\left\{\alpha|\Delta^s|^2+\gamma_1|\mathrm{grad}\,\Delta^s|^2+\gamma_2\left|\frac{\mathbf{P}}{|\mathbf{P}|}\mathrm{grad}\,\Delta^s\right|^2\right.$$

$$\left.+\delta|\mathrm{div\,grad}\,\Delta^s|^2+\beta|\Delta^s|^4+\varepsilon|\Delta^s-\Delta^{s*}|^2\right\}. \tag{30}$$

Here $\alpha=a(T-T_c)$ and the remaining coefficients of the constant and their numerical values are given in Ref. 16. Usually $\varepsilon>0$, because the effective coupling constant corresponding to a real order parameter exceeds that for an imaginary one. However, as noted in Ref. 17, the presence of impurities[17] and the spin-order interaction[18] alter the ratio of the constants in favor of the imaginary order parameter. It is easily seen that at $\varepsilon>0$, $\gamma_1>0$, and $\gamma_1+\gamma_2>0$ the minimization of the functional (30) leads to the appearance of a real homogeneous order parameter in T_c, i.e., to ferroelectricity (29). For a state with spontaneous current to be produced, the necessary (but not sufficient) condition is inhomogeneity. This calls for $\gamma_1<0$. In addition, this inhomogeneity should have a transfer character, which is realized at $\gamma_2<0$. At a temperature $T_1>T_c$ determined from the condition

$$a(T_1-T_c)=-\gamma_1^2/4\delta,\quad \gamma_1<0,\quad \gamma_2>0,\quad \delta>0,\quad \varepsilon>0, \tag{31}$$

a structure appears with a transversely inhomogeneous real order parameter with a wave vector

$$q_0=(-\gamma_1/2\delta)^{1/2}. \tag{32}$$

At the temperature T_2 this structure turns out to be unstable to the onset of an imaginary transversely inhomogeneous order parameter (i.e., of a current state). This temperature can be determined from the relation

$$a(T_2-T_c)+\varepsilon+\gamma_1 q_0^2+\delta q_0^4+{}^1/_4\beta u_{\mathrm{R}}^2=0, \tag{33}$$

where u_{R}^2 is the square of the amplitude of the real order parameter.

Specific applications of formulas (31)–(33) to a system with a semimetallic spectrum were made in Ref. 16. It turned out that in the vicinity of the Lifshitz point ($\gamma_1=0$) there actually exists on the phase diagram a region where a state with spontaneous current is realized.

The physical picture of the onset of a macroscopically inhomogeneous current in a ferroelectric can be visualized as the following sequence of transitions. First, to appear in a uniaxial system at the temperature T_1 is a ferroelectric phase with a domain (amplitude soliton) structure, in which the domains are stretched out along the polarization vector ($\mathbf{P}$) and are periodically disposed in space in a direction transverse to the polarization. In the soliton region, the phase of the order parameter Δ remains unchanged ($\mathrm{Im}\,\Delta=0$), and the amplitude reverses sign on going through zero. Next, at the temperature T_2, a nonzero imaginary value of the order parameter sets in the transition region between neighboring domains. Now both the phase and the amplitude of the order parameter vary smoothly in this region, over scales of the order of q_0(32). The soliton becomes thus gradually phase dependent and spontaneous current begins to circulate in it. The phase of the order parameter changes from 0 to π within the region of one soliton. Thus, according to (20), two opposing current loops are produced inside the soliton (Fig. 1). The soliton has therefore no magnetic moment or ferromagnetic properties, but is a magnetic multipole.

All the foregoing pertains to a system without doping ($\mu=0$). If the chemical potential $\mu\neq 0$ (there is an excess of carriers), then the possibility of appearance of a real antisymmetrical order parameter $\mathrm{Re}\,\Delta^a$(18) and of the associated current $\mathbf{j}_c$ (20) must be considered. This leads, as shown by a microscopic calculation, to an additional term in the functional (30),

$$\tau(\mathrm{Re}\,\Delta^a)^2+\sigma\{(\mathrm{Im}\,\Delta^s)$$

$$\times(\mathrm{div\,Re}\,\Delta^a)$$

$$-\mathrm{Re}\,\Delta^a(\mathrm{grad\,Im}\,\Delta^s)\} \tag{34}$$

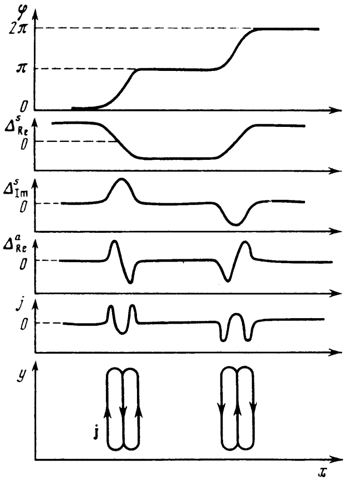

FIG. 1.

In the semimetal model, the coefficients τ and σ are given by

$$\tau=a(T-T_c^a),\qquad \sigma=\mu\frac{k_F^2T}{6\pi}\sum_{n\geqslant 0}\frac{\omega_n}{(\omega_n^2+\mu^2)^2}. \tag{35}$$

It is curious that the presence of a complex invariant in (34) of the type of the Lifshitz invariant) causes the inhomogeneous symmetrical order parameter that appears in the doped system to induce a real antisymmetrical parameter, so that the total current produced in the doped system is always a sum of two currents, $\mathbf{j}_s$ and $\mathbf{j}_a$(20). Qualitatively, however, the situation illustrated in the figure remains unchanged.

CONCLUSION

We have thus shown in present paper that in substances with allowed interband dipole transitions ($\mathbf{P}\neq 0$), which are unstable to dielectric pairing, a system of rigidly secured macroscopic currents is produced when a spatially inhomogeneous order parameter appears. The structure of these currents is such that they cannot produce a homogeneous magnetic field, so that the substance is not ferromagnetic in the state with the currents. Moreover, the free-energy functional constructed in §3 for the order parameter $\Delta(\mathbf{R})$ (the density of the exciton condensate) cannot be rewritten at all in terms of the density of the magnetic moment (even if the density is inhomogeneous), for in accord with Eqs. (20) for the currents it would acquire in this case a patently nonlocal form. For the same reason, the current can likewise not be regarded as an order parameter.[3]

Next, since the spontaneous current vanishes under conditions of thermodynamic equilibrium, when the order parameter becomes homogeneous, the statement of the Bloch theorem,[19] which is violated in Ref. 1, turns out to be satisfied, although in a nonequilibrium system there can exist a special contribution to the current due to the homogeneous order parameter.

It is of particular interest to determine the behavior of the investigated system in an external magnetic field. It turns out that the response of the system to a homogeneous magnetic field, when the temperature of the phase-transition into the current state is approached from above, has no singularities whatever, and the diamagnetic anomally obtained for the equilibrium case in Ref. 20 does not exist.

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APPENDIX

The description of a system in an excitonic-insulator state can be regarded as one of the variants of the solutions of the nonlinear Hartree-Fock equations for this system. We present below two methods of proving the absence of homogeneous spontaneous currents from equilibrium systems in the Hartree–Fock approximation. The first is due to L. V. Keldysh.

1. In the Hartree-Fock approximation, the Hamiltonian of an electron in a periodic field $U(\mathbf{r})$ is of the form

$$\hat{H}=-\frac{\nabla^2}{2m}+U(\mathbf{r})-\int d^3\mathbf{r}'\,V(\mathbf{r}-\mathbf{r}')\rho(\mathbf{r},\mathbf{r}'), \tag{A.1}$$

where $V(\mathbf{r}-\mathbf{r}')$ is the interelectron Coulomb interaction, $\rho(\mathbf{r},\mathbf{r}')=\rho^*(\mathbf{r}',\mathbf{r})$ is the density matrix, and the last term of (A.1) describes the nonlocal exchange potential. Taking the periodicity of the problem into account, the solution of the Hartree-Fock equation

$$\hat{H}\psi_{n\mathbf{k}}=E_{n\mathbf{k}}\psi_{n\mathbf{k}} \tag{A.2}$$

is sought in the form

$$\psi_{n\mathbf{k}}=u_{n\mathbf{k}}(\mathbf{r})\exp(i\mathbf{k}\mathbf{r}). \tag{A.3}$$

After substituting (A.1) and (A.3) in (A.2), cancelling the factor $\exp(i\mathbf{k}\cdot\mathbf{r})$, and differentiating the obtained equation with respect to k, we get

$$\begin{aligned}&\frac{1}{im}\nabla u_{n\mathbf{k}}+\left(-\frac{\nabla^2}{2m}-\frac{i}{m}\mathbf{k}\nabla\right)\frac{\partial u_{n\mathbf{k}}}{\partial\mathbf{k}}-\int d^3\mathbf{r}'\,V(\mathbf{r}-\mathbf{r}')\\&\times\rho(\mathbf{r},\mathbf{r}')\exp(i\mathbf{k}(\mathbf{r}-\mathbf{r}'))\frac{\partial u_{n\mathbf{k}}}{\partial\mathbf{k}}-\left(E_{n\mathbf{k}}-\frac{k^2}{2m}\right)\frac{\partial u_{n\mathbf{k}}}{\partial\mathbf{k}}\\&=\left(\frac{\partial E_{n\mathbf{k}}}{\partial\mathbf{k}}-\frac{\mathbf{k}}{m}\right)u_{n\mathbf{k}}+i\int d^3\mathbf{r}'\,V(\mathbf{r}-\mathbf{r}')\rho(\mathbf{r},\mathbf{r}')\exp(i\mathbf{k}(\mathbf{r}-\mathbf{r}'))(\mathbf{r}-\mathbf{r}')u_{n\mathbf{k}}(\mathbf{r}').\end{aligned} \tag{A.4}$$

Multiplying this relation from the left by u_{nk} and integrating with respect to $\mathbf{r}$, taking it into account that the Hamiltonian (A.1) is Hermitian, we get

$$\begin{aligned}\left\langle u_{n\mathbf{k}}\left|\frac{\hat{\mathbf{p}}}{m}\right|u_{n\mathbf{k}}\right\rangle&=\frac{\partial E_{n\mathbf{k}}}{\partial\mathbf{k}}-\frac{\mathbf{k}}{m}+i\int d^3\mathbf{r}\int d^3\mathbf{r}'\,V(\mathbf{r}-\mathbf{r}')\\&\times\rho(\mathbf{r},\mathbf{r}')u_{n\mathbf{k}}^*(\mathbf{r})e^{-i\mathbf{k}\mathbf{r}}u_{n\mathbf{k}}(\mathbf{r}')e^{i\mathbf{k}\mathbf{r}'}(\mathbf{r}-\mathbf{r}').\end{aligned} \tag{A.5}$$

The last term in (A.5) is the exchange correction to the electron velocity. In its absence, we would get the known result

$$\left\langle \psi_{n\mathbf{k}}\left|\frac{\hat{\mathbf{p}}}{m}\right|\psi_{n\mathbf{k}}\right\rangle=\frac{\partial E_{n\mathbf{k}}}{\partial\mathbf{k}}. \tag{A.6}$$

To find the average current connected with the exchange correction, we must sum the exchange term over all the occupied states. We then get

$$\langle \mathbf{j}_{\text{exch}}\rangle=i\int d^3\mathbf{r}\int d^3\mathbf{r}'\,V(\mathbf{r}-\mathbf{r}')\rho(\mathbf{r},\mathbf{r}')(\mathbf{r}'-\mathbf{r})\rho(\mathbf{r}',\mathbf{r})=0, \tag{A.7}$$

since the density matrix is Hermitian (the interchange

$\mathbf{r}\rightleftarrows r'$). Thus, the homogeneous current is determined by averaging (A. 7) over all the occupied states, and if the occupation numbers of these states depend only on their energies, then the current for all the solutions of the Hartree-Fock equation vanishes identically.

2. The second method is based on a definition of the current in the form (3) and on the requirement that the Hartree-Fock Hamiltonian (A. 1) be gauge-invariant. If it is noted that the exchange term in (A. 1) can be identically rewritten in the form

$$\int d^3\mathbf{a}\, V(\mathbf{a})\rho(\mathbf{r},\mathbf{r}+\mathbf{a})\exp(i\mathbf{a}\hat{\mathbf{p}}), \tag{A.8}$$

then it is transformed for an external field A, with the aid of the formal gauge substitution $\hat{\mathbf{p}}\rightarrow\hat{\mathbf{p}}-e\mathbf{A}/c$, into

$$\int d^3\mathbf{a} V(\mathbf{a})\rho(\mathbf{r},\mathbf{r}+\mathbf{a})\exp[i\mathbf{a}(\hat{\mathbf{p}}-e\mathbf{A}/c)]. \tag{A.9}$$

Variation of (A. 9) with respect to **A** yields for the current an exchange correction identical with (A. 7), if we set $\mathbf{A}=0$ after the variation and reverse the change of variable $\mathbf{a}=\mathbf{r}-\mathbf{r}'$. We note that this approach yields also the local-current density in the Hartree-Fock approximation. To this end it is necessary to "untangle" the shift operator in (A. 9). If the vector potential is resolved into a transverse part $\mathbf{A}_\perp(\mathrm{div}\mathbf{A}_\perp=0)$ and a longitudinal one $\mathbf{A}_\parallel$ $(\mathbf{A}_\parallel=\mathrm{grad}\varphi)$, we get

$$\hat{\mathbf{T}}=\exp\left\{i\mathbf{a}\left(\hat{\mathbf{p}}-\frac{e}{c}\mathbf{A}\right)\right\}=\exp\left\{-\frac{ie}{c}\mathbf{a}\mathbf{A}_\perp(\mathbf{r})\right\}$$
$$\times\exp\left\{-\frac{ie}{c}[\varphi(\mathbf{r}+\mathbf{a})-\varphi(\mathbf{r})]\right\}\exp(i\mathbf{a}\hat{\mathbf{p}}). \tag{A.10}$$

To prevent misunderstanding, it must be noted that the density matrix itself depends on A and the Hartree-Fock equation is in fact its self-consistent definition.

[1)] The formal expression for **d** cannot be used, since **r** does not belong to the basis of bounded functions. To calculate **d** one must use the formula

$$\mathbf{d}=\lim_{\mathbf{q}\rightarrow 0}\frac{1}{i}\frac{d}{d\mathbf{q}}\langle\varphi_{2\mathbf{k}}|e^{i\mathbf{q}\mathbf{r}}|\varphi_{1\mathbf{k}'}\rangle,$$

which is analogous to that in §22 of Ref. 12.

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