

# Stimulated thermal scattering near a resonance

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The spectrum of stimulated scattering from density fluctuations in gases near the absorption resonance has been analyzed. This type of scattering is shown to have a low threshold  $\sim 10$  W/cm<sup>2</sup>.

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1. Stimulated thermal scattering due to absorption (SSA) was predicted theoretically<sup>1</sup> and observed experimentally for the first time in Refs. 2 and 3. The mechanism of SSA can be explained in the following way. Suppose that two monochromatic waves—a strong wave  $E_l e^{i\mathbf{k}_l \mathbf{r} - i\omega_l t}$  and a weak wave  $E_s e^{i\mathbf{k}_s \mathbf{r} - i\omega_s t}$  in a test field—propagate in an absorbing medium. The absorption of light in the medium causes a release of heat  $Q(\mathbf{r}, t)$ ; this expression contains an interference term  $Q(\mathbf{r}, t) \propto E_l^* E_s e^{-i\mathbf{q}\mathbf{r} + i\Omega t}$ ,  $\mathbf{q} = \mathbf{k}_l - \mathbf{k}_s$ ,  $\Omega = \omega_l - \omega_s$ . This leads to a thermal modulation and a buildup of acoustic waves, which, in turn, causes a scattering. In Refs. 1, 4, and 5 the onset of SSA has been attributed to modulation of the real part of the dielectric constant  $\epsilon$ . This is valid in those cases in which the relatively weak, broad band is responsible for the absorption. In this letter we show that the modulation of the imaginary part of  $\epsilon$  contributes substantially to the scattering in gases near the narrow absorption resonance. The spectrum shape in this case changes and the stimulated-scattering threshold decreases, so that this effect can be observed when the power density is  $c/8\pi|E_l|^2 \sim 10$  WT/cm<sup>2</sup>.

2. A change in  $\epsilon$  gives nonlinear corrections to the linear absorption (amplification) coefficient  $\alpha_L$  of a weak wave  $\alpha = (\omega/c)e''(\omega_s)$  which can be represented in the form

$$\alpha(\omega_s) = \alpha_L(1 + g), \quad g = g_1 + g_2. \quad (1)$$

The quantities  $g_1$  and  $g_2$  are associated with thermal scattering and with scattering by acoustic waves, respectively.

First, let us consider the thermal scattering. The modulation  $\epsilon$  in this case is caused by the temperature variation  $\delta\epsilon = (\partial\epsilon/\partial T)_p \delta T$ , and  $\delta T$  obeys the equation (see, for example, Ref. 5)

$$\frac{\partial(\delta T)}{\partial t} - \kappa \Delta(\delta T) = \frac{1}{Nc_p} Q(\mathbf{r}, t), \quad (2)$$

where  $\kappa$  is the thermal conductivity coefficient,  $N$  is the gas density, and  $c_p$  is the specific heat at constant pressure. We have for the absorbed heat

$$Q(r, t) = \frac{\pi \hbar \omega}{8} \left( \frac{d_0}{\hbar} \right)^2 N_0 E_l^* E_s e^{-iqr + i\Omega t} f''(\Delta\omega_0), \quad (3)$$

where  $d_0$  is the dipole moment of the absorbed transition,  $N_0$  is the ground-state population,  $\hbar$  is Planck's constant,  $\Delta\omega_0 = \omega_0 - \omega_l$  is the separation from resonance, and  $f(\Delta\omega_0) = f'(\Delta\omega_0) + if''(\Delta\omega_0)$  is the absorption (dispersion) line shape.

We shall assume that the dielectric constant of gas is

$$\epsilon = 1 + 4\pi^2 \sum_j \frac{d_j^2}{\hbar} N_j f(\Delta\omega_j), \quad \Delta\omega_j = \omega_j - \omega_l, \quad (4)$$

hence, we obtain for  $(\partial\epsilon/\partial T)_p$

$$\left( \frac{\partial\epsilon}{\partial T} \right)_p = - \left( 4\pi^2 / \hbar T \right) \sum_j d_j^2 A_j N_j f(\Delta\omega_j); \quad A_j = 1 - \frac{E_j - \langle E_j \rangle}{kT},$$

where  $\langle E_j \rangle$  is the average internal energy of molecules and  $k$  is the Boltzmann constant. Using these relations and solving (2), we find  $\epsilon''(\omega_s)$  and  $\alpha(\omega_s)$ . We obtain for

$$g_1 = - \frac{\pi \hbar \omega}{2c_p T} \frac{1}{N} \sum_j A_j N_j \left( \frac{d_j E_l}{\hbar} \right)^2 \frac{-\Omega f'(\Delta\omega_j) + \Gamma_1 f''(\Delta\omega_j)}{\Omega^2 + \Gamma_1^2}, \quad (5)$$

where  $\Gamma_1 = \kappa q^2$  is the width of the central part of the Rayleigh component of the scattering spectrum.

The quantity  $g_2$  can be determined in a similar way. Using (4) and following the procedure of Ref. 5, we obtain

$$g_2 = \frac{\pi \hbar \omega}{2c_p T} \frac{1}{N} \sum_j B_j N_j \left( \frac{d_j E_l}{\hbar} \right)^2 4\Omega_{MB}^2 \frac{(\Omega_{MB} - \Omega) f'(\Delta\omega_j) + \Gamma_2 f''(\Delta\omega_j)}{(\Omega^2 - \Omega_{MB}^2)^2 + \Gamma_2^2 \Omega_{MB}^2}, \quad (6)$$

where  $B_j = A_j - c_p/c_v E_j - \langle E_j \rangle/kT$ ,  $\Omega_{MB} = qv_a$  is the frequency, and  $\Gamma_2$  is the sound-attenuation velocity.

3. Only the resonance transition  $\alpha_L = 4\pi^2 \omega / \hbar c d_0^2 N_0 f''(\Delta\omega_0)$  affects the linear absorption in Eq. (1), whereas the nonlinear components are determined by the sum over all the transitions of the molecule. It was assumed in Refs. 1, 4, and 5 that the nonresonance terms are the main contributors to this sum. In this case we can set  $f'(\Delta\omega_j) \approx 1/\pi\Delta\omega$  and  $f''(\Delta\omega) = 0$ , where  $\Delta\omega$  is the characteristic separation of the  $\omega_l$  frequency from the band frequency which is the main contribution to the dispersion (as a rule,  $\Delta\omega < 0$ ). Assuming that  $\sum_j N_j / N d_j^2 = d^2$  and  $A_j \approx B_j \approx 1$ , we obtain from (5) and (6)

$$g = g_1 + g_2 = \frac{\hbar \omega}{2c_p T} \frac{1}{\Delta\omega} \left( \frac{dE_l}{\hbar} \right)^2 \left[ \frac{\Omega}{\Omega^2 + \Gamma_1^2} - 4\Omega_{MB}^2 \frac{\Omega - \Omega_{MB}}{(\Omega^2 - \Omega_{MB}^2)^2 + \Gamma_2^2 \Omega_{MB}^2} \right], \quad (7)$$

consistent with Refs. 4 and 5, if the specific dielectric constant (4) is taken into account. The quantity  $g$  in Eq. (7) is plotted as a function of  $\Omega$  in Fig. 1a. This dependence changes radically if the resonance term  $j=0$  is the main contribution to the sums (5) and (6). For a precise resonance  $\Delta\omega_0 = 0$ ,  $f'(\Delta\omega_0) = 0$ , and  $f''(\Delta\omega_0) = 1/\pi\Delta\omega_L$ , where  $\Delta\omega_L$  is the line width of the absorption transition, and  $g$  has the form

$$g_p = \epsilon_{1p} + \epsilon_{2p} = \frac{\hbar\omega}{2c_p T} \frac{N_0}{N} \frac{1}{\Delta\omega_L} \left( \frac{d_0 E_l}{\hbar} \right)^2 \left[ -A_0 \frac{\Gamma_1}{\Omega^2 + \Gamma_1^2} + B_0 \frac{4\Omega_{MB}^2}{(\Omega^2 - \Omega_{MB}^2)^2 + \Gamma_2^2 \Omega_{MB}^2} \right]. \quad (8)$$

A typical plot of  $g_p$  vs  $\Omega$  is shown in Fig. 1b. A dip appears in the frequency region  $|\Omega| \leq \Gamma_1$  and the region  $|\Omega \pm \Omega_{MB}| \leq \Gamma_2$  has a peak in the dependence of the absorption of a weak wave  $\alpha(\omega_s)$  on the frequency  $\omega_s = \omega_l - \Omega$  (see Fig. 2). If the central dip is sufficiently strong, then the absorption will change to amplification. Notice that the amplification peak occurs at  $\Omega = 0$ , i.e., the stimulated scattering in this case is not accompanied by a frequency drift. The  $\Omega$  dependence of  $g_p$  can be changed by tuning to the line wing, and  $f'(\Delta\omega_0) \gg f''(\Delta\omega_0)$  is in qualitative agreement with Eq. (7) in the remote wing.

4. The ratio of the contributions of resonance and nonresonance terms to the sums (5) and (6) in order to magnitude is  $g_p/g \approx N_0/N \Delta\omega/\Delta\omega_L d_0^2/d^2 \approx N_0/N \Delta\omega/\Delta\omega_L \times A_0/A(\omega/\omega_0)^3$ , where  $A_0$  and  $A$  are probabilities of the absorption transition and of the optically resolved transition, and  $\omega_0$  and  $\omega$  are their frequencies. Bearing in mind that the vibrational-rotational molecular transitions are responsible for absorption, we shall assume that  $A_0/A \approx 10^{-7}$ ,  $\Delta\omega/\Delta\omega_L \approx 10^7$ , and  $\omega/\omega_0 \approx 10^3$ . Finally, we can see that the ratio  $g_p/g \approx N_0/N \times 10^3$  may be appreciably larger than

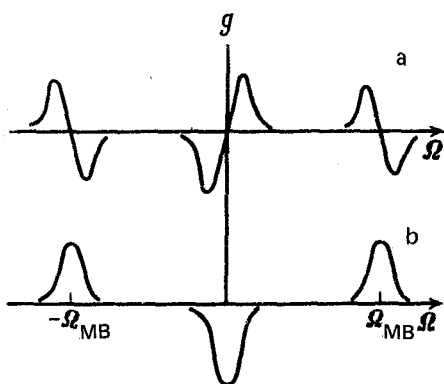


FIG. 1. Qualitative dependence of  $g$  on the frequency separation  $\Omega$ ; (a) the main contribution to  $g$  comes from the nonresonance terms [Eq. (7)]; (b) the main contribution to  $g$  comes from the absorption transition [Eq. (8)].

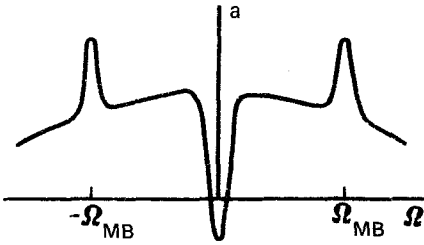


FIG. 2. Qualitative dependence of the absorption coefficient of a weak wave  $\alpha(\omega_s)$  on the separation frequency  $\Omega$  provided that the main contribution to  $g$  comes from the absorption transition.

unity.

Let us estimate the power density  $P_D$  for which the nonlinear terms in Eq. (8) are close to unity. We find  $\hbar\omega_0/c_p T N_0/N(d_0 E_1/\hbar)^2 1/\Delta\omega_\pi \Gamma = 1$  from the condition  $P_{II} = \pi/8 c_p T/\hbar\omega_0 \hbar\omega_0 N T/\alpha$ .  $\Gamma \sim 1/N$  for the specified scattering angle. The line width is determined by the Doppler effect  $\Delta\omega_L = \Delta\omega_D$  in the region of small pressures, and the absorption coefficient  $\alpha \sim N$ .  $P_D$  ultimately decreases with increasing density  $P_D \sim 1/N$  and reaches a minimum in the pressure region in which the impact width of the  $\gamma$  line is larger than the Doppler width. Assuming that  $c_p T/\hbar\omega_0 = 0.1$ ,  $N = 3 \times 10^{17} \text{ cm}^{-3}$ ,  $\Gamma = 10^4 \text{ cm}^{-1}$ ,  $\hbar\omega_0 = 6 \times 10^{-20} \text{ J}$ , and  $\alpha = 2 \text{ cm}^{-1}$  (the transition  $\lambda = 3.39 \times 10^5 \text{ nm}$  in methane), we obtain  $P_D = 5 \text{ W/cm}^2$ . We note that the values of  $g \sim 10^{-2}$  can be recorded by using the simultaneous-detection method, i.e., the structure of (8) can be observed when the signal power is of the order of  $5 \times 10^{-2} \text{ W/cm}^2$ .

In deriving Eq. (3) we have assumed that the energy absorbed by the molecule relaxes instantaneously into heat. Equation (3) must be multiplied by the factor  $(1 + i\Omega\tau)^{-1}$  if the time  $\tau$  needed to establish this process is taken into account.<sup>4</sup> Expression (8), therefore, holds for  $\Omega \ll 1/\tau$ . For the opposite case  $\Omega > 1/\tau$ , both the absolute and relative values of  $g_{1p}$  and  $g_{2p}$  and their spectral form vary. This makes it possible to measure, for example, the quenching time of molecular vibrations.

Having a low threshold, the SSA process may also play an important role in the propagation of light in a resonant medium, thereby changing the angular and frequency components of light.

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