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STIMULATED RAMAN SCATTERING IN ANISOTROPIC MEDIA

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Stimulated Raman scattering in crystals is considered. It is shown that each scattering component (besides the first Stokes component) consists of a fundamental and supplementary radiation. Both radiations propagate at definite (but different) angles with respect to the directions of the incident wave. The supplementary radiation is due to interaction of various types of waves in a crystal and can be observed only in a certain range of orientations of the crystal relative to the direction of the incident wave. Equations are deduced for the angles of emission of the stimulated Raman scattering components, and the corresponding angular widths are estimated.

In this paper we consider stimulated Raman scattering (SRS) in anisotropic media. It is assumed that a plane monochromatic wave of the scattered radiation (pump) from a laser is incident on the investigated medium (which is located outside the laser cavity). It is known that the scattered radiation contains in this case a number of components (both in the Stokes and in the anti-Stokes region), and all the components (except the first Stokes component) are emitted at definite angles to the direction of the pump wave (see, for example, [1,2]). We shall show below that in crystals we can observe for each scattering component additional radiation, connected with the interaction of different types of waves and propagating at a different angle (compared with the main radiation).

The angular distribution of the intensities of the components of the scattered radiation for isotropic media was considered in a number of papers [3-7][1)]. In particular, in [4] we investigated the properties of the first Stokes and first anti-Stokes scattering components, and in [3,5-7] we investigated the properties of the Stokes and anti-Stokes components of arbitrary orders. The results of [4] and [3,5-7] in the ratio of the first Stokes to the first anti-Stokes components coincide. These investigations are based on an analysis of the radiation field in an active medium consisting of individual (independent) dipole molecules (whose amplitudes are determined by the usual Placek cross section and whose frequency is equal to the first Stokes frequency). The active medium is the investigated substance itself in the presence of an intense pump wave (from the quantum point of view, allowance for the activity of the medium corresponds to the presence of a stimulated part in the probability of the two-quantum transition of the molecule).

It was shown in [5,6] that the emission angles of SRS components for an isotropic medium (gas or liquid) can be obtained from the following conditions for the wave vectors:

$$\mathbf{k}_m + m\mathbf{k}_{-1}^{(m)} = (m+1)\mathbf{k}_0, \quad \mathbf{k}_{-m} + (m-1)\mathbf{k}_0 = m\mathbf{k}_{-1}^{(-m)}, \tag{1}$$

where $k_l = \omega_l c^{-1} n(\omega_l)$, ω_l is the frequency of the component of order l, n is the refractive index of the medium, the index model pertains to the anti-Stokes component of order of m, and the index $-m$ pertains to the Stokes component of order m ($l = \pm m$). With the aid of (1) we easily obtain explicit expressions for these angles[2)]

$$2\sin\frac{\vartheta_m}{2} = \left\{\frac{[k_m + mk_{-1} - (m+1)k_0][mk_{-1} + (m+1)k_0 - k_m]}{(m+1)k_0 k_m}\right\}^{1/2},$$

[1)]Here and below we are referring to volume radiation in stimulated Raman scattering. We do not deal with radiation of the surface type[8].

[2)]It must be borne in mind that for any component of order $l \neq -1$ there are two emission angles (one inside the active region and the other outside). This is valid even when the refractive indices of the active medium and of the ambient coincide[6]. By ϑ_l we mean the emission angle (with respect to the pump wave vector $\mathbf{k}_0$) outside the active region, and in the case when the refractive index does not change on going from the active region into the surrounding medium. As shown in [6], ϑ_l is the angle between the vectors $\mathbf{k}_l$ and $\mathbf{k}_0$.

$$2\sin\frac{\vartheta_{-m}}{2} = \left\{\frac{[k_{-m}+(m-1)k_0-mk_{-1}][k_{-m}+(m-1)k_0+mk_{-1}]}{(m-1)k_0k_{-m}}\right\}^{1/2}. \quad (2)$$

The total widths $\Delta\vartheta_l$ corresponding to these angles depend on the values of ϑ_l themselves and, if

$$|l\varepsilon_{-1}''| \ll \frac{k_l+lk_{-1}-(l+1)k_0}{k_{-1}} \ll 1 \quad (3)$$

they are determined by the expressions [4,6]

$$\Delta\vartheta_l = \frac{lk_{-1}^2}{(l+1)k_0k_l}\frac{|l\varepsilon_{-1}''|}{n^2}\frac{1}{\vartheta_l}, \quad (4)$$

where ξ''_{-1} is the imaginary part of the dielectric constant of the medium (in the presence of pumping) at the first Stokes frequency (in the absence of pumping, the medium is assumed nonabsorbing).

We shall generalize below the equations (1), (2), and (4) to the case of anisotropic media. It is in fact necessary to determine radiation from a point dipole at the first Stokes frequency in an active medium (which is a transparent non-magnetic crystal in the absence of pumping)[3]. This problem can be solved by the method developed in [5,6]. However, its complete solution is quite cumbersome. At the same time, the greatest interest attaches to the expressions for the emission angles of the SRS components and to the widths corresponding to them. These scattering characteristics can be obtained by a simpler method. It is merely necessary to modify (1) and (4).

We assume below for simplicity that in the investigated crystal the pump wave and the first Stokes component (in the wave zone) are of the same type α ($\alpha = 1, 2$). Then any other dipole emission component (within the limits of the active region) represents a sum of two terms, each of which has its own direction of maximum intensity. If we again assume that the refractive indices ($\sqrt{\epsilon^{(x)}}$, $\sqrt{\epsilon^{(y)}}$, $\sqrt{\epsilon^{(z)}}$) do not change on going from the active region into the surrounding medium, then the sought radiation directions will be determined from the following conditions for the wave vectors:

$$\mathbf{k}_{m\beta}+m\mathbf{k}_{-1\alpha}^{(m,\beta)} = (m+1)\mathbf{k}_{0\alpha}, \quad \mathbf{k}_{-m\beta}+(m-1)\mathbf{k}_{0\alpha} = m\mathbf{k}_{-1\alpha}^{(-m,\beta)} \quad (5)$$

($\beta = 1, 2$). For $\alpha = \beta$ we have the fundamental radiation, and for $\alpha \neq \beta$ we obtain the supplementary radiation. We choose an arbitrary plane passing through the dipole in question and the pump wave vector $\mathbf{k}_0$. In this plane, the sought directions make angles $\vartheta_l^{\alpha\beta}$ with the vector $\mathbf{k}_0$. With the aid of (5) we can easily obtain in explicit form a system of equations for the angles $\vartheta_l^{\alpha\beta}$:

$$2\sin\frac{\vartheta_m^{\alpha\beta}}{2} = \left\{\frac{[k_{m\beta}(\vartheta_m^{\alpha\beta})+mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(m)})-(m+1)k_{0\alpha}]}{(m+1)k_{0\alpha}k_{m\beta}(\vartheta_m^{\alpha\beta})} \times [mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(m)})+(m+1)k_{0\alpha}-k_{m\beta}(\vartheta_m^{\alpha\beta})]\right\}^{1/2},$$

$$2\sin\frac{\vartheta_{\alpha\beta}^{(m)}}{2} = \left\{\frac{[k_{m\beta}(\vartheta_m^{\alpha\beta})+mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(m)})-(m+1)k_{0\alpha}]}{m(m+1)k_{0\alpha}k_{-1\alpha}(\vartheta_{\alpha\beta}^{(m)})} \times [k_{m\beta}(\vartheta_m^{\alpha\beta})+(m+1)k_{0\alpha}-mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(m)})]\right\}^{1/2},$$

$$2\sin\frac{\vartheta_{-m}^{\alpha\beta}}{2} = \left\{\frac{[k_{-m\beta}(\vartheta_{-m}^{\alpha\beta})+(m-1)k_{0\alpha}-mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(-m)})]}{(m-1)k_{0\alpha}k_{-m\beta}(\vartheta_{-m}^{\alpha\beta})} \times [k_{-m\beta}(\vartheta_{-m}^{\alpha\beta})+(m-1)k_{0\alpha}+mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(-m)})]\right\}^{1/2},$$

$$2\sin\frac{\vartheta_{\alpha\beta}^{(-m)}}{2} = \left\{\frac{[k_{-m\beta}(\vartheta_{-m}^{\alpha\beta})-mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(-m)})+(m-1)k_{0\alpha}]}{m(m-1)k_{0\alpha}k_{-1\alpha}(\vartheta_{\alpha\beta}^{(-m)})} \times [k_{-m\beta}(\vartheta_{-m}^{\alpha\beta})+mk_{-1\alpha}(\vartheta_{\alpha\beta}^{(-m)})-(m-1)k_{0\alpha}]\right\}^{1/2}. \quad (6)$$

Here $k_{l\alpha}(\vartheta) = \omega_l c^{-1} n_\alpha(\omega_l, \vartheta)$. This system includes also the absorption angles in the first Stokes component ($\vartheta_{\alpha\beta}^{(l)}$). Inasmuch as in crystals the lengths of the wave vectors depend on the directions, the equations in (6) constitute a rather complicated system (with respect to the two unknowns $\vartheta_l^{\alpha\beta}$ and $\vartheta_{\alpha\beta}^{(l)}$). We shall therefore assume below for simplicity that the pump wave vector $\mathbf{k}_0$ lies in the plane of the optical axis of the crystal, and consider the pattern of the phenomenon only in this plane. This plane is of greatest interest with respect to the supplementary radiation.

Let the axis of the Cartesian system of coordinates x, y, and z coincide with the principal axis of the crystal ($\epsilon^{(x)} < \epsilon^{(y)} < \epsilon^{(z)}$). Then both optical axes lie in the xz plane. We denote by μ the angle between the optical axis and the x axis and by ν the angle between the x axis and the vector $\mathbf{k}_0$ (see the figure). As is well known, the refractive indices $n_{1,2}(\omega, \vartheta)$ are determined by the following formulas (see, for example [9]).

[3]It must be borne in mind here that in ordinary Raman scattering in a crystal, an appreciable correlation can exist between different dipoles as a result of the collective lattice vibrations. The presented analysis is valid only for the case when the radius of this correlation is small compared with the wavelength of the scattered radiation.

$$n_1(\omega, \vartheta) \equiv \sqrt{\varepsilon^{(y)}}(\omega),$$

$$n_2(\omega, \vartheta) = \left(\frac{\sin^2(\vartheta+\nu)}{\varepsilon^{(x)}(\omega)} + \frac{\cos^2(\vartheta+\nu)}{\varepsilon^{(z)}(\omega)}\right)^{-1/2} \quad (7)$$

We now assume that $\alpha = 1$. From (6) we easily see that when $\beta = 1$ the angles ϑ_l^{11} are determined by the same expressions as in an isotropic medium with refractive index $n(\omega) \equiv \sqrt{\epsilon^{(y)}(\omega)}$. When $\beta = 2$ we obtain additional angles ϑ_l^{12}, which are determined, for example when $l = m$ (i.e., for the anti-Stokes components), by the equations

$$2\sin\frac{\vartheta_m{}^{12}}{2} = \left\{\frac{[k_{m2}(\vartheta_m{}^{12}) + mk_{-11} - (m+1)k_{01}]}{(m+1)k_{01}k_{m2}(\vartheta_m{}^{12})} \times [mk_{-11} + (m+1)k_{01} - k_{m2}(\vartheta_m{}^{12})]\right\}^{1/2} \quad (8)$$

($k_{m2}(\vartheta) = \omega_m c^{-1} n_2(\omega_m, \vartheta)$). This equation contains one unknown ϑ_m^{12} (the values of k_{-11} and k_{01} do not depend on ϑ). We are interested only in its real roots. An investigation of Eq. (8) shows that it always has a real solution only for a definite interval of crystal orientations relative to the direction of the pump wave. We can verify that the additional radiation angle ϑ_m^{12} (in the anti-Stokes component of the order m) always exists if

$$0 < \mu - \nu < \vartheta_m{}^{11}. \quad (9)$$

Furthermore, this angle must lie in the range $\mu - \nu < \vartheta_m^{12} < \vartheta_m^{11}$. In the opposite case (i.e., either when $\nu > \mu$ or when $\mu - \nu > \vartheta_m^{11}$) there are also small intervals of ν in which (8) has a real solution. However, the limits of these intervals are determined in a more complicated manner. We note that under normal dispersion of the refractive index, the approximate equations $\vartheta_m^{11} \approx m\vartheta_1^{11}$ is valid.

Let us estimate the widths $\Delta\vartheta_l^{\alpha\beta}$ corresponding to the emission angles $\vartheta_l^{\alpha\beta}$. To this end we write formula (4) in a form that will be suitable for the quantities k_{-1} and k_l which depend on ϑ. Essentially, formula (4) denotes that the angular widths are connected with absorption (amplification) in the medium of the wave at the first Stokes frequency. It is easy to see that the expression (4) follows from the condition

$$|\Delta[k_l - |(l+1)\mathbf{k}_0 - l\mathbf{k}_{-1}|]| = k_{-1}\frac{|l\varepsilon_{-1}''|}{n^2}$$

for values of k_{-1} and k_l independent of ϑ. In the anisotropic case we must stipulate

$$|\Delta[k_{l\beta} - |(l+1)k_{0\alpha} - lk_{-1\alpha}|]| = k_{-1\alpha}\frac{|l\varepsilon_{-1}''|}{n^2}$$

and take into account at the same time the dependence of $k_{l\beta}$ on $k_{-1\alpha}$ on ϑ. This gives a more general expression, which determines the widths $\Delta\vartheta_l^{\alpha\beta}$ of the SRS components in an anisotropic medium,

$$\Delta\vartheta_l{}^{\alpha\beta} = \frac{k_{-1}|l\varepsilon_{-1}''|}{n^2|p_1+p_2+p_3|}, \quad (10)$$

where

$$p_1 = \frac{(l+1)k_{0\alpha}k_{l\beta}\vartheta_l{}^{\alpha\beta}}{lk_{-1\alpha}}, \quad p_2 = -\frac{dk_{l\beta}}{d\vartheta}(\vartheta_l{}^{\alpha\beta}),$$

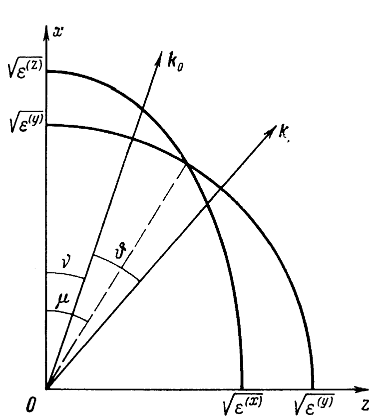
$$p_3 = -\frac{k_{l\beta}}{k_{-1\alpha}}\frac{dk_{-1\alpha}}{d\vartheta}(\vartheta_{\alpha\beta}{}^{(l)}). \quad (11)$$

In particular, for the angles ϑ_m^{11} considered above, the widths $\Delta\vartheta_m^{11}$ are determined by the same expressions as in an isotropic medium with refractive index $n(\omega) \equiv \sqrt{\epsilon^{(y)}(\omega)}$. For the widths $\Delta\vartheta_m^{12}$ of the supplementary radiation we obtain from this

$$p_2 = \frac{k_{l\beta}}{2n}\left(\frac{\varepsilon^{(z)}(\omega_l)}{\varepsilon^{(x)}(\omega_l)} - 1\right)\sin[2(\vartheta_l{}^{\alpha\beta} + \nu)], \quad p_3 = 0 \quad (12)$$

(For simplicity we have assumed here $\epsilon^{(z)} - \epsilon^{(x)} \ll \epsilon^{(z)}$). We see that in general the angular widths of the additional radiation are smaller than, or of the order of, the corresponding angular widths of the fundamental radiation. We note that all the conditions for the observation of the additional radiation angles of SRS components can be satisfied, for example, with a $CaCO_3$ crystal, for which $\mu \approx 8.7°$, $\vartheta_1^{11} \approx 1.4°$, and $\vartheta_m^{11} \approx m\vartheta_1^{11}$.

The foregoing calculations are based on an analysis of the radiation field of individual dipoles in an active medium. The justification for this approach in the theory is explained in [3-7]. At the present time there exists also another point of view, stipulating that the components of the scattered radiation are waves that are plane in the entire active volume. Such an approach was developed in several papers, starting with [10].



These two approaches lead to different expressions for the emission angles of the second, third, etc. Stokes and anti-Stokes scattering components. However, for normal dispersion the refractive indices (which possess in the visible region all the most active substances with respect to Raman scattering) the differences in the predicted angles turned out to be much smaller than the corresponding angular widths. This in turn, does not make it possible to identify experimentally the correct point of view concerning this phenomenon. Observation of the supplementary radiation angles in crystals can afford such an opportunity, inasmuch as the difference between the calculated angles now turns out to be much larger than the corresponding angular widths.

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