THREE-PHOTON LIGHT SCATTERING IN DIELECTRIC CRYSTALS EXCITED WITH RADIATION OF A COPPER-VAPOR LASER

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1. Introduction

The specific properties of laser radiation make it possible to observe a number of nonlinear optical effects in crystals, in particular, three-photon light scattering. An example of such effects are hyper-Rayleigh and hyper-Raman scattering of light. Hyper-Raman scattering (HRS) is observed in the region of the second optical harmonic and is characterized by the frequency \( \omega_s = 2\omega_l \pm \Omega \), where \( \omega_l \) is the excitation wavelength and \( \Omega \) is one of the possible frequencies of long-wavelength vibrations of a crystal lattice \([1]\). Hyper-Rayleigh scattering is observed exactly at the frequency of the second optical harmonic. According to the phenomenological theory \([2]\), the second optical harmonic may be observed not only in transmitted light, but in light reflected from a crystal surface as well. In \([3]\), this effect was observed for GaAs, InSb, Te, and other crystals. It is clear from general considerations that in addition to the nonlinear signal reflected from a crystal surface at a frequency of \( 2\omega_l \), one may observe the reflection signal at the frequency \( \omega_s = 2\omega_l \pm \Omega \) as well. It is natural to call this phenomenon the hyper-Raman reflection of light.

The interest in effects of this kind is due to the fact that the study of nonlinear reflection of light gives valuable information on the properties of a crystal surface. In particular, HRS makes it possible to observe those modes of crystal lattices which are forbidden for ordinary Raman scattering (RS) and study lattice modes without the use of double and triple monochromators.

2. Experimental Technique

In this work, HRS and hyper-Rayleigh scattering were excited with radiation of a copper-vapor laser (\( \lambda = 0.5106 \) and 0.5782 \( \mu \)m). The laser operated in a quasi-CW regime with a pulse repetition rate of about 8 kHz and produced pulses of 20 ns duration with a peak power of 20 kW. The average laser power reached several watts in each of the two spectral lines. To decrease the angular divergence of laser radiation, we used an unstable resonator of the telescopic type \([4]\). It should be noted that in view of the fact that the intensity of scattered light is proportional to \( 1/\lambda^4 \), the copper-vapor laser must be more efficient for HRS excitation than the yttrium-aluminum garnet laser (\( \lambda = 1.064 \) \( \mu \)m) with the same values of peak and average power. Moreover, the copper-vapor laser seems to be promising for studying HRS in crystals with absorption in the near-UV spectral region. This is due to the fact that the HRS cross-section increases in the region of resonant absorption.

HRS and hyper-Rayleigh scattering in crystals were observed in the reflection-type configuration without polarization arrangements. Exciting radiation was focused on a sample into a small-size spot 0.1 mm in diameter. Its minimum size was determined by the threshold of surface laser-induced damage. The depth of
the scattering medium in crystals transparent for scattered light was determined mainly by the length of the waist of the focusing lens and was of the order of $10^{-1}$ cm.

The spectrum of scattered radiation was continuously recorded by using a MDR-2 monochromator and a dc amplifier or a photon-counting system [7]. Scattering was observed in the direction close to the direction of specular reflection of exciting radiation from the crystal surface. In the detection system, exciting laser radiation was suppressed with a UFS-1 absorption filter. The solid angle in which the scattered radiation was collected could be varied with an iris diaphragm placed in front of a condenser lens (Fig. 1) and was set equal to $3 \cdot 10^{-2}$ srad. Spectra were recorded with a resolution of $10^{-50}$ cm$^{-1}$ determined by the width of the entrance monochromator slit. Experiments were carried out at a temperature of 300°C.

3. Experimental Results and Discussion

Here, we present the results of studying HRS in a LiTaO$_3$ crystal in the region of the second optical harmonic at $\lambda = 0.2891$ $\mu$m excited with laser radiation at $\lambda = 0.5782$ $\mu$m (the radiation at $\lambda = 0.5106$ $\mu$m was suppressed with an OS-12 filter) and HRS in the region of the summary frequency ($\lambda = 0.2711$ $\mu$m) excited by a laser operating at two spectral lines ($\lambda = 0.5106$ and 0.5782 $\mu$m). Moreover, we studied three-photon scattering in a KCl crystal in the region of the second optical harmonic ($\lambda = 0.2891$ $\mu$m). The excitation intensity for LiTaO$_3$ and KCl crystals was about $I_t = 10^5$ and $2 \cdot 10^8$ W/cm$^2$, respectively. Hyper-Rayleigh scattering was studied in CaCO$_3$, KCl, SrTiO$_3$, LiNbO$_3$, LiTaO$_3$, and LiI$O_3$ crystals excited with laser radiation at $\lambda = 0.5782$ $\mu$m with intensity of $10^8$ W/cm$^2$.

Figure 2 presents the HRS spectrum of a LiTaO$_3$ crystal in the region of the second optical harmonic. One can see the maximum corresponding to the second harmonic (in Fig. 2, it corresponds to the zero frequency). Moreover, the spectrum contains Stokes (marked by the arrow) and anti-Stokes HRS components which are caused mainly by totally symmetric vibrations of the LiTaO$_3$ crystal lattice. A similar spectrum presented in the left upper corner is recorded with a lower gain and illustrates the relative intensities of spectral lines of the second optical harmonic and the HRS spectrum.

To ensure that the spectrum observed in the experiment was not associated with ordinary RS excited with the second optical harmonic, we performed a special experiment. We used the second optical harmonic...