Experimental study of wave-front reversal in stimulated temperature and Mandel'shtam-Brillouin scattering and stimulated temperature scattering in liquids

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The dependences of the reflectances and the degree of wave-front reversal (WFR) of single-mode and multimode beams on the power of the exciting radiation under conditions of lens focusing into a liquid that is active with respect to stimulated Mandel'shtam-Brillouin scattering (SMBS mirror) and to stimulated temperature scattering (STS mirror) are obtained. A WFR effect is observed for the STS mirrors. It is shown that the WFR effect is disturbed for STS mirrors upon increase in the power of the exciting radiation, and that an intense transfer of energy from strong angular modes to weak ones takes place in multimode beams. In this case, the STS mirror does not reverse the wave front but preserves the set of angular pumping modes in the reflected radiation. Possible applications of the STS mirror are discussed.

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From the moment of discovering of the effect of wave-front reversal (WFR),[1] a large number of experimental and theoretical papers have appeared, devoted to the study of the properties of stimulated Mandel'shtam–Brillouin scattering (SMBS mirror) and its applications (see Refs. 2, 3, 4 and the references cited in them). However, quantitative investigations of the dependences of the reflectances and of the degree of WFR on the power of the exciting radiation in the case of a different angular spectrum of the pumping in different media are very few.

From the viewpoint of several applications, greatest interest attaches to observation of the phenomenon of WFR also in stimulated temperature scattering (STS), since the frequency shift in STS is extremely small. So far as we know, WFR in the case of STS had not been previously observed. In experiments on dynamic holography with a thermal recording method[5] the interaction of the light beams bore a four-photon character.

At the present time, the dependences of the reflectances and degree of WFR on the power of the exciting radiation have been obtained experimentally for SMBS and STS mirrors in the focusing-lens regime. The degree of WFR has been defined as the ratio of the power of the exciting radiation for SMBS and STS mirrors under lens focusing conditions. The degree of WFR was defined as the ratio of the power reflected into an angle equal to the angle of divergence of the undistorted exciting radiation to the power reflected into the total aperture of the focusing lens.

Linearly polarized second-harmonic radiation of a neodymium laser was used in the experiments, with the following output parameters: wavelength $\lambda = 0.527\mu$, energy $E \approx 0.3$ *J*, diameter of beam $d \approx 7$ mm, pulse duration at half height, $\tau \approx 50$ nanosec, width of spectrum $\delta\nu \approx 0.01$ cm^{-1}, divergence $\theta \approx 0.5$ mrad.

A diagram of the experimental setup is shown in Fig. 1. The spectral composition of the incident and scattered radiation was monitored in the experiments. For this purpose, part of the incident and part of the scattered radiation were diverted to a Fabry-Perot etalon. The plane of polarization of the scattered radiation was turned by 90° relative to the plane of polarization of the incident radiation with the aid of a quartz plate of thickness 3.2 mm, cut perpendicular to its optic axis. In front of the film, was located a compound analyzer whose halves passed radiation with mutually perpendicular polarizations, so that only the incident radiation passed through one half and only the reflected radiation passed through the other. By means of coaxial photocells FEK-09 and the oscilloscope 6LOR-04 (resolution time of the system ≈1 nanosec) there were recorded the pulse of exciting radiation, the pulse reflected into the total aperture of the lens, and the pulse of radiation reflected into an angle equal to the angle of divergence of the undistorted incident radiation. A spatial filter (objective with focal length 92 cm and diaphragm diameter 0.8 mm in the focal plane) was placed in front of the film that recorded the radiation reflected into the reproducing angle. The photocells were first calibrated against the readings of the calorimeters IKT-1M.

The following liquids were used in the research: acetone, carbon tetrachloride, distilled water, ethyl alcohol, acetic acid, benzene, toluene, and nitrobenzene. A focusing lens with focal length $f = 4$ cm was used in all the experiments. The enumerated liquids could be divided into two classes, according to the type of stimu-

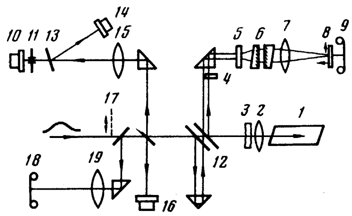

FIG. 1. Diagram of experiment: 1—vessel with SMBS- or STS-active liquid; 2—lens; 3—phase plate; 4—quartz plate; 5—frosted glass; 6—Fabry-Perot etalon; 7, 15, 19—objectives with $f = 92$ cm; 8—polarization analyzer; 9, 18—photographic apparatus; 10, 14, 16—coaxial photocells; 11—diaphragm; 12, 13—beam-splitting plates; 17—screen.

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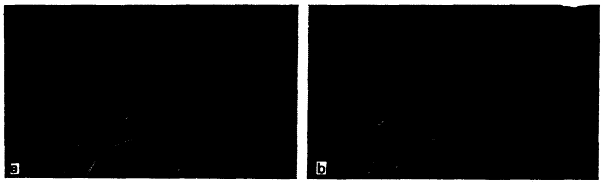

FIG. 2. Spectrograms of exciting (left) and scattered (right) radiations; a) SMBS scattering; b) STS scattering (free spectral range of the Fabry-Perot etalon, 0.5 cm^{-1}).

lated scattering that was observed in our experiments. Thus, a shift of the frequency of the scattered radiation, of magnitude corresponding to 180° SMBS, was observed in all these liquids except toluene and nitrobenzene [see Fig. 2(a)]. Two of the measured liquids, toluene and nitrobenzene, behaved like STS mirrors when the exciting radiation was focused in them. In the limits of resolving power of the Fabry-Perot etalon used, the no shift in the frequency of the radiation scattered by these liquids could be recorded [see Fig. 2(b)]. Inasmuch as even the direction of the frequency shift of the scattered radiation is unknown, the answer to the question as to whether this scattering is STS-I (electrocaloric effect) or STS-II (an effect due to light absorption) cannot be obtained from analysis of the spectrograms. It is impossible to exclude the hypothesis on the STS-1 mechanism, since especially pure liquids were used, although additional distillation was not carried out. Some arguments concerning the scattering mechanism are given at the end of the paper.

Under the conditions of our experiments, the time of establishment of the STS was determined by the width of the pump line $\delta\nu \approx 10^{-2}$ cm^{-1}, which appreciably exceeds the natural line width of the thermal scattering, $\delta\Omega = \chi q^2 \approx 5 \cdot 10^{-4}$ cm^{-1} (χ is the coefficient of temperature conductivity, $q = k_1 - k_2$ is the difference in the wave vectors of the incident and back-scattered radiation). Therefore the duration of the pump pulse ~50 nanosec is much greater than the time of establishment ~3 nanosec and the scattering took place in a stationary regime.

Figure 3 shows typical oscillograms of pulses reflected by SMBS and STS mirrors. In the oscillograms: 1—pulse of exciting radiation, 2—pulse of radiation reflected from an SMBS or STS mirror into the total aperture of the lens, 3—pulse reflected into an angle equal to the angle of divergence of the undistorted incident radiation. Pulses 1, 2, and 3 were obtained in the same experiment, and all the pulses were reduced to a single scale. In Figs. 3(a) and 3(b), typical results are given of experiments on SMBS in acetone. Similar shapes of pulses were obtained also for the other active SMBS liquids mentioned above.

In the analysis of the physical mechanisms leading to the observed modulation of the reflected component, it is obviously significant that the intensity distribution of the incident radiation at the focal spot of the focusing lens is strongly inhomogeneous. The inhomogeneities in the beam arise after doubling of the frequency in a KDP crystal. In the development of the stimulated radiation from the various "hot spots" with different phases, the appearance of oscillations of intensity of the Stokes

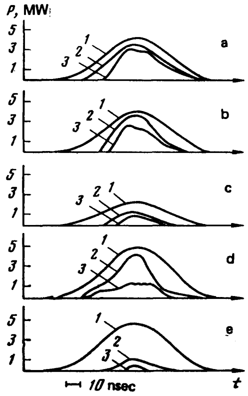

FIG. 3. Oscillograms of pulses reflected by SMBS and STS mirrors: a) SMBS mirror without phase plate, acetone, divergence of the exciting radiation $\theta = 0.5$ mrad; b) SMBS mirror with phase plate, acetone, $\theta = 10$ mrad; c) STS mirror without phase plate, toluene, $\theta = 0.5$ mrad; regime without transfer of energy from the axial mode to weak angular modes; d) STS mirror without phase plate, toluene, $\theta = 0.5$ mrad, regime with transfer of energy from axial mode to angular modes; e) STS mirror with phase plate, toluene, $\theta = 10$ mrad, regime with reversal of wave front.

wave is possible, due to the beating of the Stokes beams with the uncorrelated phases that fluctuate in time. Correlation in the phases of the priming Stokes waves is absent if the sources are separated by at least the sound-attenuation length. Under our experimental conditions, the latter condition was satisfied in order of magnitude.

As is seen from the obtained oscillograms, the pulses reflected by the STS mirror have a shape that is similar to that of pulses reflected by the SMBS mirror. In the presence of a phase plate, steepening of the leading edge is observed and a shortening of the pulse duration. However, the thresholds for the STS mirror are somewhat higher and the reflection coefficients lower. A feature of the STS mirror is the increase in the threshold and a decrease in the reflection coefficient after a large number of pulses of irradiation of the liquid, until the reflection of the STS vanishes completely. An SMBS line was sometimes observed in back scattering from such a "spent" liquid.

Figure 4 shows the dependences of the intensity re-

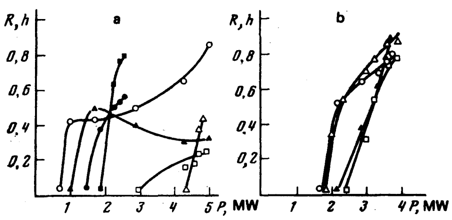

FIG. 4. Dependence of the reflection coefficients in the complete lens aperture (R) and degree of WHO (h) of STS and SMBS mirrors on the power of the incident radiation. a) STS mirror: ●, □, ○ correspond to R and $\theta = 0.5$, 10, 0.5 mrad (regime with transfer): ■, △, ▲ correspond to h and $\theta = 0.5$, 10, 0.5 mrad (regime with transfer); b) SMBS mirror: ○, △ correspond to R and $\theta = 0.5$, 10 mrad; ▲, □ correspond to h and $\theta = 0.5$ and 10 mrad.

flection coefficients (R) of SMBS and STS mirrors and of the degree of WFR (h) for single-mode and multimode pump beams on the power of the exciting radiation. The multimode pump beam was produced by means of a phase plate which uniformly widened the angular spectrum up to ~10 mrad at half intensity.

In reflection from the STS mirror, a selection of angular modes in the focal region takes place in a system with focusing in the case of small intensities. This selection is analogous to that observed earlier in the case of SMBS.[6] However, at high powers of pumping, the phenomenon of transfer of energy from strong to weak angular modes is observed. (The effect of redistribution of the intensity of light beams in alcohol with addition of a dye, with the help of reflection dynamic holograms, employing a thermal method of recording, was observed in the work of Dukhovnyĭ and Stasel'ko[7]). When several plane waves of different intensities, traveling at small angles with respect to one another, were focused in an STS liquid, it was observed that the strong angular mode is weakened in the reflected radiation, while the weak modes are materially strengthened. In our experiments, it was necessary for the appearance of the effect that the zone in which the beams intersect be not far from the focal constriction of the lens, i.e., in the zone of intersection of the beams there should be a high density of pump power. For this, the geometry of the experiments was so chosen that the angular modes did not manage to diverge spatially ahead of the focusing lens, and the lenses that were used were of short focal length. Such a transfer of the energy from the strong angular mode to the weak modes was observed not only in back reflection of the radiation, but also in passage of radiation through the STS liquid, even when, after many irradiations, this liquid ceased to operate as an STS mirror.

The effect of transfer of the energy from the strong angular mode to the weak one was observed qualitatively in the case of photography of the Fourier spectrum of a multimode pump beam passing through an STS liquid or reflected back. The scheme of observation was the following. A rectangular grating, prepared by photoetching, was placed in the pump beam (see Fig. 1). The grating period was 250 μ, the dimensions of the cell 220×220 μ. The plane pump wave, passing through the grating, was split into a system of plane waves. The angular separation of the modes amounted to ~2.5 mrad. The intensity in the successive angular modes fell off as 1 : 0.034 : 0.024 : 0.015 : 0.0044 : 0.0015. Figure 5 shows the Fourier spectra of the pump in two regimes. The regime with transfer corresponds to Fig. 3(d), and, in Fig. 4(a), to the curves denoted by open circles and black triangles. The observed transfer of energy can be explained by the mechanism described previously.[5]

Just as in SMBS mirror, there exists a WFR effect for the STS mirror. This property of the STS mirror is demonstrated by Fig. 6. It follows from the drawing that, after the second passage through the phase plate, the angular spectrum of the incident radiation is restored to a significant degree. Preservation of the ordered structure in the near zone along with the picture of the further zone (Fig. 6) leads to a conclusion that the STS mirror has reversing properties.

Still another feature of the STS mirror was observed. Modulation of the intensity was observed in the pulse of radiation reflected by the STS mirror into an angle equal to the angle of divergence of the undistorted radiation. This modulation had a characteristic time $t_0 \sim (\delta\nu)^{-1}$, where $\delta\nu$ is the width of the spectrum of the exciting radiation [see Fig. 3(d), curve 3]. One can attempt to explain this effect in the following way. Under the conditions of our experiments, the time of the thermal-grating spreading due to the thermal conductivity, is much greater than the pulse length of the exciting radiation and, hence, greater than the reciprocal width of the spectrum. If the scattering mechanism is STS-II, i.e., formation of the thermal grating takes place because of light absorption, then when the time of switching-on of the amplitude of the heating wave is sufficiently fast in comparison with $(\delta\nu)^{-1}$, a standing thermal grating is formed in the medium. The source of the heating is the slowly moving wave formed as a result of interference of the incident and reflected waves (the latter shifted by the width of the spectrum). As each instant of time, the standing thermal lattice, which was formed at the initial instant of time ($t \leq (\delta\nu)^{-1}$) interferes with the thermal grating which is formed by the moving heating wave at the present instant of time. The contrast of the traveling thermal grating from which the reflection occurs then changes periodically. Estimates show that the change in contrast of the traveling thermal lattice at $\tau = (1-2)t_0$ (where τ is the time of switching on of the heaving wave, $t_0 = \Lambda/v$ is the time of smearing out the lattice, Λ is the thermal-grating wavelength, and v is the heating-wave velocity) can amount to 5–10%, which in turn produces in the reflection coefficient of the traveling thermal grating a relative change $\Delta R/R \approx 2\delta n/\Delta n$ with an amplitude of 10–20%; here Δn is the amplitude of the change in the index of refraction in the

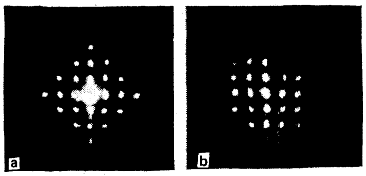

FIG. 5. Fourier spectra of radiation of a multimode beam, reflected by an STS mirror in two regimes: a) regime without transfer of energy from a strong angular mode to weak; b) regime with transfer of the energy.

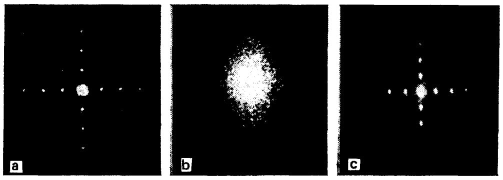

FIG. 6. Fourier spectra of radiation of multimode beam reflected by a) ordinary mirror without phase plate; b) mirror, in front of which a phase plate has been placed; c) STS mirror with the same phase plate.

traveling thermal grating and δn is the change in contrast due to interference of the standing and traveling thermal gratings.

A correlation was noted between the period and depth of modulation of the Stokes wave. The depth increases with increase in the period. In the case of constant time of switching on, this can be explained by the change in the pump spectrum. Of course, additional experimental investigations are needed to verify this explanation. This includes studies of the spectra of the incident and scattered radiation. Nevertheless, in the case of STS of sufficiently short pump pulses, it is possible in principle to distinguish STS-I from STS-II from the shape of the Stokes pulses and to draw conclusions on the width of the pump spectrum. The presence of a characteristic modulation of the intensity of the scattered radiation, and the correlation of the period with the modulation depth in the present experiment, conform to the mechanism described above.

The STS mirror can find application in those cases in which the required shift of frequency in the reflection with reversal of the wave front is minimal. For example, in the scheme of reversal of the wave front in resonant four-photon interaction,[8] the STS mirror can replace the ordinary mirror which requires careful alignment, and the SMBS mirror can also find application in well known systems for decoupling amplifying stages in laser systems with a narrow amplification band, where the frequency shift of the SMBS leads the radiation out of the amplification band.

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Autoionizing band of electron–exciton complexes and their role in reflection of slow electrons from the surface of a solid

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The model of multichannel small-radius interactions is used to obtain an analytical solution of the problem of electron reflection from the surface of a two-dimensional ordered lattice of two-level atoms. The positions of singularities of the reflection coefficient on a complex energy plane are used to reconstruct the energy spectrum of electron–exciton states. The conditions for the existence of autoionizing bands of electron–exciton complexes are determined and it is shown that in the presence of such bands the energy and angular dependences of the elastic reflection coefficient have characteristics Fano-Feshbach singularities. Numerical calculations are carried out for a monolayer of Xe atoms adsorbed physically on the surface of niobium.

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1. INTRODUCTION

In the last 10–15 years the experiments on low-energy electron diffraction have provided direct confirmation that monolayers of adsorbed atomic particles form regular structures (two- and one-dimensional crystals) at sufficiently low temperatures.[1,2] The properties of periodic low-dimensional structures are not identical with the properties of bulk crystals composed of the same atoms. Investigations of the geometry and spectra of ordered adsorbed monolayers are among the most interesting current topics in the physics of surface phenomena. Systems of this kind may exhibit, in particular, collective states of basically

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