SPECTRAL BRIGHTNESS OF SRS RADIATION EXCITED WITH A XeCl LASER BEAM IN METAL VAPORS

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Spectral brightness of SRS radiation in lead vapor pumped with a broadband (two lines 3 cm^{-1} wide) and a narrowband (0.1 and 0.01 cm⁻¹) XeCl laser radiation with the divergence close to the diffraction limit has been studied experimentally. It was revealed that SRS linewidth is Doppler limited. For a narrowband pump the SRS efficiency increases two times, and the SRS beam divergence determined by the beam energy within the diffraction angle is three to four times as large as that of the pump beam.

Stimulated Raman scattering (SRS) excited with an excimer laser radiation in metal vapors is one of the most promising ways of obtaining high-power coherent radiation in the visible.^{1–5} The highest efficiency of the conversion was achieved in lead vapor.^{4–6} However as to the degree of coherence of the radiation converted, it remains to be studied.

Rieger⁵ used a XeCl-laser narrowband pump beam with 0.1 cm^{-1} linewidth. According to his findings, linewidth of the Stokes signal was somewhat narrower than that of the pump radiation. Line broadening due to Doppler effect or the presence of different lead isotopes was not observed. Djeu² used XeCl-laser beam with 0.03 cm^{-1} spectral width to obtain the inverse SRS in lead vapor and to compress the Stokes signal. Unfortunately, in his paper he does not report about a spectral width of the radiation converted. Works, dealing with the study of divergence of SRS radiation in metal vapors, are now unavailable in literature.

In this paper we present some results of the experiments on studying the divergence and spectral linewidth of SRS radiation in lead vapor at 458 nm wavelength in order to determine their limit values.

Geometry of the experiment is shown in Fig. 1. The pump beam was generated by a laser system comprising a master oscillator (MO) and a XeCl laser operating in the injection synchronization (IS) mode. The master oscillator was built around the electric-discharge XeCl laser with $0.7 \times 2 \times 60$ cm³ active volume. Its active medium was excited with a 100 nF storage capacitor assembled of K15-10 ceramic condensers (40 kV, 10 nF). The capacitor was charged to 30 kV voltage and then applied to the laser gap through six spark gaps connected in parallel. The 6 nF capacitor comprising 13 K15-4 ceramic condensers (40 kV, 470 pF) was connected in parallel to the laser gap. This capacitor was charged via the discharge on the surface of a dielectric functioning as one of the electrodes of the discharge gap. The laser, operating in the mixture Ne:Xe:HCl = 800:10:1 at a pressure of 4 atm with a plane-parallel cavity, generated the radiation pulse of 100 ns duration and 0.3 J energy. The MO transverse modes were selected with two diaphragms 1.4 mm in diameter. Narrowing of the spectral line was provided by two diffraction gratings (2400 grooves/mm) and the solid-state etalon (free dispersion range 0.5 cm⁻¹, sharpness 12). One grating operated in the grazing incidence (first diffraction order), and the other one operated in the autocollimation mode. Radiation comes out of the cavity through the zeroth diffraction order of the first grating. In such a scheme, the MO output beam had a divergence close to the diffraction limit, spectral line width of 0.01 cm⁻¹, degree of polarization close to unity, pulse length of 50 ns and energy of 4 mJ (Ref. 7). Without the solid-state etalon, MO radiation linewidth was 0.1 cm⁻¹.



FIG. 1. Optical arrangement of the experiment: mirrors with aluminium coating 1, 7, 12, and 16; solidstate etalon 2; a diaphragm 3; active media of MO and laser under control 4 and 9; diffraction gratings with 2400 grooves/mm 5, 6, and 16; a semitransparent meniscus 8; a supergaussian mirror 10; positive lenses (F = 2 and 3 m) 11 and 13; a dielectric mirror with 95% reflection coefficient at $\lambda = 308$ nm and 25% at $\lambda = 458$ nm 14; a cell with metal vapor 15; and, a recording system 17.

The second laser included two $1.5 \times 3.5 \times 32 \text{ cm}^3$ active volumes set in the same cell on the optical axis.⁸ One volume could be switched on with a lag with respect to another, that allowed us to increase the laser pulse length. As a preionization, as in MO, the discharge on the dielectric surface was used. The laser with a plane-parallel cavity, operating by a mixture Ne:Xe:HCl = 1000:5:1 at a pressure of 4 atm, generated a pulse of 110 ns FWHM duration and energy of 120 mJ. The frequency spectrum of radiation in this case consisted of two lines 3 cm^{-1} wide being 28 cm^{-1} apart from each other.

In experiments on radiation conversion, an unstable cavity was used which was formed by a meniscus convex face (reflection coefficient 80%) with the curvature radius of 134 cm and a supergaussian mirror coated on a plane quartz plate. The size of mirror surface was 3-8 mm, the maximum reflection coefficient at the center of mirror was 37%, and the supergaussian order was 4.3. To suppress the noise component of a beam, the diaphragm 10 mm in diameter was set near the convex mirror. With such a cavity, laser generated a radiation pulse up to 100 ns duration and having the energy of 8 mJ. Radiation from MO was injected into the cavity of the second laser through the semitransparent meniscus 8. Both laser were synchronized within ±5 ns. The radiation from the second laser was collimated with the lens 11 and then focused into the cell filled with metal vapor. The working volume of the cell, made of beryllium ceramics, 50 cm long and 2 cm in diameter, can be heated up to 1400°C with an electric furnace. In order to keep metal vapor in the cell, it was filled with neon up to pressure of 10 kPa.

Radiation parameters of the pump beam were recorded using the portion of radiation reflected from the input window of the SRS cell. Concurrent SRS radiation and pump radiation were separated with the diffraction grating 16 and then recorded. Parameters of backward SRS radiation were recorded once it has passed through the semitransparent (for $\lambda = 458$ nm) mirror 14. The shape of radiation pulses was recorded using FEK 22–SPU photodiodes and a 6LOR–04 oscilloscope, the energy was measured with an IMO–2N power and energy meter, and spectral characteristics were recorded with an IT–28–30 interferometer. Radiation energy distribution in the far zone was found using a lens (F = 3 m) and calibrated diaphragms.

As was shown in Ref. 5 as well as in our previous work,⁶ the efficiency of XeCl laser radiation conversion with SRS in lead vapor increases sharply when IS mode is used. However, it was unclear why it happens, since in the IS mode the width of pump line decreases but its intensity increases due to lower divergence. In our experiments the beam divergence was practically constant for the mode of free generation (without MO) and the IS mode, and about 60% of energy was within the diffraction angle. Thus, it was possible to observe SRS radiation with the parameters different in the

values of the linewidth of the pump radiation, with its intensity remaining unchanged. Measurements of the energy of SRS radiation showed that in the IS mode with the linewidth of 0.01 and 0.1 cm⁻¹ the efficiency of radiation conversion at an optimal temperature was approximately twice as large as that in the free generation mode.

Figure 2 presents the interferograms of pump radiation for two values of linewidth (0.01 and 0.1 cm⁻ 1) and the corresponding interferograms of SRS radiation. In both cases the broadening of the spectral line of converted radiation was observed. Thus in the case the linewidth at $\lambda = 458$ nm first was $0.05{-}0.06\;\mathrm{cm^{-1}}$ that, according to our estimates, is close to the value of Doppler broadening equal to 0.045 cm^{-1} . In the second case the line broadening up to 0.2 cm^{-1} was likely due to the Doppler effect too. Variation in the buffer gas pressure from 2 to 70 kPa had no effect on the SRS radiation linewidth, that allows us to speak about insignificant role of the collisional broadening. For the pump beam with the linewidth of 0.01 cm⁻¹, the backward SRS was observed In this case the linewidth of the Stokes signal was practically the same as in the forward SRS.

Monitoring of the divergence of the pump beam passed through the cell showed that it depends on the metal vapor density. Thus, at temperature of 850°C, that corresponded to the SRS beginning, the divergence of the pump beam having passed through the cell changed insignificantly. However, with the further increase in temperature, its marked worsening was observed (see Fig. 3). In addition, the size of the pump beam at the output of the cell increased by three to four times and its shape transformed from rectangular to oval.

As follows from Fig. 3, where the angular distribution of SRS radiation is presented as well, the divergence of the radiation converted does not follow that of the pump beam: the diffraction core disappears, and 60% energy is confined within the angle of $4 \cdot 10^{-4}$ rad which is four times greater than the diffraction angle.

Shown in Fig. 4 are the waveforms of pulses of the narrowband pump pulse radiation, backward and forward SRS radiation. As is seen from the figure, SRS radiation is a set of short pulses with different lengths, that most likely is connected with the effect of SRS process saturation due to small concentration of metal vapor in the cell.

Additional study of SRS in barium vapor has shown that the radiation converted consists of a set of short pulses too, and the values of divergence and linewidth of SRS radiation are comparable with those measured in lead vapor. Thus, in this paper we have shown that the efficiency of XeCl-laser radiation conversion with SRS in lead vapor is independent of the pump radiation linewidth unless it exceeds the value of Doppler broadening. The latter one determines the minimum possible spectral width of scattered radiation. When the pump radiation linewidth exceeds the value of Doppler broadening the conversion efficiency decreases. The divergence of the radiation converted far exceeds that of the pump radiation if determined by the amount of energy confined within the diffraction core, however it is one order of magnitude less than the geometric one determined by the shape of caustic of the beam focused.



FIG. 2. Fragments of interferograms of the pump beam with the linewidth of 0.01 (a) and 0.1 cm⁻¹ (b) and the corresponding SRS radiation (c and d). The etalon free dispersion region was 0.07 (a) and 0.25 cm⁻¹ (b, c, and d).



FIG. 3. Angular distribution of the pump beam energy passed through the cold cell (1), the cell with metal vapor at T = 850 (2) and 1300°C (3) as well as the SRS radiation (4); j denotes the portion of the radiation energy within the corresponding angle Q with respect to the total energy, Q_d denotes the diffraction angle.



FIG. 4. Waveforms of radiation pulses of the incident pump beam (a) and that passed through the cell (b) as well as forward (c) and backward (d) SRS radiation. The period of calibration was 20 ns.

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