

STIMULATED SCATTERING AND SPECTRAL BROADENING OF A HIGH-POWER LASER PULSE IN GASES

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A spectral investigation of stimulated Mandel'shtam-Brillouin scattering in compressed nitrogen as a function of exciting light intensity is performed under various observation conditions. Stimulated Mandel'shtam-Brillouin scattering and stimulated thermal (entropy) scattering of light in gaseous hydrogen are detected in the low pressure range. An anti-Stokes spectrum broadening of the laser emission that passed through the interaction region in various gases is also detected. The obtained results are compared with the predictions of the existing theories.

INTRODUCTION

THE availability of ruby lasers capable of emitting giant pulses of light within a narrow spectral interval allowed us to return to the experimental investigation of the fine structure in gases. Prior to the appearance of lasers the problem of fine structure observation remained unsolved.^[1]

The fine structure in the stimulated Mandel'shtam-Brillouin scattering (SMBS) in compressed gases was recently detected and published almost simultaneously in several papers.^[2-4] The detection of the fine structure was of itself of principal importance, since it confirmed the predictions of classical theory^[1] and provided new tools for the study of optical and acoustic properties of the gaseous medium.

The detection of SMBS was soon followed by the discovery and investigation of the fine structure of the Raleigh line in thermal scattering^[5, 6] excited by the $\lambda = 6328 \text{ \AA}$ line of the gas laser. The determination of hypersound velocity from Mandel'shtam-Brillouin components (MBC) of thermal scattering of light in nitrogen and in a number of other gases showed that the hypersound velocity at $\sim 10^9 \text{ Hz}$ has an adiabatic value. At the same time even the early experimental investigation of SMBS in compressed gases revealed the hypersound velocity to be closer to the isothermal value^[2, 3] regardless of the fact that under the experimental conditions the frequency at which sound propagation changed from adiabatic to isothermal ($\sim 10^{11} \text{ Hz}$) was much higher than that of significance in light scattering ($\sim 10^9 \text{ Hz}$).^[1]

Various hypotheses were advanced to explain the observed velocity values.^[2, 3, 7-9] Some of these can be rejected at this time in the light of the subsequent experimental research.

On the other hand the diversity of experimental conditions showed that in some cases of SMBS an adiabatic value of velocity is observed while in other cases this velocity is close to the isothermal value. This problem is discussed in detail in Sec. 1 of this paper.

This paper develops the method of additional amplification of stimulated scattering based on the utilization of the superregenerative amplification or, in other words, trigger amplification of a laser resonator mode.

The use of this method enabled us to investigate SMBS that could not be directly detected at comparatively low pressures. We studied the MBC shift as a function of hydrogen pressure. The results of this study are given in Sec. 2.

Section 3 presents the results of studying stimulated thermal (entropy) scattering of light in gaseous hydrogen. The detection of this effect in gases was also made possible thanks to the trigger amplification method. The obtained results are compared with theoretical predictions.

The fourth and last section of this paper reports on the results of observing the frequency broadening of laser pulse spectrum in the anti-Stokes direction in gases and presents a discussion of the possible causes of such a broadening.

1. MBC SHIFT AS A FUNCTION OF EXCITING LIGHT INTENSITY

It is as yet unclear why the hypersound velocity in compressed gases as computed from the SMBS component shift is below the adiabatic value under certain conditions. Therefore it was of interest to examine at least qualitatively the location of MBC as a function of the exciting light intensity.

We investigated SMBS in gaseous nitrogen at 125 atm using the setup shown in Fig. 1. The laser emission

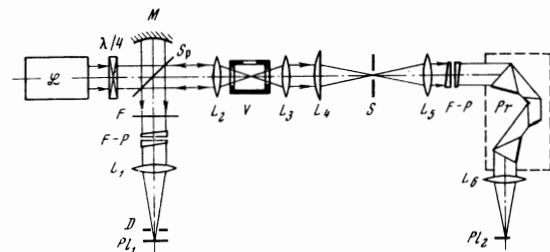


FIG. 1. Diagram of the setup. \mathcal{L} – ruby laser^[16]; $\lambda/4$ – quarter-wave mica plate for $\lambda = 6943 \text{ \AA}$ emission; M – spherical mirror ($f = 6 \text{ m}$, $R \sim 96\%$); S_p – glass beam splitting plate; L_2 and L_3 – circular lenses; V – high-pressure gas cell; L_4 – cylindrical lens ($f = 10 \text{ cm}$); $F-P$ – Fabry-Perot interferometer; L_1 – lens ($f = 160 \text{ cm}$); F – red light filter; D – diaphragm; S , L_5 , and P – slit, lens, and prisms of ISP-51 spectrograph; L_6 – lens ($f = 80 \text{ cm}$); Pl_1 and Pl_2 – photographic plates.

Table I

Focal length of lens, f, cm	MBC shift, cm ⁻¹			Velocity of sound, v m/sec
	I *		II	
	$\Delta\nu$	$\Delta\nu$	$\Delta\nu_{ar}$	
30	0,038	0,040 0,038	0,039	393 ± 20
20	0,039	0,037 0,037 0,035	0,037	372 ± 20
10	0,035	0,035 0,034 0,033	0,033	331 ± 25
5	0,032	0,028 0,029 0,034	0,030	300 ± 25
3	0,031	0,025 0,027 0,033 0,037	0,030	300 ± 25

*I – SMBS investigation was performed with the feedback between the laser and the scattering volume suppressed by a polarizing shutter ($\lambda/4$). II – consecutive generation of several MBC.

contained one axial and possibly several transverse modes with a total width $\Delta\nu_l = (0.5-1) \times 10^{-2} \text{ cm}^{-1}$ (without excluding the instrument width of the Fabry-Perot interferometer). The investigation was performed both with feedback between the scattering volume and the laser (consecutive generation of several MBC) and when the feedback was disrupted by a $\lambda/4$ plate. In the latter case we observed only a single Stokes component of SMBS.

Table 1 shows the measured shifts of MBC and the hypersound velocities in compressed nitrogen computed from these shifts. The exciting radiation of $\sim 100 \text{ MW}$ was focused with lenses of various focal lengths.

We see from the measurement results that the MBC shifts obtained with the aid of lenses having focal lengths of 30 and 20 cm correspond to the adiabatic sound velocity at $T = 300^\circ \text{K}$ and a pressure of 125 atm (386 m/sec).^[8] The average values of MBC shifts in consecutive scattering correspond to the Stokes MBC in the no-feedback regime with an accuracy of an experimental error ($\pm 0.002 \text{ cm}^{-1}$). The hypersound velocity decreases with decreasing focal length, i.e., with increasing intensity of the exciting light in the interaction volume. The difference between consecutive SMBS component shifts observed when short-focus lenses were used in some cases exceeded the experimental error as well as the distance between the axial laser modes (0.005 cm^{-1}).

The measurement of shifts of each consecutive MBC in the feedback regime shows that in the case of long-focus lenses ($f = 20-30 \text{ cm}$) the MBC are equidistant and the sound velocity computed from the shifts is adiabatic. As the focal length decreases the shift of the consecutive MBC is not equidistant; it decreases at first and then increases (the sound velocity computed from the average value of MBC shifts is less than adiabatic).

Table 2 shows some results of measuring the hypersound velocity from MBC in compressed nitrogen obtained by various authors under various experimental conditions. According to the table, hypersound velocities computed from MBC shifts at 30–150 atm fall below the adiabatic values. The authors of these reports point out that they used the highest of the laser powers indicated in Table 2 to detect SMBS in this range of pressures.

Table II

Laser characteristics*	focal length of lens, f, cm	gas pressure p, atm	sound velocity v, m/sec		literature
			exp	theor. [8]	
U = 0.8 J $\tau = 8-35 \text{ nsec}$	15, 20	25–50	370–290	353–362	[11]**
U = 0.5 J $\tau = 6-15 \text{ nsec}$					
I = 10–100 MW/cm ²	5	70–150	337–385	370–399	[3]
I = 250 MW/cm ²	30	125	356 ± 20***	386	[4]**
I = 50 MW/cm ²	3	125	290 ± 10	386	[2]
I = 100 MW/cm ²	3	150	320 ± 20	399	[10]
	4	250	435 ± 20	450 ± 5	[12]
		500	560 ± 25	590 ± 5	

*I – intensity, U – energy, τ – laser emission pulse length.

**SMBS was observed when the feedback between the laser and the interacting volume was suppressed by a polarizing shutter ($\tau/4$).

***The result of measuring hypersound velocity made more precise in [8].

The hypersound velocity at gas pressures of 150 atm and over reported in [3, 4, 10] is close to the adiabatic value. As pressure decreases the hypersound velocity deviates from the adiabatic, passes through a minimum at $\sim 50 \text{ atm}$, and again increases reaching the adiabatic value at pressures of $\sim 25 \text{ atm}$.^[3, 11] Thus the hypersound velocity, according to Table 2 and our results given in Table 1, is determined by the exciting light intensity in the interaction region as well as by gas pressure.

When long-focus lenses are used ($f = 20-30 \text{ cm}$) the hypersound velocity becomes adiabatic (^[4] and Table 1) and decreases with decreasing focal length of the lenses (^[2, 11] and Table 1).¹⁾ Similar results were obtained in the case of other gases.^[2, 3, 12] Madigosky and others^[8] tend to explain the discrepancy of experimental results obtained in [2–4] (see Table 2) by the presence of several modes in the exciting radiation. This should cause errors in the determination of MBC shift capable of increasing or decreasing $\Delta\nu_{MB}$.^[13, 14] However, these considerations are not applicable to our results that were obtained with a single axial laser mode and in the absence of feedback between the scattering medium and the laser.²⁾ The presence of transverse modes in our case could change the value of $\Delta\nu_{MB}$ by $\sim 10^{-4}-10^{-5} \text{ cm}^{-1}$ and could not serve as an explanation of the observed values of $\Delta\nu_{MB}$.

We note that in the case of short-focus lenses used under our experimental conditions plasma was generated in the interaction region that disrupted the SMBS process;^[11] the observed MBC thus occurred in an increasing laser emission field.

The mechanism by which intensity affects the value of $\Delta\nu_{MB}$ is not yet known. However we can point out some possible causes of such an effect. A high intensity can give rise to a nonlinear hypersonic wave that is absorbed more than a strong linear sonic wave and the absorption of hypersound leads to a reduction in $\Delta\nu_{MB}$.^[11] Whether such a mechanism of varying $\Delta\nu_{MB}$ exists can be determined only after a simultaneous solution of nonlinear Maxwell equations and hydrodynamic equations, a procedure that has not yet been performed. Another cause of reduction of $\Delta\nu_{MB}$ can be ascribed to

¹⁾We note that inhomogeneous distribution of laser emission intensity over the beam cross section is not taken into account in the comparison of data obtained by various authors.

²⁾In [13, 14], where adiabatic velocity of hypersound was observed in liquids with a single-mode laser emission, the exciting light intensity seems to be comparatively moderate.

the strong interaction ("bunching") of molecules in a growing field of an intense light wave.^[9]

The above research yielded a practically significant result: we determined the experimental conditions that make it possible to obtain the correct values of hypersonic velocity from the measured SMBS components.

2. STIMULATED MANDEL'SHTAM-BRILLOUIN SCATTERING IN HYDROGEN AT LOW PRESSURES

According to the results of our measurements (Sec. 1) the adiabatic velocity of hypersonic wave propagation is obtained with long-focus lenses, i.e., with comparatively low exciting light intensity in the interaction region regardless of the presence or absence of feedback between the laser and the scattering volume. This order of intensity in the investigation of SMBS in the low-pressure region calls for a consecutive amplification of the scattered light to be registered on the photographic film.

If the scattering is observed in the feedback regime the additional amplification of a Stokes MBC scattered at the angle of $\theta = 180^\circ$ can be obtained by superregenerative amplification by the pumping generator mode. The superregenerative amplification by the pumping generator mode, or the trigger amplification mechanism, consist of the fact that the pump power excess over generation threshold and the additional amplification in the gas cell change the single-mode laser regime into a two-mode (biharmonic) regime in which the second mode appears at a frequency that is closest to the maximum gain of the stimulated light scattering. The process of amplification of the pumping generator modes can be controlled, according to^[15], either by the choice of pumping intensity or by a suitable selection of the mode Q-factor in a specified spectral region within the limits of the ruby luminescence linewidth (R_1).

Using the trigger mechanism of MBC amplification in the pumping generator we performed spectral investigation of SMBS in gaseous hydrogen within the pressure range of 2–100 atm.

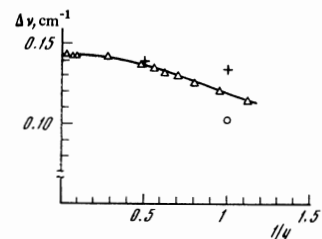
At high pressures (70–100 atm) we observed two equidistant Stokes MBC; a single Stokes component was observed as a rule at the pressures of 2–70 atm.

A giant pulse of ruby laser \mathcal{L} was used as the pumping light; the setup and operating principle of this laser were described earlier.^[16] In order to vary the pump power within wide limits we used eight flash lamps instead of the reported four.^[16] Each of two rubies was placed in a separate reflector consisting of four cylinders with elliptical cross section. The mode separator consisted of two plane parallel plates of fused quartz 2.5 mm thick separated by a ring 5 mm thick.

By varying the initial transmission of the saturable filters used as the laser Q-switch and selecting the corresponding pump power we were able to obtain a giant pulse of 50–150 MW, at the same time retaining a narrow emission spectrum of $\Delta\nu \leq 0.01 \text{ cm}^{-1}$ with a 50% pumping power excess over threshold. The SMBS was investigated with a 25–30% excess over generation threshold.

The emission was focused by a lens ($f = 20 \text{ cm}$) into a cell filled with gaseous hydrogen. The emission power was picked so as to avoid the formation of plasma in

FIG. 2. Shift of Stokes component of SMBS as a function of $1/y = \pi^{3/2}l/\Lambda$; + — location of spectral intensity maximum of fine structure of thermal scattering in adiabatic approximation; O — the same in isothermal approximation computed in [7].



the focal region.³⁾ The frequency-shifted light scattered by 180° from the direction of laser propagation excited the ruby laser resonator mode that was closest in frequency to the maximum gain of the SMBS. Glass beam-splitting plate S_p (Fig. 1) and spherical mirror M ($f = 6 \text{ m}$) directed a portion of the light through light filter F to Fabry-Perot interferometer $F-P$ and photographic plate Pl_1 . Photographs were obtained with the aid of Fabry-Perot etalons having dispersion regions 0.166; 0.333; and 0.5 cm^{-1} .

Figure 2 shows the MBC shift $\Delta\nu$ as a function of the ratio \bar{l}/Λ , or of the quantity $1/y$ ^[17] ($y = \pi^{-3/2}\Lambda/\bar{l}$). Here \bar{l} is the mean free path of the molecules and Λ is the hypersound wavelength. The experimental points plotted in the graph were obtained by averaging the results of measuring several series of photographs which showed a smooth variation of the MBC location with varying gas pressure. The variation smoothness and accuracy of the results are determined by the distance between the axial modes of the laser resonator ($\Delta\nu = c/2L \approx 5 \times 10^{-3} \text{ cm}^{-1}$).

The previously obtained^[18] sound absorption coefficient as a function of the ratio of sound frequency f to hydrogen pressure p leads to the conclusion that already at $f/p \approx 10 \text{ MHz/atm}$ the entire absorption is determined by shear viscosity. According to the relaxation theory^[19] the hypersonic velocity should in this case be close to v_∞ . Consequently under our experimental conditions ($f/p \gtrsim 50$) the propagation of hypersonic waves can be described by gas kinetic Boltzmann equation for ideal gas^[17] since the energy of compression has no time to convert into the energy of the rotational degrees of freedom.

According to Fig. 2 the MBC shift at the pressures of 10–100 atm ($1/y < 0.5$) corresponds to $0.141 - 0.138 \text{ cm}^{-1}$. The sound velocity computed from this shift ($1480-1436 \pm 60 \text{ m/sec}$) corresponds to the adiabatic value of $v_\infty \approx 1400 \text{ m/sec}$.

With decreasing pressure $1/y \geq 0.5$ the MBC shift decreases faster than predicted by the adiabatic analysis. The mean free path of molecules in this pressure region becomes comparable to the hypersound wavelength. Consequently the reduction of MBC shift in hydrogen when $1/y \geq 0.5$ can be ascribed to the gradual transition of the adiabatic process of sound propagation into the isothermal process.

3. STIMULATED THERMAL LIGHT SCATTERING IN GASEOUS HYDROGEN AT LOW PRESSURES

The stimulated thermal (entropy) scattering of light (STS) that was recently detected^[20] in benzene and in

³⁾As we know, plasma not only absorbs the exciting light but also tends to cause undesirable reflections.

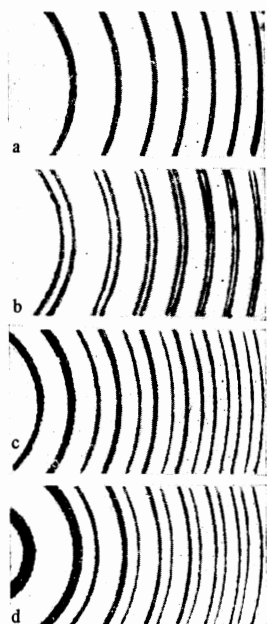


FIG. 3. Spectrograms of amplified STS for various gaseous hydrogen pressures. a — laser emission; b — $p = 6.5$ atm; c — $p = 2.5$ atm; d — $p = 1.5$ atm; dispersion region of Fabry-Perot etalon is 0.166 cm^{-1} .

principle occurs in all liquids (except for water at 4°C) should also be observed in gases. Therefore simultaneously with ^[20] stimulated light scattering was investigated in gaseous hydrogen.

Gaseous hydrogen is the most convenient object of investigation since the width of its scattering line is markedly larger than in other gases.^[11]

According to the STS theory, valid in the hydrodynamic approximation,^[21] the anti-Stokes component of scattered light should damp out, while the Stokes component intensity $E^2(x)$ should increase according to

$$E^2(x) = E_1^2(0) \exp \{g_{1T}x\}, \quad (1)$$

where $E_1(0)$ is the field intensity of the initial thermal scattering at the half-width of the line and g_{1T} is the gain:

$$g_{1T} = -2K_\omega + B|K_1| \times \frac{\Omega/\Omega_m}{1 + (\Omega/\Omega_m)^2} E_0^2. \quad (2)$$

Here E_0 is the field intensity of the exciting radiation, $2K_\omega$ is the light extinction coefficient,

$$B = \frac{T(\partial\epsilon/\partial T)^2_p}{16\pi n^2 \rho c_p}$$

$\Omega_m = \chi q^2$ is the frequency at the half-width of the scattering line, χ is the thermal conductivity coefficient, $q = 2\pi/\Lambda$ is the wave vector, and the remaining symbols are customary.^[11]

It follows from (2) that the gain maximum of STS occurs at the half-width (Ω_m) of the thermal scattering line.

In addition to the central line liquids always produce Mandel'shtam-Brillouin components (MBC). In gases MBC are present only when $\bar{l}/\Lambda < 1$. In the case of $\bar{l}/\Lambda \gg 1$ there are no MBC and only the central line remains. At high pressures the central component ($\bar{l}/\Lambda \ll 1$) has a Lorentz shape and at low pressures, in the limit $\bar{l}/\Lambda > 1$ the line shape of thermal scattering assumes Doppler character. Accordingly, the half-width

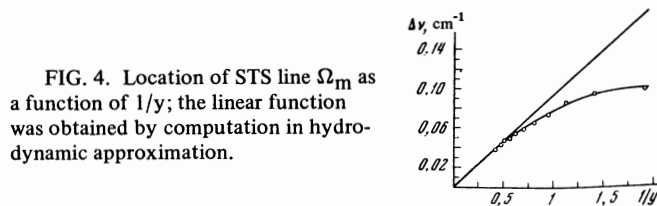


FIG. 4. Location of STS line Ω_m as a function of $1/y$; the linear function was obtained by computation in hydrodynamic approximation.

varies from a small magnitude at high pressures to the limit of a large Doppler half-width at low pressures.

In the case of stimulated scattering in gases within the pressure range from 2 to 10 atm and a pumping pulse power of 100–150 MW (by integrating over the interaction region) we have the factor of $\sim e^3$ in front of $E_1^2(0)$ in (1); however in order to observe the effect the exponent should be larger by an order of magnitude.^[4] It follows then that the scattered light should be additionally amplified with a high gain to make the observation of the sought phenomenon possible.

To observe the scattered light at the angle of $\theta = 180^\circ$ the amplification was achieved in the same manner as in the preceding section, using the superregenerative amplification of a laser mode whose frequency occurs at the gain maximum in (2). In order to make the STS amplification the most effective in the laser we picked the mode separator so as to practically exclude SMBS. The mode separator consisted of two plane parallel quartz plates 13 and 2.5 mm thick held apart by a spacer ring 5 mm thick. The photographs were obtained with a Fabry-Perot etalon with dispersion regions of 0.166, 0.333, and 0.5 cm^{-1} .

Figure 3 shows the spectrograms of amplified stimulated thermal scattering at various gas pressures.

Figure 4 shows the position of the STS line (or thermal scattering half-width) as a function of the ratio \bar{l}/Λ or of the quantity $1/y$ (see Sec. 2).

According to the graph (Fig. 4) the experimental values of the half-width of the unshifted fine structure line are in agreement with the computed values when $y \geq 2$. In this case the spectrum has a Lorentz form whose width is proportional to $1/y$. Deviation from the linear law occurs when $y < 2$. As y decreases the shift of STS maximum tends to approach $\Delta\nu \approx 0.11\text{--}0.12 \text{ cm}^{-1}$. This is in qualitative agreement with the predictions of the gas kinetic theory^[17, 22] for ideal gas with Maxwellian velocity distribution of molecules.

4. BROADENING OF THE FREQUENCY SPECTRUM OF HIGH POWER LIGHT PULSES IN GASES

We detected the anti-Stokes broadening of the laser emission spectrum in nitrogen and hydrogen within the pressure range of 1–150 atm, using the setup shown in Fig. 1. In this experiment we suppressed the feedback between the laser and the radiation-gas interaction region with a polarization shutter (a $\lambda/4$ plate).

A light pulse of ~ 150 MW was focused by a lens with $f = 3$ cm into the gas cell. The observation was performed in both the forward and reverse directions with respect to the direction of propagation of laser emission.

The light that passed through the gas cell was focused on the slit of an ISP-51 spectrograph with lens L_3

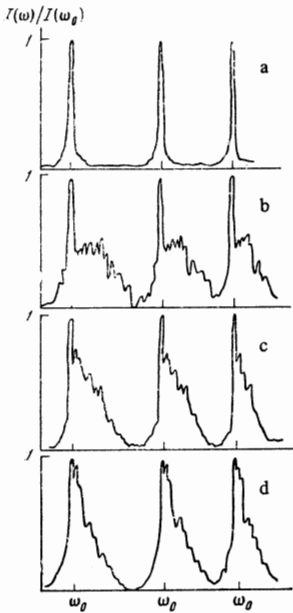


FIG. 5. Microphotographs of laser emission spectrum that passed through the focal region $I(\omega)/I(\omega_0)$ in gaseous nitrogen at various pressures. a — unfocused laser emission; b, c, d — spectrum of light that passed through the focal volume at 15, 35, and 100 atm respectively; the dispersion region of the interferometer is 0.625 cm^{-1} .

($f = 3 \text{ cm}$) and cylindrical lens L_4 ($f = 10 \text{ cm}$). The Fabry-Perot interferometer (with a dispersion region of 0.6 cm^{-1} or 1.66 cm^{-1}) was crossed with a prism spectrograph by the internal parallel beam method.^[23] The observation of the scattering at 180° angle was performed similarly to the observation of SMBS (see Sec. 2). In this case the dispersion region of the interferometer was 0.166 ; 0.5 ; and 1.66 cm^{-1} . The anti-Stokes broadening of the frequency spectrum was detected in the forward direction. Such a broadening was not observed in scattered light in the reverse direction. Only the Stokes component of SMBS was observed in the reverse direction in nitrogen at 50 – 150 atm .

Figure 5 shows microphotographs of the spectrum of laser emission that passed through the focal region in gaseous nitrogen at various pressures. According to the spectrogram the anti-Stokes broadening reveals a structure rather than a monotonic background. The smaller the frequency shift and the higher the pressure the greater is the intensity of the maxima. The spectral broadening increases with decreasing pressure.

Figure 6 shows the averaged spectral half-width as a function of nitrogen pressure. We see that the spectrum broadening is approximately tripled when pressure drops from 100 to 1 atm . When the laser emission intensity was attenuated in half the broadening also decreased by a half.

This broadening of the laser pulse spectrum can apparently be ascribed to the variation of the nonlinear part of the refraction index of the gas in the time-varying laser emission field; as a result the phase of the light wave changes in time.

Several theoretical^[24–26] and experimental^[26, 27] papers appeared in the last few years showing that multiple broadening of the frequency spectrum of optical pulses 10^{-8} – 10^{-9} sec long can take place in the self-focusing channels in liquids or generally in a nonlinear medium. Such a spectral broadening is possible without significant distortion of the amplitude envelope of the initial pulse.^[24]

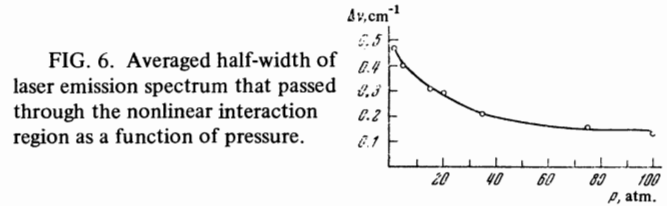


FIG. 6. Averaged half-width of laser emission spectrum that passed through the nonlinear interaction region as a function of pressure.

Let us determine if this interpretation of the spectral broadening observed in our experiments is feasible. In this analysis we use first-approximation formulas describing the propagation of a quasi-harmonic plane wave in a nonlinear medium.^[24]

If the maximum phase shift Φ_{max} exceeds π , the pulse spectrum width, according to^[24], should be close to the maximum frequency deviation:

$$\Delta\omega = \frac{\partial\Phi}{\partial t} = -\frac{kx}{n_0} \left(\frac{\partial n'}{\partial t} \right)_{\text{max}}, \quad (3)$$

where $n = n_0 + n'(E^2)$ is the refraction index and n' is its nonlinear part.

The "critical" distance x_{cr} at which Φ_{max} is of the order of π equals

$$x_{\text{cr}} = \pi n_0 / kn'_{\text{max}}. \quad (4)$$

The shape of the spectral line $I(\omega)$ of the emission pulse that passed through the nonlinear medium depends on the time variation of pulse intensity. Thus for trapezoidal and parabolic shapes we expect a maximum at a distance $\Delta\omega$ from the unshifted line of the laser emission; the distance is determined by (3)⁴⁾ both in the Stokes and anti-Stokes regions. The broadening process in the Stokes and anti-Stokes regions is separated in time. If $\partial n'/\partial t < 0$ when the pulse intensity increases, then $\Delta\omega > 0$ and, conversely, when the intensity decreases $\partial n'/\partial t > 0$ and $\Delta\omega < 0$.

In our experiment the focal region gave rise to plasma known^[28] to be opaque to laser radiation. Under such conditions the breakdown takes place as a rule with increasing intensity of laser radiation.^[10] Consequently the spectral broadening process could be recorded in our case only when the laser pulse intensity was increasing.

In order to clarify the observed anti-Stokes spectral broadening accompanying the increasing light intensity we assume that the variation of the nonlinear part of the gas index of refraction is due to the electro-calorimetric effect.^[21] Then

$$n' \approx \frac{\epsilon'}{2n_0} \approx \left(\frac{\partial\epsilon}{\partial T} \right)_p \frac{\Delta T}{n_0}, \quad \Delta T = -\frac{T(\partial\epsilon/\partial T)_p E_0^2}{8\pi\rho c_p}. \quad (5)$$

In this case $\partial n'/\partial t < 0$ and $\Delta\omega > 0$.

Assuming that there is no self-focusing in the course of focusing a high-power light pulse in a gas and the focusing region remains constant and is determined by the lens, we evaluate the light intensity required for the observation of the effect under consideration. If the length of the interaction region is taken as 10^{-1} cm and

⁴⁾Equation (3) does not account for the fine structure of the line within intervals of the order of the initial width of the laser emission spectrum.

x_{CR} is at least an order smaller (10^{-2} cm), then considering (4) and (5) we find that spectral broadening in nitrogen at the pressures of 1–100 atm should be observed at electric field intensities of the light wave of $\sim 10^7$ – 10^8 V/cm.⁵⁾ The spectral broadening then is, according to (3), 10^{-1} cm⁻¹ which agrees in the order of magnitude with the observed spectral broadening (see Fig. 6).

The structure of spectral broadening (see Fig. 5) can apparently be ascribed to the space–time development of the laser pulse generation; it propagates transversely (from the center outwards) in a time comparable with the pulse length. The pulses generated by the individual sectors of the ruby end face can be different in shape and shorter than the giant pulse generated by the entire end face of the crystal.^[30]

Consequently the growth of intensity in time proceeds differently in different regions of the lens focus. Since the recorded spectral broadening of the light that passed through the nonlinear medium is integrated over the entire volume, and since the shape of the broadened spectral line depends, according to (3), on the time–variation of intensity in each focal region, several maxima are observed rather than one.

At low gas pressures the breakdown occurs with a longer delay than at high pressures.^[29] Consequently the spectral broadening of laser emission can be larger at low pressures since the derivative $\partial n'/\partial t$ at low pressure is larger than the same derivative at high pressure; this is further confirmed qualitatively by the experimental results shown in Figs. 5 and 6.

It is of interest to note that (see^[29]) equal laser threshold powers are required to cause breakdown in nitrogen and in hydrogen at the pressures of 1 atm and 60–70 atm; the breakdown powers are 1.5×10^6 and 2×10^5 W respectively.

If we assume that the maximum local fields are proportional to the volume–average threshold breakdown fields, i.e., $E_{\max \text{ loc}} \sim E_{\text{br}}$, then under these pressures the broadening in hydrogen should be approximately six times lower than the broadening in nitrogen. According to our measurements however the broadening is the same in hydrogen as in nitrogen at 1 atm and the broadening in hydrogen is only twice as small as in nitrogen at 70 atm. This discrepancy seems to be typical of the focusing conditions in various gases. The fact that the laser spectrum broadening is larger in hydrogen indicates possibly the presence of self-focusing at light intensities that are lower than the threshold intensities for plasma generation.

An indirect confirmation of this assumption is the fact that we observed stimulated Raman scattering (SRS) in hydrogen in which laser emission was focused by lenses with focal lengths of 20 and 10 cm in a gas cell 40 cm long at pressures of 30–150 atm. The SRS was not observed when the same emission power was focused by the same lenses in a cell only 10 cm long.

The stimulated Mandel'shtam–Brillouin scattering and the thermal (entropy) scattering in gases were detected only a few years ago and yet this research has

already yielded many physical results of interest. It is expected that the further development of this branch of nonlinear optics will bring new data on the relaxation processes in gases.

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