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THEORY OF STIMULATED RAMAN SCATTERING OF LIGHT IN ANISOTROPIC CRYSTALS

V. L. STRIZHEVSKIĬ, V. V. OBUKHOVSKIĬ, and A. M. PANARIN

Kiev State University

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The general theory of SRS on vibrations of arbitrary symmetry in anisotropic crystals is developed in the given-field approximation. In the absence of resonator effects, the problem of generation of the first Stokes SRS component is reduced to the problem of the radiation from dipoles of fluctuation origin in a medium pumped by a linearly polarized exciting wave. The pump deforms the dielectric constant of the medium, and at low anisotropy of the tensor of the proper dielectric constant, the symmetry of the pumped medium corresponds in most cases of practical importance to the symmetry of a uniaxial crystal. Accordingly, an "ordinary" and an "extraordinary" wave (relative to the pumped medium) are produced in the scattered field, and are characterized by separate gain coefficients. Explicit expressions for the gains are obtained in terms of the components of the corresponding spontaneous-scattering tensor and other parameters of the problem. The question of the degree of depolarization of the scattered radiation is discussed. The case when additional illumination from an external source is produced at the Stokes frequency is also briefly discussed. The indicatrix of SRS by fully-symmetrical vibrations in cubic crystals is investigated for the case of a rectangular scattering volume (with square cross section) and a cylindrical volume.

INTEREST has recently increased in the investigation of stimulated Raman scattering (SRS) of light in crystals. A systematic theory of such scattering was developed by Lugovoĭ[1]. However, in analyzing the SRS intensities, Lugovoĭ confined himself to fully-symmetrical vibrations. Yet cases of SRS by not-fully-symmetrical vibrations in anisotropic crystals have also been registered[2].

The purpose of the present paper is to develop a theory describing (under certain simplifying assumptions) the intensities of SRS by vibrations of arbitrary symmetry. We consider both isotropic and ansiotropic crystals and reveal the distinguishing features of non-fully-symmetrical vibrations.

We confine ourselves to the first Stokes component. It is assumed that there is no resonator effect (it can be eliminated by using a suitable experimental observation technique[3]). The approximations of the given pump field and of the stationary character of the problem are employed. Polariton effects[4] are neglected. The medium is assumed to be non-magnetic and spatially homogeneous.

1. GENERAL THEORY

In the macroscopic SRS theory, as in any other macroscopic scattering theory, the scattering field must be formed by averaging the microscopic field over values that are physically infinitesimally small. At the same time, the second averaging operation, carried out on going from the microscopic theory to the macroscopic description, namely averaging over the particle motion, is postponed until the scattering intensities are determined[5]. Accordingly, all the quantities characterizing the field are macroscopically smooth, but the connection between the induction and the electric field of the scattered wave

$$\mathbf{D} = \hat{\varepsilon}\mathbf{E} + 4\pi\mathbf{d} \qquad (1)$$

contains a specific polarization of fluctuation origin

$$\mathbf{d}(\mathbf{r}, t) = \beta(\mathbf{r}, t)\mathbf{E}_0(\mathbf{r}, t) = \int \mathbf{d}(\mathbf{r}, \omega)\exp(-i\omega t)\,d\omega$$

($\mathbf{E}_0$ is the pump field).

The first term in (1) describes the propagation of the scattered wave in the medium, and the second describes the occurrence of the wave. The relation (1) is formally analogous to the corresponding expression from[5] (formula (93.2)), but the SRS considered here differs in essence in the fact that the dielectric constant ϵ_{ij} of a medium pumped by an intense laser wave

$$\mathbf{E}_0(\mathbf{r}, t) = \mathbf{A}_0 \exp[i(\mathbf{k}_0\mathbf{r} - \omega_0 t)] + \text{к.с.} \qquad (\mathbf{A}_0 = A_0\mathbf{e}_0,\ e_0 = 1),$$

contains a term μ_{ij} that depends on $\mathbf{E}_0$: $\epsilon_{ij} = \epsilon^0_{ij} + \mu_{ij}$. Here ϵ^0_{ij} is the dielectric constant of the medium in the absence of the field $\mathbf{E}_0$, and $\mu_{ij} = 4\pi|A_0|^2\chi_{ijkl}e_{0k}e_{0l}$, with $\chi_{ijkl} = \chi_{ijkl}(\omega = \omega + \omega_0 - \omega_0)$ the corresponding nonlinear polarizability[6].

The scattered-wave field $\mathbf{E}(\mathbf{R}, t)$ satisfies the equation

$$\operatorname{rot}\operatorname{rot}\mathbf{E} + \frac{\hat{\varepsilon}}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = -\frac{4\pi}{c^2}\frac{\partial^2\mathbf{d}}{\partial t^2}.$$

In view of its linearity, the field

$$\mathbf{E}(\mathbf{R}, t) = \int \mathbf{E}(\mathbf{R}, \omega)\exp(-i\omega t)\,d\omega$$

at the point R on the surface of the scattering volume v can be regarded as a superposition of fields produced by elementary sources located at different points of the volume v. A distinguishing feature of the radiation problem is in this case not only the fact that ϵ is complex, but that it is essentially a tensor. Even if the medium were isotropic prior to the irradiation, a linearly polarized pump wave would deform ϵ^0 and give rise to anisotropy.

We confine ourselves here to the most interesting cases, when the symmetry of the pumped medium corresponds to a uniaxial crystal. This includes, first, liquids and cubic crystals, and also uniaxial crystals with e_0 parallel to the optical axis C of the unpumped crystal. For most oscillations in uniaxial crystals (for details see Sec. 4) this category also includes cases when $e_0 \perp C$, if one neglects in the calculation of the gain coefficients the usually negligible anisotropy of the tensor ϵ^0_{ij}.1) A similar stipulation can be made also with respect to vibrations in biaxial crystals of monoclinic and rhombic syngony with $e_0 \parallel C_2$ or $e_0 \perp C_2$ (in the class C_s, $e_0 \parallel \sigma$ or $e_0 \perp \sigma$, where σ is the symmetry plane). For simplicity, we assume that ϵ^0_{ij} is real (the generalization to the case of complex ϵ^0_{ij} is obvious). In connection with the foregoing, we shall put henceforth $\epsilon^0_{ij} = \epsilon^0\delta_{ij}$.

The radiation of a dipole in a uniaxial crystal with complex ϵ was considered in[1] on the basis of a procedure developed in[7]; we shall use here the results obtained in these investigations. The z axis will be aligned with the preferred principal axis of the tensor μ_{ij} corresponding to a principal value $\mu_{\parallel}$ (the remaining two principal values are equal to $\mu_{\perp}$). The z axis determines the "optical" axis of the pumped crystal C_p. It is important that in the region of applicability of the given-field approximation we have $|\mu_{\parallel,\perp}| \ll \epsilon^0$, and therefore it is necessary to neglect the difference between the quantities $\epsilon_{\parallel,\perp} = \epsilon^0 + \mu_{\parallel,\perp}$ and ϵ^0 when they do not enter under the exponential sign. Taking the results of[1] into account, we get

$$\mathbf{E}(\mathbf{R},\omega) = k^2 \int \frac{\exp(ik\nu\rho)}{\rho}\{\mathbf{a} - \mathbf{b}[1 - \exp ik(\sqrt{\varepsilon_\perp} - \nu)\rho]\}\,d^3r,$$

$$\nu = (\varepsilon_\perp \cos^2\theta + \varepsilon_\parallel \sin^2\theta)^{1/2} \qquad k = \frac{\omega}{c}, \quad \mathbf{a} = [\boldsymbol{\Omega}[\mathbf{d}(\mathbf{r},\omega)\boldsymbol{\Omega}]], \quad \boldsymbol{\Omega} = \frac{\boldsymbol{\rho}}{\rho}$$

$$\boldsymbol{\rho} = \mathbf{R} - \mathbf{r} = (\rho, \theta, \varphi), \quad \mathbf{b} = [\mathbf{s}[\mathbf{d}_\perp(\mathbf{r},\omega)\mathbf{s}]], \quad \mathbf{s} = \boldsymbol{\Omega}_\perp/\Omega_\perp, \quad (2)^*$$

where $\boldsymbol{\Omega}_\perp$ and $\mathbf{d}_\perp$ are the components of the vectors $\boldsymbol{\Omega}$ and d in the xy plane; ρ, θ and φ are the spherical coordinates of the vector ρ. The Fourier components of the magnetic field can be obtained with the aid of the formula $H(R, \omega) = (ik)^{-1}\,\mathrm{curl}_R E(R, \omega)$.

We shall henceforth be interested in the component $E_e = e(e \cdot E)$ of the field E; the polarization of this component is specified by the unit vector e. In an anisotropic crystal, e determines one of the natural directions of polarization of the unpumped crystal. All the quantities considered below correspond to a fixed polarization e of the field E.

It is convenient to relate the final result to the luminosity of the surface bounding the volume v, namely $\Phi(R) = \langle S, N_R \rangle$, where N_R is the unit vector normal to the surface at the given point R, and $S = cE_e \times H_e/4\pi$ is the Poynting vector. The symbol $\langle \ldots \rangle$ denotes the still-to-be-performed operation of averaging over the motion of the particles of the medium. In the calculation of $\Phi(R)$ we shall need the correlators $\langle d_i(r_i, \omega_1) d_j(r_2, \omega_2)\rangle$, which reduce to linear combinations of the correlators $\langle \beta_{ik}(r, \omega')\beta_{jm}(r_2, \omega'')\rangle$ with $\omega' = \omega_1 \pm \omega_0$ and $\omega'' = \omega_2 \pm \omega_0$. The latter are proportional to $\delta(\omega' + \omega'')\delta(r_1 - r_2)$. Discarding terms with $\delta(\omega_1 + \omega_2 \pm 2\omega_0)$, which lead to $\Phi(R)$ terms that oscillate at frequencies $2\omega_0$ and vanish after the time averaging corresponding to the experimental-registration process, we obtain

$$\langle d_i(\mathbf{r}_1\ \omega_1) d_j(\mathbf{r}_2, \omega_2)\rangle = \tilde{\gamma}_{ijkm}(\omega_1) e_{0k} e_{0m} |A_0|^2 \delta(\omega_1 + \omega_2)\delta(\mathbf{r}_1 - \mathbf{r}_2).$$

In the case of homogeneous broadening of the scattering line, the tensor $\tilde{\gamma}_{ijkm}(\omega_1)$ can be represented in the form $\tilde{\gamma}_{ijkm}(\omega_1) = F(\omega)\gamma_{ijkm}$, where γ_{ijkm} does not depend on ω and determines the integrated intensity of the spontaneous Raman scattering, while $F(\omega)$ is a form factor for which we can put, in the case of an isolated scattering line,

$$F(\omega) = \frac{1}{\pi}\frac{\gamma}{(|\omega| - \tilde{\omega})^2 + \gamma^2}, \quad \int_0^\infty F(\omega)\,d\omega \approx 1.$$

Here $\tilde{\omega}$ is the frequency of the center of the line and 2γ is its half-width.

By comparison with the known results on the integrated line intensities in crystals[8] and liquids[9], we can readily verify that in the case of crystals

$$\gamma_{ijkm} = N_0 \sum_\nu \alpha^{(\nu)}_{ik}\alpha^{(\nu)}_{jm},$$

and in the case of liquids

$$\gamma_{ijkm} = N_0 L \nu_j \langle \alpha^{(\nu)}_{ik}\alpha^{(\nu)}_{jm}\rangle_{\mathrm{rot}}.$$

Here $\alpha^{(\nu)}_{ik}$ is the tensor of spontaneous Raman scattering per unit cell[8] or per molecule[9], and the index ν numbers the mutually degenerate vibrations with given frequency ω_j; N_0 is the number of cells or scattering molecules per unit volume; $L = L(\tilde{\omega}) = \frac{1}{27}[\epsilon(\omega_0) + 2]^2[\epsilon(\tilde{\omega}) + 2]$ is the Lorentz factor of the effective field; the symbol $\langle \ldots \rangle_{\mathrm{rot}}$ denotes averaging over the rotations of the molecule. The result of the averaging does not depend on ν; ν_j is the degeneracy multiplicity.

Taking the foregoing into account, the calculation of $\Phi(R)$ no longer entails any difficulty. We present the final result:

$$\Phi(\mathbf{R}) = \int d\Omega \int_0^\infty d\omega \cos\vartheta B(\mathbf{R}, \boldsymbol{\Omega}; \omega), \tag{3}$$

$$B(\mathbf{R}, \boldsymbol{\Omega}; \omega) = M[w_1 u(g_1 R_0) + w_2 u(g_2 R_0)], \tag{4}$$

$$M = \left(\frac{\omega}{c}\right)^4 \frac{n}{n_0} R_0 N_0 F(\omega) I_0, \quad w_1 = \pi_1 w_1^0, \quad w_1^0 = \sum_\nu |(\mathbf{e}_1, \alpha^{(\nu)}\mathbf{e}_0)|^2,$$

$$w_2 = \pi_2 w_2^0, \quad w_2^0 = \sum_\nu |(\mathbf{e}_2, \alpha^{(\nu)}\mathbf{e}_0)|^2, \quad \pi_{1,2} = (\mathbf{e}, \mathbf{e}_{1,2})^2, \quad I_0 = \frac{c}{2\pi} n_0 |A_0|^2 \tag{5}$$

Here I_0 is the pump intensity and n and n_0 are the refractive indices at the frequencies ω and ω_0. The unit vectors e_1 and e_2 are perpendicular to $\boldsymbol{\Omega}$2), with e_1 lying in the plane passing through C_p and $\boldsymbol{\Omega}$ ("principal cross section") and e_2 perpendicular to it. Formulas (3)–(5) have been written out for the case of crystals. In the case of a liquid it is necessary in addition to average $w_{1,2}$ over the rotations of the molecule and to introduce the factor $L(\omega)$ into M (see below). Further, $R_0 = R_0(\boldsymbol{\Omega})$ is the length of the seg-

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1) At arbitrary directions of e_0, the symmetry of the uniaxial crystal is lost by the tensor μ_{ij}.

*$[\Omega[d\Omega]] \equiv \Omega \times [d \times \Omega]$.

2) The unit vector Ω determines the observation direction in the investigation of the angular distribution of the SRS intensity.

ment drawn inside the volume v through the point with radius vector **R** in the direction of **Ω**, and ϑ is the angle between Ω and $\mathbf{N_R}$ defined inside the scattering medium and connected with θ by the relation $\cos\vartheta = \sin\theta\cos\varphi'$, where φ' is the angle between $\mathbf{N_R}$ and $\mathbf{\Omega}_\perp$. Finally,

$$u(x)=\frac{e^x-1}{x},\quad g_1=\frac{k}{n}(\tau_\perp+\Delta\tau\sin^2\theta),\quad g_2=\frac{k}{n}\tau_\perp,\tag{6}$$

$$\tau_{\parallel,\perp}=-\operatorname{Im}\mu_{\parallel,\perp},\quad \Delta\tau=\tau_\parallel-\tau_\perp.$$

The quantity $B(R, \Omega; \omega)$ has the meaning of the spectral density of the surface brightness of the radiation, i.e., the spectral density of the light flux emitted by a unit surface in a unit solid angle near the direction of **Ω**. The integration with respect to dΩ is within the solid angle 2π.

Formulas (3)–(6) have a simple physical meaning. The quantities $Mw^0_{1,2}$ constitute the spectral density of the intensity of the spontaneous Raman scattering polarized along the directions $e_{1,2}$. The latter determine the polarizations of the "extraordinary" and "ordinary" waves relative to the pumped medium. If the polarization e is intermediate between e_1 and e_2, the scattering field $\mathbf{E_e}$ must be expanded in terms of e_1 and e_2, and the propagation of both components, with separate gains (with respect to intensity) g_1 and g_2, must be considered. In the general case (unlike the case of fully-symmetrical oscillations considered in[1]), both coefficients $g_{1,2}$ differ from zero.

By choosing as the perturbation-theory basis, for example, mechanical excitons[10] and performing a microscopic calculation of $\chi_{ijkl}(\omega = \omega + \omega_0 - \omega_0)$ in the dipole approximation using the well-known procedure[11], we can verify that in the case of homogeneously broadened isolated lines in arbitrary anisotropic crystals, the following representation is valid:

$$\chi_{ijkl}=\frac{1}{\hbar v_0(\omega\ -\Delta\omega+i\nu_f)}\sum_\nu \alpha_{ik}^{(\nu)}\alpha_{jl}^{(\nu)},\quad \Delta\omega=\omega_0-\omega,\tag{7}$$

$v_0 = N_0^{-1}$, ω_f is the frequency of the corresponding mechanical phonon. Using (6) and (7), we get

$$\tau_{\parallel,\perp}=K\lambda_{\parallel,\perp},\quad K=Q\gamma_f,\quad Q=\frac{8\pi^2I_0}{cn_0\hbar v_0[(\omega_f-\Delta\omega)^2+\gamma_f^2]},\tag{8}$$

where $\lambda_{\parallel,\perp}$ are the principal values of the tensor

$$\lambda_{ij}=\sum_\nu \alpha_{ik}^{(\nu)}\alpha_{jl}^{(\nu)}e_{0k}e_{0l}.\tag{9}$$

We note also that

$$w_1^0=\lambda_\perp+\Delta\lambda\sin^2\theta,\quad w_2^0=\lambda_\perp,\quad \Delta\lambda=\lambda_\parallel-\lambda_\perp.\tag{10}$$

In his SRS observations, Bobovich[12] used a pump at the scattering frequency, produced by the auxiliary luminescence spectrum, as a result of which the initial intensity level greatly exceeded the spontaneous Raman scattering intensity. This made it possible, in particular, to register SRS by vibrations with a high threshold; one can hope that such a procedure will also yield SRS from non-fully symmetrical vibrations. Formulas of type (3) and (6) also remain valid for this case, but their explicit form depends on the static properties of the pump. Assuming the pump to be natural light, we can easily verify that the result reduces to replacing $Mw^0_{1,2}$ in (4) by $B_0/2$, where B_0 is the spectral density of the pump energy flux per unit solid angle near the given direction. In the case of a pump wave linearly polarized along the unit vector e′, we have

$$B=B_0[w_1'u(g_1R_0)+w_2'u(g_2R_0)+2w_3'\tilde{u}(g_3R_0,g_4R_0)],$$

where

$$w_1'=\pi_1(\mathbf{e}_1,\mathbf{e}')^2,\quad w_2'=\pi_2(\mathbf{e}_2,\mathbf{e}')^2,\quad w_3'=(\mathbf{e}\mathbf{e}_1)(\mathbf{e}\mathbf{e}_2)(\mathbf{e}'\mathbf{e}_1)(\mathbf{e}'\mathbf{e}_2),$$

$$\tilde{u}(x,y)=\frac{e^x\cos(y+\eta)-\cos\eta}{\sqrt{x^2+y^2}},\quad \operatorname{tg}\eta=\frac{y}{x},\quad g_3=\frac{1}{2}(g_1+g_2),$$

$$g_4=\frac{k}{2n}\Delta\tilde{\tau}\sin^2\theta,$$

$$\Delta\tilde{\tau}=\tilde{\tau}_\parallel-\tilde{\tau}_\perp,\quad \tilde{\tau}_{\parallel,\perp}=\operatorname{Re}\mu_{\parallel,\perp}=\tilde{K}\lambda_{\parallel,\perp},\quad \tilde{K}=Q(\omega_f-\Delta\omega).$$

The term containing $\tilde{u}$ describes interference effects due to the fact that the pump-field components along $e_{1,2}$ are coherent in this case. If $g_{1,2,3}$ do not differ greatly from one another, these effects can lead, in particular, to a dependence of the SRS line shape on its polarization e, to an oscillatory dependence of B on R_0 (with a period $2\pi g_4^{-1}$) on going from the center of the line into the region where $g_4 \neq 0$, etc. We shall not stop to discuss this in greater detail.

The quantity $B(R, \mathbf{\Omega}, \omega)$ contains complete information on the angular and frequency dependences of the SRS intensity, on the polarization effects, on the influence of the symmetry of the oscillations, etc. We shall apply the results obtained below to the investigation of SRS in crystals. We first make a few remarks pertaining to liquids.

Averaging over the molecule rotations, we find

$$w_1^0=\frac{\nu_f}{15}\sigma_2^2+\frac{\nu_f}{45}(5\sigma_1^2+\sigma_2^2)\sin^2\theta,\quad w_2^0=\frac{\nu_f}{15}\sigma_2^2,$$

where σ_1 and σ_2 are the trace and the anisotropy[13] of the tensor α. Further, in liquids we have $\chi_{ijkl} = a_0\delta_{ij}\delta_{kl} + b_0\delta_{ik}\delta_{jl} + c_0\delta_{il}\delta_{jk}$; on the other hand, $\chi_{ijkl} = N_0L(\omega)\langle\kappa_{ijkl}\rangle_{\mathrm{rot}}$, where $\kappa_{ijkl}(\omega = \omega + \omega_0 - \omega_0)$ is the corresponding tensor per molecule; it satisfies a representation similar to (7). Using the results of[14] or the simpler method of invariants[15], we can easily express the phenomenological parameters a_0, b_0, and c_0 in terms of $\sigma_{1,2}$:

$$a_0=c_0=G_0\sigma_2^2,\quad b_0=\frac{G_0}{3}(5\sigma_1^2-2\sigma_2^2),$$

$$G_0=G_1+iG_2=\frac{\nu_fN_0L(\omega)}{15\hbar}\frac{\omega_f-\Delta\omega+i\gamma_f}{(\omega_f-\Delta\omega)^2+\gamma_f^2}.$$

We then readily obtain, using (6) and (8),

$$g_1=G[(5\sigma_1^2+\sigma_2^2)\sin^2\theta+3\sigma_2^2],\quad g_2=3G\sigma_2^2,$$

$$g_3={}^1\!/\!_2G[(5\sigma_1^2+\sigma_2^2)\sin^2\theta+6\sigma_2^2],$$

$$g_4=\tilde{G}(5\sigma_1^2+\sigma_2^2)\sin^2\theta,\quad G=\frac{8\pi^2kI_0G_2}{3n_0^2c},\quad \tilde{G}=\frac{4\pi^2kI_0G_1}{3n_0^2c}.$$

When $\theta \neq 0$, we have $g_1 > g_2$ and $w_1^0 > w_2^0$, so that the SRS line, as is well known, is usually polarized along e_0. The degree of depolarization is

$$\eta=B_\perp/B_\parallel=\exp[-R_0G(5\sigma_1^2+\sigma_2^2)\sin^2\theta],$$

where $B_{\parallel,\perp}$ are the values of B when e lies respectively in the plane of the "principal cross section" and perpendicular to it. η depends quite strongly on θ, and at $\theta = 0°$ (SRS perpendicular to the exciting beam) we have $\eta = 1$. For non-fully-symmetrical vibrations ($\sigma_1 = 0, \sigma_2 \neq 0$) at $\theta = 90°$ (SRS in the direction of the exciting beam) we have $g_1 = 4G\sigma_2^2$, $g_2 = 0.75g_1$, $g_3 = 0.88g_1$, and at $\theta = 0°$ we have $g_1 = g_2 = g_3 = 3G\sigma_2^2$, so that the investigation of SRS with pumping would make it possible to observe here a number of new effects, some of which were mentioned above.

2. CASE OF CUBIC CRYSTALS

We proceed to consider SRS in crystals. The use of the procedure formulated above is possible only if the pumped crystal has the symmetry of a uniaxial crystal. This includes, first, the case of a crystal that is cubic in the unpumped state; this case will be considered in the present section. The "optical axis" C_p is in this case always parallel to e_0. As is clear from (3)–(6) and (8), the problem reduces to finding the principal values $\lambda_{\parallel,\perp}$ of the tensor λ_{ij} (9).

In the case of fully-symmetrical vibrations[3] we have $\lambda_\parallel = a^2$ and $\lambda_\perp = 0$; this case was considered earlier in[1]. For doubly degenerate vibrations $\lambda_\parallel = 4b^2$ and $\lambda_\perp = 0$. Finally, for triply degenerate vibrations $\lambda_\parallel = 0$ and $\lambda_\perp = d^2$. Accordingly, for fully-symmetrical vibrations

$$g_1 = 2g_3 = \frac{k}{n} Ka^2 \sin^2\theta, \qquad g_2 = 0, \qquad g_4 = \frac{k}{2n}\tilde{K}a^2 \sin^2\theta;$$

for doubly degenerate vibrations

$$g_1 = 2g_3 = \frac{4k}{n} Kb^2 \sin^2\theta, \qquad g_2 = 0, \qquad g_4 = \frac{2k}{n}\tilde{K}b^2;$$

and for triply degenerate vibrations

$$g_1 = -\frac{k}{n} Kd^2 \sin^2\theta, \qquad g_2 = \frac{k}{n} Kd^2, \qquad g_3 = \frac{k}{2n} Kd^2 \cos^2\theta,$$

$$g_4 = -\frac{k}{2n}\tilde{K}d^2 \cos^2\theta.$$

Let us consider in detail the angular dependence (the indicatrix) of SRS for fully-symmetrical vibrations. It is expedient to connect it with the spectral power density of the SRS radiation in a fixed direction $\Psi(\Omega, \omega) = \cos\vartheta \int B(R, \Omega; \omega)ds$, where the integration is over the surface of the radiating volume v. The value of Ψ depends, obviously, on the shape of the volume v. Let us consider the case of a volume of rectangular or cylindrical form, and confine ourselves to the angular dependence in the xy plane[4].

a) Volume of rectangular shape with height L and square cross section l on the side; $k_0 \parallel L$, e_0 parallel to one of the sides of the square. The concrete formulas for Ψ can be obtained by using relations (4) and (5). We present only the final results. Assume that a square diaphragm with side l is placed at the end face of the scattering volume v in such a way that only the radiation emerging from the end face is registered, whereas the radiation of the side walls is blocked. In this case

$$\frac{\Psi}{\Psi_0} = u(q_1)(q_1 + q_2) - q_2 e^{q_1}, \qquad \vartheta < \vartheta' = \operatorname{arc\,tg}\frac{l}{L}, \tag{11}$$

$$\frac{\Psi}{\Psi_0} = u(\varphi) - 1, \qquad \vartheta \geqslant \vartheta',$$

where

$$q_1 = \frac{k}{n}\frac{Ka^2L}{\cos\vartheta}, \qquad q_2 = \frac{\operatorname{tg}\vartheta}{\operatorname{tg}\vartheta'}, \qquad \varphi = \frac{q_1}{q_2},$$

$$\Psi_0 = \frac{Llk^3}{K} n_0 N_0 F(\omega) I_0 \cos^2\varphi_1;$$

φ_1 is the angle between e and e_0.

If there is no diaphragm, so that the radiation of the side walls is not blocked, then

$$\frac{\Psi}{\Psi_0} = u(q_1)(q_1 + 2q_2 + q_1q_2) - 2q_2 e^{q_1}, \qquad \vartheta < \vartheta',$$

$$\frac{\Psi}{\Psi_0} = u(\varphi)\frac{1}{q_2}(q_1 + 2q_2 + q_1q_2) - 2e^{\varphi}, \qquad \vartheta \geqslant \vartheta'. \tag{12}$$

b) Volume of cylindrical form with height L and diameter l, with $k_0 \parallel L$ and $e_0 \perp L$. In this case

$$\frac{\Psi}{\Psi_0} = \frac{2}{\pi}\left[\left(\arccos q_2 - q_2\sqrt{1 - q_2^2}\right)e^{q_1} + \left(\frac{\pi}{2} - \arccos q_2\right)\frac{2I_1(\varphi)}{\varphi} - \frac{\pi}{2}\right.$$

$$\left. + \frac{4}{\varphi}\sum_{n=1}^{\infty}\alpha_n \frac{\sin(n\pi/2) - \sin n \arccos q_2}{n}\right], \qquad \vartheta < \vartheta' = \operatorname{arc\,tg}\frac{l}{L},$$

$$\frac{\Psi}{\Psi_0} = \frac{\pi}{2}\left(\frac{2I_1(\varphi)}{\varphi} - 1\right) + \frac{4}{\varphi}\sum_{n=1}^{\infty}\alpha_n\frac{\sin(n\pi/2)}{n}, \qquad \vartheta \geqslant \vartheta'; \tag{13}$$

$$\alpha_n = I_{n-1}(\varphi) - \frac{n(n+1)}{\varphi} I_n(\varphi), \qquad \varphi = \frac{q_1}{q_2};$$

I_n are modified Bessel functions of the first kind.

In the region $\vartheta \ll \vartheta'$ we can use the approximation

$$\Psi = \Psi_0 e^{q_1}\left\{1 - \frac{2q_2}{\pi}\left[2 - \frac{2}{q_1} - q_2^2\left(\frac{1}{2} - \frac{3}{q_1} + \frac{3}{q_1^2}\right)\right]\right\}$$

and in the absence of a diaphragm we have

$$\Psi = \Psi_0\left\{(e^{q_1} - 1)\left[1 + \frac{2q_2}{\pi}\left[\pi + \frac{\pi - 2}{q_1} + \frac{1}{\varphi}\left(\frac{4}{q_1} - 2 - \frac{3q_2}{q_1^2}\right)\right]\right]\right.$$

$$\left. - \frac{2q_2}{\pi}e^{q_1}\left[2 + \pi + q_2\left[\frac{2}{q_1} - \frac{q_2}{2}\left(1 - \frac{3}{q_1} + \frac{6}{q_1^2}\right)\right]\right]\right\}$$

(when $q_2 < 0.6$, the accuracy of the last formula is already ~ 3%).

In the case of either a rectangular or a cylindrical volume, we have in the region $\vartheta < \vartheta'$ for the half-width Δ of the SRS line (in the case of sufficient gain)

$$\Delta = \Delta_0\left(\frac{\ln 2\cos\vartheta}{gL}\right)^{1/2}, \qquad g = \frac{8\pi^2 a^2 k I_0}{n n_0 c\hbar v_0 \gamma_f}, \tag{14}$$

where Δ_0 is the half-width of the spontaneous Raman scattering line, and g is the value of g_1 at $\theta = 90°$. From (14) it follows, in particular, that at sufficiently high temperatures (when k_0T exceeds the energy of those phonons whose interaction leads to a broadening energy level $\hbar\omega_f$ of the given optical phonon), in view of the fact that $\gamma_f \sim T$[17], we have $\Delta_0 \sim T$, $g \sim T^{-1}$, and $\Delta \sim T^{3/2}$. The SRS line shape is approximately Gaussian[1] and the intensity at the maximum is

$$\Psi_{max} = \tilde{\Psi}_0 \exp(gL/\cos\vartheta).$$

When $\vartheta \ll \vartheta'$, we have $\tilde{\Psi}_0 = \Psi$, and in the region $\vartheta \lesssim \vartheta'$ we can easily express Ψ_0 from (11)–(13). The temperature dependence of Ψ_{max} reduces mainly to

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[3] For nonzero components of the tensor α (the quantities a, b, c, d, e, f) we use here the notation of [16].

[4] Student Yu. Yashkir of the Kiev State University took part in these calculations.

the factor $\exp(CT^{-1})$, where C is a constant. At $\vartheta > \vartheta'$, using the rectangular volume as an example, we conclude that the formulas given above for Δ and $\Psi_{\max}$ remain in force, with cos ϑ replaced by sin ϑ $(\tan\vartheta')^{-1}$.

3. CASE OF UNIAXIAL AND BIAXIAL CRYSTALS

Let us proceed to consider uniaxial and biaxial crystals. We confine ourselves here to an investigation of the cases of greatest interest, when the pump polarization vector e_0 is parallel or perpendicular to the symmetry axes C_2, C_3, C_4, or C_6[5]. Crystals of triclinic syngony, in which there are no symmetry elements, will not be considered. In the case of uniaxial crystals with $e_0 \perp C$ and in the case of biaxial crystals, as already noted, we neglect the anisotropy of the tensor $\overset{0}{\epsilon}_{ij}$ of the unpumped crystal. Since the results of Sec. 1 correspond to a μ_{ij} symmetry of the uniaxial-crystal type, we shall omit cases when all three principal directions of the tensor μ_{ij} are different. The number of such cases is small: five out of 25 modes for different classes of uniaxial crystals with $e_0 \perp C$ (see below). For $e_0 \parallel C$, all 25 modes are considered. There are likewise no exceptions for biaxial crystals (if we disregard the fact that we do not consider crystals of triclinic syngony). We shall discuss separately the cases of uniaxial and biaxial crystals (in the unpumped state) with $e_0 \parallel C$ and $e_0 \perp C$.

1. Uniaxial crystals, $e_0 \parallel C$. For all fully-symmetrical vibrations we have $\lambda_\perp = 0$ and $\lambda_\parallel = b^2$. For non-fully-symmetrical vibrations $\lambda_\parallel = 0$ and the values of $\lambda_\perp$ are listed in Table I. For those nonfully-symmetrical vibrations not included in Table I we have $\lambda_\perp = 0$. Consequently, such vibrations do not go into SRS[6]. C_p coincides with C in all cases.

Accordingly, for all fully-symmetrical vibrations we have $g_1 = 2g_3 = k/n\, Kb^2\sin^2\theta$, $g_2 = 0$, $g_4 = k/2n\, \tilde{K}b^2\sin^2\theta$. For non-fully-symmetrical vibrations with $\lambda_\perp \neq 0$ we have

$$g_1 = \frac{k}{n}K\lambda_\perp\cos^2\theta, \qquad g_2 = \frac{k}{n}K\lambda_\perp, \qquad g_3 = \frac{k}{2n}K\lambda_\perp(1+\cos^2\theta),$$

$$g_4 = -\frac{k}{2n}\tilde{K}\lambda_\perp\sin^2\theta.$$

2. Uniaxial crystals, $e_0 \perp C$. For all fully-symmetrical vibrations we have $C_p \parallel e_0$, $\lambda_\perp = 0$, $\lambda_\parallel = a^2$. The values of $\lambda_{\parallel,\perp}$ and the orientation of C_p for non-fully-symmetrical vibrations are contained in Table II. Five cases, $E(C_3, D_3, C_{3v})$ and $E_g(C_{3i}, D_{3d})$ have been disregarded, since they lead to symmetry of the biaxial-crystal type in the presence of pumping.

Accordingly, for fully-symmetrical vibrations

$$\sigma = 2g_3 = \frac{k}{n}Ka^2\sin^2\theta, \quad g_2 = 0, \quad g_4 = \frac{k}{2n}\tilde{K}d^2\sin^2\theta.$$

For all non-fully symmetrical vibrations contained in Table II, with the exception of $E_2(C_6, D_6, C_{6v})$, $E'(C_{3h}, D_{3h})$ and $E_{2g}(C_{6h}, D_{6h})$, we have

$$g_1 = 2g_3 = \frac{k}{n}K\lambda_\parallel\sin^2\theta, \qquad g_2 = 0, \qquad g_4 = \frac{k}{2n}\tilde{K}\lambda_\parallel\sin^2\theta.$$

For the last seven, for which $\lambda_\parallel = 0$ and $\lambda_\perp \neq 0$, we get

$$g_1 = \frac{k}{n}K\lambda_\perp\cos^2\theta, \qquad g_2 = \frac{k}{n}K\lambda_\perp, \qquad g_3 = \frac{k}{2n}K\lambda_\perp(1+\cos^2\theta),$$

$$g_4 = -\frac{k}{2n}\tilde{K}\lambda_\perp\sin^2\theta.$$

3. Biaxial crystals; e_0 is parallel or perpendicular to one of the axes C_2. For both $e_0 \parallel C_2$ and $e_0 \perp C_2$ we have $\lambda_\perp = 0$. The values of $\lambda_\parallel$ differ from zero and depend on the orientation e_0. They are contained in Table III, where the orientation of C_p is also indicated.

The results solve the problem, and express the coefficients g in terms of the components of the tensor α, which can be determined from spontaneous Raman scattering experiments, and in terms of other parame-

Table I. Values of $\lambda_\perp$ for non-fully-symmetrical vibrations of uniaxial crystals with $e_0 \parallel C$. Only with vibrations with $\lambda_\perp \neq 0$ are given. In all cases $\lambda_\parallel = 0$

Types of vibration	$\lambda_\perp$	Types of vibration	$\lambda_\perp$
$E(C_3)$, $E_g(C_{3i})$	e^2+f^2	$E(C_{4v}, D_4, D_{2d})$, $E_g(D_{4d})$	e^2
$E(D_3, C_{3v})$, $E_g(D_{3d})$	d^2	$E_1(C_6)$, $E''(C_{3h})$, $E_{1g}(C_{6h})$	c^2+d^2
$E(C_4, S_4)$, $E_g(C_{4h})$	e^2+f^2	$E_1(D_6, C_{6v})$, $E''(D_{3h})$, $E_{1g}(D_{6h})$	c^2

Table II. Values of $\lambda_{\parallel,\perp}$ and orientation of the "optical axis" C_p of the pumped crystal for non-fully-symmetrical vibrations in uniaxial crystals with $e_0 \perp C$

Types of oscillations	$\lambda_\parallel$	$\lambda_\perp$	Orientation of C_p
$B(C_4, S_4)$, $B_g(C_{4h})$	c^2+d^2	0	$C_p \perp C$*
$E(C_4, S_4)$, $E_g(C_{4h})$	e^2+f^2	0	$C_p \parallel C$
$B_1(C_{4v}, D_4, D_{2d})$, $B_{1g}(D_{4d})$	c^2	0	$C_p \parallel e_0$
$B_2(C_{4v}, D_4, D_{2d})$, $B_{2g}(D_{4d})$	d^2	0	$C_p \parallel e_0$
$E(C_{4v}, D_4, D_{2d})$, $E_g(D_{4d})$	e^2	0	$C_p \parallel C$
$E_1(C_6)$, $E''(C_{3h})$, $E_{1g}(C_{6h})$	c^2+d^2	0	$C_p \parallel C$
$E_2(C_6)$, $E'(C_{3h})$, $E_{2g}(C_{6h})$	0	e^2+f^2	$C_p \parallel C$
$E_1(D_6, C_{6v})$, $E''(D_{3h})$, $E_{1g}(D_{6h})$	c^2	0	$C_p \parallel C$
$E_2(D_6, C_{6v})$, $E'(D_{3h})$, $E_{2g}(D_{6h})$	0	d^2	$C_p \parallel C$

*The angle ϵ between C_p and e_0 is given by $\tan\epsilon = c/d$.

Table III. Values of $\lambda_\parallel$ and orientation of the "optical axis" C_p of the pumped crystal for vibrations in biaxial crystals at $e_0 \parallel C_2$ and $e_0 \perp C_2$*. In all cases $\lambda_\perp = 0$

Types of oscillations	$e_0 \parallel C_2$		$e_0 \perp C_2$	
	$\lambda_\parallel$	Orientation of C_p	$\lambda_\parallel$	Orientation of C_p
$A(C_2)$, $A'(C_s)$, $A_g(C_{2h})$	c^2+d^2	$\mathrm{tg}\,\varepsilon = d/c$, ε in $\sigma(x', z')$	a^2+d^2	$\mathrm{tg}\,\varepsilon = a/d$, ε in $\sigma(C_2, e_0)$
$B(C_2)$, $A''(C_s)$, $B_g(C_{2h})$	f^2	$C_p \parallel y'$	e^2	$C_p \perp \sigma(C_2, e_0)$
$A(D_2)$, $A_1(C_{2v})$, $A_g(D_{2h})$	c^2	$C_p \parallel C_2$	a^2	$C_p \parallel e_0$
$B_1(D_2)$, $A_2(C_{2v})$, $B_{1g}(C_{2h})$	0	—	d^2	$C_p \perp \sigma(C_2, e_0)$
$B_1(C_{2v})$, $B_2(D_2)$, $B_{2g}(C_{2h})$	e^2	$C_p \parallel x'$	$e^2\cos^2\psi$	$C_p \parallel C_2$
$B_2(C_{2v})$, $B_3(D_2)$, $B_{3g}(C_{2h})$	f^2	$C_p \parallel y'$	$f^2\sin^2\psi$	$C_p \parallel C_2$

*We use the following notation: x′, y′, z′—principal axes of the tensor $\overset{0}{\epsilon}_{ij}$, with $z' \parallel C_2$ (in the group C_s, the z′ axis is perpendicular to the symmetry plane σ); σ(x′, z′) and σ(C_2, e_0) are the planes passing through x′, z′ and C_2, e_0, respectively; Ψ is the angle between e_0 and x′.

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[5] In crystals of class C_s, the preferred direction is not the symmetry axis but the normal to the symmetry plane.

[6] This includes vibrations with symmetry types $B(C_4, S_4)$, $B_g(C_{4h})$, $B_1(C_{4v}, D_4, D_{2d})$, $B_{1g}(D_{4d})$, $B_2(C_{4v}, D_4, D_{2d})$, $B_{2g}(D_{4d})$, $E_2(C_6, D_6, C_{6v})$, $E'(C_{3h}, D_{3h})$ and $E_{2g}(C_{6h}, D_{6h})$.

ters that are usually known. We note that for all the cases contained in the tables, one of the quantities $\lambda_{\parallel,\perp}$ vanishes. Accordingly, one of the two quantities $g_{1,2}$ vanishes at $\theta = 90°$ (and the same pertains to $w_{1,2}$), so that the SRS lines are either practically completely polarized along e_0 (the case $g_1 \neq 0$, $g_2 = 0$) or are completely depolarized ($g_1 = 0$, $g_2 \neq 0$).

In the general case, the SRS process is described by two quantities $g_{1,2}$ which are simultaneously different from zero. But the degree of polarization is

$$\eta = \exp\zeta, \qquad \zeta = -\frac{k}{n} KR_0\Delta\lambda \sin^2\theta_1$$

and therefore the vibrations with $\lambda_{\parallel} \neq 0$ are predominantly polarized in the plane of the ''principal cross section,'' and the vibrations with $\lambda_{\perp} \neq 0$ are polarized perpendicular to it. This rule is violated either in the vicinity of the threshold or for sufficiently small θ, for in both cases $|\zeta|$ is small.

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