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Observation of stimulated thermal scattering of light upon nonsteady-state interaction between a laser pulse and a medium

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Abstract. **The induced thermal scattering of light has been experimentally investigated for short laser pulses of different durations. It is shown that the observed anomalously large anti-Stokes shift ($\Omega \gg 1/(2\pi\tau)$, where τ is the thermal grating relaxation time) in the reflected signal depends on the pump pulse duration.**

Keywords: *stimulated thermal scattering of light, large anti-Stokes shift.*

1. Introduction

Nonsteady-state interaction between a laser pulse and a medium occurs when the pulse duration τ_p is comparable with the relaxation time of the thermal grating written by this pulse in the medium: $\tau = 1/(\chi q^2)$, where χ is the thermal diffusivity of the medium and q is the grating wave vector. A series of studies have been published recently by He et al. [1–3], where they ascertained that observed a new type of stimulated light scattering, which was referred to as stimulated Mie scattering (SMS) by them. The conditions under which this scattering was observed coincide with the conventional conditions for stimulated thermal scattering (STS) of light. The main arguments in favour of the classification of the scattering observed in [1–3] as a non-STS one are as follows:

(i) Under conditions of STS of broadband pump radiation with a linewidth $\Delta\omega_p \gg 1/\tau$, one should observe an anti-Stokes shift of the scattered wave by $\Omega \approx \Delta\omega_p/2$.

(ii) Under conditions of STS of broadband pump radiation, the gain increment for the anti-Stokes component of scattered wave g (and, correspondingly, the threshold pump power) should be inversely proportional to the pump linewidth $\Delta\omega_p$: $g \sim 1/\Delta\omega_p$.

(iii) In the case of STS of light, counterpropagating waves with equal frequencies should not interact, whereas, according to [1–3], this interaction was experimentally observed.

(iv) As was observed in [1–3], the scattered radiation spectrum reproduces the pump spectrum.

The authors of [1–3] adopted the conclusions about the validity of the first two conditions from [4, 5]. Herman and Gray [4] were the first to make these erroneous statements. By analogy with classical linear spectroscopy, they stated that the total spectral response of this process should be a convolution of the STS spectral response and the spectral profile of the excitation laser beam. Then the anti-Stokes shift of the spectral line of scattered radiation under STS conditions should be $\Omega \approx {}^1/_2[\Delta\omega_p + 1/(\pi\tau)]$. Indeed, the validity of this formula was as though confirmed in the first experiments on observation of the STS of a short laser pulse [5].

The statement [4] about the presence of convolution in this process was an automatic extraction of this concept from linear spectroscopy, where it is undoubtedly valid. However, as was shown by us in 1969, the threshold of stimulated Raman scattering (SRS) is independent of the excitation linewidth in very wide limits [6]. Thus, the processes of stimulated light scattering are not described by the convolution of the corresponding spectral profiles, as was also demonstrated in a number of subsequent studies (see, for example, [7]). In our opinion, the conclusions drawn in [5] are based on a random coincidence, because the observed shift of spectral line is comparable with the instrumental resolution and, therefore, is determined with a large error.

That fact that no spectral shift of scattered radiation was observed by He et al. [1–3] has a very simple explanation. Since they used very broadband laser sources and low-resolution spectral equipment, they could not observe in principle the spectral shift occurring in this process. Indeed, our experiments [8], performed under the same conditions as in [1–3] but with a narrow-band laser source having a linewidth $\Delta\omega_p \leqslant 1/\tau$, showed typical laser beam STS but with one unique feature. The spectral shift Ω of the scattered component exceeded by almost an order of magnitude the value $1/\tau$, which stems from the steady-state theory. We showed that the experimental value Ω is on the order of 100 MHz, i.e., is much larger than $1/(2\pi\tau) \approx 16$ MHz for the toluene used by us and the excitation wavelength of 532 nm.

Concerning the third argument adduced by the authors of [1–3] (that the scattering observed by them was non-STS), it is valid for only the steady-state regime of wave interaction. As was shown by us in [9], under conditions of shock excitation of a medium by counterpropagating short laser pulses with equal frequencies, they efficiently interact even on a Brillouin nonlinearity. In this case, the scattered-component frequency shift (which is of great importance for this process) under the SBS conditions significantly exceeds the frequency shift occurring during STS. Hence, counterpropagating short laser pulses (even those with equal frequencies) nicely interact during STS in the nonsteady-state regime.

In regards to the fourth condition (the scattered radiation spectrum reproduces the pump spectrum), it in fact directly

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disproves the presence of convolution in the process under consideration. Convolution always distorts the true line profile; therefore, a profile obtained as a result of convolution in classical linear spectroscopy is a Voigt one.

As was mentioned above, we observed the STS of a laser beam with an anomalously large anti-Stokes shift of scattered component in [8]. The purpose of the present study was to reveal the physical nature of this phenomenon. It was theoretically shown in [8] that, in the case of quasi-steady-state scattering of broadband pump radiation (when $\Delta\omega_{\rm p} \gg 1/(2\pi\tau)$), the frequency shift is independent of the pump spectrum width and is equal to that observed under monochromatic pumping conditions. Note that Faris et al. [10] recorded a profile of a gain line caused by acoustic and thermal fluctuations of medium under steady-state conditions of wave interaction. They obtained excellent agreement with the steady-state theory, having shown that the maximum gain for an external signal under the STS conditions is implemented at the anti-Stokes frequency $\Omega = 1/(2\pi\tau) = \chi q^2/(2\pi)$. The significant difference of our experimental conditions [8] from those of [10] is that the duration $\tau_{\rm p}$ of the excitation laser pulse used by us is comparable with the relaxation time of the corresponding thermal grating ($\tau = 1/(\chi q^2)$) for the toluene we used. In other words, the STS under our experimental conditions occurs in the essentially nonsteady-state regime of wave interaction, and the gain profile, being spectrally asymmetric, has a form of a dispersion curve. In our opinion, specifically this is the physical reason for the anomalous frequency shift of scattered component observed us under conditions of STS of short laser pulses.

To verify this hypothesis, we measured the frequency shift of the scattered component under STS of single-frequency excitation laser pulses of different durations.

A schematic of the experimental setup is shown in Fig. 1. Here, P is the second-harmonic radiation of a single-mode single-frequency passively Q-switched neodymium laser. The laser pulse duration was 40 ns. Frequency conversion was implemented in a caesium dihydroarsenate (CDA) crystal 10×10×40 mm in size, placed in a sealed capsule equipped with protective glasses. A converter based on an LTI-401 laser was used. After passing through thick glass plate (*1*), radiation P was focused by lens (*2*) into cell (*3*) filled with an active medium (pure toluene). The focal length of lens (*2*) was 3 or 15 cm. Lens (*4*) with a focal length f = 5 cm was positioned confocally with lens (*2*). The profile of the pump pulse transmitted through the cell was recorded by photodiode (*5*), and the pulse energy was measured by calorimeter (*7*). The energy of pump radiation P was measured by calorimeter (*8*), and the pulse profile was recorded by photodiode (*10*). The energy of the reflected (backscattered) radiation S was measured by calorimeter (*21*), and the pulse profile was recorded by photodiode (*19*). The pump radiation P and scattered radiation S were directed by mirrors (*11*), (*12*), and (*18*) to lens (*14*) (f = 35 cm), with their cross sections cut off by half at the edge of opaque thin screen (*13*). The screen edge was located in the focus of lens (*14*). In this spectral device, consisting of screen (*13*), lens (*14*), Fabry–Perot etalon (*15*) (length d = 9 cm, dispersion range $\Delta\nu = 1/(2d)$ = 0.0556 cm^{-1}), and objective (*16*) (f = 43 cm), a spectral pattern was formed in the objective focus. One half of the pattern contained the pump radiation spectrum (a semicircle), and the other half contained the scattered radiation spectrum. CCD array (*17*) (WinCamD-UCM, Data Ray), located in the focus of objective (*16*), recorded a spectral pattern, which was observed and measured on a computer. The pulse time parameters were recorded by high-speed S9055-01 photodiodes with a transmission band of 2 GHz on a four-channel GDS73504A oscilloscope (GW Instek) with a bandwidth of 500 MHz.

Pure toluene does not absorb the second-harmonic radiation of the neodymium laser with a wavelength of 532 nm but has a nonzero two-photon absorption coefficient at this wavelength. Hence, depending on the excitation conditions, we could record either SBS or STS. SBS is observed when the excitation beam is focused into a cell with an active medium by the lens with $f \geqslant$ 15 cm. STS is observed when focusing the pump radiation by the lens with $f \approx$ 3–5 cm, a case where the excitation intensity in the focal region is so high that the two-photon absorption becomes dominant.

The experiments were performed using excitation pulses of two different shapes and durations (Fig. 2). The first pulse had smooth leading and trailing edges and the FWHM value

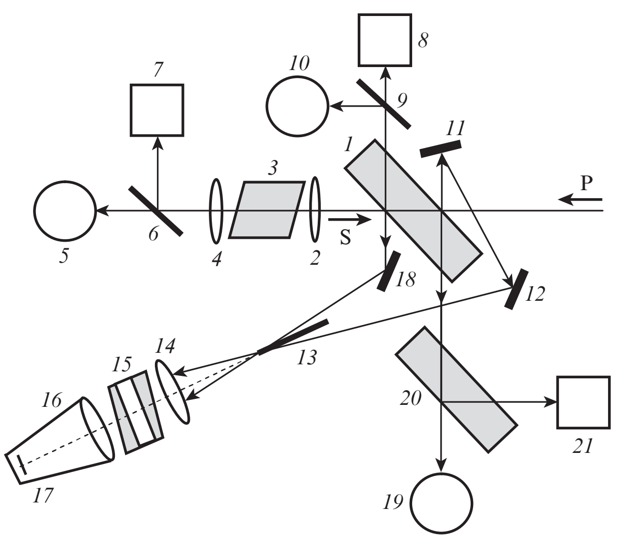

Figure 1. Schematic of the experimental setup:
(*1*, *20*) thick glass plates; (*2*, *4*, *14*) lenses; (*3*) cell with toluene; (*5*, *10*, *19*) photodiodes; (*6*, *9*) thin glass plates; (*7*, *8*, *21*) calorimeters; (*11*, *12*, *18*) highly reflecting mirrors; (*13*) screen; (*15*) Fabry–Perot etalon; (*16*) objective; (*17*) CCD array.

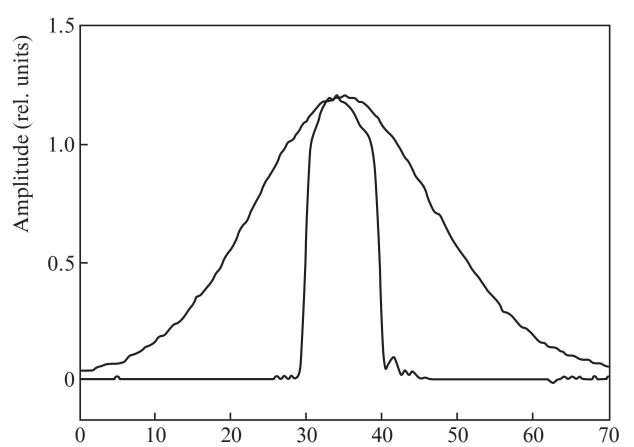

Figure 2. Profiles of pump pulses with durations of 30 and 10 ns.

$\tau_p \approx 30$ ns. This pulse is the result of second-harmonic conversion of a pulse of a Nd : glass master oscillator with a duration of ~40 ns. The second pulse had a duration $\tau_p \approx 10$ ns. This pulse was cut off from the master oscillator pulse using an electro-optic Pockels cell shutter. Under our STS experimental conditions, the spectral shift of the anti-Stokes component of scattered radiation only slightly exceeded the resolution of the Fabry–Perot etalon in use. Therefore, a special computer programme was developed for processing interference patterns; this programme will be described in a separate paper.

The experimental results for pulses with durations $\tau_p \approx 30$ ns and $\tau_p \approx 10$ ns are presented in Fig. 3 (top and bottom rows, respectively). Note that the experiments were performed at a four- to fivefold excess of excitation pulse power above the threshold of the effect observed. A computer processing of the interference patterns showed that, for the excitation pulses with durations $\tau_p \approx 30$ ns and $\tau_p \approx 10$ ns, the anti-Stokes shift of the spectral component of the STS laser radiation is $\Omega = 50 \pm 20$ and 100 ± 20 MHz, respectively. The presence of the second maximum in the scattering spectrum for the 10-ns pump pulse is most likely due to the presence of SBS Stokes component, whose shift greatly exceeds the Fabry–Perot etalon dispersion range.

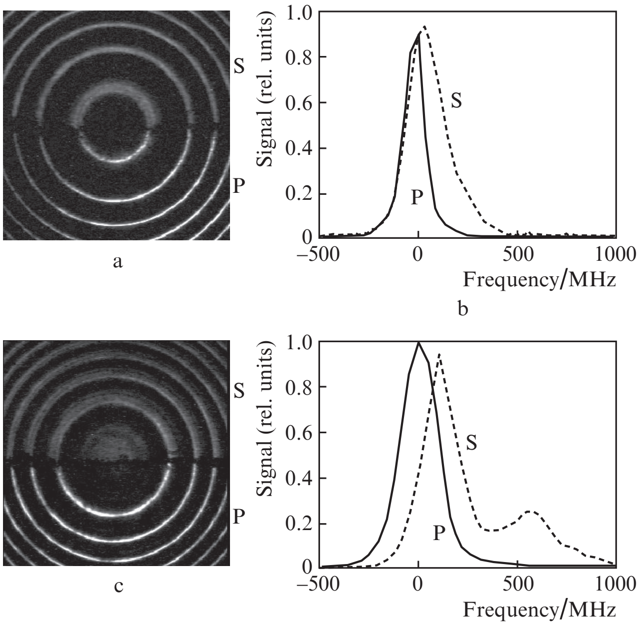

Figure 3. (a, c) Typical interference patterns of pump radiation P and reflected signal S and (b, d) the results of computer processing of interference patterns for excitation pulses with durations of (a, b) 30 and (c, d) 10 ns. Zero frequency in the plots corresponds to the maximum in the pump spectrum. The results are reported for one pattern period.

Thus, the results of our experiments showed that, in the case of STS of short single-frequency laser pulses with a duration comparable with the relaxation time of the corresponding thermal grating, one can observe an anomalously large anti-Stokes shift of the scattered laser beam component, which multiply exceeds the shift caused by the spectral response of the thermal grating for toluene (16 MHz), and depends on the excitation pulse duration.

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