# Nature of the anomalously strong cubic optical nonlinearity of a gaseous plasma

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An explanation is offered for the circumstance that the efficiency of nonresonant four-wave processes (coherent anti-Stokes Raman scattering and third-harmonic generation) in the plasma of the laser breakdown of a gas is much higher than in a neutral gas. The explanation is based on a quasiresonant increase in the cubic susceptibility as the atoms and ions decay through highly excited states.

**1**. We wish to offer an explanation for the anomalous increase in the nonresonant optical nonlinearity of third order during the laser breakdown of a gas. This effect was first discovered and studied by Brodnikovskiĭ *et al.*[1] by the method of coherent anti-Stokes Raman scattering (CARS), in the scheme $\omega_a = 2\omega_1 - \omega_2$, where $\omega_1$ and $\omega_2$ are the frequencies of the pump waves. This increase is universal in nature, manifested during the laser breakdown of any gas and also in a surface breakdown of many metals and insulators.

We believe that the reason for the sharp increase (by two or three orders of magnitude) in the slightly dispersive cubic optical susceptibility during laser breakdown is a decay of the gas particles (primarily atoms and atomic ions) through high-lying electronic states. Since these states become more closely spaced with increasing degree of excitation of the gas, the stage is set for a quasiresonance of the frequencies of the pump beams (in the visible range) and electronic transitions in the excited atoms and ions. As is shown below, this factor and also the increase in the oscillator strengths in transitions between excited states of atomic particles lead to a quantitative explanation of both the anomalously high intensity of the scattered anti-Stokes radiation and its anomalous polarization, observed experimentally. None of the models which were proposed in Ref. 1 can explain these two anomalous features simultaneously.

2. Let us examine in more detail this new mechanism for the effect. The four-wave CARS process, which starts in an unexcited neutral gas from the electronic ground state, is a nonresonant process because the pump frequencies $\omega_1$ and $\omega_2$ and their linear combinations are quite far from the frequencies of electronic transitions [see the inset in Fig. 1(a)]. When a plasma is produced, the process becomes quasiresonant, since high-lying levels of atoms and ions are populated, and transitions between these levels correspond to the visible range. As a result, the frequencies $\omega_1$ and $\omega_2$ themselves and also their combinations $\omega_1 - \omega_2$, $2\omega_1$, $2\omega_1 - \omega_2$ turn out to lie near

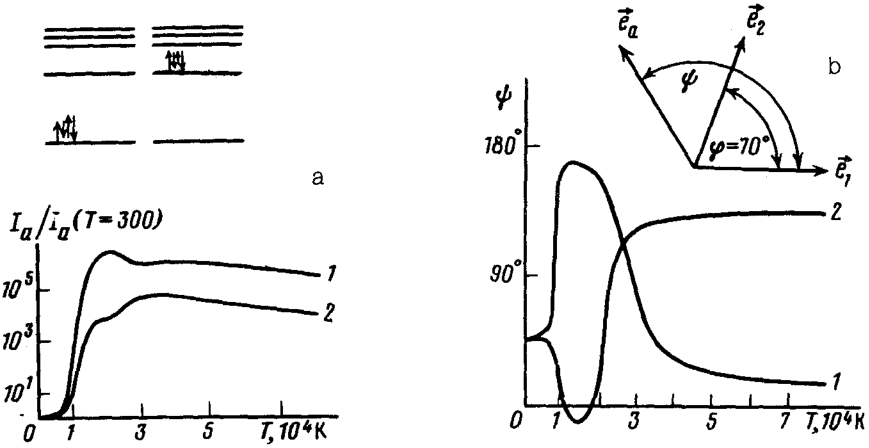

FIG. 1. (a): Calculated temperature dependence of the intensity of the CARS signal. (b): Calculated temperature dependence of the angle $\psi$, which is the angle between the unit polarization vectors of the pump light, $e_1$, and of the anti-Stokes signal, $e_a^{(\text{plasma})}$. The calculations were carried out for equilibrium heating of a "gas" of hydrogen atoms. 1—$\omega_1/2\pi c = 16\,000$ cm$^{-1}$, $\omega_2/2\pi c = 15\,000$ cm$^{-1}$; 2—$\omega_1/2\pi c = 17\,000$ cm$^{-1}$, $\omega_2/2\pi c = 16\,000$ cm$^{-1}$. The inset in Fig. 1(a) illustrates the reason for the intensification of the CARS signal when excited electronic states are filled, by virtue of a quasiresonance of the four-wave process starting from excited states.

frequencies of the numerous allowed transitions between populated excited levels. The cubic susceptibility thus undergoes a quasiresonant increase.

For a quantitative evaluation of the importance of this mechanism in the effect, we have calculated the components of the nonresonant cubic susceptibility tensor $\chi^{(3)}_{ijkl}(\omega_a;\omega_1,\omega_1,-\omega_2)$ for a model medium—an excited gas of hydrogen atoms—from the expression derived by Yuratich and Hann.[3] As the initial states we adopt states with principal quantum numbers $n$ from 1 to 6; these states are "virtual" states in perturbation-theory calculations. The relative number of ionized atoms is estimated from the Saha equation. The distribution of particles among discrete levels is assumed to be a Boltzmann distribution. It has been shown experimentally[4] that only the first six principal quantum numbers prevail in a dense hydrogen plasma at a temperature of a few electron volts.

The details of the calculation procedure will be published separately; we proceed immediately to the basic results on the temperature dependence of the intensity and the orientation of the polarization vector of the CARS signal from a highly excited gas [Fig. 1, (a) and (b)]. From Fig. 1(a) we see that the intensity of the coherent scattering increases sharply at $T > 7000$ K, in accordance with a filling of states with $n > 1$. The polarization direction of the scattered signal, which in the ordinary nonresonant case makes an angle $\cong 43°$ with the unit vector ($\mathbf{e}_1$) of the light at frequency $\omega_1$, goes outside the angle bounded by the unit vectors $\mathbf{e}_1$ and $\mathbf{e}_2$ [Figs. 1(b) and 3], becoming anomalous. We also see that the mechanism proposed here can lead to an intensification of the CARS signal of up to four or five orders of magnitude in an excited gas.

**3.** To verify the validity of this model, we have carried out several experiments. We use an apparatus similar to that described in Ref. 1. Optical breakdown is induced in air and at a metal surface by an independent laser source, a TEA-$CO_2$ laser or a Nd:YAG laser, synchronized with the CARS spectrometer. Figure 2 shows the degree of increase of the nonresonant CARS signal from the air breakdown region as a function of the time delay between the breakdown and the probing. The increase of two or three orders of magnitude achieved here is multiplied by another factor of ten when we consider an erosion plasma. Figure 3 shows a transmission characteristic of the increase anti-Stokes signal through an analyzer corresponding to linear polarization. We see that an anomalous polarization state prevails experimentally; this state cannot be

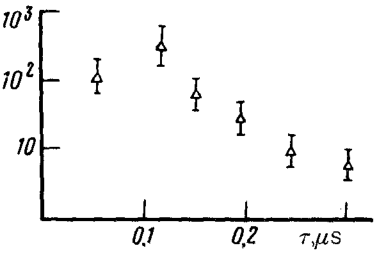

FIG. 2. Extent of the increase in the CARS signal from the plasma of the laser breakdown of air at atmospheric pressure above the level of the corresponding signal from a neutral gas versus the time delay ($\tau_d$) between the probing pulse and the laser-breakdown pulse. The breakdown is caused by an independent Nd:YAG laser with an energy of 0.1 J and a pulse length $\tau_p \cong 20$ ns.

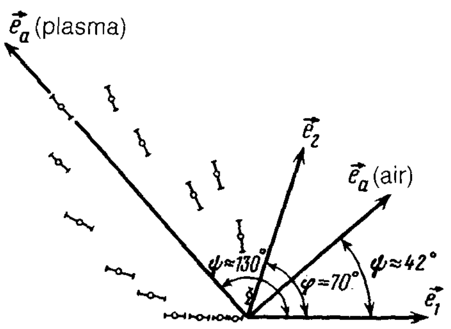

FIG. 3. Results of measurements of the polarization state of the anomalously intense CARS signal at the frequency $\omega_a = 2\omega_1 - \omega_2$ ($\lambda_1 = 532$ nm, $\lambda_2 = 583$ nm, $\lambda_a = 489$ nm) from the plasma of the laser breakdown of air (points, polar diagram). Also shown here are the orientations of the unit polarization vectors of the CARS pump waves, $\mathbf{e}_1$ ($\lambda_1 = 532$ nm) and $\mathbf{e}_2$ ($\lambda_2 = 583$ nm). Here $\mathbf{e}_a^{(\text{plasma})}$ is the polarization vector of the nonresonant CARS signal from the plasma, and $\mathbf{e}_a^{(\text{air})}$ is the same, for the CARS signal from air.

explained by any of the mechanisms proposed in Ref. 1. We also observe a change in the angle through which the polarization vector of the anti-Stokes signal is rotated as a function of the degree of excitation of the medium, as is predicted by the mechanism proposed above.

**4.** This new mechanism also explains several other experimental results. For example, it explains the increase in the efficiency of third-harmonic generation during self-breakdown of picosecond laser pulses.[5] We have been successful in observing a similar effect in the self-breakdown of a gas by pulses of nanosecond length, as well as second-harmonic generation. A similar mechanism may underlie the generation of higher harmonics of the beams from high-power neodymium and $CO_2$ lasers during breakdown at a solid target.[6]

Because of its high optical nonlinearity, a dense gaseous plasma may be a suitable medium for efficient frequency multiplication of laser beams and/or phase conjugation.

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