STIMULATED CONCENTRATION SCATTERING OF LIGHT

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In this paper we report the first indication of observation of stimulated concentration scattering of light in a gas. The phenomenon was investigated in a helium-xenon gas mixture (Fig. 1).

The stationary theory of stimulated concentration scattering [1] shows that the intensity of the Stokes stimulated concentration scattering is described by the formula

$$I = I_0 \exp\{g_c |E_L|^2 \ell\}, \quad (1)$$

where I_0 is the light intensity of the thermal scattering due to the concentration fluctuations [2], ℓ is the length of the interaction region of the Stokes "concentration" wave with the laser wave E_L, and the gain g_c, for a binary mixture in which the concentration of one of the components is small, equals

$$g_c = B_c |R_1| \frac{\Omega/\Omega_c}{1 + \Omega^2/\Omega_c^2}, \quad B_c = \frac{(\partial\epsilon/\partial c)^2 c\, m_2}{16\pi n^2 \rho k T}.$$

Here $\vec{k}_1$ is the wave vector of the Stokes wave with frequency $\omega_1 = \omega_L - \Omega$; $\Omega_c = D(\vec{k}_L - \vec{k}_1)^2$, D is the diffusion coefficient, and m_2 - the particle mass of the component with the larger concentration c. The remaining notation is standard [2]. In analogy with stimulated thermal scattering (STS) [3], g_c has a maximum at half the line width of the thermal scattering due to the concentration fluctuations. In a mixture of atomic inert gases at a pressure $P \sim 10$ atm and $D \sim 0.1$ cm^2/sec, the settling time is $T = 2\pi/\Omega_c \sim 10^{-9}$ sec.

In addition to the stimulated concentration scattering, stimulated Mandel'shtam-Brillouin scattering (SMBS) and STS are also possible in the gas mixture [3]. Estimates for a helium-xenon mixture with a xenon concentration c = 0.1 at P = 10 atm and T = 300° K give for the gain ratio of the indicated processes $g_c/g_{MB} \sim 0.1$ and $g_c/g_T \sim 7$. STS is a process with higher inertia, since $D/\chi \approx 4$ (χ - temperature-conductivity coefficient).

In fields $|\vec{E}_L| \sim 5 \times 10^7$ V/cm and at $\ell \sim 0.1$ cm, the argument of the exponential in (1) is of the order of unity. It can, however, be much larger, since the development of stimulated concentration scattering can be preceded by excitation of the xenon atoms, the polarizability of which can increase appreciably [4], leading to a growth of $\partial\varepsilon/\partial c$.

To observe the stimulated concentration scattering, we used the method of superregenerative amplification at the laser mode [3]. A ruby-laser pulse of power 150 - 200 MW and duration $\sim 10^{-8}$ sec was focused inside a cell with a helium-xenon mixture (10:1) by a lens of focal length $f_1 = 20$ cm or $f_2 = 40$ cm. A Fabry-Perot interferometer was used to register the spectrum of the radiation scattered by the mixture backward and then amplified in the laser. In all cases, breakdown was observed in the mixture.

In the pressure range 3 - 7 atm the spectra revealed a Stokes line with a shift ~ 0.055 cm^{-1}, corresponding to the calculated shift of the MB component (~ 0.056 cm^{-1}). This

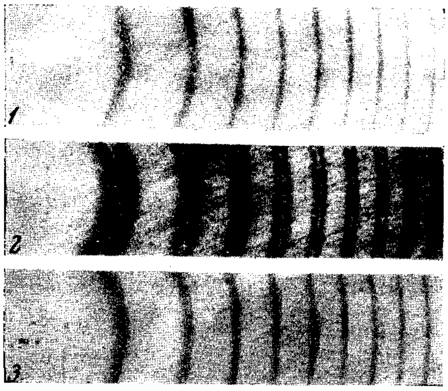

Fig. 1. Spectra: 1) of laser emission and stimulated backward scattering, 2) in helium-xenon mixture (10:1) at 3 atm pressure, 3) in pure helium at the same pressure. Interferometer dispersion 0.167 cm^{-1}.

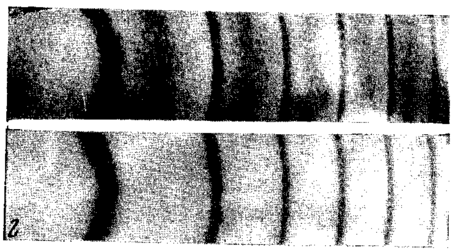

Fig. 2. Spectra of laser emission after passing trough focal region: 1) in helium-xenon mixture (100:1), 2) in pure helium. Interferometer dispersion 0.28 cm^{-1}.

line was therefore interpreted as a SMBS component amplified at the laser mode.

At a pressure 3 - 4 atm, a Stokes line with a displacement 0.033 - 0.042 cm^{-1} was observed in the spectra, and was interpreted as a stimulated concentration scattering line. This line, together with the SMBS line, was missing from the spectrum of the laser emission and from the scattering spectrum in pure helium at a pressure equal to the mixture pressure (see Fig. 1). Inasmuch as the position of the stimulated-concentration-scattering line in the spectrum was uncertain by an amount at least equal to the distance between the longitudinal modes of the laser (0.005 cm^{-1}), one cannot conclude uniquely that the stimulated-concentration-scattering line shift depends on the pressure. The absence of a stimulated concentration scattering line in the spectrum at pressures higher than 4 atm can be attributed to the earlier occurrence of the breakdown in the mixture, when the stimulated concentration scattering does not have time to develop, and also to the possible competition on the part of the SMBS. The absence of a stimulated-concentration-scattering line in the spectrum at pressures lower than 3 atm can be attributed to the smallness of g_c. From the shift of the component of the stimulated concentration scattering we obtain for the helium-xenon mixture $D \approx 0.7$ cm^2/sec and P = 1 atm, assuming that $D \sim 1/P$.

It is possible, however, that the observed shift $\Delta\nu$ is the result of the interaction of the SMBS and the stimulated concentration scattering in the scattering volume, and also of amplification in the laser. This question calls for a detailed theoretical discussion.

To clarify the role of the concentration mechanism in nonlinear interaction in mixtures, we investigated the spectrum of the light of a pulse passing through the focal region of a lens with f = 3 cm. The nonlinear increment n' of the refractive index n is negative [1] and is given by

$$n' = -B_c |\mathbf{E}_L|^2 n^2.$$

When $|\vec{E}_L| \approx 5 \times 10^7$ V/cm, $n' \approx -2 \times 10^{-4}$ in the investigated mixture. Prior to the breakdown, the laser pulse will experience a phase self-modulation, which leads to an anti-Stokes shift of the laser frequency by an amount

$$\Delta\nu = (-|\mathbf{k}_L|\ell/2\pi n)(\partial n'/\partial t) \quad [5]$$

$(\Delta\nu \sim 10^{-2}\ \text{cm}^{-1})^{1)}$.

The anti-Stokes shift observed by us experimentally in the laser-emission spectrum turned out to be larger by approximately one order of magnitude than the estimate value (Fig. 2). In the spectrum of the back-scattered light we observed only the laser emission. It is interesting that the shifted diffuse band (Fig. 2) was observed even at low xenon concentrations $c \approx 3 \times 10^{-3}$. It did not appear in pure helium up to 10 atm and in xenon up to 0.5 atm (the breakdown did occur in this case). At $c \approx 0.1$, a shift $\Delta\nu \approx 0.09 - 0.07\ \text{cm}^{-1}$ of the band maximum was observed, and at $c \approx 2 \times 10^{-2} - 1 \times 10^{-2}$ the shift was $\Delta\nu \approx 0.120\ \text{cm}^{-1}$.

The observed values of the broadening of the laser line can be apparently explained by assuming again that excited xenon atoms with noticeably large polarizability are produced in the interaction volume [4].

In conclusion, we are grateful to I. L. Fabelinskii for interest in the work, valuable hints, and discussions, to V. S. Starunov for useful discussions, to D. I. Mash for interest in the work, and to M. A. Vysotskaya, V. P. Zaitsev, and V. A. Morozov for help with the work.

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E R R A T A

In the article by I. M. Arf'ev and V. V. Morozov, Vol. 9, No. 8, "stimulated thermal scattering" on lines 19-20 of p. 269 should read "stimulated temperature scattering.'

In the article by V. B. Semikoz, Vol. 9, No. 9, "approximately smaller than" on line 3 of p. 326 should read "approximately smaller by a factor of two than."