

- kristalla (Dynamics of Elastic Half-Space with Moving Dislocations. II. Emission of Sound by Dislocations Emerging on the Surface of a Crystal), Preprint No. 12-77, Physicotechnical Institute of Low Temperatures, Academy of Sciences of the Ukrainian SSR, Kiev, 1977.
- <sup>10</sup>A. M. Kosevich, *Dislokatsii v teorii uprugosti* (Dislocations in the Theory of Elasticity), Naukova Dumka, Kiev, 1978.
- <sup>11</sup>J. P. Hirth and J. Lothe, *Theory of Dislocations*, McGraw-Hill, New York, 1968 (Russ. Transl., Atomizdat, M., 1972).
- <sup>12</sup>A. M. Kosevich and V. S. Boiko, *Usp. Fiz. Nauk* **104**, 201 (1971) [Sov. Phys. Usp. **14**, 286 (1971)].
- <sup>13</sup>S. G. Lekhnitskii, *Teoriya uprugosti anizotropnogo tela* (Theory of Elasticity of an Anisotropic Body), Nauka, M., 1977.
- <sup>14</sup>V. D. Natsik and K. A. Chishko, *Fiz. Tverd. Tela* (Leningrad) **14**, 3126 (1972) [Sov. Phys. Solid State **14**, 2678 (1973)].

Translated by A. Tybulewicz

## Exciton insulator in an alternating electric field

É. G. Batyev

*Institute of Semiconductor Physics, Siberian Branch of the Academy of Sciences of the USSR, Novosibirsk*

V. A. Borisyuk

*L. V. Kirenski Institute of Physics, Siberian Branch of the Academy of Sciences of the USSR, Krasnoyarsk*

(Submitted 12 June 1980)

*Zh. Eksp. Teor. Fiz.* **80**, 262–273 (January 1981)

An analysis is made of the linear response of an exciton insulator with an allowed dipole transition to a homogeneous alternating electric field. It is shown that the electron subsystem of such an insulator has two low-frequency branches of natural oscillations, one of which corresponds (in the homogeneous case) to phase oscillations, and the other to oscillations of the modulus of the order parameter. The frequency of the latter oscillation exceeds the dissociation threshold but nevertheless the oscillation is weakly damped. Both oscillations are accompanied by the flow of a current and, therefore, the response (i.e., the dependence of the current on the field) is resonant.

PACS numbers: 72.20. – i

### 1. INTRODUCTION

Exciton insulators with an allowed dipole transition are attracting interest because unusual properties have been predicted for them (including spontaneous currents and superdiamagnetism<sup>1–3</sup>). We shall not discuss these properties: we shall be interested in the linear response of such an exciton insulator to a homogeneous alternating electric field. We shall show that the response is resonant, by analogy to substances which are active in the infrared part of the spectrum; this is due to the fact that in an exciton insulator with an allowed dipole transition even a homogeneous field affects the magnitude of the order parameter (gap in the spectrum) already in the linear approximation and, therefore, there is a relationship between the field and free oscillations of the system. In contrast to the substances mentioned above, whose infrared activity is due to the lattice, we shall consider the case when this activity is entirely due to the properties of the electron subsystem.

We shall use the simplest model<sup>2,3</sup>; the Hamiltonian of the system considered in the two-band approximation can be written in the form of a matrix:

$$\hat{H} = \begin{pmatrix} \frac{1}{2m} \left( \hat{k} - \frac{e}{c} \mathbf{A} \right)^2 - \mu, & \frac{1}{m_0} p_{12} \left( \hat{k} - \frac{e}{c} \mathbf{A} \right) + \Delta \\ \frac{1}{m_0} p_{21} \left( \hat{k} - \frac{e}{c} \mathbf{A} \right) + \Delta^*, & -\frac{1}{2m} \left( \hat{k} - \frac{e}{c} \mathbf{A} \right)^2 + \mu \end{pmatrix}. \quad (1)$$

Here,  $\hat{\mathbf{k}} = -i\mathbf{v}$ ;  $\mathbf{A}$  is the vector potential;  $\Delta$  is the order parameter;  $m_0$  is the electron mass;  $m$  is the effective mass, assumed to be the same for an electron and a hole; the constant  $\mu$  represents the degree of overlap of the original bands ( $\mu > 0$ );  $p_{12}$  is the matrix element of

the momentum operator, calculated using Bloch functions corresponding to the extrema of the bands under consideration; these functions should be selected to be real for the extrema located at zero quasimomentum and then  $p_{12}$  is a purely imaginary quantity; in the other cases all this can be done by a canonical transformation. The Hamiltonian (1) is independent of the spin indices: singlet pairing is assumed. All the calculations will be carried out for zero absolute temperature.

In the presence of a homogeneous electric field the vector potential may be assumed to be independent of the coordinates and we can then use the momentum representation, so that Eq. (1) can be modified to

$$H(\mathbf{k}) = \begin{pmatrix} \frac{1}{2}(\mathbf{k}-\mathbf{A})^2 - \mu & v_{12}(\mathbf{k}-\mathbf{A}) + \Delta(\mathbf{k}) \\ v_{21}(\mathbf{k}-\mathbf{A}) + \Delta^*(\mathbf{k}), & -\frac{1}{2}(\mathbf{k}-\mathbf{A})^2 + \mu \end{pmatrix}; \quad (2)$$

the following simplifications and the notation are used above:

$$p_{12}/m_0 = v_{12}, \quad e\mathbf{A}/c \rightarrow \mathbf{A}, \quad m=1. \quad (3)$$

The problem is to find the linear response of the system described by the Hamiltonian (2), i.e., to find the relationship between the current and the field. In a weak field the problem can naturally be solved by perturbation theory. More precisely, we shall use the perturbation theory to solve the equation for the Green function; in this case the Green function is a  $2 \times 2$  matrix, whose elements are defined as follows:

$$\{G_{\mathbf{k}}(t, t')\}_{nm} = -i \langle T a_{n\mathbf{k}}(t) a_{m\mathbf{k}}^*(t') \rangle, \quad (4)$$

where  $a_{n\mathbf{k}}(t)$ ,  $a_{m\mathbf{k}}^*(t)$  are the operators (in the Heisenberg representation) of the annihilation and creation of an electron with a quasimomentum  $\mathbf{k}$  in a band  $n$  ( $n=1$  or 2).

The equation for the Green function has its usual form:

$$\left( i \frac{d}{dt} - H(\mathbf{k}) \right) G_{\mathbf{k}}(t, t') = \delta(t - t'). \quad (5)$$

We also need the self-consistency equation for the order parameter:

$$\Delta(\mathbf{k}) = \sum_{\mathbf{k}'} U(\mathbf{k}-\mathbf{k}') \{iG_{\mathbf{k}'}(t, t)\}_{12} \quad (6)$$

(throughout our discussion we shall assume that the volume is unity). Here,  $U(\mathbf{k})$  is the Fourier component of the potential of the interaction between electrons belonging to different bands.

We shall complete the formulation of the problem by including the expression for the current density  $j$ ; this can be written in the form

$$j = -i \sum_{\mathbf{k}} \text{Sp} \left\{ \frac{\partial H(\mathbf{k})}{\partial \mathbf{k}} G_{\mathbf{k}}(t, t') \right\}_{t=t+0}. \quad (7)$$

The above expression can be tested most simply as follows. In the  $\mathbf{k}$  representation ( $\mathbf{r} = i\partial/\partial \mathbf{k}$ ) the velocity operator  $\mathbf{r} = i(H\mathbf{r} - \mathbf{r}H)$  is given by  $\mathbf{r} = \partial H(\mathbf{k})/\partial \mathbf{k}$  and hence we obtain Eq. (7).

## 2. PRINCIPAL RELATIONSHIPS

We shall begin with some information on an exciton insulator in the absence of a field ( $A = 0$ ). In this case the Hamiltonian is

$$H_0 = \begin{pmatrix} \xi_{\mathbf{k}} & \mathbf{k}\mathbf{v}_{12} + \Delta_0(\mathbf{k}) \\ \mathbf{k}\mathbf{v}_{21} + \Delta_0^*(\mathbf{k}) & -\xi_{\mathbf{k}} \end{pmatrix}, \quad (8)$$

where  $\xi_{\mathbf{k}} = \mathbf{k}^2/2 - \mu$ . The Green function  $G^{(0)}$  of this problem has the following form in the frequency representation:

$$G^{(0)}(\mathbf{k}, \epsilon) = \frac{a(\mathbf{k}, \epsilon)}{(\epsilon - E_{\mathbf{k}} + i\delta)(\epsilon + E_{\mathbf{k}} - i\delta)}, \quad (9)$$

where the matrix  $a$  is given by the relationship

$$a = \epsilon + H_0, \quad (10)$$

and  $\pm E_{\mathbf{k}}$  is the energy of a particle in the upper and lower (filled) bands of an exciton insulator:

$$E_{\mathbf{k}} = [\xi_{\mathbf{k}}^2 + |\mathbf{k}\mathbf{v}_{12} + \Delta_0(\mathbf{k})|^2]^{1/2}. \quad (11)$$

Finally, the self-consistency equation is

$$\Delta_0(\mathbf{k}) = \sum_{\mathbf{k}'} U(\mathbf{k}-\mathbf{k}') \frac{\mathbf{k}'\mathbf{v}_{12} + \Delta_0(\mathbf{k}')}{2E_{\mathbf{k}'}}. \quad (12)$$

We shall now return to the problem. First, we shall transform Eq. (5). We shall assume that the quantity  $\Delta$  and, therefore, the Hamiltonian (2) and the Green function depend on the momentum which occurs in  $\mathbf{k} - \mathbf{A} \equiv \mathbf{p}$ , i.e.,  $\Delta = \Delta(\mathbf{p}, t)$ . The total derivative with respect to time in Eq. (5) can be written in the form

$$\frac{dG_{\mathbf{p}}(t, t')}{dt} = \left\{ \frac{\partial G_{\mathbf{p}}(t, t')}{\partial t} \right\}_{\mathbf{p}=\text{const}} - \mathbf{A} \frac{\partial G_{\mathbf{p}}(t, t')}{\partial \mathbf{p}}.$$

We shall now introduce new notation:

$$\Delta(\mathbf{p}, t) = \Delta_0(\mathbf{p}) + \Delta_1(\mathbf{p}, t); \\ H = H_0 + H_1, \quad H_1 = \begin{pmatrix} 0 & \Delta_1 \\ \Delta_1^* & 0 \end{pmatrix}. \quad (13)$$

Employing all these expressions, we can rewrite Eq.

(5) in the form

$$i \left( \frac{\partial G}{\partial t} \right)_{\mathbf{p}} - H_0 G - i \mathbf{A} \frac{\partial G}{\partial \mathbf{p}} = \delta(t - t'). \quad (14)$$

This equation is so far exact. In a weak field the last two terms on the left-hand side of Eq. (14) can be regarded as a perturbation and in these terms we can replace  $G$  with the zeroth-order Green function which is identical with  $G^{(0)}$  [see Eq. (9)] if the substitution  $\mathbf{k} \rightarrow \mathbf{p}$  is made. Thus, the correction  $G^{(1)}$  to the Green function

$$G = G^{(0)} + G^{(1)}$$

is described by the equation

$$i \left( \frac{\partial G^{(1)}}{\partial t} \right)_{\mathbf{p}} - H_0 G^{(1)} = H_1 G^{(0)} + i \mathbf{A} \frac{\partial G^{(0)}}{\partial \mathbf{p}}. \quad (15)$$

The advantages of rewriting Eq. (5) in the form (14) are obvious: Eq. (14) contains explicitly not the vector potential but the electric field, and in solving this equation with the aid of perturbation theory there is no need to ensure the gradient invariance of the results. In the homogeneous case the gradient invariance implies invariance relative to arbitrary constant correction to the vector potential; it is understood that the initial equation (5) has this property. For example, if  $\mathbf{A} = \mathbf{A}_0 = \text{const}$ , then a direct solution of Eq. (5) together with the self-consistency condition (6) gives  $\Delta(\mathbf{k}) = \Delta_0(\mathbf{k} - \mathbf{A}_0)$  for the order parameter and the expression for the energy is identical with Eq. (11) if  $\mathbf{k}$  is replaced everywhere with  $(\mathbf{k} - \mathbf{A}_0)$ ; thus, the spectrum remains unchanged, as expected. We can easily see that this is also obtained from Eq. (14).

Equation (14) can also be obtained by a shorter procedure if we begin with a different form of the Hamiltonian (1). In fact, if we allow for the existence of a homogeneous electric field by using the scalar potential  $-e\xi\mathbf{r}$  instead of the vector potential, and then adopt the momentum representation ( $\mathbf{r} \rightarrow i\partial/\partial \mathbf{k}$ ), we obtain directly Eq. (14) for the Green function. However, it is more convenient to deal with the Hamiltonian (1), as is usual in problems of this kind, so as to avoid unnecessary complications.

There is no difficulty in writing down the solution of Eq. (15) for the correction to the Green function:

$$G_{\mathbf{p}}^{(1)}(t, t') = \int dt'' G_{\mathbf{p}}^{(0)}(t, t'') \left\{ H_1(t'') + i \mathbf{A}(t'') \frac{\partial}{\partial \mathbf{p}} \right\} G_{\mathbf{p}}^{(0)}(t'', t'). \quad (16)$$

We shall be interested in this correction only for coincident times  $t' = t$ . Going over to the  $\omega$  representation in Eq. (16) at  $t' = t$ , we obtain

$$G^{(1)}(\mathbf{p}, \omega) = \int \frac{de}{2\pi} G^{(0)}(\mathbf{p}, \epsilon) \left\{ H_1(\omega) + \omega \mathbf{A} \frac{\partial}{\partial \mathbf{p}} \right\} G^{(0)}(\mathbf{p}, \epsilon - \omega); \quad (17)$$

$$H_1(\omega) = \begin{pmatrix} 0 & \Delta_1(\omega) \\ \Delta_1^*(-\omega) & 0 \end{pmatrix}. \quad (18)$$

Here, each of the Fourier amplitudes,  $G^{(1)}(\mathbf{p}, \omega)$  and  $H_1(\omega)$ ,  $\mathbf{A}$ , is the coefficient in front of  $\exp(-i\omega t)$ ; we shall assume that the amplitude  $\mathbf{A}$  is real.

The correction  $G^{(1)}$  contains a part depending explicitly on  $\Delta_1$  and a part depending explicitly on  $\mathbf{A}$ . Denoting these parts by  $G_{\Delta}^{(1)}$  and  $G_{\mathbf{A}}^{(1)}$ , we shall write down the result obtained from them after integration with respect to  $\epsilon$  in Eq. (17); for simplicity, we shall omit the

indices and arguments; we thus find that

$$G^{(1)} = G_A^{(1)} + G_\Delta^{(1)},$$

$$G_\Delta^{(1)} = \frac{i}{E(4E^2 - \omega^2)} \left\{ aH_1 a - \left( E + \frac{\omega}{2} \right) aH_1 - \left( E - \frac{\omega}{2} \right) H_1 a \right\}, \quad (19)$$

$$G_A^{(1)} = \frac{i\omega A}{E(4E^2 - \omega^2)} \left( H_0 + \frac{\omega}{2} \right) \left( \frac{\partial H_0}{\partial p} - \frac{\partial E}{\partial p} \frac{H_0}{E} \right).$$

Here,  $a = a(E) = E + H_0$  [see Eqs. (10) and (8)], the matrix  $H_1$  is given by Eq. (18), and we have allowed for the fact that  $H_0^2 = E^2$ .

We shall now obtain expressions for  $\Delta_1$  and for the current. Substituting the relationships (19) in Eq. (6), we obtain

$$\Delta_1(p) - \sum_{p'} U(p-p') \left\{ \frac{(2E^2 - |\Delta|^2 + \omega\xi)\Delta_1 - \Delta^2 \Delta_1^*}{E(4E^2 - \omega^2)} \right\}_{p'},$$

$$= - \sum_{p'} U(p-p') \left\{ \left( \xi + \frac{\omega}{2} \right) \frac{\partial \Delta}{\partial p} - \Delta p - \frac{\omega \Delta}{2E} \frac{\partial E}{\partial p} \right\}_{p'} \frac{\omega A}{E_{p'}(4E_{p'}^2 - \omega^2)}. \quad (20)$$

The following notation is used above:  $\Delta \equiv \Delta_0 + p v_{12}$  and  $\Delta_1^* \equiv \Delta_1^*(-\omega)$ . The equation for  $\Delta_1^*$  is obtained from Eq. (20) by complex conjugacy and by the substitution  $\omega \rightarrow -\omega$ .

We shall carry out specific calculations for the simplified model in which the interaction is independent of the angle between  $p$  and  $p'$ ; moreover, since integrations in Eq. (20) are real mainly near the Fermi surface, the interaction can in general be regarded as independent of  $p$  and  $p'$ . Thus, our simplification is essentially

$$U(p-p') \rightarrow U(\xi_p, \xi_{p'}) \rightarrow g = \text{const}, \quad (21)$$

i.e., we have here a model of the BCS type.

In this case it is known that  $\Delta_0$  is constant and real (fixation of the phase of the order parameter occurs because of the term  $p \cdot v_{12}$  in the original Hamiltonian). This circumstance and the constancy of the function  $U$  allow us to simplify greatly Eq. (20) so that we can drop the terms containing  $\partial \Delta_0 / \partial p$ , which are odd in respect of the momentum. Moreover, we shall consider moderately high frequencies:

$$|\omega/2| \leq \Delta_0 \ll \mu. \quad (22)$$

The right-hand side of this condition is a consequence of the assumed smallness of the coupling constant  $g$ . As far as the quantity  $|p v_{12}|$  is concerned, we shall assume that  $|p v_{12}| \leq \Delta_0$  (for too high values of  $|p v_{12}|$ , the order parameter  $\Delta_0$  vanishes, i.e., this happens when  $|p v_{12}|$  is the order of that value of the gap  $\Delta_0$  which would be obtained for  $v_{12}=0$ ).

Consequently, instead of Eq. (20), we now obtain

$$\Delta_1 - g \sum_p \frac{(2E^2 + \omega\xi)\Delta_1 - \Delta_0^*(\Delta_1 + \Delta_1^*) - |p v_{12}|^2(\Delta_1 - \Delta_1^*)}{E(4E^2 - \omega^2)} \\ = g \sum_p \frac{\omega(pA)(p v_{12})}{E(4E^2 - \omega^2)}. \quad (23)$$

Only the principal term is retained on the right-hand side of this equation, whereas the left-hand side is given in full because this is necessary in the subsequent discussion.

It is clear from Eq. (23) that it contains the field even in the linear approximation, which is not true if  $v_{12}=0$ .

This is one of the reasons why the problem under consideration is of interest.

Finally, we shall analyze the expression for the current (7). We shall consider first the zeroth approximation. At first sight it might seem that even in this approximation there is some contribution to the current because all the quantities in the summation in Eq. (7) contain fields explicitly, i.e., they depend on the momentum in the combination  $k - A$ . In fact, there is no contribution to the current at all. This can be demonstrated by the following elementary consideration. The quasimomentum  $p$  of a particle in a field changes in accordance with the equation of motion  $p = -A$ , and hence  $p(t) = k - A(t)$  ( $k$  is a constant of integration). Nevertheless, it is known that the total current for a completely filled band is zero. This shows why all the quantities depend specifically on  $k - A(t)$  (in the zeroth approximation there are no additional time dependences) and why in the zeroth approximation we can regard the current as zero.

The current may appear only if we include, for example, interband transitions. This contribution is found in the next approximation. We should mention immediately that the whole contribution to the current is acquired, as shown later, near the Fermi surface ( $\xi=0$ ) so that we can go over to summation with respect to the variable  $p = (k - A)$  and then the additional dependence on  $t$  disappears completely.

In the expression for the current, we shall identify the components  $j_\Delta$  and  $j_A$  which depend explicitly on  $\Delta_1$  and  $A$ , as has been done earlier for the correction  $G^{(1)}$  to the Green function given by Eq. (19). We shall not give detailed procedures, which are generally not very complex. Instead, we shall mention a small point relating to the derivation of the expression for  $j_\Delta$ . If this expression is obtained using Eqs. (18) and (19), it might seem that it does not vanish in the limit  $\omega \rightarrow 0$ ; in fact, the expression under the summation sign contracts—as is easily shown—to the total derivative with respect to  $p$  and, therefore, the current  $j_\Delta$  vanishes in the limit  $\omega \rightarrow 0$ , as expected. This can be used to transform the expression for  $j_\Delta$  even in the case when  $\omega \neq 0$ : it is necessary to separate under the summation sign the part independent of the frequency and to drop it.

In this way we obtain the following expressions for the current:

$$j = j_\Delta + j_A;$$

$$j_\Delta = \sum_p \frac{\omega(\Delta_1 + \Delta_1^*)}{E(4E^2 - \omega^2)} [-p(p v_{12}) + \xi v_{12}]$$

$$+ \sum_p \frac{\omega^2(\Delta_1 - \Delta_1^*)}{2E^3(4E^2 - \omega^2)} [(\xi^2 + \Delta_0^2)v_{12} - \xi(p v_{12})p],$$

$$j_A = \sum_p \frac{\omega^2}{E(4E^2 - \omega^2)} \left\{ p(pA) - v_{12}(Av_{12}) - \frac{1}{E} \left( A \frac{\partial E}{\partial p} \right) [p\xi - v_{12}(p v_{12})] \right\}.$$

We shall assume the model represented by Eq. (21); the terms with the derivative  $\partial \Delta_0 / \partial p$  containing the additional smallness are dropped. Retaining the principal terms in these expressions [using the condition (22)],

we finally find that

$$j = \sum_p \frac{1}{E(4E^2 - \omega^2)} \left\{ \frac{\Delta_0^2 + |\mathbf{p}v_{12}|^2}{E^2} \omega^2 p(\mathbf{p}A) - \omega(\Delta_1 + \Delta_1^*) p(\mathbf{p}v_{12}) + \frac{\omega^2(\Delta_1 - \Delta_1^*)}{2E^2} [(\xi^2 + \Delta_0^2)v_{12} - \xi p(\mathbf{p}v_{12})] \right\}. \quad (24)$$

The quantity  $j_A$  represents the contribution of the virtual (when  $|\omega| < 2\Delta_0$ ) transitions of particles between the bands of an exciton insulator, which by definition is completely analogous to the corresponding contribution in the case of an ordinary insulator; part of the current  $j_A$  has no analog; this is the direct contribution of an electron-hole pair condensate associated probably with the polarization of this condensate.

We can thus see that Eqs. (23) and (24) give, in principle, the solution of the problem of determining the response (to an alternating electric field) of an exciton insulator with an allowed dipole transition. We shall analyze these expressions in the sections below.

### 3. FREE OSCILLATIONS

The system of equations for  $\Delta_1$  and  $\Delta_1^* \equiv \Delta_1^*(-\omega)$  [the latter is obtained from Eq. (23) by complex conjugacy and the substitution  $\omega \rightarrow -\omega$ ] can be conveniently rewritten for the quantities  $\Delta_1 \pm \Delta_1^*$ ; for these quantities we have

$$\begin{aligned} x &= \Delta_1 + \Delta_1^*, \quad y = \Delta_1 - \Delta_1^*; \\ \sum_p \frac{\omega^2 - 4\Delta_0^2}{E(4E^2 - \omega^2)} x + \sum_p \frac{2\xi\omega}{E(4E^2 - \omega^2)} y &= -4 \sum_p \frac{\omega(pA)(\mathbf{p}v_{12})}{E(4E^2 - \omega^2)}; \\ \sum_p \frac{2\xi\omega}{E(4E^2 - \omega^2)} x + \sum_p \frac{\omega^2 - 4|\mathbf{p}v_{12}|^2}{E(4E^2 - \omega^2)} y &= 0. \end{aligned} \quad (25)$$

The system (25) is obtained using the equation for the gap (12) in the model (21), which allows us to replace the first term in Eq. (23) with

$$\Delta_1 g \sum_p \frac{1}{2E}.$$

We shall determine natural (eigen) frequencies by considering the system (25) when  $A=0$  (restrictions of this approach are discussed in the next section). All further calculations will be made on the assumption that

$$|\mathbf{p}v_{12}| \ll \Delta_0. \quad (26)$$

We can see directly that the coefficient in front of  $y$  in the first equation of the system (25) (or the coefficient in front of  $x$  in the second equation) is generally small compared with the coefficient in front of  $x$  (or the coefficient in front of  $y$  in the second equation); if, in the first approximation, we assume that the density of states near the Fermi surface is constant, we find that this coefficient vanishes. In this approximation we find that free oscillations have the following frequencies:

$$\omega_1 \approx 2p_F v_0 / \sqrt{3}, \quad x=0, \quad y \neq 0; \quad (27.1)$$

$$\omega_2 = 2\Delta_0, \quad x \neq 0, \quad y=0 \quad (27.2)$$

( $v_{12} = i\mathbf{v}_0$ , where  $\mathbf{v}_0$  is a real vector). The first frequency is calculated using the condition (26); here,  $p_F$  is the Fermi momentum ( $\xi_{p_F} = 0$ ).

It is known<sup>4</sup> that a conventional exciton insulator (with  $v_{12}=0$ ) exhibits an acoustic oscillation branch whose frequency in the case  $q=0$  of interest to us is thus zero.

When the phase of the order parameter is fixed, the corresponding frequency naturally becomes finite and is given by Eq. (27.1) for the phase fixation mechanism (because of  $\mathbf{p} \cdot \mathbf{v}_{12}$ ) considered here. The same result follows also from the expressions given in Ref. 5.

In general, the origin of the low-frequency branch which begins from the frequency (27.1) and goes over to an acoustic branch at sufficiently high values of  $q$  (Ref. 5) is quite obvious. However, the appearance of a new branch whose frequency is close to  $2\Delta_0$  near  $q=0$ , as given by Eq. (27.2), is fairly unexpected. Let us consider what occurs in the case of such oscillations. Both oscillations are accompanied by the current, as indicated by Eq. (24); the direction of the current ("polarization" of the wave) is given by the vector  $\mathbf{v}_0$ ; thus, the polarization is constant in the limit  $q \rightarrow 0$  and is independent of the direction of wave propagation. In the case of the order parameter it is seen from Eqs. (27.1) and (27.2) that the frequency  $\omega_1$  corresponds to a phase oscillation and the frequency  $\omega_2$  to an oscillation of the modulus of  $\Delta$ . We can thus see that these oscillations have no analogs in superfluid systems (we recall that we are speaking here of oscillations with  $q=0$ ).

The situation becomes somewhat clearer if we look from the other (microscopic) point of view. This approach is aided by the use of a pseudospin model proposed for superconductors by Anderson,<sup>6</sup> in which each point in the  $\mathbf{p}$  space is attributed a spin  $1/2$ . Naturally, this model applies also to exciton insulators. Collective excitations can be investigated by this approach; naturally, the equations then obtained are completely identical with the system (25) if  $A=0$ . These excitations are simply spin waves in a pseudospin system or, in other words, excitons in a system of Cooper pairs. Hence, it is clear that this is hardly possible in the case of structure-free particles.

We shall now consider more accurate calculation of the oscillation frequency (27.2). The point is this: this frequency is slightly greater than  $2\Delta_0$ , as is indicated by the initial system (25), i.e., an oscillation quantum may dissociate into two one-particle excitations. The problem is to find the attenuation and frequency shift which result from the inclusion in the system (25) of the terms rejected earlier.

We shall proceed as follows. We shall first calculate the determinant  $D(\omega)$  of the system (25) in the sub-threshold range  $|\omega| < 2\Delta_0$ . We shall then find an analytic continuation of  $D(\omega)$  via the upper half-plane to the region beyond the threshold and find the frequency and attenuation in the usual way from the equation  $D(\omega)=0$ . It is then found that in our case it is sufficient to calculate the integrals in Eq. (25) subject to the condition

$$|(\omega/2)^2 - \Delta_0^2| \ll (p_F v_0)^2. \quad (28)$$

The calculation is a simple process. The results are:

$$\begin{aligned} \sum_p \frac{1}{E(4E^2 - \omega^2)} &\approx \frac{\pi\gamma}{4\Delta_0 p_F v_0} L, \\ L &= \ln \{2p_F v_0 / (\Delta_0^2 - (\omega/2)^2)^{1/2}\}; \\ \sum_p \frac{\xi}{E(4E^2 - \omega^2)} &\approx \frac{\gamma}{4\lambda\mu}, \end{aligned} \quad (29)$$

where  $\gamma = p_F/2\pi^2$  is the density of states on the Fermi surface and  $\lambda = g\gamma$  is the dimensionless coupling constant. In the calculation of the second integral in the system (29) we have to allow for the difference between the density of states near the Fermi surface  $\gamma(\xi)$  and the constant density  $\gamma$ , namely,

$$\gamma(\xi) \approx \gamma(1 + \xi/2\mu).$$

The integral is then logarithmically divergent and this gives rise to a coupling constant  $\lambda$ .

We shall now introduce the following dimensionless quantities:

$$v = \frac{(\omega/2)^2 - \Delta_0^2}{4(p_F v_0)^2}, \quad \alpha^2 = \left( \frac{\Delta_0}{2\pi\lambda\mu} \right)^2. \quad (30)$$

The determinant of the system (25), including Eqs. (29) and (30), can be written (apart from an unimportant factor) in the form

$$D \sim v L^2 - \alpha^2. \quad (31)$$

The constant  $\alpha$  is small and it amounts to  $|\ln \alpha| \gg 1$  in the weak interaction limit. This makes it possible to find relatively simply the zero value of  $D$  by first determining the analytic continuation of Eq. (31) from the negative half-axis  $v$  via the upper half-plane to the positive half-axis and then to the lower half-plane. The zero value of  $D$  is located at the point  $v_0$ , where

$$v_0 = |v_0| e^{i\varphi}, \quad \varphi \approx 2\pi/\ln|v_0|, \quad |v_0| (\ln|v_0|)^2 = 4\alpha^2. \quad (32)$$

It should be noted that the phase  $\varphi$  is negative and, as expected, small. Solving approximately the relationships (32), we finally obtain the following expression for the oscillation frequency and attenuation

$$\omega_2 - 2\Delta_0 \approx 4 \frac{(p_F v_0)^2}{\Delta_0} \left( \frac{\alpha}{\ln \alpha} \right)^2 \left( 1 + \frac{i\pi}{\ln \alpha} \right). \quad (33)$$

The frequency shift is so small that we can use the condition (28) and the attenuation is even less. The calculations are carried out subject to the condition (26), but in respect of the order of magnitude the result (33) is valid also if  $p_F v_0 \sim \Delta_0$ ; even in this case the attenuation remains weak.

The origin of this attenuation weakness in Eq. (33) can be understood partly if we assume that the amplitude  $W$  of decay of an oscillation quantum into two Fermi excitations depends on the angle  $\theta$  between the wave polarization represented by the vector  $v_0$  and the excitation momentum in accordance with the law  $W = W_0 \cos \theta$ . Then, the probability of decay per unit time, calculated from the familiar formula, is

$$\frac{1}{\tau} = 2\pi \sum_p |W|^2 \delta(\omega_p - 2E_p) = 2\pi |W_0|^2 \left( \gamma \frac{\pi \Delta_0}{p_F v_0} \right) v$$

[in the notation of Eq. (30)]. Here, parentheses are used to identify the quantity which acts as the effective density of states of the spectrum

$$E = [\xi^2 + \Delta_0^2 + (p_F v_0)^2]^{1/2}$$

near the edge of the spectrum. The quantity  $v$  is approximately  $(\alpha/\ln \alpha)^2$  for a free oscillation and, therefore, we can now understand how the small factor in  $i\omega_2$  appears from Eq. (33).

#### 4. RESPONSE

The most interesting are the natural and adjoining frequencies. Consequently, we shall consider low frequencies [where the natural frequency  $\omega_1$  of Eq. (27.1) is located] and high frequencies (near  $2\Delta_0$ ). It follows from the system (25) that in the low-frequency limit

$$x = \left[ 1 + \left( \frac{\Delta_0}{2\lambda\mu} \right)^2 \frac{\omega^2}{\omega_1^2 - \omega^2} \right] \frac{p_F^2 \omega(Av_{12})}{3\Delta_0^2}, \\ y = \frac{\omega^2}{\omega_1^2 - \omega^2} \frac{2(Av_{12})}{3\lambda}. \quad (34)$$

We shall now consider what happens in the limit of a static electric field ( $\omega \rightarrow 0, \omega A = \text{const}$ ). It follows from Eq. (34) that the correction to the order parameter is

$$\Delta_1 = \frac{\mu}{3\Delta_0} \frac{e\mathcal{E}v_0}{\Delta_0} \quad (35)$$

(in terms of conventional units with  $\hbar = 1$ ). Here,  $\mathcal{E}$  is the electric field.

Having made some simple calculations of the integrals in Eq. (24) subject to the conditions represented by Eq. (26) and by  $|\omega| \ll \Delta_0$ , and substituting in the resultant expression the resonant terms  $\Delta_1 \pm \Delta_1^*$  from Eq. (34), we find that approximately

$$j \approx \frac{\gamma p_F^2}{9} \frac{\omega^2}{\Delta_0^2} \left\{ A + \frac{\omega_1^2}{\omega_1^2 - \omega^2} \frac{v_0(Av_0)}{2\lambda^2 p_F^2} \right\},$$

or in terms of conventional units

$$j \approx \frac{\omega^2}{4\pi c} \frac{\epsilon - \epsilon_0}{2} \left\{ A + \frac{\omega_1^2}{\omega_1^2 - \omega^2} \frac{v_0(Av_0)}{2\lambda^2 p_F^2} \right\}, \quad (36)$$

where  $v_F = p_F/m$  and

$$\epsilon = \epsilon_0 \left[ 1 + \frac{16}{9\pi} \frac{e^2}{\epsilon_0 v_F} \left( \frac{\mu}{\Delta_0} \right)^2 \right]. \quad (37)$$

The expression for the current (36) is simplified by dropping the term  $(p_F v_0 / \Delta_0)^2$  compared with the non-resonant part and also dropping the contribution  $\Delta_1 - \Delta_1^*$  containing an additional smallness  $\lambda$ .

We shall now explain the notation introduced in Eqs. (36) and (37). The quantity  $\epsilon$  is the static permittivity of an exciton insulator in the limit  $v_{12} = 0$ , whereas  $\epsilon_0$  is the same permittivity but without allowance for the contribution of the electron-hole subsystem considered here. The required expression for the current can be obtained by relating it to the polarization  $P$  in the same subsystem:

$$j = P, \quad P = \frac{\epsilon - \epsilon_0}{4\pi} \mathcal{E},$$

which gives

$$j = \frac{\omega^2}{4\pi c} (\epsilon - \epsilon_0) A \quad (v_{12} = 0).$$

The factor  $1/2$  in Eq. (36) appears because this is the contribution made to the current only by electrons with one projection of the spin. This explains the meaning of the notation. The expression (37) is in agreement with the results obtained by Baklanov and Chaplik.<sup>7</sup>

It is appropriate to make here the following comment. The method used in the preceding section to find the natural frequencies gives, strictly speaking, the correct results only in the case of transverse oscillations ( $q \perp v_0$ ) and only in the limit  $c \rightarrow \infty$  ( $cq/\epsilon^{1/2} \gg \omega_1$ ); these

restrictions can be lifted, as is known, by solving [instead of Eq. (25) with  $A=0$ ] the Maxwell equations and using the response (36). It is generally found that the spectrum is of the polariton type; in particular, the frequency of a transverse oscillation in the limit  $cq/\epsilon^{1/2} \ll \omega_1$  is identical with the frequency  $\omega_{11}$  of a longitudinal oscillation ( $\mathbf{q} \parallel \mathbf{v}_0$ ), and in the latter case we have

$$\frac{\omega_{11}^2 - \omega_1^2}{\omega_1^2} = \frac{1}{2\lambda^2} \left( \frac{v_0}{v_F} \right)^2. \quad (38)$$

In accordance with our assumptions, this ratio is small.

We shall now go over to a study of the response at high frequencies ( $|\omega| \approx 2\Delta_0$ ). We shall first consider the case

$$\Delta_0^2 \gg |(\omega/2)^2 - \Delta_0^2| \gg (p_F v_0)^2.$$

In this limit, we obtain

$$j = \frac{\pi(p_F)^2}{12\Delta_0} \frac{\omega^2}{(\Delta_0^2 - (\omega/2)^2)^{1/2}} \left\{ A + \frac{p_F^2}{3} \frac{v_0(Av_0)}{\Delta_0^2 - (\omega/2)^2} \right\}. \quad (39)$$

The result is qualitatively valid up to

$$|(\omega/2)^2 - \Delta_0^2| \sim (p_F v_0)^2,$$

when the resonant component becomes important. We can see that the contribution of the additional degrees of freedom (i.e., the contribution of the condensate) is large in a fairly wide range of frequencies.

We shall now consider the frequency range (28). A considerable contribution to  $j_A$  originates from  $\Delta_1 + \Delta_1^*$  and, therefore, we shall give the expression only for this quantity:

$$\Delta_1 + \Delta_1^* \approx -\frac{\omega(Av_{12})}{8v_0^2} \frac{L(1+4vL)}{vL^2 - \alpha^2}, \quad (40)$$

where the notation introduced in Eqs. (29) and (30) is used. We shall give the separate expressions for  $j_A$  and  $j_\Delta$ :

$$\begin{aligned} j_\Delta &= \frac{\pi(p_F)}{8v_0\Delta_0} \omega^2 \left\{ \left( L - \frac{1}{2} \right) [A - n(An)] + n(An) \right\}, \\ j_\Delta &= -\frac{\pi(p_F)}{8v_0\Delta_0} \omega^2 \left\{ \frac{L}{8} \frac{n(An)}{vL^2 - \alpha^2} + n(An) \right\}, \end{aligned} \quad (41)$$

where  $n = v_0/v_0$ . The conversion to conventional units is made by multiplying by  $e^2/mc$ .

In the calculation of Eqs. (39)–(41) it is assumed that  $v$  is negative. The expression for the positive values of  $v$  is obtained, as before, by analytic continuation across the upper half-plane.

All that we have said so far on the determination of natural frequencies near  $\omega_1$  clearly remains valid also in the present case. Longitudinal oscillations obviously disappear because of the strong attenuation.

The expressions (39) and (41) generally resemble Eq. (36); naturally, they are somewhat more complex because the absorption edge has its effect. The common feature of these expressions is the fairly unusual angular dependence of the coefficient for the resonant part. This dependence appears because the polarization of free oscillations is constant (as pointed out above) and it is governed by the quantity  $v_0$ .

We have thus solved the problem formulated above: we have found the response [the results (36), (39), and

(41) should be multiplied only by 2 to represent both projections of the spin]. One of the intermediate results, Eq. (35), which gives the changes in the order parameter in a static electric field, is interesting.

The results obtained are typical of exciton insulators with an allowed dipole transition. Since even in the most standard part [far from the absorption edges given by Eq. (36)] the response has its specific features (an unusual angular dependence and particularly the behavior near the absorption edge), we may expect that relatively simple optical methods will enable identification of this type of substance.

## 5. CONCLUSIONS

We shall consider once again the problem of derivation of the expression for the current which has been subject recently to some controversy. Our derivation is given in Sec. 1. In fact, we have used there the effective mass approximation. The fact that there are two bands has the formal effect that the total set of the functions of the problem (apart from the spin factor) can be represented in the form

$$\begin{pmatrix} 1 \\ 0 \end{pmatrix} e^{i\mathbf{k}\mathbf{r}}, \quad \begin{pmatrix} 0 \\ 1 \end{pmatrix} e^{i\mathbf{k}\mathbf{r}},$$

which follows from the form of the Hamiltonian. It would seem that such an approach cannot cause any misunderstanding. However, Volkov and Kopaev<sup>1</sup> have suggested an explicit allowance for the fact that the electron eigenfunctions are of the Bloch type, which may give rise to an additional contribution to the current apart from that obtained in the effective mass approximation. This approach results in a contradiction, as shown in Ref. 8, where the solution to this problem is suggested.

It should be stressed that if we use a definition of the current employed in Ref. 8 and apply it particularly to the problem under discussion, the result is Eq. (7), which gives the usual expression for the electron velocity  $\mathbf{v} = \pm dE_k/d\mathbf{k}$  ( $\pm E_k$  are the energies of the upper and lower bands of an exciton insulator). Irrespective of the phase of  $\Delta$ , the total current for a filled band is zero, which formally follows from the fact that  $\mathbf{v}$  is a total derivative. This circumstance has been used earlier to analyze the expression for the current. The same conclusion is reached by Volkov *et al.*,<sup>3</sup> but they speak of cancellation of the interband and intraband contributions [see comments after Eq. (4) in Ref. 3]. Apart from the terminology, the point of view of Volkov *et al.*<sup>3</sup> is basically in agreement with ours in respect of the absence of a spontaneous homogeneous current.

We shall now consider the role of other (apart from  $\mathbf{p} \cdot \mathbf{v}_{12}$ ) mechanisms of fixation of the phase of the order parameter  $\Delta$  ignored by us. For an exciton insulator with  $v_{12}=0$  these mechanisms also have the effect that the acoustic branch begins from a finite frequency<sup>4,5</sup>; however, if  $q=0$  such an oscillation cannot be accompanied by the current because the vector cannot be directed anywhere and, therefore, the response is not sensitive to such an oscillation. However, in the case of a substance with a finite value of  $v_{12}$  we can have a

current, as shown above, and consequent resonant behavior of the response. Therefore, it is clear that other mechanisms of phase fixation can hardly change anything in principle, apart from resulting in, for example, some renormalization of the frequency  $\omega_1$  of free oscillations. The frequency  $\omega_2$  is found to be relatively insensitive to the additional interaction fixing the phase, because this frequency corresponds (as pointed out in Sec. 3) to an oscillation mainly of the modulus of  $\Delta$ .

The authors are grateful to I. A. Gilinskii for valuable comments. One of the authors (E.G.B.) is grateful to B. A. Volkov and Yu. V. Kopaev for discussing the preliminary results.

<sup>1</sup>B. A. Volkov and Yu. V. Kopaev, Pis'ma Zh. Eksp. Teor. Fiz. 27, 10 (1978) [JETP Lett. 27, 7 (1978)].

- <sup>2</sup>B. A. Volkov, Yu. V. Kopaev, and V. V. Tugushev, Pis'ma Zh. Eksp. Teor. Fiz. 27, 615 (1978) [JETP Lett. 27, 582 (1978)].
- <sup>3</sup>B. A. Volkov, Yu. V. Kopaev, M. S. Nunuparov, and V. V. Tugushev, Pis'ma Zh. Eksp. Teor. Fiz. 30, 317 (1979) [JETP Lett. 30, 293 (1979)].
- <sup>4</sup>A. N. Kozlov and L. A. Maksimov, Zh. Eksp. Teor. Fiz. 49, 1284 (1965) [Sov. Phys. JETP 22, 889 (1966)].
- <sup>5</sup>A. V. Klyuchnik and Yu. E. Lozovik, Zh. Eksp. Teor. Fiz. 76, 670 (1979) [Sov. Phys. JETP 49, 335 (1979)].
- <sup>6</sup>P. W. Anderson, Phys. Rev. 112, 1900 (1958) (Russ. Transl. in: Teoriya sverkhprovodimosti, III, M., 1960, p. 285).
- <sup>7</sup>E. V. Baklanov and A. V. Chaplik, Fiz. Tverd. Tela (Leningrad) 7, 2768 (1965) [Sov. Phys. Solid State 7, 2240 (1966)].
- <sup>8</sup>E. G. Batyev, Pis'ma Zh. Eksp. Teor. Fiz. 29, 381 (1979) [JETP Lett. 29, 345 (1979)].

Translated by A. Tybulewicz

## Experimental determination of the critical exponent and of the asymmetric and nonasymptotic corrections to the equation of the coexistence curve of Freon-113

E. T. Shimanskaya, I. V. Bezruchko, V. I. Basok, and Yu. I. Shimanskii

Kiev State University

(Submitted 17 June 1980)

Zh. Eksp. Teor. Fiz. 80, 274–292 (January 1981)

The coexistence curve (CC) of Freon-113 was experimentally investigated in a wide range of temperatures, including the vicinity of the critical point, for the purpose of checking on new theories of critical phenomena, the results of which are presented in the form of extended expansions. It is shown that the CC can be described by a formula with not less than four terms, having different forms for the liquid and gas branches. Statistical reduction of the experimental data on the CC yields the exponents and the coefficient of such a formula. It is established that the expressions for the symmetric and asymmetric terms are the same for the liquid and gas branches. The validity of zero-order symmetric scaling is proved. A singularity is found in the CC "diameter." From a comparison of the experimental data with the extended nonanalytic theories it is deduced that the existing theories agree with experiment only qualitatively.

PACS numbers: 64.60.Fr

### 1. INTRODUCTION

In the last decade much progress was made in the theoretical research into the critical state<sup>1</sup> and this stimulated a number of experimental studies, which are reviewed in Refs. 2 and 3. Methods were developed<sup>4–10</sup> for the construction of an extended nonanalytic theory of critical phenomena. The results are presented in the form of series whose leading terms correspond to zero-order scaling. The asymmetric terms take into account the difference between a real liquid + gas system and idealized models, and when used together with nonasymptotic symmetrical terms they enlarge the describable vicinity of the critical points. Various suggestions are encountered in the theoretical papers concerning the numerical values of the exponents of the correction terms, but the values of the coefficients do not lend themselves so far to theoretical calculation.

To find the true equation of state in a large vicinity of the critical point it becomes urgent, besides the experimental determination of the leading (limiting, zero-order) terms, to obtain by experiment the succeeding terms of the expansion, and this calls for a highly accurate study of the behavior of the medium in a larger vicinity of the critical point. In particular, in the study of the liquid–vapor coexistence curve (CC), besides the determination of the leading terms,<sup>2,3,11–15</sup> attempts were already made to find the first nonasymptotic and asymmetric corrections.<sup>16–20</sup> It is obvious that the experimentally determined form of the correction terms can be strongly influenced by even an insignificant error made when choosing the leading term of the expansion. However, in the experimental determination of the limiting laws frequent use is made of finite intervals of the state parameters, so that as a rule one obtains not the limiting zero-order scaling exponents, but cer-