Quantum electrodynamics of a dispersive medium and multiphoton processes at an impurity center

G. V. Vikhnina and S. I. Pekar

Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR
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The probabilities of multiphonon light absorption, emission, and Raman scattering of different orders at an impurity center in an isotropic medium with frequency dispersion but no spatial dispersion are calculated in the region of transparency. In the first part of the paper, thanks to a consistent quantization of the field in the dispersive medium [S. I. Pekar, Sov. Phys. JETP **41**, 430 (1975)], for the probabilities of most of the processes general formulas are obtained that differ from those applied previously. In the second part of the paper all the formulas are specialized for specific states of the phonon subsystem and its spectrum. The explicit dependence of the probabilities on the temperature and on the evolved heat accompanying a multiphoton process is obtained.

Multiphoton processes at an impurity center in a crystal have been considered long ago and by many authors; the polarizability of the medium has been taken into account macroscopically by the introduction of the dielectric permittivity ε. However, the dispersion (the dependence of ε on the frequency ω) has not been taken into account in most cases, and in those papers in which it has been taken into account this has been done inconsistently. Most commonly, the authors have assumed that the whole calculation can be performed with neglect of the dependence of ε on ω, and only in the final formulas is ε assumed to be a function of ω. But it is clear that in this way we lose all the derivatives of ε with respect to ω and it is impossible to obtain even the classical expression for the energy of the field of a monochromatic wave:

$$W=\frac{1}{16\pi}\int_V\left\{\mathbf{E}^*\frac{\partial}{\partial\omega}(\omega\varepsilon)\mathbf{E}+\mathbf{H}^*\frac{\partial}{\partial\omega}(\omega\mu)\mathbf{H}\right\}d^3r. \tag{1}$$

Here V is the volume of the basic repeat unit of the crystal, E and H are the intensities of the electric and magnetic fields, and $\mu(\omega)$ is the magnetic permeability of the medium.

All authors have postulated the form $\sum_k\hbar\omega_{\mathbf{k}}a_{\mathbf{k}}^+a_{\mathbf{k}}$ for the field-energy operator, without being able to prove the commutation law for the operators $a_{\mathbf{k}}^+$ and $a_{\mathbf{k}}$ and referring only to the analogy with the case of a nondispersive medium. These difficulties were connected with the fact that, up to 1975, a sound method of quantizing the field in a dispersive medium did not exist. The point is that, by itself, the expression (1) for the field energy is still not adequate for the quantization of the field, just as the classical expression for the energy of a harmonic oscillator or rotator in terms of its amplitude is not adequate for *its* quantization. In the expression for the energy it is still necessary to distinguish the canonically conjugate quantities, with a known commutation law. It is necessary to have a general expression for the energy of the system in terms of the canonically conjugate quantities, assuming the latter to be independent, not coupled by the equations of motion, and not expressed classically in terms of the time. All this was done in 1975 for the case of no dissipation, i.e., when the electromagnetic field lies in the spectral region of transparency of the crystal.[1] Also in Ref. 1, the electromagnetic field was quantized and the photons in a dispersive medium were obtained consistently, with macroscopic allowance for the polarization.

Below we consider multiphoton processes at an impurity center in a transparent isotropic medium with frequency dispersion but no spatial dispersion of ε and with $\mu=1$.

According to Ref. 1, the operator of the energy of the electromagnetic field in the absence of sources has the form

$$\hat{W}=\sum_{\mathbf{k}}\sum_{\nu=1}^{2}\sum_{l=0}^{s_k}\hbar\omega_{lk}a_{l\mathbf{k}\nu}^+a_{l\mathbf{k}\nu}, \tag{2}$$

where $\mathbf{k}$ are the wave vectors, the quasi-discrete spectrum of which is determined by the cyclic boundary conditions, and l labels the roots of the dispersion equation

$$\varepsilon(\omega)\omega^2/c^2=k^2 \tag{3}$$

for a fixed value of k. We shall denote these roots by ω_{lk}^2, where $l=1,2,\ldots,s_k$. The polarizations of a mode are labeled by the index $\nu=1,2$; $a_{l\mathbf{k}\nu}^+$ and $a_{l\mathbf{k}\nu}$ are Bose creation and annihilation operators for a photon in the mode $l\mathbf{k}\nu$.

The operator of the vector potential of the field is equal to

$$\hat{\mathbf{A}}(\mathbf{r})=\sum_{l\mathbf{k}\nu}\mathbf{e}_{\mathbf{k}\nu}e^{i\mathbf{k}\mathbf{r}}\left(\frac{\hbar}{Vm(\omega_{lk})\omega_{lk}}\right)^{1/2}(a_{l\mathbf{k}\nu}+a_{l,-\mathbf{k},\nu}^+), \tag{4}$$

where $\mathbf{e}_{\mathbf{k}\nu}$ is the unit polarization vector of the mode $l\mathbf{k}\nu$, and

$$m(\omega_{lk})\equiv m_{lk}\equiv\frac{1}{2\pi c^2}\frac{\partial}{\partial\omega_{lk}^2}[\omega_{lk}^2\varepsilon(\omega_{lk})]. \tag{5}$$

The operator of the energy of interaction of the impurity center with the electromagnetic field has the form

$$\hat{V}=-\frac{1}{c}\mathbf{D}\hat{\mathbf{A}}(0)+\frac{1}{c^2}N_e\frac{e^2}{2\mu}\hat{\mathbf{A}}^2(0), \tag{6}$$

where $\mathbf{D}$ is the operator of the dipole moment produced by the electrons of the center, which is positioned at the coordinate origin, e and μ are the charge and mass

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of the electron, and N_e is the number of electrons on the center.

In a number of papers (see, e.g., Refs. 2 and 3), the energy of interaction of a molecule with light is written in the form $V=-DE$, where E is the intensity of the electric field of the light wave. This inexact expression does not lead to errors in the calculation of the probabilities of single-photon processes, but it is not valid for the treatment of multiphoton processes. Almost all authors have considered multiphoton processes not at impurity centers but at molecules of the host substance, assuming V to be a perturbation. But they have written the unperturbed operator of the energy of the photons taking the permittivity $\varepsilon(\omega)$ into account, i.e., with the assumption that the dispersion law of the photons has the form (3) and not $k^2=\omega^2/c^2$. It is necessary to emphasize that such an operator for the photon energy W already partly contains the interaction V of the molecules with the light (this is why the dispersion law of these photons differs from that in the case of the vacuum). Therefore, the perturbation of the operator W will be not V but some part of V that is difficult to separate. In this paper, which considers the interaction of light with impurity centers, these difficulties do not arise, since the contribution of the centers to the polarizability is not included in $\varepsilon(\omega)$.

We shall denote the Schrödinger electronic-vibrational states of the impurity center by $\psi_i, \psi_j, \ldots$, and the corresponding energy levels by $E_i E_j, \ldots$. We shall denote the state of the whole system by the set of indices $i, \ldots, n_{lk\nu}, \ldots$, where $n_{lk\nu}$ is the number of photons in a mode (in the volume of the repeat unit). In this state the average value of the Poynting vector is equal to

$$S=\frac{\hbar}{2\pi Vc}\sum_{k\nu=1}^{2}\sum_{l=0}^{\infty}\frac{\omega_{lk}\varepsilon^{1/2}(\omega_{lk})n_{lk\nu}}{m(\omega_{lk})}\frac{\mathbf{k}}{|\mathbf{k}|}. \tag{7}$$

The number of values of the vector k in the frequency interval $d\omega$ and inside the solid angle $d\Omega$ is equal to

$$\rho(\omega)d\omega d\Omega=V(\varepsilon^{1/2}(\omega)/4\pi^2c)m(\omega)\omega^2 d\omega d\Omega. \tag{8}$$

The formulas (7) and (8) are written for the frequency region in which $\varepsilon(\omega)>0$. In the region in which $\varepsilon(\omega)<0$, in an infinite medium plane light waves do not exist ($S=0, \rho=0$).

The Einstein relations for single-photon transitions have the form

$$B_{j\to i}=B_{i\to j}, \quad A_{j\to i}=\frac{2\hbar}{\pi c}\varepsilon^{1/2}(\omega_{ji})m(\omega_{ji})\omega_{ji}^3 B_{j\to i}, \tag{9}$$

where $B_{j\to i}$ and $B_{i\to j}$ are the coefficients determining the probabilities of induced emission and absorption, $A_{j\to i}$ is the coefficient determining the probability of spontaneous emission, and $\omega_{ji}=(E_j-E_i)/\hbar$.

Below we give a brief catalog of the expressions for the probabilities of single-photon and multiphoton processes in unit time. These expressions are easily obtained by the usual methods of quantum electrodynamics. The following system of notation will be used. The symbols $\omega_1^+, \omega_2^-, \ldots$, signify that, in the process under consideration, one photon of frequency ω_1 and polarization $\mathbf{e}_1$ is produced, one photon of frequency ω_2 and polarization $\mathbf{e}_2$ is absorbed, and so on. If factors $d\omega_1, d\Omega_1$ are present, this means that the photon ω_1 is produced in the frequency interval $d\omega_1$ and inside the solid angle $d\Omega_1$. The original incident light is always assumed to be exactly specified in direction. For example, the probability of spontaneous two-photon emission is denoted as $P(j\to i, \omega_1^+, \omega_2^+)d\omega_1 d\Omega_1 d\Omega_2$, and the probability of Raman scattering as $P(i\to j, \omega_1^-, \omega_2^+)d\Omega_2$.

Single-photon processes. The probability of absorption of a photon from the mode ω_1 in unit time is

$$P(i\to j, \omega_1^-)=\frac{2\pi|\mathbf{D}_{ij}|^2\omega_1 n_1}{3c^2V\hbar m(\omega_1)}\delta(\omega_{ji}-\omega_1), \tag{10}$$

where $\mathbf{D}_{ij}=\langle i|\mathbf{D}|j\rangle$ and n_1 is the number of photons in the mode ω_1. If in the absorption the mechanical system undergoes a transition between discrete levels E_i and E_j, the incident light should be nonmonochromatic. In this case,

$$P(i\to j, \omega_1^-)=\frac{4\pi^2|\mathbf{D}_{ij}|^2 J(\omega_1)}{3c\hbar^2\varepsilon^{1/2}(\omega_1)}, \quad \omega_1=\omega_{ji}, \tag{11}$$

where the spectral density of the intensity

$$J(\omega_1)=\int S(\omega_1)\rho(\omega_1)d\Omega_1, \tag{12}$$

and $S(\omega_1)$ is the intensity of the light in the one mode ω_1 [cf. (7)].

If the mechanical system undergoes a transition from a discrete level to one of the levels of the continuum, the incident light may be monochromatic and the corresponding probability is obtained from (10) by integrating over E_j:

$$P(i\to j, \omega_1^-)=\frac{2\pi|\mathbf{D}_{ij}|^2\omega_1 n_1}{3c^2Vm(\omega_1)}r(E_j), \quad \omega_1=\omega_{ji}, \tag{13}$$

where $r(E_j)dE_j$ is the number of energy levels of the mechanical system in the energy interval dE_j. In formulas (10), (11), and (13), averaging over all possible, equally probable orientations of the vector $\mathbf{D}_{ij}$ has been performed. The probability of emission of a photon of definite polarization into the solid angle $d\Omega_1$ in unit time is

$$P(j\to i, \omega_1^+)d\Omega_1=\frac{\omega_1^3\varepsilon^{1/2}(\omega_1)|\mathbf{D}_{ij}|^2}{6\pi c^3\hbar}(\bar{n}_1+1)d\Omega_1, \quad \omega_1=\omega_{ji}. \tag{14}$$

Here $\bar{n}_1$ is the average number of photons in the mode within the angle $d\Omega_1$.

Two-photon processes. The probability of Raman scattering of light is

$$P(i\to j, \omega_1^-, \omega_2^+)d\Omega_2=\frac{\omega_2\varepsilon^{1/2}(\omega_2)n_1(\bar{n}_2+1)}{2\pi c^2Vm(\omega_1)\omega_1}|M(\omega_1^-, \omega_2^+)|^2 d\Omega_2, \quad \omega_2=\omega_1-\omega_{ji}, \tag{15}$$

where

$$M(\omega_1^-, \omega_2^+)=\frac{1}{\hbar}\sum_{i'}\left\{\frac{(\mathbf{D}_{ji'}\mathbf{e}_2)(\mathbf{D}_{i'i}\mathbf{e}_1)}{\omega_{ii'}+\omega_1}+\frac{(\mathbf{D}_{ji'}\mathbf{e}_1)(\mathbf{D}_{i'i}\mathbf{e}_2)}{\omega_{ii'}-\omega_2}\right\}+\frac{N_e e^2}{\mu}\cos\theta\delta_{ij}, \tag{16}$$

where θ is the angle between the unit vectors $\mathbf{e}_1$ and $\mathbf{e}_2$.

When $i=j$, only the last term in (16) is nonzero and the formula (15) determines the probability of Rayleigh scattering; $\bar{n}_2$ is the average value of n_2 for the modes within the angle $d\Omega_2$.

From (15) and (7), putting $\bar{n}_2=0$, we can obtain the effective Raman-scattering cross section of the center:

$$d\sigma=\frac{\varepsilon^{1/2}(\omega_2)\omega_2}{c^4\varepsilon^{1/2}(\omega_1)\omega_1}|M(\omega_1^-,\omega_2^+)|^2 d\Omega_2. \tag{17}$$

The probability of two-photon absorption of light from two fixed modes is

$$P(i\to j,\omega_1^-,\omega_2^-)=\frac{2\pi n_1 n_2}{c^4V^2\omega_1\omega_2 m(\omega_1)m(\omega_2)}|M(\omega_1^-,\omega_2^-)|^2\delta(\omega_{ji}-\omega_1-\omega_2), \tag{18}$$

$$M(\omega_1^-,\omega_2^-)=\frac{1}{\hbar}\sum_{i'}\left\{\frac{(\mathbf{D}_{ji'}\mathbf{e}_2)(\mathbf{D}_{i'i}\mathbf{e}_1)}{\omega_{ii'}+\omega_1}+\frac{(\mathbf{D}_{ji'}\mathbf{e}_1)(\mathbf{D}_{i'i}\mathbf{e}_2)}{\omega_{ii'}+\omega_2}\right\}. \tag{19}$$

If in the two-photon absorption the mechanical system undergoes a transition from a discrete to a discrete energy level, then one of the waves, e.g., that possessing frequency ω_1, should be nonmonochromatic. In this case, from (18), taking (8) and (12) into account, we obtain

$$P(i\to j,\omega_1^-,\omega_2^-)=\frac{4\pi^2 n_2 J(\omega_1)|M(\omega_1^-,\omega_2^-)|^2}{c^3\hbar V\omega_2 m(\omega_2)\omega_1^2\varepsilon^{1/2}(\omega_1)},\quad \omega_{ji}=\omega_1+\omega_2. \tag{20}$$

If the mechanical system undergoes a transition from a discrete level to the continuum, both waves may be monochromatic and the corresponding probability is obtained from formula (18) by integrating over E_j:

$$P(i\to j,\omega_1^-,\omega_2^-)=\frac{2\pi\hbar n_1 n_2}{c^4V^2\omega_1\omega_2 m(\omega_1)m(\omega_2)}|M(\omega_1^-,\omega_2^-)|^2 r(E_j),\quad \omega_{ji}=\omega_1+\omega_2. \tag{21}$$

In a transition of the mechanical system from a discrete to a discrete level the probability of two-photon emission in unit time is equal to

$$P(j\to i,\omega_1^+,\omega_2^+)d\Omega_1 d\Omega_2 d\omega_2=\frac{\omega_1\omega_2(\varepsilon(\omega_1)\varepsilon(\omega_2))^{1/2}|M(\omega_1^+,\omega_2^+)|^2}{8\pi^3c^6}d\Omega_1 d\Omega_2 d\omega_2,\quad \omega_{ji}=\omega_1+\omega_2. \tag{22}$$

Here $M(\omega_1^+,\omega_2^+)$ is obtained from (19) by changing the signs in front of ω_1 and ω_2.

Multiphoton processes. The probability of absorption of two photons from a mode ω_1 and emission of a photon ω_2 into angle $d\Omega_2$ for $n_2=0$ is equal to

$$P(i\to j,2\omega_1^-,\omega_2^+)d\Omega_2=\frac{\hbar\varepsilon^{1/2}(\omega_2)n_1^2\omega_2}{2\pi c^7V^2m^2(\omega_1)\omega_1^2}|M(2\omega_1^-,\omega_2^+)|^2 d\Omega_2, \tag{23}$$

where $(\omega_{ji}=2\omega_1-\omega_2)$

$$M(2\omega_1^-,\omega_2^+)=\frac{1}{\hbar^2}\sum_{i',j'}\left\{\frac{(\mathbf{D}_{jj'}\mathbf{e}_2)(\mathbf{D}_{j'i'}\mathbf{e}_1)(\mathbf{D}_{i'i}\mathbf{e}_1)}{(\omega_{ii'}+\omega_1)(\omega_{ij'}+2\omega_1)}+\frac{(\mathbf{D}_{jj'}\mathbf{e}_1)(\mathbf{D}_{j'i'}\mathbf{e}_2)(\mathbf{D}_{i'i}\mathbf{e}_1)}{(\omega_{ii'}+\omega_1)(\omega_{ij'}+\omega_1-\omega_2)}+\frac{(\mathbf{D}_{jj'}\mathbf{e}_1)(\mathbf{D}_{j'i'}\mathbf{e}_1)(\mathbf{D}_{i'i}\mathbf{e}_2)}{(\omega_{ii'}-\omega_2)(\omega_{ij'}+\omega_1-\omega_2)}\right\}. \tag{24}$$

The probability of absorption of three photons from a mode ω_1 and emission of a photon ω_2 into angle $d\Omega_2$ for $n_2=0$ is equal to

$$P(i\to j,3\omega_1^-,\omega_2^+)d\Omega_2=\frac{\hbar^2\varepsilon^{1/2}(\omega_2)\omega_2 n_1^3}{2\pi c^9V^3m^3(\omega_1)\omega_1^3}|M(3\omega_1^-,\omega_2^+)|^2 d\Omega_2, \tag{25}$$

where $(\omega_{ji}=3\omega_1-\omega_2)$

$$M(3\omega_1^-,\omega_2^+)=\frac{1}{\hbar^3}\sum_{i',j',k'}\left\{\frac{(\mathbf{D}_{jj'}\mathbf{e}_2)(\mathbf{D}_{j'i'}\mathbf{e}_1)(\mathbf{D}_{i'k'}\mathbf{e}_1)(\mathbf{D}_{k'i}\mathbf{e}_1)}{(\omega_{ij'}+3\omega_1)(\omega_{ii'}+2\omega_1)(\omega_{ik'}+\omega_1)}+\frac{(\mathbf{D}_{jj'}\mathbf{e}_1)(\mathbf{D}_{j'i'}\mathbf{e}_2)(\mathbf{D}_{i'k'}\mathbf{e}_1)(\mathbf{D}_{k'i}\mathbf{e}_1)}{(\omega_{ij'}+2\omega_1-\omega_2)(\omega_{ii'}+2\omega_1)(\omega_{ik'}+\omega_1)}+\frac{(\mathbf{D}_{jj'}\mathbf{e}_1)(\mathbf{D}_{j'i'}\mathbf{e}_1)(\mathbf{D}_{i'k'}\mathbf{e}_2)(\mathbf{D}_{k'i}\mathbf{e}_1)}{(\omega_{ij'}+2\omega_1-\omega_2)(\omega_{ii'}+\omega_1-\omega_2)(\omega_{ik'}+\omega_1)}+\frac{(\mathbf{D}_{jj'}\mathbf{e}_1)(\mathbf{D}_{j'i'}\mathbf{e}_1)(\mathbf{D}_{i'k'}\mathbf{e}_1)(\mathbf{D}_{k'i}\mathbf{e}_2)}{(\omega_{ij'}+2\omega_1-\omega_2)(\omega_{ii'}+\omega_1-\omega_2)(\omega_{ik'}-\omega_2)}\right\}. \tag{26}$$

We now compare the formulas obtained above with those applied previously, which were written in analogy with the nondispersive case and took the dependence of ε on ω into account only in the final results; this was done, e.g., in the monographs of Bloembergen[2] and Kovarskiĭ,[4] in the textbook by Loudon,[3] and in numerous other books and articles. It is easy to see that the formulas obtained above go over into those applied previously if, in the right-hand side of (5), we omit the derivative of ε with respect to ω, i.e., replace $m(\omega)$ by $\varepsilon(\omega)/2\pi c^2$. Thus, the formulas (11), (14), (17), and (22), in which the factor $m(\omega)$ has cancelled, coincide with those previously. But the formulas (10), (13), (15), (18), (20), (21), (23), and (25), which contain factors $m(\omega)$, differ from those used previously.

We turn now from the general exact formulas for the probabilities of multiphoton processes to consider special models and approximations that make it possible to push the calculations further and obtain explicit frequency and temperature dependences of the probabilities of multiphoton processes. We shall assume that the state of the electrons of the local center follows adiabatically the comparatively slow vibrations of the ions of the crystal. In this case the multidimensional quantum numer j defining the state of the mechanical system decomposes into a set of quantum numbers s_j of the electronic subsystem and a set of oscillator quantum numbers... $N_\varkappa^j$... defining the state of the lattice vibrations ($\varkappa$ is the phonon wave-vector). Denoting the set of coordinates of the electrons by $\mathbf{r}$ and the normal coordinates of the vibrations by $\mathbf{q}_\varkappa$, we can write according to Refs. 5 and 6 the wavefunction of the mechanical system in the form

$$\Psi_j(r,\ldots\mathbf{q}_\varkappa\ldots)=\psi_{s_j}(r)\prod_\varkappa\Phi_N(\mathbf{q}_\varkappa-\mathbf{q}_\varkappa^j), \tag{27}$$

where Φ_N is the wavefunction of a one-dimensional harmonic oscillator in the state $N_\varkappa^j$ and $q_\varkappa^j$ is its equilibrium position, which depends on the state s_j of the electrons. In this approximation the Bohr frequency of the transition is equal to

$$\omega_{ji}=\omega_{ji}^0+\sum_\varkappa(\omega_\varkappa^j N_\varkappa^j-\omega_\varkappa^i N_\varkappa^i), \tag{28}$$

where ω_{ji}^0 is the frequency of the no-phonon transition and $\omega_\varkappa^j$ is the normal frequency of the lattice vibrations when the electronic subsystem is in the state s_j.

We assume further that in the sums (16), (19), (24), and (26) the principal contribution is given by terms in which in ω_{ji} we can neglect the second term in the

right-hand side of (28) in comparison with ω_{ij}^0, i.e., the heat evolved in the transition is small compared with $\hbar\omega_{ji}^0$. A small Stokes shift compared with $\hbar\omega_{ji}^0$ can be approximate criterion for these cases.[6] In this case the matrix elements (16), (19), (24), and (26) decompose into two factors:

$$M=M_sM_N. \tag{29}$$

Here M_s is obtained from M by the replacements $\omega_{km}\to\omega_{km}{}^0$, $D_{km}\to D_{s_k s_m}=\langle s_k|D|s_m\rangle$ and $\sum_m\to\sum_{s_m}$. For example, in the case (19),

$$M_s(\omega_1^-,\omega_2^-) = -\frac{1}{\hbar}\sum_{s_{i'}}\left\{\frac{(D_{s_j s_{i'}}e_2)(D_{s_{i'}s_i}e_1)}{\omega_{ii'}^0+\omega_1}+\frac{(D_{s_j s_{i'}}e_1)(D_{s_{i'}s_i}e_2)}{\omega_{ii'}^0+\omega_2}\right\}\omega_{ji'}^0\omega_{i'i}^0. \tag{30}$$

Introducing for the overlap integral of the oscillator functions the notation

$$F_{ji}=\prod_\varkappa\int\Phi_N^*(q_\varkappa-q_\varkappa^j)\Phi_{N'}(q_\varkappa-q_\varkappa^i)\,dq_\varkappa, \tag{31}$$

where $N\equiv N_\varkappa^j$ and $N'\equiv N_\varkappa^i$, for processes with r intermediate states we can write M_N in the form

$$M_N=\sum_{N_\varkappa^1N_\varkappa^2\ldots N_\varkappa^r}F_{j1}F_{12}\ldots F_{r-1,r}F_{ri}. \tag{32}$$

Using the relations

$$\sum_{N_\varkappa^i}\Phi_N(q_\varkappa-q_\varkappa^i)\Phi_N(q_\varkappa'-q_\varkappa^i)=\delta(q_\varkappa-q_\varkappa'),\qquad \sum_{N_\varkappa^m}F_{j'm}F_{mi'}=F_{j'i'}, \tag{33}$$

we can reduce the expression (32) to the form

$$M_N=F_{ji}. \tag{34}$$

We note that M_s and , in particular, the expression (30) depend only on the properties of the electronic subsystem; M_s does not depend on the oscillator quantum numbers $N_\varkappa$ or, consequently, on the temperature. It is possible to calculate M_s only after the form of the electron center and its energy spectrum and wavefunctions have been specified, which lies beyond the scope of this article. According to (34) and (31), M_N does not depend on r, i.e., it is the same for processes of any order and coincides with the M_N of simple single-photon processes.

In most cases, immediately before a multiphonon process the vibrational subsystem is in thermal equilibrium, i.e., the $N_\varkappa^i$ of the initial state are determined by Planck's formula. In this case $|M_N|^2$ depends on the temperature, and this determines the entire temperature dependence of the probability in the process. In the context of the theory of single-photon processes M_N has been calculated in many papers, beginning with Refs. 5, 7, and 8 with neglect of the dispersion of $\omega_\varkappa$ and with Refs. 9 and 10 with this dispersion taken into account. As in these papers, we shall assume that the initial state s_i and final states s_j of the electronic subsystem are fixed. However, it is not possible to fix the corresponding vibrational states...$N_\varkappa^i$... and...$N_\varkappa^j$... because of the infinite number and uncontrollability of the quantum numbers $N_\varkappa$. Therefore, it is not possible to use the detailed formulas given above for the probabilities $P(i\to j,\ldots)$ of processes for comparison with experiment. But we can determine and compare with experiment the probabilities $P(s_i\to s_j,\ldots)$ of multiphoton processes occurring with fixed s_i and s_j and fixed energies and numbers of all the photons produced and absorbed. This also fixes ω_{ji}. Such a process can proceed from any state of the vibrational subsystem and is accompanied by an infinite number of variants of the numbers $N_\varkappa$, restricted only by the condition that the sum over $\varkappa$ in the right-hand side of (28) be fixed.

According to the theorem of addition of probabilities, the probability of such a process can be represented in following way:

$$P(s_i\to s_j,\ldots)=\sum_{\substack{\ldots N_\varkappa^j\ldots\\ \omega_{ji}=\text{const}}}\bar{P}(i\to j,\ldots). \tag{35}$$

Here the summation is performed over...$N_\varkappa^j$...for a fixed initial vibrational state...$N_\varkappa^i$...and for a given fixed value of the frequency (28) [i.e., a fixed value of $\sum_\varkappa$ in (28)]. The bar denotes averaging over all possible initial states...$N_\varkappa^i$...of the vibrational subsystem, under the assumption that this subsystem is in thermal equilibrium.

The formulas given above for $P(i\to j,\ldots)$ and the properties of M_s mentioned above show that everything except the factor $|M_n|^2$ can be taken outside the summation sign in the sum (35). In fact, the sum

$$\sum_{\substack{\ldots N_\varkappa^j\ldots\\ \omega_{ji}=\text{const}}}|M_N|^2\equiv R. \tag{36}$$

was calculated in Refs. 5, 7, and 9. In particular, if one neglects the dispersion of $\omega_\varkappa$ and puts $\omega_\varkappa=\omega_0$, it was found there that

$$R(\omega_{ji},T)=e^{-(N+1/2)a}\left(1+\frac{1}{N}\right)^{l/2}I_l(z), \tag{37}$$

$$\omega_{ji}=\omega_{ji}{}^0+l\omega_0,\quad l=\sum_\varkappa(N_\varkappa^j-N_\varkappa^i),\quad N=\frac{1}{\exp(\hbar\omega_0/kT)-1}, \tag{38}$$

$$a=\sum_\varkappa(q_\varkappa^i-q_\varkappa^j)^2,\quad z=a((N+1)N)^{1/2}, \tag{39}$$

where $I_l(z)=J_l(iz)/i^l$, where J_l is the Bessel function of order l.

The dependence of $R(\omega_{ji},T)$ on ω_{ji} and T is exactly the same as the well known[5,11,12] frequency and temperature dependence of the probability of single-photon absorption by an impurity center in a nondispersive medium.

It is now not difficult to calculate the probabilities (35) for all the types of multiphoton processes considered above. For this it is necessary to use the formula (29) and the prescription for obtaining M_s that follows it, and also formulas (36)–(39). In all cases the results of the calculation can be written compactly in the form

$$P(s_i\to s_j,\ldots)=R(\omega_{ji},T)\,[P(i\to j,\ldots)]_{M\to M_s}, \tag{40}$$

where the expressions for $P(i\to j,\ldots)$ have already been given above. In place of...one must write the frequencies and number of the emitted and absorbed

photons. For example, for two-photon absorption, from (20) we obtain

$$P(s_i \to s_j, \omega_1^-, \omega_2^-) = R(\omega_{ji}, T) \frac{4\pi^2 n_2 J(\omega_1) |M_s(\omega_1^-, \omega_2^-)|^2}{c^3 \hbar V \omega_2 m(\omega_2) \omega_1^2 \varepsilon^{1/2}(\omega_1)}, \quad \omega_{ji} = \omega_1 + \omega_2, \tag{41}$$

where M_s has the form (30).

Thus, in the approximation used above, when the heat evolved in the transitions is small compared with $\hbar\omega_{km}^0$, in the probabilities (40) of all multiphoton processes the same factor $R(\omega_{ji}, T)$ is separated out and includes the entire dependence of the probability on the temperature and on the evolved heat $l\hbar\omega_0$ accompanying the process. The second factor in (40) has the form it would have if we were considering the electronic subsystem alone, not interacting with the vibrations. It can be calculated only after the form of the electronic center and its energy spectrum and wavefunctions, and also the dispersion law $\varepsilon(\omega)$ of the light in the medium, have been specified.

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Theory of high-frequency and thermodynamic properties of iron garnets

V. N. Krivoruchko and D. A. Yablonskiĭ

Donets Physicotechnical Institute, Academy of Sciences, Ukrainian SSR

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The high-frequency magnetic susceptibility tensor is found for iron garnets. For yttrium-iron garnet, expressions are obtained for the temperature renormalization and for the damping of both the acoustical and the optical branches of the spectrum. The temperature renormalization of the acoustic branch of the spectrum differs considerably from the case of a ferromagnet. Thus in a ferromagnet, the energy of a spin wave with a given wave vector decreases with rise of temperature; in the ferrite, it increases. Corrections to the thermodynamic potential and magnetization of the ferrite, resulting from spin-wave interaction, are also found; and it is shown that these corrections have the opposite sign to those for a ferromagnet.

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1. INTRODUCTION

The study of the high-frequency and thermodynamic properties of ferrites has been the object of a large amount of experimental and theoretical research. In a theoretical description of the observed results, a ferrite is, as a rule, treated within the framework of the single-sublattice model. Although this approach does allow one to obtain a number of results in a simple manner, nevertheless the question of the limits of its applicability remains open. This is due to the fact that a ferrite is a many-sublattice system. As is shown in the present paper, a more consistent description of an iron garnet, within the framework of a two-sublattice model, leads to some conclusions that are in direct contradiction to those that follow from the single-sublattice model. Among these must be included, in particular, the conclusion that the energy of the acoustic branch of the spectrum of spin waves with a given wave vector increases, not decreases, with rise of temperature. This result is in agreement with experiment.[1]

Precision experiments have recently been conducted in the study of the dependence of the damping of spin waves on the wave vector and on the temperature in yttrium-iron garnet (YIG).[2] These experiments showed that the conclusions obtained within the framework of the single-sublattice model,[3,4] do not describe the observed results; specifically, at temperatures 200–300 K the experimental data are systematically higher than the theoretical values.[2] As is shown in the present paper, increase of the damping coefficient oc-

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