Observation of interference of direct and cascade processes in active spectroscopy of polaritons

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It is observed in experiment that the interference of three- and four-photon processes leads to deformation of the line shape and to the appearance of a doublet structure in the spectra obtained by active spectroscopy of Raman scattering of light by polaritons. The sign and the magnitude of the effective nonlinear susceptibility of third order $\chi_{\text{eff}}^{(3)} \approx 1.9 \times 10^{-13}$ cgs esu are determined for the LiIO$_3$ crystal.

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In ordinary active spectroscopy of Raman scattering (ASRS) of light, the signal generation goes through an intermediate non-propagating state.[1] In crystals without a symmetry center, however, ASRS is possible also on dipole-active oscillations, which because of the strong coupling with the transverse electromagnetic field are transformed into polaritons whose natural frequencies depend on the magnitude and the direction of the wave vector.[2–4] The generation of the ASRS signal goes therefore in this case through a nonlocal intermediate and the signal depends not only on the difference between the frequencies of the exciting fields, but also on the difference between the wave vectors. Observation of this dependence was called by the De Martini[2] "spectroscopy in k-space."

In crystals without symmetry center, the intensity of the ASRS signal receives contributions both from direct four-photon processes as well as from cascade three-photon processes that go via polariton states. In view of the coherence of the scattering, the different contributions are not additive but interfere, leading generally speaking to a considerable deformation of the ASRS spectra in both ω space and k space.

We shall show that cascade three-photon processes in ASRS of light on polaritons constitute two successive three-photon processes: a) excitation of polaritons of frequency ω_p in the field of two laser beams with frequencies ω_1 and $\omega_2 (\omega_1 - \omega_2 = \omega_p)$, and b) scattering of the sounding wave of frequency ω_1 by
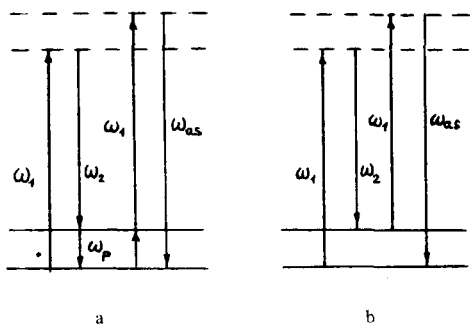
FIG. 1. Scheme illustrating the cascade three-photon process (a) and direct four-photon (b) processes occurring in ASRS spectroscopy on polaritons.

polaritons excited in this manner ($\omega_{as} = \omega_p + \omega_1 = 2\omega_1 - \omega_2$) (Fig. 1a). Each of these processes is due to the quadratic nonlinearity of the crystal $\chi^{(2)}$, and they are most effective when the synchronism conditions $\mathbf{k}_p - (\mathbf{k}_1 - \mathbf{k}_2) \equiv \Delta\mathbf{k}_p = 0$ and $\mathbf{k}_p - (\mathbf{k}_{as} - \mathbf{k}_1) \equiv \Delta\mathbf{k} = 0$, are satisfied, where $\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_p$, and $\mathbf{k}_{as}$ are the corresponding wave vectors. A direct four-photon process constitutes a mixing of the radiation fields of three lasers ($\omega_{as} = 2\omega_1 - \omega_2$), due to the cubic nonlinear susceptibility of the crystal $\chi^{(3)}$ (Fig. 1b). The maximum effectiveness of this process is reached under the condition $\mathbf{k}_{as} - (2\mathbf{k}_1 - \mathbf{k}_2) \equiv \Delta\mathbf{k}_{as} = 0$.

This paper is devoted to the first experimental investigation of the influence of interference effects between direct and cascade processes on the ASRS spectra on polaritons.

The investigation of the spectra of ASRS of light by polaritons was carried out in the crystal $LiIO_3$, the polariton spectra of which had been thoroughly investigated. [5] The polaritons were excited by the second harmonic (frequency ω_1) of an yttrium-aluminum-garnet Q-switched laser operating in the TEM_{00} mode with a repetition frequency up to 20 Hz, and by radiation of a tuneable dye laser (rhodamine-6G, frequency ω_2) with generation line widths 0.2 and 1 cm^{-1} respectively. Their radiation at the frequency $\omega_{as} = 2\omega_1 - \omega_2$ was registered with the aid of an FÉU-79 photomultiplier. The signals from the photomultiplier were integrated and their logarithms were fed to an automatic recorder, the chart of which was moved in synchronism with the variation of the dye-laser wavelength or with the rotation of the investigated crystal. The use of this registration scheme has enabled us to record directly the ASRS spectra in ω and k space.

Figure 2 shows the spectrum of ASRS of light by polaritons in k space, obtained by varying the direction of the wave vectors of the laser emission relative to the optical axis of the crystal; this is accomplished by rotating the investigated sample without changing the geometry of the scattering. In this case the width of the synchronism in k space for direct four-photon processes greatly exceeded the width of the synchronism for three-photon cascade processes. The spectrum was obtained under the condition that the synchronism of centers coincided for both processes.

In ω space, however, the width of the synchronisms for the direct and cascade processes are close in magnitude and therefore the effects of the interference should manifest themselves weakly for the considered processes. [6] To observe the interference effects in ω space we have therefore obtained a spec-
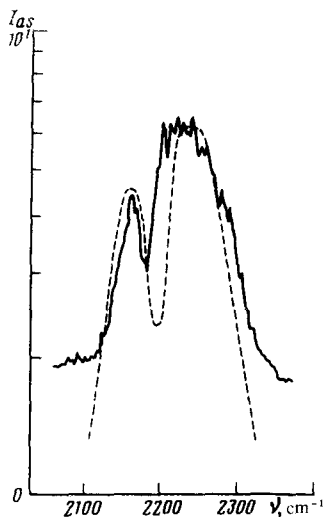
FIG. 2. ASRS spectrum in k space: solid curve—experiment, dashed—results of calculation, θ_L—angle between the wave vector of the radiation of frequency ω_1 and the otpical axis of the crystal. The spectrum was obtained at a fixed value of ν (≈ 2300 cm^{-2}); I_{as} is the intensity of the ASRS signal in relative units, $\nu \equiv \omega/2\pi c = (\omega_1 - \omega_2)/2\pi c$.

trum (Fig. 3) at a negligible mismatch of the synchronism centers (i.e., within the limits of the synchronism width).

The strong asymmetry and the presence of dips in the spectra shown in Figs. 2 and 3 indicate that interference between the direct and cascade processes influence the spectra of ASRS of light by polaritons.

On the basis of the results of the theoretical paper[6] we have also calculated the ASRS spectra. The results of the calculations are shown in the figures by
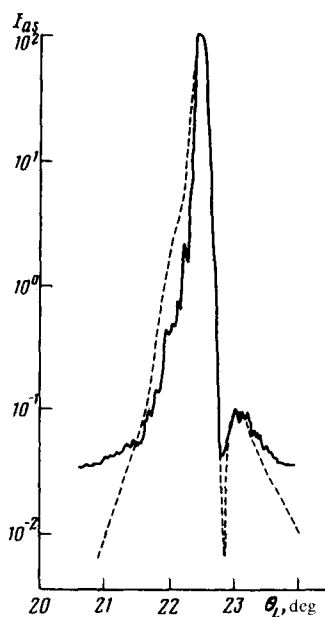

FIG. 3. ASRS spectrum in ω-space: solid curve—experimental spectrum at $\theta_L = 22.5°$, dashed—result of calculation.

dashed curves. We used relations corresponding to the case of weak pumping and weak polariton absorption, conditions which were realized in the experiment. Data on the polariton dispersion and on the refractive indices of the crystal $LiIO_3$ were taken from.[5,7] We note that the form of the spectrum depends substantially on the relative contributions of the direct and cascade processes, which are determined in[6] by the parameter ρ'. It is obvious, for example, that the interference vanishes if one of the scattering mechanisms is eliminated. Therefore the parameter ρ' was chosen such as to obtain the best agreement between the experimental and calculated data. Starting from the value $\rho' \approx 0.15$ obtained in this manner and the known value of the nonlinear second-order constant $d_{31} \approx 1.6 \times 10^{-8}$ cgs esu,[8] we estimated the effective nonlinear third-order susceptibility $\chi_{\text{eff}}^{(3)} = (c_{11}/3) \cos^2\theta_L + c_{16} \sin^2\theta_L$[9]: $2.1 \times c_{11} + c_{16} \approx +7.3 \times 10^2 d_{31}^2 = +1.9 \times 10^{-13}$ cgs esu. We note that since $\chi_{\text{eff}}^{(3)}$ is determined relative to the square of d_{31}, we determined the absolute sign of $\chi_{\text{eff}}^{(3)}$ from the sign of ρ.

In conclusion, we emphasize once more that according to the experimental results, a correct interpretation of the ASRS of light by polaritons calls for allowance for the possible appearance of several maxima, on account of the interference of the three- and four-photon mechanism of formation of the scattered waves.

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