SELF-ACTION OF LIGHT ARISING FROM STIMULATED RAMAN SCATTERING

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Received 24 August 1970

A discussion is given for the dynamics of the self-focusing due to stimulated Raman scattering and the self-bending of the Stokes component beam in the case of an asymmetrical distribution of the field in the pumping beam.

1. In the many experimental investigations concerned with the study of the self-focusing of light, it is noted that it is accompanied by stimulated Raman scattering (SRS) [1, 2]. As we have pointed out [3], SRS can give rise to a nonlinear addition to the polarizability of the substance, which is due to a redistribution of the populations of the levels of the transition on which the SRS takes place and to the Stark shift of these levels. The magnitude of this contribution can be comparable with, or even exceed, the magnitude of the nonlinear polarizability of the substance due to the Kerr effect. It can therefore be expected that SRS can produce self-action of the light. The conditions for observing SRS self-focusing have been discussed in a note [4]. Here we shall discuss the dynamics of the self-focusing due to SRS and also the self-bending of the beam of the Stokes component when there is an asymmetrical distribution of the field in the pumping beam. As will be shown, the self-action of SRS is practically always weak, i.e., for example, when self-focusing is present the layer in which a nonlinear susceptibility \( \chi_{nl} \) of significant magnitude is formed can be considered as a thin lens.

2. In [3] a general expression was obtained for the polarizability of the substance in the presence of SRS as a function of pumping field \( E_p \) and the field \( E_S \) of the Stokes component and of the frequency detuning, \( \delta = \omega_p - \omega_S - \omega_\text{r} \), where \( \omega_p \) and \( \omega_S \) are the moduli of the complex amplitudes of the fields, \( \delta \), and \( \omega_\text{r} \) is the frequency of the transition on which the SRS takes place. In the approximation of relatively weak fields, when the population is far from saturation and if the Stark effect is neglected and the detuning is small, i.e., \( \delta \ll \tau \), where \( \tau \) is the line width of the 2–1 transition *, the nonlinear contribution to the dielectric susceptibility can be written in the form (cf. Eq. (18b) in [3])

\[
\chi_{nl} = \chi_{nl}^\text{p} \chi_{nl}^\text{S},
\]

where \( \chi_{nl}^\text{p} \) and \( \chi_{nl}^\text{S} \) are the dimensionless intensities of the pumping and Stokes components, normalized on the saturation intensity,

\[
\chi_{nl}^\text{p} = \frac{\chi_{nl}^\text{p}}{4\pi}, \quad \chi_{nl}^\text{S} = \frac{\chi_{nl}^\text{S}}{4\pi}.
\]

Here \( \tau \) is the lifetime of the molecule in the excited state 2, \( \gamma_{21} \) is the matrix element characterizing the cross section of the SRS for the transition in question [5]. The quantity \( \epsilon_{nl} \) has the form

\[
\epsilon_{nl} = \frac{\gamma_{21}}{\tau} (1 - \epsilon_{nl}),
\]

where \( \epsilon_{nl} \) and \( \epsilon_{nl} \) are the linear polarizabilities of the states 2 and 1, and \( N \) is the number density of the particles.

The difference of the linear polarizabilities for SRS involving a vibrational transition can be estimated quite simply as [4]

\[
\omega_{21} = \frac{\omega_0^2}{\omega_p} - \omega_0 \frac{\omega_0}{\omega_p} \frac{\omega_0}{\omega_p}.
\]

Here \( \omega_{21} \) is the frequency of the vibrational transition, and \( \omega_0 \) is the frequency of the nearest electronic transition.

3. It is known [5] that the spatial development of SRS with the threshold taken into account can be described sufficiently accurately by the equations,

\[
\frac{d^2m_p}{dz^2} = - \frac{1}{L_0} m_p \gamma_{12}, \quad \frac{d^2m_0}{dz^2} = - \frac{1}{L_0} m_0 \gamma_{21} m_p \omega_p - \omega_0 \frac{m_p}{L_0},
\]

where \( L_0 \) is the attenuation length of the Stokes component, and \( L_0 \) is the characteristic length of the SRS that is inversely proportional to the coefficient of transformation of the pumping component into the Stokes component,

\[
L_0 = \frac{\gamma_{12}}{k_p n_p \gamma_{21}},
\]

where \( k_p = \omega_p / c \) is the wavenumber.

It is seen from Eqs. (5) that the length in which the pumping energy is transformed into the Stokes component for \( m_p \) (initial) \( \ll m_p \) (initial) is
where $I_{\text{SRS}}$ is the threshold pumping intensity for SBS,

$$I_{\text{SRS}} = \frac{L_0}{\mu^2} \left( \frac{1}{L_0} \right)^{\gamma_{\text{SRS}}} \frac{1}{\gamma_{\text{SRS}}}.$$  

The nonlinear layer can be considered here as a phase corrector (thin lens or prism) formed by the pumping waves with an axis yet undistorted geometry. This allows us to use in the calculation of the lens the magnitudes of the fields in the emerging plane wave. If the lens is described by its optical thickness $\Delta l$ at each point of the transverse cross section $p$ of the wave,

$$\Delta l = \frac{1}{2} \frac{\epsilon_{0}^2}{\epsilon_{0}^2 + \epsilon_{0}} \frac{1}{(m_{\Delta} - m_{0})},$$

where $m_{\Delta}$ and $m_{0}$ are the entrance and exit pumping intensities, substituting the values of $\epsilon_{4}$ (3), $L_0$ (6), and $m$ (2) we find

$$\Delta l = \frac{1}{2} \frac{\epsilon_{0}^2}{\epsilon_{0}^2 + \epsilon_{0}} \frac{1}{(m_{\Delta} - m_{0})}.$$  

where $\epsilon_{0} = \hbar / [2 \tau (x - X_1)]$ is the characteristic field of the resonance cross polarization [cf. (3)].

In the absence of absorption ($L_{\text{att}} = \infty$, $m_{\text{SRS}} = 0$), all the pump photons are transformed in Stokes photons, i.e., $m = 0$; in this case we have

$$\Delta l = \text{constant}.$$  

It is interesting that, although the initial nonlinearity [Eq. (1)] is proportional to $E_{2}^2 E_{2}$, i.e., to the fourth power of the field, the optical thickness of the nonlinear layer [Eq. (12)] is here proportional only to the pumping intensity; this explains the integral nature of the effect [cf. Eq. (9)]. This also explains the fact that in the absence of a SRS threshold ($L_{\text{att}} = \infty$), $\Delta l$ depends neither on the concentration $N$ nor on the value of $\tau$ that characterizes the nonlinearity.

Actually, an increase in $N$ and $\tau$ leads to an increase in the nonlinear addition to the polarisability $\chi_{n}^2 \propto N \tau^2$, cf. Eqs. (2) - (3), but at the same time it leads to a decrease in the thickness of the interaction layer $L_{\text{SRS}} \propto 1/N \tau^2$, cf. Eqs. (6) and (7); in the integration these effects compensate each other ($\Delta l \propto \chi_{n}^2 L_{\text{SRS}}$). The latter is justified only as long as the geometry of the wave can be assumed to be unchanged, i.e., if $L_{\text{SRS}}$ is smaller than the diffraction length of the beam.

As will be shown below, this condition is automatically fulfilled if the threshold for self-focusing of the SRS is only slightly exceeded. The calculation of the SRS threshold in Eqs. (5) for the condition $m_{\Delta} \ll m$ (initial), which is usually fulfilled (the Stokes component grows out of the spontaneous emission at the entrance), leads to the following equation for the quantity $m - m_{\infty} = \nu$, which enters into Eq. (10) for $\Delta l$:

$$m \rightarrow \nu \left[ 1 - \exp \left( -\gamma_{\text{SRS}} \right) \right].$$

A graph of $\nu(m_{\Delta})$ can be constructed as a function of $m_{\Delta}$ [cf. the figure]: it is easy to see that $\nu(m_{\Delta})$ can be approximated quite accurately by the functions

$$\nu = \begin{cases} 0 & \text{for } m_{\Delta} \leq m_{\text{SRS}}, \\ 2 \left( m_{\Delta} - m_{\text{SRS}} \right) & \text{for } m_{\text{SRS}} \leq m_{\Delta} \leq 2 m_{\text{SRS}}, \\ m_{\Delta} & \text{for } m_{\Delta} > 2 m_{\text{SRS}}. \end{cases}$$

The dashed line in the figure shows the variation of $\nu(m_{\Delta})$ corresponding to Eq. (14). Thus, Eqs. (11), (12), and (14) determine $\Delta l$ when the SRS threshold is neglected as well as it is taken into account.

We shall discuss now SRS self-focusing when the SRS threshold is zero. This condition can be neglected, for example, if $m_{\Delta} \gg m_{\text{SRS}}$. As we shall see, for this case the properties of SRS self-focusing can be most clearly discussed. It is not difficult to see from Eq. (12) that, just as in the case of ordinary self-focusing, a quadratic distribution of the pumping intensity over the cross section of the beam should correspond to an aberrationless lens,

$$m = \frac{m_{\text{SRS}}}{2} \left( 1 - \frac{1}{N \tau^2} \right),$$

where $m_{\text{SRS}}$ is the pumping intensity in the center of the beam. Here the focal distance of the lens (in the geometrical approximation) is equal to

$$X_{s} = \frac{\left[ 2 L_{0} \left( m - m_{\infty} \right) \right]^{1/2}}{\epsilon_{0}^2},$$

or, after substituting Eqs. (12) and (15),

$$X_{s} = L_{0} \frac{m_{\Delta} - m_{0}}{\epsilon_{0}^2} L_{\text{SRS}}.$$

where $L_{0} = k_{p} a^2$ is the diffraction length of the beam in the medium.

Since the beam propagates behind the SRS lens in a practically linear medium, the whole situation can be considered as focusing by a thin nonlinear layer (external self-focusing). Here, as was shown in [6], the threshold power corresponds to $X_{\phi} L_{0}$; substituting Eq. (17) in this, we obtain
5. We shall calculate now the threshold for SRS self-focusing taking into account the threshold for the stimulated Raman scattering itself. If the pumping beam on entering is distributed quadratically [Eq. (16)], from Eqs. (10), (11), and (14) and taking account of Eqs. (16) and (17) we obtain the following result. If the pumping intensity in the center is greater than the SRS threshold, but not by more than a factor of \(2 (m_{SRS} < m_{th} < 2m_{SRS})\), a single nonlinear lens of radius \(a_1\) is formed where

\[
e^2_1 - a^2 = 4a^4 \left(1 - n_{SRS} / n_{th} \right)
\]

and the new focal length is \(X_1 = 0.5 X_0 / \left[ n_{SRS} \right] \) from Eq. (17).

However, when the pumping intensity is greater than twice the SRS intensity \(n_{th} > 2m_{SRS} \), in the center of this lens there appears a second lens with radius \(a_2\),

\[
e^2_2 - a^2 = 4a^4 \left(1 - 2m_{SRS} / m_{th} \right)
\]

and focal length \(X_2 \) [Eq. (17)]. As the pumping intensity is further increased the area of the first (ring) lens decreases, and the area of the second increases, tending toward the cross-sectional area of the pumping beam, the power therefore goes over from the first near focus \(X_1 \) to the more distant focus \(X_2 \). In those cases in which all, or most of, the radiation goes through one lens, one can introduce the idea of a threshold for SRS self-focusing. If only a lens with radius \(a_1\) [cf. Eq. (19)] and focal length \(X_0 \) is formed, we obtain from the relation \(k_1 a^2 = X_0 \)

\[
e^2_1 + 0.5 a^2 = X_0
\]

(21)

This case is realized if

\[
e^2_1 + 0.5 a^2 < 2 \frac{m_{SRS}}{m_{th}}
\]

(22)

which is fulfilled for \(e^2 < 2 \frac{m_{SRS}}{X_0} \).

However, if we have

\[
e^2_1 + 0.5 a^2 > 2 \frac{m_{SRS}}{X_0}
\]

(23)

the area of the ring lens near the threshold intensity will be negligibly small, and the whole nonlinear layer can be represented as a lens of radius \(a_2 \) [cf. Eq. (20)]; in this case, from the condition \(k_2 a^2 = X_0 \) we obtain

\[
e^2_2 > a^2 + 2 \frac{m_{SRS}}{X_0}
\]

(24)

The inequality (23) is fulfilled when \(X_1 \gg 2 \frac{m_{SRS}}{X_0} \). And, as might be expected, the value of \(e^2 \) (thres) [Eq. (24)] is practically the same as in Eq. (23), which was computed with the SRS threshold neglected. In the intermediate case, \(2e^2 \) (thres) \(= e^2 \), it is not possible to define a threshold in a meaningful way.

6. We shall make an estimate of the SRS self-focusing threshold intensities. For benzene the threshold power for SRS is \(m_{SRS} \approx 18 \) MW cm\(^{-2}\), and the value of \(e^2 \) is \(l_{th} \approx 1 \) GW cm\(^{-2}\); therefore, the SRS self-focusing threshold according to (23) and Eq. (24) is \(l_{th} \approx 1 \) GW cm\(^{-2}\). For hydrogen at a pressure of 30 atm \(m_{SRS} \approx 12 \) MW cm\(^{-2}\) and \(e^2 \) corresponds to a flux of \(l_{th} \approx 75 \) kW cm\(^{-2}\); i.e., the threshold is given by Eq. (21), and self-focusing should begin practically at the same time as the SRS threshold is exceeded; it is necessary of course to assume that the pumping pulse in this case is long enough, since the lifetime of the hydrogen molecule in the excited vibrational state at 30 atm is \(t \approx 3 \times 10^{-3} \) sec. We note that at the present time with lasers operating in a quasi-continuous regime, it is possible to obtain flux densities of light that are high enough to exceed the SRS threshold in a gas; it should therefore be possible to observe the effect that we have discussed.

It is interesting that in the case of compressed hydrogen, if the pumping intensity exceeds the SRS threshold by a factor of \(2 (m_{th} = 2m_{SRS}) \), the focal distance \(X_1 \approx 0.5 X_0 \) is quite small \((X_0 = L_0 / \lambda_0) \), or for \(k_0 = \frac{10^8}{cm} \), \(a \approx 0.3 \) cm and \(X_0 = 10 \) cm). For \(m_{th} / m_{SRS} = 10 \), the value of \(X_0 \) is \(\approx 6 \) cm. It is evident from this that in calculations of SRS for multiple cascade transformations, it is necessary even under ordinary conditions to take the effect of SRS self-focusing into account.

7. We shall now clarify the conditions for the validity of the thin lens approximation. We shall require that the thickness of the lens \(\approx \frac{1}{m_{SRS}} \) [Eq. (7)] be less than the focal distance \(X_0 \) [Eq. (17)]; from this it follows that the conditions on the initial radius \(a \) of the beam are

\[
e^2 > a^2 + \frac{l_{th}}{k_0} = \frac{l_{th}}{k_0 X_0} (a_1 - a) = \frac{m_{SRS}}{l_{th} X_0}
\]

(25)

and the condition on the initial intensity \(m_{th} \) in the center of the pumping beam is

\[
m_{th} > a^2 + \frac{m_{SRS}}{1 - a / a_1}
\]

(26)

If any one of these conditions is violated, the thin lens approximation is inappropriate. However, a calculation gives, for example, for benzene \(a \approx 10^{-4} \) cm, and for compressed hydrogen \(a \approx 10^{-3} \) cm, so that Eq. (25) can be assumed in practice to be fulfilled with a wide margin. Also, Eq. (26) can be assumed to be always fulfilled except in the narrow region for which \(m_{th} \approx m \approx M \) [if \(m_{th} \approx M \)]: even if the power is only a little greater than the values of \(m \) (thres) and \(M \), the SRS lens is a thin one.

If the initial pumping intensity greatly exceeds the SRS threshold, one can expect the appearance of a succession of equidistant SRS lenses formed by successive transformations of one Stokes component into others of higher orders. This lens system is similar to a quasi-optical waveguide capable of canalizing narrow-light beams.

8. We shall now discuss the self-refraction of light when SRS is present; this is analogous to the self-bending of asymmetric beams of light in ordinary nonlinear media [7, 8]. In the present case the effect results in the following. If the pumping intensity at the entrance is distributed over the cross section not quadratically but linearly,

\[
m_{th} = \begin{cases} 0.5m_{th} & \text{if } |y| < a \\ \frac{1}{4} m_{th} & \text{if } |y| > a \end{cases}
\]

(27)

(the intensity of the beam is assumed to be changing only in one direction, i.e., along \(y \) in the transverse cross section), in place of an SRS lens there is formed an SRS prism. As a consequence there should be observed, not a focusing, but a deviation of the beam of the Stokes component by the angle \(\phi \); with respect to the initial beam,

\[
\phi = \frac{\Delta l}{\lambda a}
\]

(28)

where \(\Delta l \) is determined as before from Eqs. (10), (11), and (14). Substituting Eq. (27) in Eq. (10), we obtain that for \(m_{SRS} = 0 \) the angle of SRS refraction is equal to
where $\varphi_d = (ka)^{-1}$ is the half-value of the diffraction divergence of the initial beam. A calculation of the SRS threshold (similar to that carried out for the SRS self-focusing) shows that, for $m_{\text{SRS}} < m_{\text{th}} < 2m_{\text{SRS}}$, the part of the beam in which the SRS threshold is exceeded ($m_{\text{th}} > m_{\text{SRS}}$) is deviated by the angle $\varphi = \varphi_d$, and the subthreshold part ($m_{\text{th}} < m_{\text{SRS}}$) does not change its direction. For $m_{\text{th}} > 2m_{\text{SRS}}$, two deviated beams are formed from the Stokes component, one ($m_{\text{th}} > 2m_{\text{SRS}}$) by the angle $\varphi$, the other ($m_{\text{th}} < m_{\text{SRS}}$) by the angle $\varphi_1$. Finally, the subthreshold part of the beam is propagated as before without any change. As the initial pumping intensity is increased the whole power transformation is concentrated in the first beam.

If one wishes to speak of a threshold for the SRS refraction with the threshold defined as the point at which the angle of refraction $\varphi$ is greater than the order of magnitude of the angle of diffraction (say, $\approx (1/4 \varphi_d)$), one easily deduces that the threshold pumping intensity for SRS refraction is the same as the threshold for SRS self-focusing. [Eqs. (18) and (21)].

We are grateful to G. A. Askaryan for useful discussion and assessment of the results of this investigation.

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UDC 535.37-548.0

STRUCTURE OF THE PHONON WING IN SPECTRA OF THE $O_2$ AND $S_2$ MOLECULAR IONS IN A KI CRYSTAL

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Received 2 February 1971

The structure of the phonon wing in the spectra of molecular ions in KI crystals was investigated. It is shown that the intensity distribution in the phonon wing reflects the characteristics of the phonon density functions of the undistorted crystal.

The spectra of the molecular ions $O_2$ and $S_2$ have a rich vibrational structure [1-3]. Part of the vibrational structure, dependent on the interaction of the electronic transition with localized (intramolecular) vibrations, is subject to complete interpretation: the energy of the purely electronic transition, the frequencies and anharmonic constants of the localized vibrations, and the magnitudes of the Stokes losses at the localized vibrations have been determined [2, 4, 6].

Information on the interaction of the electronic transition in the impurity center with vibrations of the crystal lattice is contained in the intensity and form of the zero phonon lines and in the structure of the wing which accompanies each zero phonon line in the spectrum. The width and form of the zero phonon lines for $O_2$ in KBr was investigated. It is shown that mixing with the normal coordinates of the crystalline vibrations during an electronic transition provides a significant contribution to the line width. A reduction of the Stokes losses also was observed in crystalline vibrations according to the degree of excitation of the local vibration in $O_2$ [7].

The phonon wing corresponds to excitation of crystalline vibrations during an electronic transition and contains information both about the phonon spectrum of the pure crystal and about the characteristics of the electron-phonon interaction. The degree of the interaction with the crystalline vibrations characterizes the magnitude of the total Stokes losses to the crystalline vibrations [8]. In Table 1 are given data of measurements of the magnitude of the total Stokes losses for the spectra of the $O_2^-$ and $S_2^-$ centers. The $O_2^-$ center has the weakest interaction with phonons in the CsCl and KI lattices.

Next in order of increasing interaction are NaCl, KBr, KCl, RbBr, and RbCl [9]. For the $S_2$ molecular ion, isoelectronic with $O_2$, the Stokes losses to phonons in these same lattices are significantly larger. The position and number of the zero phonon lines for which Stokes losses were determined and the half-widths of some zero phonon lines are also indicated in the table.

The detailed interpretation of the form and structure of the phonon wing is a rather complex problem. The most easily understood interpretation is in the case where one is limited to taking into account only the change of the equilibrium positions of the nuclei during the electronic transition. In this case, the determination of the intensity in the single phonon part of the wing is given by the Stokes loss function [8] $p(\nu) = \alpha^2 (\nu) \varphi(\nu)$, where $\varphi(\nu)$ is the phonon density function of the undistorted crystal and $a(\nu)$ is a function of the displacements of the equilibrium normal coordinates. In the given case, the function $a(\nu)$ reflects all the features of the interaction of the center with phonons.

In Fig. 1, the phonon wing in the luminescence spectrum of $O_2$ in KI (minimum Stokes losses) is compared with phonon density function $p(\nu)$ of the pure KI crystal (according to the data of [5]). The intensity in the wing 0.13 from the maximum [sic] corresponds to the limiting frequency of the KI phonon spectrum, equal to 142 cm$^{-1}$, i.e., in the case of KI-O$_2$ the wing was formed fundamentally from single phonon transitions. It is evident from Fig. 1 that the distribution of intensity in the wing is reflected rather well by the features of the function $p(\nu)$ of the undistorted crystal, and this demonstrates the possibility...