"Anomalies" of stimulated Raman scattering

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It is possible to explain in a consistent way the asymmetry of the scattering indicatrix and also structural features on the energy and spectral characteristics of stimulated Raman scattering which have been observed experimentally but not explained theoretically.

Experiments on stimulated Raman scattering have yielded several results which do not conform to the existing theory[1–3] for this process. These features have been termed "anomalies" of stimulated Raman scattering. Foremost among them is the asymmetry of the scattering index; specifically, the intensity of the forward scattering is considerably higher than that of the backscattering.[1] Second, the backscattering may reach saturation at an intensity of the Stokes component two or three orders of magnitude below the pump intensity,[4] and this saturation is generally unrelated to a depletion of the pump or to the excitation of higher-order components of the stimulated Raman scattering. Third, the spectrum of the stimulated Raman scattering is considerably more complicated than predicted by the theory. Specifically, the spectrum has a quasiregular fine structure, and the total width of the spectrum in the saturation region increases with increasing pump energy.[5]

In this paper we propose a physical mechanism which leads to a consistent explanation of all of these features. This mechanism is the appearance of self-induced frequency shifts of electromagnetic waves in the course of the stimulated Raman scattering and a disruption of the phase-matching conditions over the entire nonlinear-interaction region.

To describe the stimulated Raman scattering of a pump wave E_0, accompanied by the formation of Stokes components $E_{\pm 1}$, which correspond to scattering forward and backward, respectively, we work from the following system of equation:

$$\pm \frac{\partial}{\partial z} E_{\pm 1} - i \frac{\omega_s}{2nc} \delta\epsilon E_{\pm 1} = i \frac{\omega_s}{nc} \pi N \left(\frac{\partial \alpha}{\partial Q}\right) q^*_{\pm 1} E_0 ,$$

$$\left(\frac{\partial}{\partial t} + \frac{1 + i\Delta_{\pm 1}}{T_2}\right) q_{\pm 1} = \frac{i}{4M\Omega} \left(\frac{\partial \alpha}{\partial Q}\right) E_0 E^*_{\pm 1} , \qquad (1)$$

$$\frac{\partial}{\partial t} \delta\epsilon = - \frac{\omega_s}{\omega_0} A \frac{\partial}{\partial z} |E_0|^2 \equiv A \frac{\partial}{\partial z} (|E_1|^2 - |E_{-1}|^2).$$

System (1) uses the standard notation and differs from the system of equations which is ordinarily used to describe stimulated Raman scattering[3] in that the dielectric constant of the medium has a nonlinear increment $\delta\epsilon$. To show that the effects discussed

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above occur at a scattering intensity well below the pump intensity, we ignore pump depletion everywhere except in the equation for $\delta\epsilon$, where this depletion determines the number of stimulated-Raman-scattering photons which appear per unit volume per unit time.

The increment $\delta\epsilon$ arises by two mechanisms. During stimulated Raman scattering, an energy flux $i(\Omega N/8)(\partial\alpha/\partial Q)[E_0 E^*_{-1} q^*_{-1} + E_0 E^*_1 q^*_1 - \text{c.c.}]$ is transferred from the pump wave to the medium. This energy is expended on exciting molecules from the ground state into an excited state. In a medium with a short vibrational-energy relaxation time ($T_1 \ll t_p$, where t_p is the length of the pump pulse), $\delta\epsilon$ is caused by a heating of the medium, so that we have $A = (\partial n/\partial T)cn^2\Omega/4\pi\omega_s\rho c_p$). In a medium with a long relaxation time ($T_1 \gg t_p$), on the other hand, $\delta\epsilon$ stems from the different ($\Delta\alpha$) between the polarizabilities (α) of the molecule in its ground and excited states, so that we have $A = (\Delta\alpha/\alpha)\cdot(n^2-1)cn/4\pi\hbar\omega_s N$. The existence of a time-varying increment in the dielectric constant of the medium, $\delta\epsilon$, causes the waves E_0 and $E_{\pm 1}$ to acquire shifts of their instantaneous frequencies as they propagate through the nonlinear medium. In the case of steady-state scattering, in which the intensities of the waves E_0 and $E_{\pm 1}$ do not depend on the time, this effect corresponds to the following amplitudes for the interacting waves:

$$E_0(z,t) = \mathcal{E}_0(z)\exp\left[i\frac{\omega_0 A}{2nc}\int_0^z dz'\frac{\partial}{\partial z'}(|\mathcal{E}_1(z')|^2 - |\mathcal{E}_{-1}(z')|^2)t\right],$$

$$E_{\pm 1}(z,t) = \mathcal{E}_{\pm 1}(z)\exp\left[i\frac{\omega_s A}{2nc}\int_{l,0}^{z} dz'\frac{\partial}{\partial z'}(|\mathcal{E}_1(z')|^2 - |\mathcal{E}_{-1}(z')|^2)t\right], \tag{2}$$

where the amplitude $\mathcal{E}_{0,\pm 1}$ depend on only the coordinate; the integration in the expression for $E_{\pm 1}$ is carried out over the interval (l, z) and that in the expression for E_{-1} over the interval $(0, z)$; and $z = 0$ and $z = l$ are the boundaries of the nonlinear medium. The pumping is carried out from the $z = 0$ side.

It follows from (2) that the nonlinear increments in the frequencies of the electromagnetic waves, which occur in the course of the stimulated Raman scattering, depend explicitly on the coordinate. As a result, there is a frequency deviation from the center of the gain line, and this deviation varies in space. This deviation cannot be canceled by any choice of $\Delta_{\pm 1}$, so that the scattering efficiency must fall off. To prove this assertion, we analyze the solution of system (1). We first consider stimulated Raman scattering accompanied by the excitation of only one of the scattered waves, for definiteness, E_{-1}. From the solution of (1) we find the following relation, which determines the backscattering intensity which is maximized in Δ_{-1}, $I_{-1} = cn|E_{-1}(0)|^2/8\pi$, as a function of the pump intensity, $I_0 = cn|E_0|^2/8\pi$:

$$\ln I_{-1}(0)/I_n + \frac{1}{2}h_b^2 I_{-1}^2(0) = gI_0 l. \tag{3}$$

Here I_n is the intensity of the noisy nucleation; $g = (4\pi^2 N\omega_s T_2/c^2 M\Omega n^2)(\partial\alpha/\partial Q)^2$; and $h_b = (4\pi A\omega_s T_2/c^2 n^2)(1+\omega_0/\omega_s)$. The frequency shift of the scattered radiation which is emitted is

$$\Delta_{-1} = -h_b I_{-1}(0). \tag{4}$$

It follows from (3) that the exponential intensification of the scattered radiation which occurs when a scattering level $I_{-1}(0) \sim h_b^{-1}(gI_0 l)^{1/2}$ is reached gives way to a slow increase, proportional to a root. For forward scattering we find an equation analogous to (3), but the saturation is determined by the parameter $h_f = h_b(1-\omega_0/\omega_s)(1+\omega_0/\omega_s)^{-1} \approx h_b/20$. The saturation of the backscattering thus occurs at a much lower level than that of the forward scattering, and it is generally unrelated to pump depletion. The saturation of the forward scattering may not result from the mechanism discussed here, being determined instead by pump depletion or by the parametric excitation of higher-order components of the stimulated Raman scattering.

A common solution of system (1) for the waves E_{+1} and E_{-1} under the condition that there is no saturation of the forward scattering leads to a saturation of the backscattering which is even more restrictive than that in (3):

$$I_{-1}(0) = I_n \exp(gI_0 l)[1 + h_b^2 I_n^2 \exp(2gI_0 l)]^{-1/2}. \tag{5}$$

The mechanism under discussion here thus makes it possible to explain the asymmetry of the scattering indicatrix (the forward scattering is more intense) and the saturation of the backscattering at a level $I_{-1}(0) \approx h_b^{-1}$. For liquid nitrogen, for example, with $T_1 \approx 2$ s, $\Delta\alpha/\alpha = 1.5\times 10^{-2}$, and $T_2 = 75$ ps (Ref. 3), we would have

$$h_b = \frac{\Delta\alpha}{\alpha}\frac{n^2-1}{n}\frac{T_2}{\hbar Nc}\left(1+\frac{\omega_s}{\omega_0}\right) \approx 0.5\times 10^{-4}\ \text{cm}^2/\text{MW}$$

and thus $I_{-1}^{\max}(0) = 20$ GW/cm^2. This result agrees well with the observed saturation level of stimulated Raman backscattering in nitrogen.[4,6] For media with a short T_1, and under the assumption that a thermal expansion can occur over the scale length of the variation of the field of the scattered wave in the direction across the beam, $l_\perp$ ($l_\perp/v_s < t_p$, where v_s is the sound velocity), we would have a saturated parameter $h_b = [(\partial n/\partial T)_p/\rho c_p]\Omega T_2(1+\omega_0/\omega_s)/c$. As a result, for nitrobenzene, for example, with $(\partial n/\partial T)_p/\rho c_p \approx 10^{-10}$ cm · s^2/g and $\Omega T_2 \approx 2\times 10^2$, we find a saturation intensity ~ 80 GW/cm^2, again in good agreement with the experimental results of Refs. 4 and 6.

Okladnikov *et al.*[4] offered a theory which explained the "anomalous" saturation of stimulated Raman scattering in terms of an equalization of the populations of the ground and vibrational excited states of a molecule as a result of the stimulated Raman scattering. That theory cannot be regarded as satisfactory, for two reasons: First, it applies only to media with a long T_1 (liquid nitrogen), while Okladnikov *et al.*[4] themselves observed the same saturation effect in nitrobenzene, where the condition $T_1 \ll t_p$ holds, and where the change in the population is negligible. Second, even for liquid nitrogen, where the relaxation time of the vibrationally excited state is a matter of seconds, Okladnikov *et al.*[4] assert that their theory does not reach quantitative agreement with experiment and that further assumptions are required regarding a limitation of the number of molecules which are involved in the stimulated Raman scattering. We wish to emphasize that in our case the saturation of the stimulated Raman scattering arises in the approximation of a low population of the vibrationally excited state of the molecules.

Finally, our mechanism explains the quasiregular line structure in the stimulated-Raman-scattering spectrum.[5] These structures arise because the pump intenstiy $I_0(t)$ in an experiment varies in time. In this case, the intensity of the scattered radiation, and also, according to (4), the frequency shift of the radiation will vary in time. The results of a frequency sweep of this sort are line structures in the time-integrated spectrum of the radiation, as was shown in Ref. 7. From (4) we also find an obvious explanation for the broadening of the time-integrated stimulated-Raman-scattering spectrum in the saturation region.

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