CONTRIBUTION TO THE THEORY OF NONLINEAR EFFECTS IN CRYSTALS WITH ACCOUNT TAKEN OF PARTIALLY FILLED BANDS

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Expressions are obtained for the conductivity as a function of the external fields in the form of a series in the parameter ω/Δ (ω and Δ are the characteristic frequencies of the field and of the interband transitions). The contribution of the intraband motion and of the combined terms, which contain simultaneously intraband motion and interband transition, to the cubic conductivity is investigated in detail as a function of the carrier density in the band, the polarization, and the frequencies of the external fields. Comparison with experiment is made for mixing of three frequencies in InSb and InAs in the infrared band.

1. INTRODUCTION

IN considering the nonlinear properties of a solid, due to the electron motion, it is customary to confine oneself (see, for example[1]) to allowance of the interband transitions. However, at field frequencies ω that are small compared with the distance Δ between the "centers" of the bands ($\omega \ll \Delta$), an appreciable contribution to the nonlinear characteristics is made by the intraband motion in partially filled bands[2-5], and also under definite situations ($\omega < \Delta$) the combination of the intraband motion and interband transitions. The case of low ($\omega \ll \Delta$) and intermediate ($\omega < \Delta$) frequencies has attracted particular interest in connection with the appearance of experimental work on nonlinear effects in the infrared band[6]. In the present paper we investigate, in the electric dipole approximation, the nonlinear characteristics of semiconductors with account taken of partially filled bands. We obtain expressions for the current as a function of the external fields in the form of a series in the parameter ω/Δ with coefficients that depend on the field. We consider in detail the contribution of the partially filled bands with simultaneous allowance of both the intraband motion and the interband transitions, as functions of the frequency and polarization of the external fields and of the carrier density in the band. Estimates of the conductivity of third order in the field are presented for certain semiconductors.

2. DERIVATION OF THE EFFECTIVE HAMILTONIAN AND OF THE EXPRESSION FOR THE CURRENT DENSITY

1. We consider an electron situated in a periodic field of a crystal lattice. In the self-consistent field approximation, the Hamiltonian of the electron is of the form

$$\hat{H}_0 = \frac{1}{2m}\hat{\mathbf{p}}^2 + V(\mathbf{r}), \tag{1}$$

where m is the mass of the electron, $\hat{\mathbf{p}}$ the momentum operator, and $V(\mathbf{r})$ the self-consistent potential, which is periodic with the period of the lattice. The eigenfunctions $|n\mathbf{k}\rangle$ and the eigenvalues ϵ of the Hamiltonian (1) are respectively

$$|n\mathbf{k}\rangle = u_{n\mathbf{k}}(\mathbf{r})e^{i\mathbf{kr}}, \qquad \varepsilon = \varepsilon_n(\mathbf{k}). \tag{2}$$

Here n is an index numbering the bands, hk is the quasimomentum characterizing the state of the electron in the band, and $u_{n\mathbf{k}}(\mathbf{r})$ is the modulating Bloch factor, which is periodic with the lattice period. It is assumed below that the functions $|n\mathbf{k}\rangle$ are normalized to the volume of the crystal. In the presence of a homogeneous electric field, the Hamiltonian (1) goes over into

$$\hat{H} = \frac{1}{2m}\left(\hat{\mathbf{P}} + \frac{e}{c}\mathbf{A}(t)\right)^2 + V(\mathbf{r}), \tag{3}$$

where $\hat{\mathbf{P}}$ is the canonical momentum conjugate to the coordinate $\mathbf{r}$, $\mathbf{A}(t)$ is the vector potential of the field, e is the electron charge, and c is the speed of light.

The solution of the Schrödinger equation

$$i\hbar\partial\psi/\partial t = \hat{H}\psi \tag{4}$$

with Hamiltonian (3) will be sought in the form of the expansion

$$\psi = \sum_{n\mathbf{k}} \psi_{n\varkappa}(\mathbf{r})\, c_n(\mathbf{k}t) \tag{5}$$

in the base system of functions

$$\psi_{n\varkappa}(\mathbf{r}) = e^{i\mathbf{kr}}u_{n\varkappa}(\mathbf{r}), \tag{6}$$

where

$$\varkappa = \mathbf{k} + \frac{e}{\hbar c}\mathbf{A}(t). \tag{7}$$

The chosen system of functions (6) is complete and orthonormal. Indeed, this system of functions coincide at any fixed instant of time, apart from a phase factor, with the eigenfunctions (2). We note that it is precisely this phase factor that distinguishes the functions (6) from the Houston functions[7]. Substituting (5) and (6) in (4) and taking into account the fact that the basis system (6) is orthonormal, we get

$$i\hbar\frac{\partial c_n(\mathbf{k}t)}{\partial t} = \sum_{n'}\left[\varepsilon_n(\varkappa)\delta_{nn'} + ie\Omega_{nn'}(\varkappa)\mathbf{E}(t)\right]c_{n'}(\mathbf{k}t), \tag{8}$$

where

$$\Omega_{nn'}(\varkappa) = \frac{1}{v}\int_v u^*_{n\varkappa}(\mathbf{r})\nabla_{\mathbf{k}}u_{n'\varkappa}(\mathbf{r})\,d\mathbf{r}, \tag{9}$$

and v is the volume of the unit cell of the crystal, $\mathbf{E}(t)$ the electric field, which is connected with $\mathbf{A}(t)$ in the

usual manner ($\dot{\mathbf{A}}(t) = -c\mathbf{E}(t)$). For what follows it is convenient to write (8) in operator form

$$i\hbar \frac{\partial c(\mathbf{k}t)}{\partial t} = [\hat{H}_0(\boldsymbol{\varkappa}) + \hat{H}(\boldsymbol{\varkappa}t)]\, c(\mathbf{k}t). \tag{10}$$

The form of the operators in (10) is determined by their matrix elements (8), and in matrix notation the operator wave function $c(\mathbf{k}t)$ is a column matrix, and the operators $H_0(\boldsymbol{\kappa})$ and $\hat{H}(\boldsymbol{\kappa}t)$ in (10) correspond to the first and second terms of (8). To find the effective Hamiltonian we carry out in (10) a unitary transformation

$$\hat{S}^+(\boldsymbol{\varkappa}t)\hat{S}(\boldsymbol{\varkappa}t) = \hat{1}, \tag{11}$$

such that the nondiagonal matrix elements of the operator

$$\hat{H}'(\boldsymbol{\varkappa}t) = \hat{S}^+(\boldsymbol{\varkappa}t)[\hat{H}_0(\boldsymbol{\varkappa}) + \hat{H}(\boldsymbol{\varkappa}t)]\,\hat{S}(\boldsymbol{\varkappa}t) + ie\hat{S}^+(\boldsymbol{\varkappa}t)\nabla_{\boldsymbol{\varkappa}}\hat{S}(\boldsymbol{\varkappa}t)\mathbf{E}(t) - i\hbar\hat{S}^+(\boldsymbol{\varkappa}t)\partial\hat{S}(\boldsymbol{\varkappa}t)/\partial t \tag{12}$$

vanish identically when $n \neq n'$, i.e.,

$$H'_{nn'}(\boldsymbol{\varkappa}t) = 0 \qquad (n \neq n'). \tag{13}$$

Then, introducing the wave function $\varphi_n(\mathbf{k}t)$ in the new representation in accordance with the equality

$$c_n(\mathbf{k}t) = \sum_{n'} S_{nn'}(\boldsymbol{\varkappa}t)\varphi_{n'}(\mathbf{k}t), \tag{14}$$

we find that the equation for $\varphi_n(\mathbf{k}t)$, with account of (13), takes the form

$$i\hbar\partial\varphi_n(\mathbf{k}t)/\partial t = \tilde{\varepsilon}_n(\boldsymbol{\varkappa}t)\varphi_n(\mathbf{k}t), \tag{15}$$

where $\tilde{\varepsilon}_n(\boldsymbol{\kappa}t)$ is the diagonal matrix element of the operator (12) which plays the role of the effective Hamiltonian. Its form will be determined later.

The problem has now been reduced to a determination of the explicit form of the operator $\hat{S}(\boldsymbol{\kappa}t)$. We note first that the operator equations (11) and (12), which define $\hat{S}(\boldsymbol{\kappa}t)$, are valid for all values of $\boldsymbol{\kappa}$ ($\boldsymbol{\kappa}$ enters as a parameter), and consequently also when $\boldsymbol{\kappa}$ is equal to $\mathbf{k}$. This circumstance allows to solve Eqs. (11) and (12) for $\boldsymbol{\kappa}$ equal to $\mathbf{k}$, and then find $\hat{S}(\boldsymbol{\kappa}t)$ by simple substitution $\mathbf{k} \to \boldsymbol{\kappa}$ in the solution. The solution of (12) for $\boldsymbol{\kappa} = \mathbf{k}$ will be sought in the form of an expansion in powers of the electric field $\mathbf{E}(t)$

$$\hat{S}(\mathbf{k}t) = \hat{1} + \hat{S}_1(\mathbf{k}t) + \hat{S}_2(\mathbf{k}t) + \ldots, \tag{16}$$

where $\hat{1}$ is a unit operator, and $\hat{S}_j(\mathbf{k}t)$ is an operator of order j in the field. Substituting the expansions (16) in (11), we have, omitting for brevity the arguments of $\hat{S}_j(\mathbf{k}t)$,

$$\begin{aligned} &\hat{S}_1^+ + \hat{S}_1 = 0, \quad \hat{S}_2^+ + \hat{S}_1^+\hat{S}_1 + \hat{S}_2 = 0, \\ &\hat{S}_3^+ + \hat{S}_2^+\hat{S}_1 + \hat{S}_1^+\hat{S}_2 + \hat{S}_3 = 0, \ldots, \end{aligned} \tag{17}$$

and from (12), (16), and (17) we get

$$\hat{H}_0'(\mathbf{k}t) = \hat{H}_0(\mathbf{k}), \tag{18}$$

$$\hat{H}_1'(\mathbf{k}t) = [\hat{H}_0(\mathbf{k}), \hat{S}_1] + \hat{H}(\mathbf{k}t) - i\hbar\partial\hat{S}_1/\partial t, \tag{19}$$

$$\hat{H}_2'(\mathbf{k}t) = [\hat{H}_0(\mathbf{k}), \hat{S}_2] + \hat{H}(\mathbf{k}t)\hat{S}_1 + ie\nabla_{\mathbf{k}}\hat{S}_1\mathbf{E}(t) - \hat{S}_1\{\hat{H}(\mathbf{k}t)\}_d - i\hbar\partial\hat{S}_2/\partial t, \tag{20}$$

$$\hat{H}_3'(\mathbf{k}t) = [\hat{H}_0(\mathbf{k}), \hat{S}_3] + \hat{H}(\mathbf{k}t)\hat{S}_2 + ie\nabla_{\mathbf{k}}\hat{S}_2\mathbf{E}(t) - \hat{S}_2\{\hat{H}(\mathbf{k}t)\}_d - \hat{S}_1\{\hat{H}(\mathbf{k}t)\hat{S}_1 - i\hbar\partial\hat{S}_2/\partial t\}_d - i\hbar\partial\hat{S}_3/\partial t \tag{21}$$

etc. Here $\hat{H}_j'(\mathbf{k}t)$ is the term of j-th order in the expansion of $\hat{H}'$ in the field, and the symbol $\{\ \}_d$ shows that we take the diagonal part of the corresponding operator.

It should be noted that (11) and (12) determine only the nondiagonal elements of the operator $\hat{S}$. To determine the diagonal elements it is necessary to satisfy an additional condition, which is conveniently chosen in the form

$$S_{nn}^+ = S_{nn}. \tag{22}$$

Again using (16), we get from (17) and (22) for the diagonal elements of the expansion of $\hat{S}$ in a series in the field

$$S_{1nn} = 0, \quad S_{2nn} = -\tfrac{1}{2}(\hat{S}_1^+\hat{S}_1)_{nn}, \quad S_{3nn} = -\tfrac{1}{2}(\hat{S}_2^+\hat{S}_1 + \hat{S}_1^+\hat{S}_2)_{nn}, \ldots \tag{23}$$

Equations (18)–(21) and (23) together with (13), turn out to be sufficient to find the matrices $\hat{S}_j$. For $S_{1nn'}$, for example, we have from (13) and (19)

$$i\hbar\partial S_{1nn'}/\partial t = \hbar\omega_{nn'}(\mathbf{k})S_{1nn'} + H_{nn'}(\mathbf{k}t), \tag{24}$$

where $\hbar\omega_{nn'}(\mathbf{k})$ equals $\epsilon_n(\mathbf{k}) - \epsilon_{n'}(\mathbf{k})$. The solution of (24) under condition of adiabatic application of the field and allowance for the explicit form of $H_{nn'}(\mathbf{k}t)$ is

$$S_{1nn'}(\mathbf{k}t) = -\frac{ie}{\hbar}\frac{\Omega^{\alpha}_{nn'}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) - \omega}E_{\alpha}(\omega)e^{-i\omega t}, \tag{25}$$

where α numbers the projections on the coordinate axes, and $E_{\alpha}(\omega)$ are the amplitudes of the harmonics of the field. In (25) and below we sum over the coinciding indices α and frequencies ω. We obtain in similar fashion the matrices $S_{2nn'}$, etc.:

$$S_{2nn'}(\mathbf{k}t) = -\frac{e^2}{\hbar^2}\Bigg[\sum_{n''\neq n'}\frac{\Omega^{\alpha_1}_{nn''}(\mathbf{k})\Omega^{\alpha_2}_{n''n'}(\mathbf{k})}{\omega_{n''n'}(\mathbf{k}) - \omega_2} + \nabla_{k_{\alpha_1}}\left\{\frac{\Omega^{\alpha_2}_{nn'}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) - \omega_2}\right\} - \frac{\Omega^{\alpha_1}_{n'n'}(\mathbf{k})\Omega^{\alpha_2}_{nn'}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) - \omega_2}\Bigg]\frac{1}{\omega_{nn'}(\mathbf{k}) - \omega_1 - \omega_2}E_{\alpha_1}(\omega_1)E_{\alpha_2}(\omega_2)e^{-i(\omega_1+\omega_2)t}. \tag{26}$$

Using (11), (18)–(21) with allowance for (23), (25), and (26), it is now easy to obtain the explicit form of the effective Hamiltonian

$$H'_{nn}(\mathbf{k}t) \equiv \tilde{\varepsilon}_n(\mathbf{k}t) = \varepsilon_n(\mathbf{k}) + ie\Omega^{\alpha}_{nn}(\mathbf{k})E_{\alpha}(\omega)e^{-i\omega t}$$

$$-\frac{e^2}{2\hbar}\sum_{n'\neq n}\left[\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})\Omega^{\alpha_2}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) + \omega_2} + \frac{\Omega^{\alpha_2}_{nn'}(\mathbf{k})\Omega^{\alpha_1}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) - \omega_2}\right]E_{\alpha_1}(\omega_1)E_{\alpha_2}(\omega_2)e^{-i(\omega_1+\omega_2)t}$$

$$-\frac{ie^3}{2\hbar^2}\Bigg\{\sum_{\substack{n'\neq n\\ n''\neq n}}\Bigg[\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})\Omega^{\alpha_2}_{n'n''}(\mathbf{k})\Omega^{\alpha_3}_{n''n}(\mathbf{k})}{(\omega_{nn'}(\mathbf{k}) - \omega_1)(\omega_{nn''}(\mathbf{k}) - \omega_1 - \omega_2)} + \frac{\Omega^{\alpha_3}_{nn'}(\mathbf{k})\Omega^{\alpha_2}_{n'n''}(\mathbf{k})\Omega^{\alpha_1}_{n''n}(\mathbf{k})}{(\omega_{nn'}(\mathbf{k}) + \omega_1 + \omega_2)(\omega_{nn''}(\mathbf{k}) + \omega_1)}\Bigg]$$

$$-\sum_{n'\neq n}\Bigg[\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})\Omega^{\alpha_2}_{n'n}(\mathbf{k})\Omega^{\alpha_3}_{nn'}(\mathbf{k})}{(\omega_{nn'}(\mathbf{k}) - \omega_1)(\omega_{nn'}(\mathbf{k}) - \omega_1 - \omega_3)} + \frac{\Omega^{\alpha_2}_{nn'}(\mathbf{k})\Omega^{\alpha_1}_{n'n}(\mathbf{k})\Omega^{\alpha_3}_{nn'}(\mathbf{k})}{(\omega_{nn'}(\mathbf{k}) + \omega_1)(\omega_{nn'}(\mathbf{k}) + \omega_1 + \omega_3)}$$

$$-\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) + \omega_2 + \omega_3}\nabla_{k_{\alpha_2}}\left\{\frac{\Omega^{\alpha_3}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) + \omega_3}\right\} + \nabla_{k_{\alpha_2}}\left\{\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) - \omega_3}\right\}\frac{\Omega^{\alpha_3}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k}) - \omega_2 - \omega_3}\Bigg]\Bigg\}$$

$$\times E_{\alpha_1}(\omega_1)E_{\alpha_2}(\omega_2)E_{\alpha_3}(\omega_3)\exp[-i(\omega_1+\omega_2+\omega_3)t] \tag{27}$$

It is interesting to note that for the case of zero field frequencies, any terms in (27) proportional to some power of the field coincides with the corresponding en-

ergy correction that follows from the stationary perturbation theory. Further, in order to go over from (27) to the operator entering in (15), it is sufficient to replace k in (27) by κ. With this, of course, the individual terms of (27) are no longer the terms of the expansion of a certain operator in powers of the field, since the coefficients at the powers of the field in (27) depend themselves on the field via κ, and the contribution, say, to the expression of third order in the field will be made not only by the last term of (27) but also by the first three. However, an expansion of the type (16) and an expansion in powers of the field of the coefficients in (27), which depend on the field via κ, are expansions in terms of different parameters. Indeed, comparing the different terms of the expansion in (25) and (26) when κ is equal to k, we find that the parameter in the expansion of the type (16) is

$$\delta \sim \frac{\omega}{\Delta}\frac{ea}{\hbar\omega}E. \tag{28}$$

The estimate (28) is obtained by putting $\Omega^{\alpha}_{nn'}(k) \sim a$, where a is the lattice constant.

As to the expansion of the coefficients in (27), which depend on the field via κ, the corresponding parameter differs from (28) by the factor ω/Δ. The latter can be readily verified by expanding an arbitrary coefficient $f(\kappa)$ in (27) in terms of the field at the point $\kappa = k$ and substituting division by the characteristic interval of the variation of the function (a^{-1}) for differentiation in the estimate

$$f(\varkappa) \sim f(k)\sum_{j=0}^{\infty}\frac{1}{j!}\left(\frac{ea}{\hbar\omega}\right)^{j}E^{j}. \tag{29}$$

Thus, when the substitution $k \to \kappa$ is made, each of the terms (25)—(27) contains all the powers of the field, but the relative magnitude of the terms in each of the expressions (25)—(27) is determined by the parameter ω/Δ. This result can be obtained also by successively expanding in terms of the field and subsequent summation of the terms of the same order in ω/Δ. In particular, it is precisely in this manner that we obtained earlier[2] the effective Hamiltonian in the zeroth order in ω/Δ, but containing all the powers of the field (the first two terms in (27)).

2. To consider nonlinear effects it is necessary to have, besides the effective Hamiltonian (27), also expressions for the current density in the representation of the operator $\tilde{\varepsilon}(\kappa t)$. In accordance with the usual definition of the current density, we have

$$\mathbf{j} = -\frac{e}{V}\frac{d\langle \mathbf{r}\rangle}{dt}, \tag{30}$$

where V is the volume of the system under consideration, $\langle \mathbf{r}\rangle$ is the mean value of electron-coordinate operator. In the representation (6), the matrix elements of the operator $\mathbf{r}$ is given by

$$\mathbf{r}_{nk\,n'k'} = i\delta_{nn'}\delta(\mathbf{k}-\mathbf{k}')\nabla_{\mathbf{k}} + i\Omega_{nn'}(\varkappa)\delta(\mathbf{k}-\mathbf{k}'). \tag{31}$$

Relation (31) is obtained in perfect analogy with the procedure used in[8] to find the matrix element of the coordinate in terms of the Bloch functions (2). Taking (31), (14), and (15) into account, we find from (30) that the expression for the current density can be represented in the form

$$\mathbf{j} = -\frac{e}{\hbar V}\sum_{nk} f_n(\mathbf{k})\frac{\partial\tilde{\varepsilon}_n(\varkappa t)}{\partial \mathbf{k}} + \mathbf{P}, \tag{32}$$

where $f_n(k)$ is the distribution function over the states prior to turning on the electric field. The first term in (32) is the drift current, whereas the second term can be interpreted as the polarization current. The quantity $\mathbf{P}$ in (32) is the dipole moment per unit volume, due to polarization of the medium in the presence of an external field:

$$\mathbf{P} = -\frac{ie}{V}\sum_{nn'\mathbf{k}} f_n(\mathbf{k})S^{+}_{nn'}(\varkappa t)[\Omega_{n'n''}(\varkappa) + \delta_{n'n''}\nabla_{\mathbf{k}}]S_{n''n}(\varkappa t). \tag{33}$$

If we fix κ and use the expansion (16) (expansion in the parameter (28)), then the polarization P can be represented in the form

$$\mathbf{P} = \mathbf{P}_0 + \mathbf{P}_1 + \mathbf{P}_2 + \ldots, \tag{34}$$

where

$$\mathbf{P}_0 = -\frac{ie}{V}\sum_{nk} f_n(\mathbf{k})\Omega_{nn}(\varkappa), \tag{35}$$

$$\mathbf{P}_1 = -\frac{ie}{V}\sum_{nn'\mathbf{k}} f_n(\mathbf{k})[S^{+}_{1nn'}\Omega_{n'n}(\varkappa) + \Omega_{nn'}(\varkappa)S_{1n'n}], \tag{36}$$

$$\mathbf{P}_2 = -\frac{ie}{V}\sum_{nn'n''\mathbf{k}} f_n(\mathbf{k})\Big[S^{+}_{2nn'}\Omega_{n'n}(\varkappa)\delta_{n''n} + \delta_{nn'}\Omega_{nn'}(\varkappa)S_{2n'n} + S^{+}_{1nn'}\Omega_{n'n''}(\varkappa)S_{1n''n} + \frac{1}{2}(S^{+}_{1nn'}\nabla_{\mathbf{k}}[S_{1n'n}] - \nabla_{\mathbf{k}}[S^{+}_{1nn'}]S_{1n'n})\delta_{n''n}\Big] \tag{37}$$

$$\mathbf{P}_3 = -\frac{ie}{V}\sum_{nn'n''\mathbf{k}} f_n(\mathbf{k})\Big[S^{+}_{3nn'}\Omega_{n'n}(\varkappa)\delta_{n''n} + \Omega_{nn'}(\varkappa)S_{3n'n}\delta_{n''n} + S^{+}_{2nn'}\Omega_{n'n''}(\varkappa)S_{1n''n} + S^{+}_{1nn'}\Omega_{n'n''}(\varkappa)S_{2n''n} + \frac{1}{2}(S^{+}_{2nn'}\nabla_{\mathbf{k}}[S_{1n'n}] + S^{+}_{1nn'}\nabla_{\mathbf{k}}[S_{2n'n}] - \nabla_{\mathbf{k}}[S^{+}_{1nn'}]S_{2n'n} - \nabla_{\mathbf{k}}[S^{+}_{2nn'}]S_{1n'n})\delta_{n''n}\Big]. \tag{38}$$

etc.

The explicit form of expressions (34)—(38) can be obtained by using (23), (25), and (26). For example, for (36) we have

$$\mathbf{P}_1 = \frac{e^2}{\hbar V}\sum_{nn'\neq n\mathbf{k}} f_n(\mathbf{k})\left[\frac{\Omega_{nn'}(\varkappa)\Omega^{\alpha_1}_{n'n}(\varkappa)}{\omega_{nn'}(\varkappa)+\omega_1} + \frac{\Omega^{\alpha_1}_{nn'}(\varkappa)\Omega_{n'n}(\varkappa)}{\omega_{nn'}(\varkappa)-\omega_1}\right]E_{\alpha_1}(\omega_1)e^{-i\omega_1 t}, \tag{39}$$

where $\mathbf{P}_1$ contains, owing to the dependence on κ, all the powers of the field, starting with the first, but is a term of order ω/Δ compared with $\mathbf{P}_0$. This can be readily verified by comparing identical terms of the expansion in powers of the field in $\mathbf{P}_0$ and $\mathbf{P}_1$. Comparing also (36) with (37), (37) with (38), etc., we can easily verify that

$$P_1/P_0 \approx P_2/P_1 \approx \ldots \approx P_n/P_{n-1} \approx \omega/\Delta, \tag{40}$$

where P_j are the moduli of the corresponding vectors $\mathbf{P}_j$.

Thus, the expression (32) for the current density with allowance for the expressions (27) and (34)—(38) is represented in the form of an expansion in the parameter ω/Δ, the coefficients of the series containing all the powers of the electric field, owing to their dependence on κ, and being exact sums of the series of the type (29), which represent expansion in the parameter $\hbar\omega/ea$. Consequently, if we adhere to the well-known definition of the conductivities[1,4] as tensor coefficients at the corresponding powers of the field, then we find, by expanding (32) in powers of the field, that the conductivity linear in the field is given by

$$\sigma_{\alpha\alpha_1}(\omega)=\frac{ie^2}{V\hbar^2\omega}\sum_{n\mathbf{k}} f_n(\mathbf{k})\frac{\partial^2\varepsilon_n(\mathbf{k})}{\partial k_\alpha\partial k_{\alpha_1}}+\frac{ie^2}{V\hbar}\sum_{n\mathbf{k}} f_n(\mathbf{k})\left(\frac{\partial\Omega^{\alpha}_{nn}(\mathbf{k})}{\partial k_{\alpha_1}}-\frac{\partial\Omega^{\alpha_1}_{nn}(\mathbf{k})}{\partial k_\alpha}\right)$$

$$-\frac{ie^2\omega}{V\hbar}\sum_{nn'\neq n\mathbf{k}} f_n(\mathbf{k})\left[\frac{\Omega^{\alpha}_{nn'}(\mathbf{k})\Omega^{\alpha_1}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k})+\omega}+\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})\Omega^{\alpha}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k})-\omega}\right]. \quad (41)$$

The interband transitions (the third sum) in (41) are not connected in any way with the intraband motion represented by the first two terms, and the second of them vanishes in the most important case of a distribution function that is even in $\mathbf{k}$, inasmuch as $\boldsymbol{\Omega}_{nn}(\mathbf{k}) = \boldsymbol{\Omega}_{nn}(-\mathbf{k})$. It is particularly easy to show, with the linear conductivity as an example, that the contribution to the conductivity, connected with the intraband motion, vanishes identically for completely filled bands, as follows from the fact that $\epsilon_n(\mathbf{k})$ and $\boldsymbol{\Omega}_{nn'}(\mathbf{k})$ are periodic in $\mathbf{k}$. Such a situation takes place also in the general case, the dependence of the effective Hamiltonian and of the polarization on κ becomes insignificant, and all the expressions for the conductivity go over into the well known results obtained neglecting the intraband motion.

As to the tensor of second order in the field, without writing it out explicitly[1], we note only that besides the terms connected with the intraband motion and the interband transitions, it is determined also by the terms containing simultaneously both interband transitions and intraband motion. An analogous structure is possessed also by the third-order conductivity.

In connection with experiment[6], it is of interest to consider the contribution made to the cubic conductivity by partially filled bands. Leaving out the cumbersome expression for $\sigma_{\alpha\alpha_1\alpha_2\alpha_3}(\omega_1\omega_2\omega_3)$, we confine ourselves here to the terms that are determined by the intraband motion[2]

$$\sigma_{\alpha\alpha_1\alpha_2\alpha}(\omega_1,\omega_2,\omega_3)=-\frac{ie^4}{V\hbar^4 3!}\sum_{n\mathbf{k}} f_n(\mathbf{k})\frac{\partial^4\varepsilon_n(\mathbf{k})}{\partial k_\alpha\partial k_{\alpha_1}\partial k_{\alpha_2}\partial k_{\alpha_3}}\frac{1}{\omega_1\omega_2\omega_3} \quad (42)$$

and by the combination of the intraband motion and the interband transitions

$$\sigma_{\alpha\alpha_1\alpha_2\alpha_3}(\omega_1,\omega_2,\omega_3)=\frac{ie^4}{\hbar^3V3!}\sum_P P(\alpha_1\omega_1,\alpha_2\omega_2,\alpha_3\omega_3)\sum_{nn'\neq n\mathbf{k}} f_n(\mathbf{k})$$

$$\times\left\{\frac{\Omega^{\alpha_2}_{nn'}\Omega^{\alpha_3}_{n'n}\nabla_{k_\alpha}[\nabla_{k_{\alpha_1}}[\omega_{nn'}(\mathbf{k})]]}{(\omega_{nn'}(\mathbf{k})+\omega_2)(\omega_{nn'}(\mathbf{k})-\omega_3)\omega_1}+\left[\frac{\Omega^{\alpha_2}_{nn'}\Omega^{\alpha_3}_{n'n}}{(\omega_{nn'}(\mathbf{k})+\omega_3)(\omega_{nn'}(\mathbf{k})-\omega_1-\omega_2)}\right.\right.$$

$$\left.-\frac{\Omega^{\alpha_1}_{nn'}\Omega^{\alpha_2}_{n'n}}{(\omega_{nn'}(\mathbf{k})-\omega_3)(\omega_{nn'}(\mathbf{k})+\omega_1+\omega_2)}\right]\frac{\nabla_{k_{\alpha_1}}[\omega_{nn'}(\mathbf{k})]\nabla_{k_\alpha}[\omega_{n'n}(\mathbf{k})]}{\omega_1^2}$$

$$+\frac{(\omega_1+\omega_2+\omega_3)}{2\omega_2\omega_3}\left[\frac{\Omega^{\alpha}_{nn'}\Omega^{\alpha_1}_{n'n}}{(\omega_{nn'}(\mathbf{k})+\omega_1)(\omega_{nn'}(\mathbf{k})+\omega_1+\omega_2+\omega_3)}\right.$$

$$\left.+\frac{\Omega^{\alpha_1}_{nn'}\Omega^{\alpha}_{n'n}}{(\omega_{nn'}(\mathbf{k})-\omega_1)(\omega_{nn'}(\mathbf{k})-\omega_1-\omega_2-\omega_3)}\right]\nabla_{k_{\alpha_2}}\nabla_{k_{\alpha_3}}[\omega_{n'n}(\mathbf{k})]$$

$$-\frac{(\omega_1+\omega_2+\omega_3)}{\omega_2\omega_3}\left[\frac{\Omega^{\alpha_1}_{nn'}\Omega^{\alpha}_{n'n}\nabla_{k_{\alpha_2}}[\omega_{nn'}(\mathbf{k})]\nabla_{k_{\alpha_3}}[\omega_{n'n}(\mathbf{k})]}{(\omega_{nn'}(\mathbf{k})-\omega_1)(\omega_{nn'}(\mathbf{k})-\omega_1-\omega_2)(\omega_{nn'}(\mathbf{k})-\omega_1-\omega_2-\omega_3)}\right.$$

$$\left.\left.+\frac{\Omega^{\alpha}_{nn'}\Omega^{\alpha_1}_{n'n}\nabla_{k_{\alpha_2}}[\omega_{nn'}(\mathbf{k})]\nabla_{k_{\alpha_1}}[\omega_{n'n}(\mathbf{k})]}{(\omega_{nn'}(\mathbf{k})+\omega_1)(\omega_{nn'}(\mathbf{k})+\omega_1+\omega_2)(\omega_{nn'}(\mathbf{k})+\omega_1+\omega_2+\omega_3)}\right]\right\}. \quad (43)$$

Here $\sum_P P(\ldots)$ is the sum of permutations of the quantities $\alpha_1\omega_1$, $\alpha_2\omega_2$, and $\alpha_3\omega_3$. The expression (43) is written out in the approximation in which the matrix elements $\boldsymbol{\Omega}_{nn'}(\mathbf{k})$ depend little on $\mathbf{k}$, and the distances between the bands are small compared with the width of the bands, as is the case for a semiconductor of InSb type.

3. Deferring the detailed discussion of expressions (42) and (43) to the next section, we note here that in the case of rectification, in an arbitrary order in the field (the sum of the external frequencies is equal to zero), direct current is produced in substances with partly filled bands, and this current is expressed only in terms of $\epsilon_n(\kappa t)$ and its derivatives. Indeed, the dc component of the polarization makes no contribution to the current (32) (it vanishes upon differentiation with respect to time), and the expression for the rectified current in arbitrary order in the field takes the form

$$\mathbf{j}=-\frac{e}{\hbar V}\sum_{n\mathbf{k}} f_n(\mathbf{k})\frac{\partial\varepsilon_n(\kappa t)}{\partial\mathbf{k}},\qquad \sum_l \omega_l=0. \quad (44)$$

It is sufficient to carry out the summation in (44) over the partially filled bands, since for the filled band ($f_n(\mathbf{k}) = 1$) the sum over $\mathbf{k}$ in (44) vanishes identically since $\tilde{\epsilon}_n(\kappa t)$ is periodic with the reciprocal-lattice period.

The expression for the current which is quadratic in the field ($\omega_1 = -\omega_2$) is described by the tensor

$$\sigma_{\alpha\alpha_1\alpha_2}(\omega_1,-\omega_1)=\frac{e^3}{2\hbar^2V}\sum_{n\mathbf{k}} f_n(\mathbf{k})\left[\left(\frac{\partial^2\Omega^{\alpha_1}_{nn}(\mathbf{k})}{\partial k_\alpha\partial k_{\alpha_2}}-\frac{\partial^2\Omega^{\alpha_2}_{nn}(\mathbf{k})}{\partial k_\alpha\partial k_{\alpha_1}}\right)\frac{1}{\omega_1}\right.$$

$$\left.+\sum_{n'\neq n}\nabla_{k_\alpha}\left(\frac{\Omega^{\alpha_1}_{nn'}(\mathbf{k})\Omega^{\alpha_2}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k})-\omega_1}+\frac{\Omega^{\alpha_2}_{nn'}(\mathbf{k})\Omega^{\alpha_1}_{n'n}(\mathbf{k})}{\omega_{nn'}(\mathbf{k})+\omega_1}\right)\right]. \quad (45)$$

For degenerate semiconductors without a symmetry center, going over in (45) from summation to integration, we have after simple transformations

$$\sigma_{\alpha\alpha_1\alpha_2}(\omega_1,-\omega_1)=\frac{e}{2}\sum_n\int_{\alpha_1\alpha_2}^{|n\mathbf{k}_F\rangle}(\omega_1)\frac{v_n^{\alpha}(\mathbf{k}_F)}{|v_n(\mathbf{k}_F)|}\,dS_F^n, \quad (46)$$

where

$$\chi_{\alpha_1\alpha_2}^{|n\mathbf{k}_F\rangle}(\omega_1)=-\sigma_{\alpha_1\alpha_2}^{|n\mathbf{k}_F\rangle}(\omega_1)/\,i\omega_1$$

is the linear susceptibility of the electron in the state $|n\mathbf{k}_F\rangle$,

$$v_n^{\alpha}(\mathbf{k}_F)=\hbar^{-1}\nabla_{k_\alpha}\varepsilon_n(\mathbf{k})\,|_{\mathbf{k}=\mathbf{k}_F}$$

is the velocity of the electron (the symbol F denotes that the integration is carried out over the Fermi surface). Thus, in the case of detection the current is expressed in terms of a suitably weighted linear conductivity.

In concluding the section, it is appropriate to note that all the obtained expressions for the conductivity admit of a simple transition to the several-band approximation. Simply discarding the high-lying bands leads to errors on the order of ω/Δ^* in the susceptibility, where Δ^* is the distance from the uppermost of the populated to the discarded bands. The proof of this statement is perfectly analogous to that given in[3] and will not be given here.

3. FEATURES OF CUBIC NONLINEARITY IN SUPERCONDUCTORS IN THE INFRARED BAND

1. In the infrared region of frequencies in semiconductors of the InSb type, there was observed a dependence of the nonlinear characteristics of the carrier density[6]. Such a dependence is connected only with the

[1] This tensor, obtained by a different manner, was considered in sufficient detail earlier [9,10].

[2] Detection and doubling in a constant field, determined by intraband motion, were considered respectively by Buĭmistrov [18] and Grinberg [19].

partly filled bands (with intraband motion and with combined terms), and the contribution of the completely filled bands can be neglected[6] (at carrier densities n in the band exceeding 10^6 cm^{-3}). This result agrees with estimates (we shall henceforth use the parameters of InSb unless otherwise stipulated) for the contribution of the interband transitions[17]

$$\sigma_{\alpha\alpha_1\alpha_2\alpha_3} = \frac{e^4\Omega^4\omega}{\hbar^3\Delta^3} n_0, \qquad (47)$$

If we assume for the characteristic distance Δ between the "centers" of the conduction and valence bands[3] the perfectly reasonable value ≈ 3 eV, and recognize that the matrix elements $e\Omega_{nn'}$ for InSb reach values 10^{-16} cgs esu[4], and $n_0 \sim 6 \times 10^{21}$ cm^{-3}.

2. The contribution of the intraband motion to the characteristics of the odd order in the field is determined, as is well known[2-5], by the law of dispersion in the band. For semiconductors of the InSb and InAs type, the dispersion law can be well approximated near the bottom of the conduction band by the expression[11]

$$\varepsilon(\mathbf{k}) = {}^1/_2\hbar\omega_0[1 + \sqrt{1 + (\mathbf{k}/k_0)^2}], \qquad k_0 = \sqrt{m^*\omega_0/2\hbar}, \qquad (48)$$

where m* is the effective mass and $\hbar\omega_0$ is the width of the forbidden band. The cubic conductivity, neglecting collisions, is determined from (42) and (48):

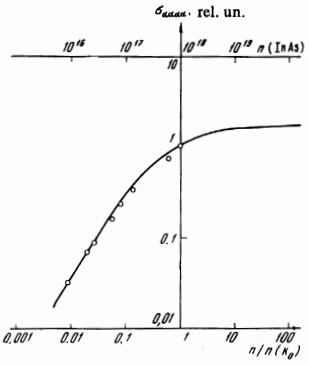
$$\sigma_{\alpha\alpha_1\alpha_2\alpha_3}(\omega_1, \omega_2, \omega_3) = -\frac{ie^4\omega_0}{2\cdot 3!\hbar^3}\frac{1}{\omega_1\omega_2\omega_3 V}\sum_{\mathbf{k}} f_n(\mathbf{k})\frac{\partial^4\sqrt{1+(\mathbf{k}/k_0)^2}}{\partial k_\alpha \partial k_{\alpha_1}\partial k_{\alpha_2}\partial k_{\alpha_3}} \qquad (49)$$

For the case of an even distribution function $f_n(\mathbf{k})$ (in particular, thermodynamic equilibrium prior to turning on the field), it follows from (49) that the current produced under the influence of the three fields is parallel to one of the fields. The possible relative polarizations of the fields and of the current are indicated by the arrows:

$$\text{a)}\ \updownarrow E_\alpha \updownarrow E_\alpha \updownarrow E_\alpha \updownarrow j_\alpha, \quad \text{b)}\ \updownarrow E_\alpha \updownarrow E_\alpha \leftrightarrow E_{\alpha_1} \leftrightarrow j_{\alpha_1}. \qquad (50)$$

All other components of the tensor (49) vanish. This result, incidentally, is not connected with the concrete form of the dispersion law (48), but follows from the parity and from the isotropy of $\epsilon_n(\mathbf{k})$ following integration in (49).

To investigate the concentration dependence, let us stop to examine the component $\sigma_{\alpha\alpha\alpha\alpha}(\omega, \omega, -\omega)$, which characterizes the wave self-action. Similar results were obtained also for the other components of the tensor and do not depend on the frequencies of the external fields. In particular, the analysis presented below pertains also to the component $\sigma_{\alpha\alpha\alpha\alpha}(\omega_1, \omega_1, -\omega_2)$, which was investigated experimentally in[6]. Assuming for simplicity



Cubic conductivity as a function of the concentration in a logarithmic scale. $n(k_0)$—value of the electron density in the band, at which $k_F = k_0$. The circles represent the experimental data [6] for InAs.

that the semiconductor is degenerate (the carrier densities are large and allowance for the finite temperature leads to the appearance of a factor on the order of unity), we obtain after going over from summation in (49) to integration

$$\sigma_{\alpha\alpha\alpha\alpha}(\omega, \omega, -\omega) = -\frac{ie^4\omega_0}{(2\pi)^2\hbar^3\omega^3 k_0} \times\left[\frac{x_F^3}{3(1+x_F^2)^{5/2}} + \frac{2x_F^5}{15(1+x_F^2)^{5/2}}\right], \qquad (51)$$

where $x_F = k_F/k_0$ and, in turn, $\hbar k_F$ is the quasimomentum corresponding to the band-filling boundary:

$$k_F = (3\pi^2 n)^{1/3}. \qquad (52)$$

It follows from (51) and (52) that when $x_F \ll 1$ the third-order conductivity increases linearly with the concentration (a fact noted in[6], whereas when $x_F \gtrsim 3$ it practically reaches its maximum value, and does not change any more with increasing concentration. A plot of the susceptibility against the density is shown in the figure, together with the experimental results on the concentration dependence, obtained for InAs in[6]5). The agreement with experiment demonstrates that for InAs in the 10-μ region the nonlinear conductivity actually is determined by the intraband motion.

It is interesting to note that the addition ϵ' to the linear susceptibility, corresponding to (51), is positive and consequently leads to self-focusing. The characteristic self-focusing length Λ is determined for a Gaussian beam in the absence of absorption by[13]

$$\Lambda = c^2/4\omega\varepsilon'\Pi, \qquad (53)$$

where Π is the radiation power flux density. Numerical estimates of the cubic conductivity and the characteristic self-focusing lengths in the 10-μ region for several semiconductors are listed in the table.

The experimental values for the susceptibility are estimated to be approximately one order of magnitude less. The agreement can be regarded as satisfactory, and possibly the discrepancy will decrease to 3 if account is taken of the spectral distribution of the radiation, which occurs in the experiment[6]. As regards self-focusing, it is seen from the table that it might be experimentally observed in InSb if a power density up to 10 MW/cm^2 is attained. The results given in the table for Ge and Si are by way of tentative estimates, since the approximation of the dispersion law by expression

[3] It is precisely with these bands that the main contribution to the nonlinear conductivity in the infrared band is connected, whereas the correction due to the bands lying higher, in accordance with the last remark of the preceding section (see also [3], does not exceed ω/Δ^*.

[4] An estimate for $e\Omega_{nn'}$ was obtained from the data of Hilsum and Rose-Innes [12] from the interband absorption in accordance with the formula

$$|e\Omega_{nn'}|^2 = \frac{\alpha(\omega)c\bar{n}\hbar}{2{,}8\omega\sqrt{\omega-\omega_0}}\left(\frac{\hbar}{m^*_{nn'}}\right)^{3/2}, \quad \omega > \omega_0,$$

where $\bar{n}$ is the refractive index, $\alpha(\omega)$ is the absorption coefficient, $\hbar\omega_0$ is the width of the forbidden band, and $m^*_{nn'}$ is the reduced mass of the bands n and n'. This formula was obtained from the standard perturbation theory, neglecting the dependence of $\Omega_{nn'}$ and $m^*_{nn'}$ on the quasimomentum.

[5] For InSb, the concentration dependence in the region $n \sim (10^{16} - 10^{18})$ cm^{-3} should be much less pronounced (according to (48) and (52), $k_F/k_0 \sim 0.6$–2.5).

Substance	$10^{-13}\omega_0$, rad/sec at 77° K	m/m^*	$10^{-4}k_0$, cm^{-1}	$10^{-6}\sigma_{max}$, cgs-esu	$10^{-14}n$, cm^{-3}	$10^{-6}\sigma$, cgs-esu	$\varepsilon' = \frac{\pi}{2}\frac{|\sigma|}{\omega}$	$\Delta\Pi$, cm-MW	$\alpha(\omega)$, cm^{-1}
InSb	0.31	0.013	1.3	5	1	1	0.8	12	0.5[15]
					10	3	2.4	4	5
					100	5	2.4	2.4	50
InAs	0.64	0.03	3	5	1	0.1	0.08	120	
					10	0.8	0.5	20	
					100	3	2.4	4	
Ge	1	0.12	7	3	100	0.5	0.4	24	70[16]
					1000	2	1.6	6	700
Si	16	0.26	1.4	2,5	100	0.05	0.04	240	
					3000	1	0.8	12	

(48) is not quite satisfactory for them, although it is convenient for estimates[14].

3. The combined terms, containing besides the intraband motion also the interband transitions, also have a concentration dependence, as can be seen from (43). The contribution of the combined terms will be considered further in comparison with the intraband motion. We shall stop first to discuss the effect of self-focusing in InSb. Using in (43) the index n pertaining to the conduction band (n-type semiconductor), and n′ to the heavy-hole band (allowance for the light-hole band reduces to the appearance of a factor on the order of unity in the final expressions), we obtain, taking the dispersion law (48) into account,

$$\sigma_{\alpha\alpha\alpha\alpha}(\omega, \omega, -\omega) = -\frac{2ie^4 k_0\omega_0\Omega^2}{9\pi^2\hbar^3\omega^3}\int_0^{x_F}\frac{x^4\,dx}{(1+x^2)(\sqrt{1+x^2}-\beta)},$$
$$\beta = \frac{4\omega_1-\omega_0}{\omega_0}. \qquad (54)$$

In the derivation of (54) it was assumed that under the conditions of[6] $\omega_0/2$ (1.5×10^{14} rad/sec) is comparable but somewhat smaller than ω (1.8×10^{14} rad/sec). This allows us to confine ourselves in (43) to terms containing small denominators $\omega_{nn'}(\mathbf{k}) - 2\omega$. In addition, it was assumed in the estimate (54) that $\omega_{nn'}(\mathbf{k}) - \omega \approx \omega$, and we also neglected the dependence of $\epsilon_{n'}(\mathbf{k})$ on $\mathbf{k}$ for heavy holes as compared with electrons.

The concentration dependence given by the integral in (54) behaves like x_F^5 in the case of small x_F and like x_F^2 in the case of large ones. At $x = \sqrt{\beta^2-1}$ there is a logarithmic singularity. We note that under the conditions of the experiment in[6], for InSb at 77° K ($\beta \sim 1.3$), the contribution of (54) to the cubic nonlinearity becomes comparable with (it turns out to be somewhat larger than (51) when $x_F > 2$, also near the logarithmic singularity $x = \sqrt{\beta^2-1}$. This circumstance gives grounds for hoping to observe the contribution of combined terms. The effect of self-action, connected with the combined terms, increases rapidly when $\omega \sim \omega_0$.

Examining the first term of (43) in conjunction with (48) under the same approximations as (54), we get

$$\sigma_{\alpha\alpha\alpha\alpha}(\omega, -\omega, \omega) = \frac{2ie^4\Omega^2 k_0}{9\hbar^3\pi^2\omega\omega_0}\int_0^{x_F}\frac{x^2[2+(1+x^2)^{-1}]\,dx}{\sqrt{1+x^2}(\sqrt{1+x^2}-\beta)^2},$$
$$\beta = \frac{2\omega-\omega_0}{\omega_0} \qquad (55)$$

The asymptotic dependence at slow and large concentrations will be respectively like x_F^3 and $\ln x_F$, and the numerical values are comparable with (larger by 2–3 times) the contribution of the intraband motion. Near $x = \sqrt{\beta^2-1}$, expression (55) has a pole-type singularity (resonance). Assuming that the smallest value of the denominator is determined by the temperature smearing of the Fermi distribution, we find that at 77° K the value of (55) already exceeds (51) by two orders of magnitude. (For concreteness, we assume both here and below that $\beta \sim \sqrt{2}$). However, the experimental observation of self action at $\omega \sim \omega_0$ is made difficult by the presence of interband absorption. This difficulty can possibly be eliminated by operating at frequencies (carrier densities) corresponding to the value $\beta^2 < 1 + x_F^2$. A similar situation takes place also when detection in a constant field is considered. In a constant field[6)], however, the doubling effect, where resonance is possible near $\omega_0/2$, is much more interesting.

Indeed, considering the next-to-last term in (43), we get

$$\sigma_{\alpha\alpha\alpha\alpha}(\omega, \omega, 0) = -\frac{2e^4\Omega^2 k_0\tau}{9\hbar^3\pi^2\omega}\int_0^{x_F}\frac{x^4\,dx}{[\sqrt{1+x^2}-\beta]^2(1+x^2)}, \beta = \frac{4\omega-\omega_0}{\omega_0} \qquad (56)$$

It should be noted that (56) turns out to be smaller by a factor $\Omega^2k_0^2x_F^2$ ($\Omega^2k_0^2 \approx 0.1$) when $x_F \ll 1$ than the corresponding contribution of the intraband motion, whereas $\sigma_{\alpha\alpha\alpha\alpha}(\omega, \omega, 0) \approx 2.55x_F\Omega^2k_0^2$ when $x_F > 1$, meaning that the contributions of the intraband motion and of the combined terms are perfectly comparable. In the case of resonance at 77° K, the value of $\sigma_{\alpha\alpha\alpha\alpha}(\omega, \omega, 0)$ already exceeds the contribution from the intraband motion by one order of magnitude, and can be discerned experimentally.

Additional possibilities of separating (56) against the background of the contribution of the intraband motion are connected with singularities of the polarization. It is seen from (43) that a current at double the frequency arises only when the alternating and constant fields are collinear.

Besides resonance at the frequency $\omega \sim \omega_0/2$, resonance takes place also at double the frequency $\omega \sim \omega_0$. Qualitatively, the concentration dependence coincides with (56). If we are interested in a situation wherein only one of the fields depends on the time (quadratic Kerr effect), then the conductivity connected with the combined terms has a pole of second order at $\omega \sim \omega_0$. Indeed, considering again the next to the last term of (43), we get after going over to integration

$$\sigma_{\alpha\alpha\alpha\alpha}(\omega, 0, 0) = -\frac{2ie^4\Omega^2 k_0}{9\hbar^3\pi^2\tau^{-2}}\frac{\omega}{\omega_0}\int_0^{x_F}\frac{x^4dx}{(1+x^2)(\sqrt{1+x^2}-\beta)^3}, \beta = \frac{2\omega-\omega_0}{\omega_0}. \qquad (57)$$

Near resonance $x = \sqrt{\beta^2-1}$ under the same conditions as for (55), the numerical value of (57) exceeds by two orders of magnitude the contribution of the intraband mo-

[6)] In the case of a constant field, the corresponding frequency ω is replaced by $-i\tau^{-1}$, where τ is the electron collision time.

tion. The value of the field at which the change of the conductivity is comparable with the conductivity itself turns out to be ~100 V/cm at $\tau \sim 10^{-11}$ sec. Just as in frequency doubling, the effect is possible if the constant and alternating fields are collinear, but the current has, generally speaking, also a component perpendicular to the field. The latter circumstance can lead to a polarization change that depends quadratically on the constant field. It should be noted that in the case of several strong fields, the analysis of various effects can be based on (32) without using a serious expansion. This question, however, will not be discussed here.

In conclusion we note that a similar analysis of nonlinear effects can be carried out also in the presence of a magnetic field **H**. Taking into account the influence of the magnetic field only on the intraband motions, we obtain the expression for the effective Hamiltonian from (27) by making the substitution

$$\mathbf{k} \to \hat{\varkappa} = \mathbf{k} + \frac{e}{\hbar c}\mathbf{A}(t) + \frac{e}{2\hbar c}[\mathbf{H} \times i\nabla_{\mathbf{k}}]$$

in a perfectly symmetrized manner. The current density is obtained by averaging the operator

$$\hat{j} = -\frac{e}{\hbar V}\frac{\partial \tilde{\varepsilon}_n(kt)}{\partial k}\Big|_{k=\hat{\varkappa}} - \frac{ie}{V}\frac{d}{dt}\left\{\left[S^+\Omega S + \frac{1}{2}\left(S^+\frac{\partial S}{\partial k} - \frac{\partial S^+}{\partial k}S\right)\right]_{k=\hat{\varkappa}}\right\}$$

over $\varphi_n(\mathbf{kt})$, defined by expression (50) with a suitable effective Hamiltonian, followed by averaging over the distribution.

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