$$I_1 = \begin{cases} \dfrac{1}{2}\ln(8\Delta/\omega) - 1, & \tau\Delta \gg 1 \\ \ln(8\Delta/\omega) - 1, & \tau\Delta \ll 1 \end{cases}$$

It follows from Eq. (42) that when the current density increases the damping of the collective oscillations grows fast and for a current density $j/j_c \sim (\Delta/T)^{1/2}$ in pure superconductors ($\tau\Delta \gg 1$) and for $j/j_c \sim 1$ in dirty superconductors ($T\tau \ll 1$) the oscillations disappear completely.

CONCLUSION

We have studied sound propagation in superconductors for arbitrary electron mean free paths. We have shown that taking into account the force connected with the transfer of momentum from the electron system to the impurities removes the anomaly in the absorption found in Vardanyan and Lisitsyn's paper[2] and that as a result one obtains in the given approximation in $(s/v)^2$ the usual BSC expression[3] for the sound absorption. We studied the effect of a current on the propagation of sound waves. When there is a current present there occurs additional dispersion for $kv\tau \sim j/j_c$ with a scale $(j/j_c)^2$ which can easily be made considerably larger than the small parameter $(s/v)^2$. For relatively pure superconductors $\tau\Delta \gg 1$ at low temperatures and for sufficiently large current densities the sound absorption increases exponentially. This growth of the absorption is connected with the diminution of the gap. Turning on the current changes not only the gap in the excitation spectrum but also the form of the Green functions.

As a result an interesting effect appears—in sufficiently pure superconductors ($\tau\Delta \gg 1$) turning on a current leads in a wide range of temperatures $T > \Delta(\tau\Delta)^{-2}$ to a decrease in the sound absorption.

We studied the effect of the current on the collective oscillations in superconductors. The presence of a current leads to a fast growth of the damping and at current densities of the order of critical there are no oscillations.

In conclusion I express my gratitude to A. I. Larkin and B. I. Ivlev for useful remarks.

[1] Yu. M. Gal'perin and V. I. Kozub, Fiz. Tverd. Tela (Leningrad) 15, 3354 (1973) [Sov. Phys. Solid State 15, 2230 (1974)].

[2] R. A. Vardanyan and S. G. Lisitsyn, Zh. Eksp. Teor. Fiz. 69, 1267 (1975) [Sov. Phys. JETP 42, 648 (1976)].

[3] J. R. Schrieffer, Theory of Superconductivity, Benjamin, New York, 1964.

[4] T. Tsuneto, Phys. Rev. 121, 402 (1962).

[5] A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. 10, 407 (1973).

[6] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskiĭ, Metody kvantovoĭ teorii polya v statisticheskoĭ fizike (Quantum field theoretical methods in statistical physics) Nauka, 1962, Ch. VII [Translation published by Pergamon Press, Oxford, 1965].

[7] A. Schmid and G. Schön, Phys. Rev. Lett. 34, 941 (1975).

[8] S. N. Artemenko and A. F. Volkov, Zh. Eksp. Teor. Fiz. 69, 1764 (1975) [Sov. Phys. JETP 42, 896 (1976)].

Translated by D. ter Haar

Quantum electromagnetic processes in condensed media and natural decay in van der Waals crystals

B. A. Grishanin

Moscow State University

(Submitted July 27, 1976)
Zh. Eksp. Teor. Fiz. 72, 783–792 (February 1977)

Quantum dynamical equations taking the variation of the populations and natural (radiative) decay into account are derived for an electromagnetic field in matter on the basis of the mean commutator approximation for the molecular transition operators. The magnitude of the decay and the corresponding natural absorption line in the crystal are calculated, and the possibility of explaining the experimental data on absorption in noble gas crystals on this basis is discussed.

PACS numbers: 78.20.Bh, 78.40.Ha

1. INTRODUCTION

In electrodynamical calculations, two basic approximations are used to simplify the exact system of equations for the system of molecules (atoms) and electromagnetic field: the semiclassical approximation widely applied in quantum electronics, [1] and the quantum approximation of the theory of excitons, [2,3] which utilizes simplification of the commutation relations for the second-quantized operators of the medium under the assumption of constancy of the populations. The semiclassical approach naturally includes the dynamics of the populations and the nonlinear effects associated with it; however, a quantum description of the field is adequate for a description of spontaneous processes. Here a new approach to an approximate description of the medium's operators is developed, allowing us also to describe on a strict quantum level the dynamics of the populations within the framework of equations which are linear in all operator variables. This approach has an exactly defined sphere of applicability in which processes involving the described quasilinear interaction of molecules and field enter and scattering processes involving a significant nonlinearity in the operator variables do not enter. The closed system of equations derived here describes, as will be evident from its structure, nonlinear processes associated with transparency transitions, the narrowing and displacement of the luminescence lines in strong fields, the dependence of the dynamics of spontaneous de-excitation on the collective state of the medium, etc., that is, a very broad spectrum of phenomena which are being intensively investigated at the present time. However, in order to clarify whether the described simplified physics of the enumerated phenomena is adequate for a description of their real manifestations, it is necessary to obtain the corresponding solutions of these equations, which are nontrivial in their mathematical aspects. In the present article only the "natural" decay determined by these equations is considered, and the corresponding absorption of light in noble gas crystals is estimated.

The essence of the "collective" approximation under investigation consists in replacing the commutators for the operators $\hat{P}_{kl}^{(\mu\alpha)}$ characterizing the $l \to k$ transitions of the molecules $(\mu\alpha)$ by their quantum mechanical averages. (Here and below μ denotes the label on an elementary crystal cell; $\alpha = 1, \ldots, \sigma$ is the label for the type of molecule in the cell.) This approximation corresponds to a correct description of only the collective variables of the medium, which depend on $\hat{P}_{kl}^{(\mu\alpha)}$ in a quasi-additive fashion. The direct classical analog of the present approximation is the Gaussian approximation for the central limit theorem in the theory of probability.

The described method is discussed here in a form which is only directly applicable to crystalline dielectrics containing localized states of molecular electrons. (In fact, it may be applied, for example, directly as a rough approximation to arbitrary crystals for processes in the x-ray regime or may be appropriately modified for the case of covalent or metallic bonds.) Only "purely electromagnetic" processes are considered; the molecules are fixed and interact dynamically only by means of van der Waals forces. It is assumed that the remaining interactions are taken into account either statically by the form of the molecular eigenfunctions $\psi_n^{(\mu\alpha)}$ or are not taken into consideration at all.

The dynamical equations corresponding to the collective approximation are written down here for the given model, with allowance for the natural radiative decay of the molecular (exciton) vibrations due to radiative energy transfer to higher spatial harmonics of the electromagnetic field. As far as we know, this decay has previously either not been taken into consideration or was introduced phenomenologically. We shall examine how plausible is the tempting conjecture that the absorption spectra of molecular crystals at low temperatures are principally determined precisely by the natural decay, which is calculated here on the basis of a general theory without the introduction of any phenomenological parameters.

2. QUASILINEAR EQUATIONS FOR MOLECULES AND FOR THE ELECTROMAGNETIC FIELD

Thanks to the assumed absence of molecular overlap, the operators $\hat{P}_{kl}^{(\mu\alpha)}$ and $\hat{P}_{mn}^{(\nu\beta)}$ commute for $(\mu\alpha) \neq (\nu\beta)$ and therefore the commutators $\hat{C}_{kl,k'l'}^{(\mu\alpha)} = [\hat{P}_{kl}^{(\mu\alpha)}, \hat{P}_{k'l'}^{(\mu\alpha)+}]$ corresponding to different molecules $(\mu\alpha)$ also commute. According to this reason the collective approximation in the present model is based on the approximation of average values for these commutators, which leads to Bose–Einstein commutation relations:

$$[\hat{P}_{kl}^{(\mu\alpha)}, \hat{P}_{k'l'}^{(\nu\beta)}] = \delta_{\mu\alpha,\nu\beta}(\delta_{ll'}\langle\hat{P}_{kk'}^{(\mu\alpha)}\rangle - \delta_{kk'}\langle\hat{P}_{ll'}^{(\mu\alpha)}\rangle).$$

A further simplification consists in neglecting the rates of the transitions $\langle\hat{P}_{nn'}^{(\mu\alpha)}\rangle$ in comparison with the maximum values obtained from the populations $n_n^{(\mu\alpha)} = \langle\hat{P}_{nn}^{(\mu\alpha)}\rangle$ and $n_{n'}^{(\mu\alpha)}$; this is valid for fields that are not too strong $(Ed_{nn'} \ll \hbar\omega_{nn'})$. Then

$$\begin{gathered}[\hat{P}_{kl}^{(\mu\alpha)}, \hat{P}_{kl}^{(\mu\alpha)+}] = n_k^{(\mu\alpha)} - n_l^{(\mu\alpha)}, \\ [\hat{P}_{kl}^{(\mu\alpha)}, \hat{P}_{k'l'}^{(\nu\beta)+}] = 0\end{gathered} \tag{2.1}$$

$((k'l') \neq (kl)$ or $(\mu\alpha) \neq (\nu\beta))$, that is, the pairs $\hat{P}_{kl}^{(\mu\alpha)}$, $\hat{P}_{kl}^{(\mu\alpha)+}$ become, apart from numerical factors, canonical variables of the type associated with the operators for the creation and annihilation of quanta (here—the operators for excitations of (kl)-transitions of the molecule). In this connection the populations can be arbitrary in magnitude and may vary in time.

Thanks to the commutativity of the operators associated with different (kl)-transitions of the medium, the approximation (2.1) allows us to consider these transitions as independent degrees of freedom, where the number of these degrees of freedom for each transition of the medium is proportional, in accordance with Eq. (2.1), to its inverse population. The possibility of such an interpretation brings the quantum mechanical description of the medium as close as possible to the classical Lorenz theory, which obviously also explains the possibility of deriving many of the results obtained in terms of the theory of excitons within the framework of this nonquantum theory (this fact was noted in[4]). The necessity of using the concept of an exciton generally does not arise in the form of the theory expounded here. In this connection the operators of the medium are described directly in a basis of unperturbed Hamiltonians of the molecules, as this is done in the theory of the laser.[5]

The quasilinear approximation consists in taking account of only the nonlinearities associated with a variation of the populations. It is obtained either by replacing the medium + field Hamiltonian with a quadratic approximation with respect to the operator variables, or by an equivalent transformation of the exact equations of motion. This amounts to replacing the quantum-number operators $\hat{n}_k^{(\mu\alpha)} = \hat{P}_{kk}^{(\mu\alpha)}$ in the Hamiltonian by a quadratic expression in terms of the transition operators based on the commutation relations

$$[\hat{n}_k^{(\mu\alpha)}, \hat{P}_{lm}^{(\mu\alpha)}] = (\delta_{kl} - \delta_{lm})\hat{P}_{lm}^{(\mu\alpha)},$$

the relationships (2.1), and the standard connection between the operations of differentiation and commutation for Bose operators. By utilizing "coordinate" operators $\hat{\chi}_s^{(\mu\alpha)} = \hat{P}_{kl}^{(\mu\alpha)} + \hat{P}_{lk}^{(\mu\alpha)}$, which are Hermitian and invariant with respect to the direction of the transition, and the "momentum" operator of the s-th (kl)-transition

$$\hat{\pi}_s^{(\mu\alpha)} = i\omega_{kl}(\hat{P}_{kl}^{(\mu\alpha)} - \hat{P}_{lk}^{(\mu\alpha)})$$

we obtain the following quadratic approximation for the Hamiltonian of the molecule $(\mu\alpha)$

$$\hat{\mathscr{H}}^{(\mu\alpha)} = \sum_m \hbar\omega_{m\alpha}\hat{n}_m^{(\mu\alpha)} \approx {}^1/_2\hbar\sum_s [(\hat{\pi}_s^{(\mu\alpha)})^2 + \omega_s^2(\hat{\chi}_s^{(\mu\alpha)})^2]/n_s\omega_s,$$

where $n_s\omega_s = (n_k^{(\mu\alpha)} - n_l^{(\mu\alpha)})(\omega_l^{(\mu\alpha)} - \omega_k^{(\mu\alpha)})$. The last nonlinearity, which is eliminated in the quasilinear approximation, is the nonlinearity associated with the terms

$$-\frac{e^2}{2mc^2}\Big[\sum_{kl}\int \hat{\mathbf{A}}^2(\mathbf{r},t)\rho_{kl}^{(\mu\alpha)}(\mathbf{r})\,dV\Big]\hat{P}_{kl}^{(\mu\alpha)}(t)$$

in the Hamiltonian of the interaction of the molecules with the transverse electromagnetic field. Here $\bar{\rho}_{kl}^{(\mu\alpha)}(\mathbf{r})$ are the matrix elements of the operator for the relative molecular charge density, which is approximately equal to the density of the electron charge. In the quasilinear approximation

$$\hat{P}_{kl}^{(\mu\alpha)} \to n_k^{(\mu\alpha)}\delta_{kl}.$$

As a result of the described procedure for the quasilinear approximation, we obtain a quasilinear Hamiltonian and a corresponding closed system of equations for the transverse electromagnetic field and for the molecular transition operators (in the derivation of the former, the Thomas–Kuhn sum rule was additionally used; with a high degree of accuracy this leads on the right hand side to a concellation of the terms that depend on $\hat{\mathbf{A}}$):

$$\Box\hat{\mathbf{A}} = -\frac{4\pi}{c}\sum_{\mu\alpha}\sum_s \frac{\mathbf{j}_{\alpha s}^{\perp}(\mathbf{r}-\mathbf{r}_{\mu\alpha})}{i\omega_{\alpha s}}\frac{d\hat{\chi}_s^{(\mu\alpha)}}{dt}, \tag{2.2}$$

$$\frac{d^2\hat{\chi}_s^{(\mu\alpha)}}{dt^2} + \omega_{\alpha s}^2\hat{\chi}_s^{(\mu\alpha)} - \frac{2\omega_{\alpha s}n_s^{(\mu\alpha)}}{\hbar}\sum_{\nu\beta}{}'\sum_{s'} V_{ss'}^{(\mu\alpha)(\nu\beta)}\hat{\chi}_{s'}^{(\nu\beta)}$$
$$= -\frac{2i}{\hbar c}\frac{\partial}{\partial t}\Big[n_s^{(\mu\alpha)}\int \mathbf{j}_{\alpha s}(\mathbf{r}-\mathbf{r}_{\mu\alpha})\hat{\mathbf{A}}(\mathbf{r},t)\,dV\Big], \tag{2.3}$$

$$\frac{dn_k^{(\mu\alpha)}}{dt} = \frac{i}{\hbar}\sum_m\Big\langle\sum_{\nu\beta}{}'\sum_{s'} V_{(km)s'}^{(\mu\alpha)(\nu\beta)}\hat{\chi}_{s'}^{(\nu\beta)}\hat{\pi}_{mk}^{(\mu\alpha)}\frac{1}{i\omega_{\alpha mk}}$$
$$+\frac{1}{c}\Big[\int \hat{\mathbf{A}}(\mathbf{r},t)\mathbf{j}_{\alpha mk}(\mathbf{r}-\mathbf{r}_{\mu\alpha})\,dV\Big]\hat{\chi}_{mk}^{(\mu\alpha)}\Big\rangle. \tag{2.4}$$

Here $\mathbf{j}_{\alpha s}(\mathbf{r})$ is the matrix element of the unperturbed current associated with the s-th $(l \to k)$ transition (with its direction taken into account) for a type α molecule with coordinate $\mathbf{r}_{\mu\alpha} = 0$; $n_s^{(\mu\alpha)} = n_l^{(\mu\alpha)} - n_k^{(\mu\alpha)}$; in the summation over s and s' these variables label the undirected (kl) transitions;

$$V_{ss'}^{(\mu\alpha)(\nu\beta)} = \int\frac{\rho_{\alpha kl}(\mathbf{r}-\mathbf{r}_{\mu\alpha})\rho_{\beta k'l'}(\mathbf{r}-\mathbf{r}_{\nu\beta})}{|\mathbf{r}-\mathbf{r}'|}\,dV\,dV' \tag{2.5}$$

denotes the Coulomb interaction energy of the transitions, which determines the effects of the internal field; the brackets $\langle\,\rangle$ denote quantum mechanical averaging. This system of equations must be supplemented by the appropriate initial conditions, which cannot be zero in a quantum-mechanical consideration, owing to the noncommutativity of coordinates and velocities. These conditions are obtained by expressing the velocity operators $d\hat{\chi}_s^{(\mu\alpha)}/dt$ and $\partial\hat{\mathbf{A}}/\partial t$ at $t = 0$ in terms of the initial coordinates and momenta of the molecular transitions and of the electromagnetic field, which by virtue of the principles of quantum mechanics are completely determined by the commutation relations (and by the law of averaging which is specified, for example, with the aid of the density matrix). The indicated expression is given by Hamilton's equations that are conjugate to Eqs. (2.2) and (2.4). For the molecules of the medium these equations take the form

$$\frac{d\hat{\chi}_s^{(\mu\alpha)}}{dt} = \hat{\pi}_s^{(\mu\alpha)} - \frac{2i}{\hbar c}n_s^{(\mu\alpha)}\int \mathbf{j}_{\alpha s}(\mathbf{r}-\mathbf{r}_{\mu\alpha})\hat{\mathbf{A}}(\mathbf{r},t)\,dV, \tag{2.6}$$

and the commutation relations determining the properties of the operators $\hat{\chi}_s^{(\mu\alpha)}$ and $\hat{\pi}_s^{(\mu\alpha)}$ are such that:

$$[\hat{\chi}_s^{(\mu\alpha)}, \hat{\pi}_{s'}^{(\nu\beta)}] = 2\delta_{\mu\alpha,\nu\beta}\delta_{ss'}i\omega_{\alpha s}n_s^{(\mu\alpha)}$$

and are equal to zero for the remaining combinations $[\chi, \chi]$ and $[\pi, \pi]$.

The described system of equations is the simplest form of the considered approximation of quantum theory for the investigated model of a medium. These equations are very similar to the corresponding equations for the classical and semiclassical theories; however, they also describe spontaneous processes and take saturation effects into account. The essential factor in Eqs. (2.2) and (2.3) is the nonlocal nature of the interaction between the molecules and the transverse electromagnetic field; as is well known, taking this factor into account leads to retardation of the waves inside the molecules and to the appearance of radiative decay of the oscillations. A calculation of this decay is given below, revealing its specific characteristics in condensed media in comparison with the case of an isolated molecule.

3. NATURAL DECAY IN CRYSTALS

Let us consider the obtained system of equations for the case of an infinite perfect crystal and for time intervals guaranteeing constancy of the populations, i.e., exact linearity of this system. In this connection sources of quantum fluctuations of the transitions will also be taken into account; these automatically cancel the decay of the molecular operators, in complete agreement with the principle of conservation of the commutation relations for dynamical canonical variables (in connection with this point see, for example, [5,6]).

Assuming spatial homogeneity of the populations $n_s^{(\mu\alpha)} = n_{\alpha s}$, let us make the following transformations in the system (2.2) and (2.3): In Eq. (2.2) a Fourier transformation with respect to $\mathbf{r}$ and a Laplace transformation with respect to t, and in Eq. (2.3) a Fourier transformation with respect to the coordinates $\mathbf{r}_\mu$ of the cell and a Laplace transformation with respect to t. We obtain

$$\left[(\varkappa+\mathbf{K})^2+\frac{p^2}{c^2}\right]\hat{\mathbf{A}}(\varkappa+\mathbf{K},p)=-\frac{4\pi}{c}p\mathbf{D}^{\perp}(\varkappa+\mathbf{K})\hat{\chi}(\varkappa,p)+\hat{\mathbf{X}}(\varkappa,\mathbf{K},p),\tag{3.1}$$

$$(p^2+W_0{}^2-W_I{}^2)\hat{\chi}(\varkappa,p)=\frac{2p}{\hbar c\Omega}NW_0\sum_{\mathbf{K}}\mathbf{D}^T(-\varkappa-\mathbf{K})\hat{\mathbf{A}}(\varkappa+\mathbf{K},p)+\hat{Y}(\varkappa,p),\tag{3.2}$$

where Ω is the volume of the elementary cell; $\hat{\chi}(\varkappa,p)$ is a column vector with elements $\hat{\chi}_{\alpha s}(\varkappa,p)$; W_0^2 is the matrix of the squares of the unperturbed frequencies with elements $(W_0^2)_{\alpha s,\alpha' s'}=\omega_{\alpha s}^2\delta_{\alpha s,\alpha' s'}$; W_I^2 is the matrix of the squares of the frequency displacements due to the internal field having matrix elements

$$(W_I{}^2)_{\alpha s,\alpha' s'}=\frac{2\omega_{\alpha s}n_{\alpha s}}{\hbar}\sum_{\nu}{}' V_{ss'}^{(0\alpha)(\nu\alpha')}\exp\{-i\varkappa(\mathbf{r}_\nu+\boldsymbol{\rho}_{\alpha'}-\boldsymbol{\rho}_\alpha)\}:\tag{3.3}$$

$\mathbf{D}(\mathbf{k})$ is a row vector of the three-dimensional vectors of the Fourier transforms of the dipole moments

$$\mathbf{d}_{\alpha s}(\mathbf{k})\exp(i\mathbf{k}\boldsymbol{\rho}_\alpha)=\mathbf{j}_{\alpha s}(\mathbf{k})\exp(i\mathbf{k}\boldsymbol{\rho}_\alpha)/i\omega_{\alpha s};$$

$N_{\alpha s,\alpha' s'}=n_{\alpha s}\delta_{\alpha s,\alpha' s'}$ is the matrix of the inverse populations; $\hat{\mathbf{X}}(\varkappa,\mathbf{K},p)$ and $\hat{Y}(\varkappa,p)$ describe the contributions of the initial conditions:

$$\hat{\mathbf{X}}(\varkappa,\mathbf{K},p)=\frac{p}{c^2}\hat{\mathbf{A}}^{(0)}(\varkappa+\mathbf{K})+\frac{1}{c^2}\hat{\mathbf{A}}^{(0)}(\varkappa+\mathbf{K})-\frac{4\pi}{c}\mathbf{D}^{\perp}(\varkappa+\mathbf{K})\hat{\chi}^{(0)}(\varkappa),\tag{3.4}$$

$$\hat{Y}(\varkappa,p)=\dot{\hat{\chi}}^{(0)}(\varkappa)+p\hat{\chi}^{(0)}(\varkappa)-\frac{2}{\hbar c\Omega}NW_0\sum_{\mathbf{K}}\mathbf{D}^T(-\varkappa-\mathbf{K})\hat{\mathbf{A}}^{(0)}(\varkappa+\mathbf{K}).\tag{3.5}$$

Out of the set of values K for the fields $\hat{\mathbf{A}}(\varkappa+\mathbf{K},p)$, only a limited number of reciprocal lattice vectors corresponding to strong coupling of the electromagnetic field with the medium are of ultimate interest: in the optical band—only the value $\mathbf{K}=0$ (for $\mathbf{K}\neq 0$ we have $\varkappa \ll K$, $c^{-2}p^2+(\varkappa+\mathbf{K})^2 \gg c^{-2}p^2+\varkappa^2$), and in the x-ray range there is the value $\mathbf{K}=0$ and those values of K for which Bragg reflection is possible. The totality of the remaining degrees of freedom may be eliminated from consideration with the aid of Eq. (3.1). As a result we obtain

$$\left(\varkappa^2+\frac{p^2}{c^2}\right)\hat{\mathbf{A}}(\varkappa,p)=-\frac{4\pi p}{c}\mathbf{D}^{\perp}(\varkappa)\hat{\chi}(\varkappa,p)+\mathbf{X}(\varkappa,0,p);\tag{3.6}$$

$$\left[p^2+W_0{}^2-W_I{}^2+\frac{8\pi p^2}{\hbar c^2\Omega}NW_0\sum_{\mathbf{K}}\frac{\mathbf{D}^T(-\varkappa-\mathbf{K})\mathbf{D}^{\perp}(\varkappa+\mathbf{K})}{(\varkappa+\mathbf{K})^2+p^2/c^2}\right]\hat{\chi}(\varkappa,p)$$
$$=\hat{Y}(\varkappa,p)+\frac{2p}{\hbar c\Omega}NW_0\sum_{\mathbf{K}}\frac{\mathbf{D}(-\varkappa-\mathbf{K})\hat{\mathbf{X}}(\varkappa,\mathbf{K},p)}{(\varkappa+\mathbf{K})^2+p^2/c^2}.\tag{3.7}$$

The term inside the square brackets in Eq. (3.7) determines the dispersion equation, and the right hand part—the sources of quantum noise. In the optical band the terms of the sum over $\mathbf{K}\neq 0$ in the dispersion equation describe the influence of the interaction with high-frequency oscillators of the field, the description of which reduces to an evaluation of the sum

$$\sum_{\mathbf{K}\neq 0}\mathbf{D}^T(-\varkappa-\mathbf{K})\mathbf{D}^{\perp}(\varkappa+\mathbf{K})/[(\varkappa+\mathbf{K})^2+p^2/c^2].$$

(Similar expressions have been investigated in the literature; however, the natural radiative decay associated with them was not investigated.)

In the cited expression, all the terms taken separately are negligible in comparison with the corresponding term with $K=0$. This indicates weak coupling of the molecular vibrations with each such high-frequency oscillator of the field. However, in totality they all give a finite effect as a consequence of the fact that the indicated sum converges only owing to the cutoff role of the molecular factor $\mathbf{D}^T(-\varkappa-\mathbf{K})\mathbf{D}^{\perp}(\varkappa+\mathbf{K})$; an illustration of the typical nature of this situation is the significant increase of the estimate for the intensity of the quasi-Čerenkov radiation from channeled charged particles in a crystal upon taking all of the terms of a similar sum into account.[7] It has an order of magnitude exceeding the order of a single term by λ/d times. As a consequence of the nonessential nature of the individual terms in this sum, with a high degree of accuracy of order $\gamma d/c$, where γ is the decay frequency, it can be replaced by an integral which converges and is equal in the dipole approximation to

$$\Omega\left[\iint\mathbf{D}^T(\mathbf{r})\Pi^{\perp}(\varkappa)\mathbf{D}(\mathbf{r}')|\mathbf{r}-\mathbf{r}'|^{-1}dV\,dV'-\frac{p}{4\pi c}\mathbf{D}_0{}^T\Pi^{\perp}(\varkappa)\mathbf{D}_0\right].$$

Here $\mathbf{D}_0$ denotes the row vector of the integral dipole moments $\mathbf{d}_{\alpha s}$ and $\Pi^{\perp}(\varkappa)$ is a projector on the plane orthogonal to $\varkappa$. The cited derivation is quite analogous to the similar derivation for the decay of an isolated molecule. The first term in the obtained expression primarily describes a frequency shift analogous to the Lamb shift for an isolated atom. The second term gives the natural, i.e., not related to additional (apart from polarization and electromagnetic) degrees of freedom, radiative decay due to the presence in the crystal of an infinite number of modes of the electromagnetic field which are interacting with the crystal. This decay is a collective characteristic of the crystal, being significantly different from the decay of an isolated molecule.

Thus, the dispersion equation takes the form

$$\det\Delta(p)=0,\tag{3.8}$$

$$\Delta(p)=p^4+\Big(c^2\varkappa^2+W_0{}^2-W_I{}^2+(p^2+c^2\varkappa^2)\frac{8\pi}{\hbar c^2}NW_0$$
$$\times\iint|\mathbf{r}-\mathbf{r}'|^{-1}\mathbf{D}^T(\mathbf{r})\Pi^{\perp}\mathbf{D}(\mathbf{r}')dV\,dV'+\frac{8\pi}{\hbar\Omega}NW_0\mathbf{D}_0{}^T\Pi^{\perp}\mathbf{D}_0\Big)p^2$$
$$+c^2\varkappa^2(W_0{}^2-W_I{}^2)-\frac{4p^3}{\hbar c^3}(p^2+c^2\varkappa^2)NW_0\mathbf{D}_0{}^T\Pi^{\perp}\mathbf{D}_0.$$

Here the last term determines the attenuation and a very small additional shift of the frequency, analogous to the shift of a classical oscillator.

Now let us simplify Eq. (3.7) that determines the quantum dynamics of the transitions in such a way that the only explicit expressions are for those quantum sources, which describe spontaneous transition processes in the system to within its vacuum fluctuations. The remaining sources of quantum noise, describing as $t\to\infty$ the equilibrium quantum noise and corresponding to the terms of the summation over $K\neq 0$ in the right-hand side, can be re-established as $t\to\infty$ from the fluctuation-dissipation theorem (at $T=0$). With Eq. (2.6) taken into account (at $t=0$) we obtain

$$\Delta(p)\hat{\chi}(\varkappa,p)=(p^2+c^2\varkappa^2)[\hat{\pi}^{(0)}(\varkappa)+p\hat{\chi}^{(0)}(\varkappa)]$$
$$+\frac{2pNW_0}{\hbar c\Omega}\mathbf{D}_0[p\hat{\mathbf{A}}^{(0)}(\varkappa)+\dot{\hat{\mathbf{A}}}^{(0)}(\varkappa)]+\hat{\xi}(p,\varkappa).\tag{3.9}$$

Here the influence of radiation damping on the quantum sources of the transition processes is neglected on the right hand side, the quantum sources being expressed in terms of the initial operators for the medium and for the field without taking the damping term into consideration. The system (3.9) and (3.6) determines the dynamics of the spontaneous processes in the approximation of constancy of the populations. If it is necessary to take their variation into account, then instead of Eq. (3.9) one should start from its temporal form with the inclusion of the additional equation (2.4) for the populations.

The complete investigation of the derived system of equations represents a technically difficult problem, although it is possible to obtain certain useful general relationships for arbitrary crystals. Thus, by allowing for the smallness of the damping term in $\Delta(p)$ one can obtain the following formula for the decay of the natural vibrations (polaritons):

$$\gamma_{i\alpha s}(\varkappa)=\frac{3}{2\pi}\frac{\Omega}{c^3}\frac{\omega^3_{i\alpha s}(\omega^2_{i\alpha s}-c^2\varkappa^2)}{2\omega^2_{i\alpha s}+c^2\varkappa^2}\sum_{\alpha's'}\frac{\partial\omega_{i\alpha s}}{\partial\mathbf{d}^\perp_{\alpha's'}}\mathbf{d}^\perp_{\alpha's'}, \qquad (3.10)$$

where $\omega_{i\alpha s}(\varkappa)$ are the eigenfrequencies in the approximation without taking account of the decay. In order of magnitude this expression gives the following result for the exciton branch

$$\gamma_{\alpha s}\sim[\varepsilon_{\alpha s}(0)-1]\Omega\omega_{\alpha s}{}^4c^{-3}$$

(since $\omega_{i\alpha s}/c\varkappa\sim\varepsilon_{\alpha s}(0)$ for $\varkappa\to 0$). In the visible band this amounts to $\sim 10^{10}$ Hz (let us turn our attention to the correctness of this estimate for the limiting transition in the case of isolated atoms: $\Omega\to\infty$, $\varepsilon(0)-1\sim\Omega^{-1}\to 0$).

4. COMPARISON OF THE NATURAL AND THE EXPERIMENTALLY OBSERVED ABSORPTION

We confine ourselves to two-level molecules and use the additional simplifying assumption that the environment of each molecule is spherically symmetrical, so that the transition under consideration is isotropic. In this connection we assume the molecules of all types to be approximately identical and we shall be interested only in in-phase oscillations of the polarization inside the cells. Then from Eq. (3.9) we obtain

$$\begin{aligned}&\left\{p^4+\left[\omega_0{}^2+c^2\varkappa^2+\frac{8\pi}{3}\nu^2\right]p^2+c^2\varkappa^2\left(\omega_0{}^2-\frac{4\pi}{3}\nu^2\right)\right.\\&\left.-\frac{2\Omega}{c^3}p^3(p^2+c^2\varkappa^2)\nu^2\right\}\hat{\mathbf{P}}^\perp(\varkappa,p)=(p^2+c^2\varkappa^2)[\hat{\dot{\mathbf{P}}}^{(0)}(\varkappa)+p\hat{\mathbf{P}}^{\perp(0)}(\varkappa)]\\&+p\frac{\nu^2}{c}[p\hat{\mathbf{A}}^{(0)}(\varkappa)+\hat{\dot{\mathbf{A}}}^{(0)}(\varkappa)]+\hat{\xi}^\perp(\varkappa,p),\end{aligned} \qquad (4.1)$$

where ν^2, $\hat{\mathbf{P}}$, $\hat{\dot{\mathbf{P}}}$, and $\hat{\xi}$ the combined variables of all the sorts of molecules. This expression is obtained in the long-wavelength limit when[8] $V_{\alpha\alpha'}(\varkappa)=[(4/3)\pi\Pi^\perp-(8/3)\pi\Pi^{\parallel}]d^2/3\Omega$, thanks to which the longitudinal and transverse vibrations are separable; ω_0 denotes the resonance frequency of the molecules without taking account of the collective analog of the Lamb shift obtained in Sec. 3: it is considerably smaller than the shift due to the square of the transition's plasma frequency, $\nu^2=fnNe^2/m$. Here $fn=-(\bar{f}_{12}n_1+\bar{f}_{21}n_2)$ denotes the strength of the transition and N denotes the density of molecules.

Calculation of the decay reduces to a determination of the eigenfrequencies of Eq. (4.1) with the damping term dropped and a subsequent calculation of γ_j according to the formula

$$\gamma_j(\varkappa)=\frac{3\sigma}{2\pi c^3}\frac{\omega_j{}^3(\omega_j{}^2-c^2\varkappa^2)}{2\omega_j{}^2+c^2\varkappa^2}\frac{\partial\omega_j}{\partial N}, \qquad (4.2)$$

which is derived in analogy with Eq. (3.10). The eigenfrequencies are given by the following ($j=+,-$):

$$\omega_j{}^2={}^1/_2\{\omega_0{}^2+{}^8/_3\pi\nu^2+c^2\varkappa^2\pm[(\omega_0{}^2+{}^8/_3\pi\nu^2+c^2\varkappa^2)^2-4c^2\varkappa^2(\omega_0{}^2-{}^4/_3\pi\nu^2)]^{1/2}\}. \qquad (4.3)$$

By utilizing (4.3) we obtain from (4.2)

$$\gamma_j(\varkappa)=\frac{\pi}{3}fn\frac{e^2}{mc^3}\frac{\omega_j{}^2}{\omega_+{}^2-\omega_-{}^2}|\omega_j{}^2-c^2\varkappa^2|. \qquad (4.4)$$

To calculate the linear absorption, it now remains to determine $\varkappa''=\operatorname{Im}\varkappa$ as the solution of the equation $\pm i\omega=\pm i\omega_j(\varkappa)-\gamma_j(\varkappa)/2$ for given frequencies ω.

For the photon branch, i.e., $j=+$, the resonant properties of the true absorption are manifested at the frequency $\omega=\bar{\omega}_+=(\omega_0^2+8\pi\nu^2/3)^{1/2}$ determining the transition to the region of total reflection. Since this frequency corresponds, according to Eq. (4.3), to the wave vector $\varkappa=0$ in whose neighborhood the above-utilized approximation with respect to the small parameter $\varkappa/K$ is certainly satisfied, the calculation of the absorption for the given branch does not encounter any additional difficulties. It leads to a narrow curve $\varkappa''(\omega)$ having a width of order $\gamma_+=(1/3)\pi fn\bar{\omega}_+^2e^2/mc^3$, localized in the neighborhood of the frequency $\bar{\omega}_+$ with a peak height $\sim(\gamma_+\bar{\omega}_+)^{1/2}/\omega$ and having a short wavelength tail $\gamma_+\nu^2/c\bar{\omega}_+^2$. These quantities are negligible in comparison with typical experimentally observed values of the absorption,[9,10] differing from them by roughly four orders of magnitude.

As to the exciton branch $j=-$, the resonant properties of its absorption appear as $\varkappa\to\infty$, i.e., actually for $\varkappa\sim k_{\rm lim}$; $\mathbf{k}_{\rm lim}$ is the limiting wave vector whose absolute magnitude and direction depend on the orientation of the crystal.

In an ideal crystal the width of the absorption line should be $\sim\gamma$ and the position of the peak should depend on the orientation since a certain effective factor depending on the orientation appears in Eq. (4.3) instead of the Lorentz factor $4\pi/3$. In this connection the position of the peak may vary within maximum limits from ω_0 to $\bar{\omega}_-=(\omega_0^2-4\pi\nu^2/3)^{1/2}$. In actual experiments on solid noble gases[9,10] one should obviously expect a rather random structure. For provisional estimates it is logical to assume that this case leads to a broadening to a value $\Delta\omega\sim\omega_0-\bar{\omega}_-$ whereas the height of the maximum is replaced by a quantity of order $2\pi(\bar{\omega}_-/\Delta\omega)^{1/2}/\lambda$ corresponding to the indicated broadening instead of the expression for an ideal resonance $\varkappa''=2\pi(\bar{\omega}_-/\gamma)^{1/2}/\lambda$. In the case of Ar for the transition $\lambda=1049$ Å and $f=0.20$, for the experiment of[9], this quantity is approximately equal to 4×10^6 cm^{-1}. The estimate given in[9] from the experimental data is equal to $\sim3\times10^6$ cm^{-1}. The width $\Delta\omega$ in our estimate ≈ 0.24 eV, and the absolute position of this peak is not predicted due to rejection from the dynamical investigation of the contribution due to non van der Waals interactions of the atoms. The shape of the absorption line for the given transition in[9] differs somewhat from that predicted by the ideas under consideration—in the first place by its width which is approximately twice as large in[9]. Nevertheless it is possible that in regard to lower temperatures ($T=20$ K in[9]) the mechanism under consideration retains a certain competitive ability with regard to the other absorption mechanisms. The experimentally observed narrowing of the lines at very low temperatures attests to this. In any case, allowance for the natural decay in the region of the resonances associated with natural absorption is essential. As to the initial general equations of the collective approximation proposed here and also possible generalizations of this method, their application to problems specific to quantum electronics such as the coherence of laser radiation, super-radiance, etc., is of particular interest.

[1] R. H. Pantell and H. E. Puthoff, Fundamentals of Quantum Electronics, Wiley, 1969 (Russ. Transl., Mir, Moscow, 1972).

[2] V. M. Agranovich, Teoriya êksitonov (Theory of Excitons), Nauka, 1968.

[3] A. S. Davydov, Teoriya molekulyarnykh êksitonov (Theory of Molecular Excitons), Nauka, 1968 (English Transl., Plenum Press, 1971).

[4] V. M. Agranovich, Usp. Fiz. Nauk **112**, 143 (1974) [Sov. Phys. Usp. **17**, 103 (1974)].

[5] H. Haken, Laser Theory, Springer-Verlag, 1970.

[6] G. A. House, Trudy IÉR **58**, 280 (1970) [Russ. Trans. of IEEE Trans.].

[7] S. A. Akhmanov and B. A. Grishanin, Pis'ma Zh. Eksp. Teor. Fiz. **23**, 562 (1976) [JETP Lett. **23**, 515 (1976)].

[8] M. Born and K. Huang, Dynamical Theory of Crystal Lattices, Oxford Univ. Press, 1954 (Russ. Transl., IIL, 1958).

[9] G. Baldini, Phys. Rev. **128**, 1562 (1962).

[10] R. Haensel, G. Keitel, C. Kunz, P. Schreiber, and B. Sontag, DESY F41-69/6, 1976.

Translated by H. H. Nickle

Sound generation in a multidomain ferromagnet

A. V. Mitin and V. A. Tarasov

Kazan' Physicotechnical Institute, USSR Academy of Sciences

(Submitted July 27, 1976)
Zh. Eksp. Teor. Fiz. **72**, 793–802 (February 1977)

The generation of sound by a radio-frequency field in a multidomain structure, formed by 180-degree Bloch walls, is treated by simultaneous solution of the equations of motion for the magnetization and for elastic waves. It is shown that the effectiveness of the generation depends strongly on the orientation of the radio-frequency field. Emphasis is placed on the large role played in the amplification of acoustic oscillations by a magnetoacoustic resonator that involves both the acoustic properties of the specimen and also the multidomain structure. From the results of the paper it follows that excitation of acoustic oscillations by Bloch walls in a multidomain structure is quite effective and that these oscillations can be detected experimentally.

PACS numbers: 43.35.Rw, 75.80.+q

1. INTRODUCTION

For detection of weak ultrasonic (US) vibrations, there has recently been increasingly broad application of the method of US modulation of Mössbauer radiation. This method offers the possibility of measuring amplitudes of the order of magnitude of a fraction of an angstrom. US Mössbauer spectroscopy has received its greatest development in the study of ultrasound excited by a radio-frequency (rf) field in ferromagnets. As has been established, the principal mechanism of excitation of sound is magnetostriction.[1] At the same time, experiment shows that the theory of excitation of sound in a single-domain structure (see, for example,[2,3]) does not fully describe the experiment of[1]. Additional clarity in the understanding of the physical picture of the effect can be achieved by use of the mechanism of excitation of sound by Bloch walls.[4] In this connection, it is of interest to investigate this mechanism on the basis of a multidomain structure of the specimen.

2. HIGH-FREQUENCY OSCILLATIONS OF A BLOCH WALL

We consider a uniaxial cubic crystal of a ferromagnetic material in the form of a plate with dimensions $L\times d_1\times d_2$. We direct the axis of easy magnetization along x; then the plane of the absorber will be the xz or yz plane when the smallest dimension of the plate is d_1 or d_2 respectively (see Fig. 1). For simplicity we restrict ourselves to the case in which the specimen consists of plane-parallel domains, separated by domain walls perpendicular to the z axis. In an equilibrium state, all the domains have the same thickness D; the wall thickness is Δ. For absorber thicknesses characteristic of Mössbauer spectroscopy, the Landau-Lifshitz[5] model of the transition layer may be considered