# Stimulated scattering following excitation by radiation having a broad angular spectrum

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It is shown theoretically that in the case of stimulated scattering in a pump field having a broad angular spectrum an appreciable parametric contribution is made to the enhancement of the Stokes radiation, and this contribution influences its angular distribution.

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In the theoretical analysis of stimulated scattering the pump radiation is usually represented in the form of a homogeneous plane wave. We indicate in the present communication interesting features of stimulated scattering that are produced in a pump field that is "nonmonochromatic" in angle. The employed calculation method is effective in the approximation of a constant pump field also under the condition that its angular spectrum is broad enough. For the sake of argument we shall deal here with the case of stimulated Raman scattering (SRS) by fully-symmetrical molecular vibrations, although the main conclusions apply equally well to stimulated Mandel'shtam-Brillouin scattering (SMBS).

We consider the simplest case when a monochromatic pumping radiation $E_L(\mathbf{r},t)=E_L(\mathbf{r})\exp(i\omega_L t)$ is incident on a layer of Raman-active medium located between the unbounded planes $z=0$ and $z=l$; The pumping radiation consists of two plane waves polarized along the $y$ axis (Fig. 1):

$$E_L(\mathbf{r}) = \mathcal{E}_{L1}e^{-i\mathbf{k}_{L1}\mathbf{r}} + \mathcal{E}_{L2}e^{-i\mathbf{k}_{L2}\mathbf{r}}, \quad (1)$$

where $|\mathbf{k}_{L1,2}|=k_L=\mu\omega_L/c$ ($\mu$ is the linear refractive index, assumed to be the same inside and outside the layer); $k_{L1x}=-k_{L2x}$, $k_{L1y}=k_{L2y}=0$, $k_{L1z}=k_{L2z}$.

Neglecting the radiation of the anti-Stokes and of the higher Stokes components, we write down the equation for the Fourier amplitudes of the field of the first Stokes component

$$\hat{L}E(\omega_S, \mathbf{r}) = \frac{4\pi\omega_S^2}{c^2}P^{sp}(\omega_S, \mathbf{r}), \quad (2)$$

where $\hat{L}$ is a linear differential operator that includes

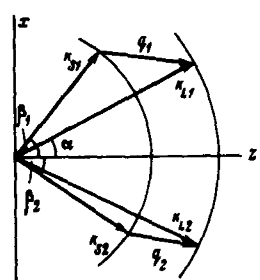


FIG. 1. Diagram of wave vectors.

  

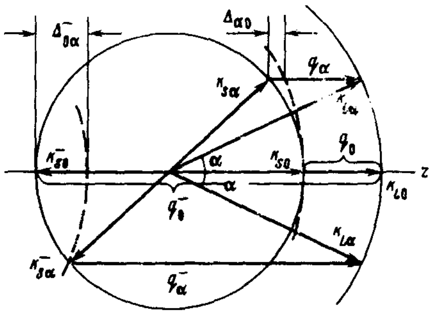


FIG. 2. Illustrating the determination of the mismatch of the phonon-wave wave vectors.

the nonlinear-polarization term responsible for the SRS, and $\mathbf{P}^{sp}(\omega_S, \mathbf{r})$ is the amplitude of the Fourier polarization produced by the spontaneous transitions.

The solution of the homogeneous equation $\hat{L}\mathbf{E}=0$ depends linearly on the boundary conditions, and therefore it suffices to consider the case when one plane wave $\mathbf{E}(\omega_S, \mathbf{r})=\vec{\mathcal{E}}_1^0 \exp(-i\mathbf{k}_{S1}\cdot\mathbf{r})$ is incident on the medium ($|\mathbf{k}_{S1}|=k_S=\mu\omega_S/c$ and $z<0$).

We assume that the angle $\alpha$ is large enough

$$\alpha >> \frac{1}{2}\sqrt{\tilde{\Delta}/k_L}\,, \tag{3}$$

where $\tilde{\Delta}$ is defined below (see (6)), and the angle between $\mathbf{k}_{S1}$ and $\mathbf{k}_{L1}$ is smaller than the angle between $\mathbf{k}_{S1}$ and $\mathbf{k}_{L2}$. The solution of the homogeneous equation can then be sought in the form

$$\mathbf{E}(\omega_S, \mathbf{r}) = \vec{\mathcal{E}}_1(\omega_S, z)e^{-i\mathbf{k}_{S1}\mathbf{r}} + \vec{\mathcal{E}}_2(\omega_S, z)e^{-i\mathbf{k}_{S2}\mathbf{r}}, \tag{4}$$

where $|\mathbf{k}_{S2}|=k_S$ and the wave-vector components perpendicular to the $z$ axis are connected by the relation $\mathbf{q}_{2\perp}=\mathbf{q}_{1\perp}$, where $\mathbf{q}_i=\mathbf{k}_{Li}-\mathbf{k}_{Si}$. We obtain for $\vec{\mathcal{E}}_1$ and $\vec{\mathcal{E}}_2$ a system of two differential equations, the solution of which depends significantly on the quantity $\Delta_{21}=q_{2z}-q_{1z}$.

We take the case $k_{S1y}=0$, $\vec{\mathcal{E}}_1^0=\mathcal{E}_1^0\mathbf{y}_1$ ($\mathbf{y}_1$ is a unit vector along the $y$ axis); then $\vec{\mathcal{E}}_1=\mathcal{E}_1\mathbf{y}_1$ and $\vec{\mathcal{E}}_2=\mathcal{E}_2\mathbf{y}_1$. At $\Delta_{21}\ll\pi/2l$ we can obtain

$$\mathcal{E}_1 = \frac{\mathcal{E}_1^0}{2}\left[e^{\kappa(1+\eta)z}+e^{\kappa(1-\eta)z}\right],$$

$$\mathcal{E}_2 = \frac{\mathcal{E}_1^0}{2}\xi\left[e^{\kappa(1+\eta)z}-e^{\kappa(1-\eta)z}\right], \tag{5}$$

where $\eta=m/(1+m^2)$, $m=|\mathcal{E}_{L2}|/|\mathcal{E}_{L1}|$, $\xi=\mathcal{E}_{L2}/\mathcal{E}_{L1}m$, $\kappa=(1/2)b(\rho)(1+i\rho)I_L/\cos\beta$ is the propagation constant of the Stokes wave without allowance for the parametric interaction, and determines the "incoherent part" of the gain, $I_L$ is the summary intensity of both pump components, $b(\rho)=b/(1+\rho^2)$, $b=b(0)$ is the gain at the central frequency $\omega_S^0$ of the Stokes line following excitation by one plane monochromatic pump wave, per unit intensity of the pump wave, $\rho=2(\omega_S-\omega_S^0)/\Delta\Omega$, and $\Delta\Omega$ is the spontaneous scattering linewidth. We have assumed $\cos\beta\approx\cos\beta_2\approx\cos\beta$.

It can be shown in the general case that if

$$\Delta_{21} < \tilde{\Delta}_{21} = \max\left\{\frac{\pi}{2l}, \tilde{g}\right\}, \tag{6}$$

where $\tilde{g}=\eta g$, $g=2\mathrm{Re}\kappa$, then the contribution of the parametric interaction to the total gain is comparable with its maximum value $\tilde{g}$, which is reached at $\Delta_{21}\ll\pi/2l$. At $\Delta_{21}>\tilde{\Delta}_{21}$, the parametric contribution decreases rapidly with increasing $\Delta_{21}$. At $\Delta_{21}\gg\tilde{\Delta}_{21}$, $\mathcal{E}_1=\mathcal{E}_1^0\exp(\kappa z)$, and $\mathcal{E}_2\approx 0$.

We turn to the inhomogeneous equation (3). The macroscopic polarization $\mathbf{P}^{sp}(\mathbf{r},t)$ can be expressed in the form $\mathbf{P}^{sp}(\mathbf{r},t)=\alpha(\mathbf{r},t)E_L(\mathbf{r})\exp(i\omega_L t)$, where $\mathbf{P}^{sp}$ and $\alpha$ are determined for a fixed averaging volume whose dimensions are small in comparison with $\lambda$. Using the expansion $\alpha(\mathbf{r},t)=\int\int_{-\infty}^{\infty}\alpha(\Omega,\mathbf{q}')\exp[-i(\Omega t-\mathbf{q}'\cdot\mathbf{r})]\,d\Omega\, d\mathbf{q}'$ and representing the Stokes field in the form $\mathbf{E}(\omega_S,\mathbf{r})=\int_{-\infty}^{\infty}\mathbf{E}_{q'}(\omega_S,\mathbf{r})\,d\mathbf{q}'$, we can easily obtain an equation for the field $\mathbf{E}_{q'}$ produced on a definite initial phonon wave. When Eq. (3) is satisfied, the solution takes the form (4) at $\mathbf{q}_{1\perp}=\mathbf{q}_{2\perp}=\mathbf{q}'_\perp$. Expressing the power spectrum of the random function $\alpha(\mathbf{r},t)$ in terms of the spontaneous Raman scattering cross section, we find that for the parametrically-active directions (see (6)) in the $xoz$ plane, at $m=1$, the spectral density of the intensity of the radiation propagating in the solid angle $d\theta$ is equal to

$$dJ(\omega_S, l) = \frac{N_a}{N_a-N_b}\frac{\hbar\omega_S}{2\pi\lambda_S^2}\left[\frac{1}{3}\left(e^{\frac{3}{2}gl}-1\right)+\frac{1}{2}(e^{gl}-1)\right]d\theta, \tag{7}$$

where $N_a$ and $N_b$ are the concentrations of the molecules on the lower and upper vibrational levels.

A similar analysis can be carried out in the case when the pump has a "noise" angle spectrum. We assume the pump radiation to be concentrated in a solid angle $\theta_L$ with aperture $2\alpha$ and to have a uniform brightness $B=I_L/\theta_L$. The data that follow pertain to "forward" scattering. The angular pump spectrum can be regarded as broad if $2\alpha\gg\sqrt{\lambda_L/2l}$. The parametrically active directions are those lying within $\theta_L$. If $B<B_{cr}=q_0/2\pi b(\rho)$, where $q_0=k_L-k_S$, then the parametric contribution to the gain $G(z)=\int_0^z\tilde{g}(z')\,dz'$ at $z>\pi/q_0\alpha^2$ approaches asymptotically the maximum value $\tilde{G}_\infty\sim\pi^2Bb(\rho)/q_0$ ($\pi^2b(\rho)B/q_0\to\pi/2$ as $B\to B_{cr}$). If $z\ll\pi/q_0\alpha^2$ or $B>B_{cr}$, then the parametric and "incoherent" parts of the gain are equal, $\tilde{g}=g$. At $b=3\times10^{-3}$ cm/MW and $(\omega_L-\omega_S)/2\pi c=992$ cm$^{-1}$ ($C_6H_6$) we have $B_{cr}=4.8\times10^5$ MW/cm$^2$ sr. The critical brightness $B_{cr}$ is the analog of the critical spectral pump density.[1]

For backscattering we get $B_{cr}^-\approx(2k_L/q_0)B_{cr}$ and $\tilde{G}_\infty^-\approx(q_0/2k_L)\tilde{G}_\infty$ (here and in Fig. 2, the minus sign denotes backscattering). The decrease in the effectiveness of the parametric interaction in backscattering is due to the increase of the mismatch of the vectors of the phonon waves, as illustrated in Fig. 2.

The obtained regularities explain the "repetition" effect investigated in[2].[1]) They determine also the presence of an appreciable "front-back" scattering asymmetry and indicate that the latter should increase sharply when the critical brightness is exceeded.

1)A similar effect in the angular distribution of the backward SMBS was discussed earlier in[3].


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