Accuracy of reproduction of the time structure of the exciting radiation in stimulated scattering of light

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A statistical investigation is carried out of the characteristic fluctuation times of the Stokes radiation in stimulated Brillouin scattering for various relative values of the width $\Delta\nu_p$ of the pumping spectrum and of the width of the spontaneous scattering line $\Delta\nu_l$. A double-beam interferometer with nonlinear optical mirrors that invert the wave front is employed in the measurements. It is shown that at $\Delta\nu_p \sim \Delta\nu_l$ the characteristic phase fluctuation times are shorter than in the cases of monochromatic or broad-band pumping. On the basis of the results one can estimate the accuracy with which the Stokes component reproduces the time structure of the exciting radiation of arbitrary spectral width.

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1. INTRODUCTION

Investigations of stimulated scattering of light have made it possible to establish many common regularities of nonlinear interaction of waves and have revealed, in particular, the possibility of reproducing the temporal structure of nonmonochromatic exciting radiation.[1] This process was investigated in detail both theoretically and experimentally (see, e.g., Ref. 2), and a corresponding physical mechanism was established as a result.[3] However, an estimate of the accuracy with which this was done was insufficient, since the estimate was based on a comparison of two broad spectra. A similar problem concerning the resultant accuracy and its connection with the characteristics of the nonlinear medium arises also when stimulated scattering is excited by monochromatic radiation. The solution of this problem is of both scientific significance (since it offers evidence of a thorough understanding of the mechanisms of nonlinear interaction of waves in stimulated scattering) and of practical interest. The point is that stimulated scattering of light has become a powerful method of obtaining coherent radiation in new spectral bands,[4] and in many applied problems it is important to retain high coherence of the initial radiation. The question of reconstructing, with the aid of Stokes radiation, the temporal structure of the pumping radiation plays an important role also in complicated laser systems based on the effect of inversion of the wave front in stimulated Brillouin scattering (SBS). Thus, for example, the temporal structure of the Stokes waves determines the efficiency of coherent summation of radiation of several laser amplifiers,[5] especially if the initiating radiation is nonmonochromatic; this situation is typical of most solid-state periodic-pulsed lasers.

We have developed a special procedure that makes it possible to compare with good accuracy the temporal structures of the scattered and exciting radiation. The preliminary results of the study, for monochromatic pumping, were published in Ref. 6. We describe below the complete results of the investigation of fluctuations of the exciting and scattered radiation at an arbitrary width of the pump spectrum and for nonlinear media with different spontaneous-scattering line widths.

In the theoretical analysis of SBS, the field of the Stokes wave is represented in the form[7]

$$E_c(t) \propto E_p(t) F(t), \tag{1}$$

where $E_s(t)$ and $E_p(t)$ are respectively the amplitudes of the Stokes signal and pump, while $F(t)$ is a certain function of time, the specific form of which, in the given-pump-field approximation, is determined by the following expression[7]:

$$F(t) = \int_0^\infty d\theta \int_0^l dz e^{-\theta/T_2} N(t-\theta, l-z) \times I_0\{[(2g\bar{I}_p z/T_2)\theta[1+\xi(t;\theta)]]^{1/2}\}, \tag{2}$$

where

$$\xi(t,\theta) = \frac{1}{\theta \bar{I}_p} \int_0^\theta I_p(t-\theta_1) d\theta_1. \tag{3}$$

Here $N(t,z)$ is the noise field, g is the gain of the active medium, T_2 is the lifetime of the acoustic oscillations, $\bar{I}_p \sim \tilde{I}_p = |E_p(t)|^2$ is the pump intensity represented as a sum of a constant and an alternating component, I_0 is a modified Bessel function, and l is the length of the active volume of the scattering medium.

In the saturation regime, which is typical of the conditions of observation of stimulated scattering, and in the case of monochromatic pumping, it was shown theoretically and experimentally[6,8,9] that the dependence of the function F on the time has the character of phase modulation:

$$F(t) \propto e^{i\varphi(t)}, \tag{4}$$

while the characteristic time of the phase change corresponds to that obtained in (2) and (3). When the SBS threshold is considerably exceeded (in our experiment, by 3–4 times) we have $|E_s(t)|^2 \approx |E_p(t)|^2$ and consequently the conditions (1) and (4) should be satisfied also for nonmonochromatic exciting radiation.

Thus, expressions (1)–(4) determine completely the time structure of the scattered field, meaning that its correlation properties are determined by the two times, τ_F and τ_p, of the respective changes of the function $F(t)$ and of the pump field

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$E_p(t)$. In particular, it is shown theoretically in Refs. 7 and 8 that in the case of monochromatic initiating radiation ($\tau_p \to \infty$) the time $\tau_F \sim T_2 \Gamma^{1/2}$ at $\Gamma \gg 1$, where $\Gamma = gI_p l$ is the gain growth rate. The same form of dependence of τ_F on T_2, with Γ replaced by $\bar{\Gamma} = g\bar{I}_p l$, holds also in the case of broad-band pumping,[7] when $\tau_p \ll T_2$. As a consequence the Stokes signal reproduces the amplitude and phase structure of the broad-band pump.[3]

The temporal structure of Stokes radiation in the case of broad-band pumping was experimentally investigated mainly by spectroscopic methods.[2,10] The characteristic times of variation of the function $F(t)$ can be investigated by direct observation of the temporal oscillations of the Stokes field, as can be seen from (1), in two limiting cases, of monochromatic and of broad-band pumping, for only in these cases do the oscillations governed by the function $F(t)$ become distinguishable from the pump fluctuations on the oscillograms. We note therefore that a correct interpretation of the scattered-field oscillations observed by the authors of Refs. 9 and 10 is difficult because the amplitude modulation of the Stokes signal can be due also to other causes: For example, even an insignificant modulation of the pump-intensity envelope in the weak-saturation regime can lead to substantial changes of the intensity of the output Stokes signal. The interferometric procedure proposed by us, jointly with a statistical analysis of a large number of experiments, is free of the indicated shortcomings. It makes it possible to investigate the characteristic times of variation of the Stokes radiation as well as of the pump at different relations between the width $\Delta\nu_p$ of the pump spectrum and the width $\Delta\nu_l = (\pi c T_2)^{-1}$ of the spontaneous scattering line, particularly in the region $\Delta\nu_p \sim \Delta\nu_l$ in which the character of the function $F(t)$ has not been investigated to this day.

2. EXPERIMENTAL SETUP

The pump source in the experiment was a YAG:Nd^{3+} laser emitting a single-mode beam with pulse duration 30 nsec and energy up to 60 mJ. Since an important factor in the analysis of the results is constancy of the duration of the reflected Stokes signal, the pump energy was chosen, depending on the experimental condition, such that the duration of the reflected pulse was always 20 nsec. The width $\Delta\nu_p$ of the laser-emission spectrum ranged from 0.001 to 0.3 cm^{-1}; this was accomplished by using various reflecting elements in the cavity and also by using different Q-switching methods: active—with a Pockels shutter, active—with stepwise Q-switching, and passive—with a 1055 dye. The value of $\Delta\nu_p$ was monitored with a Fabry-Perot interferometer, and at $\Delta\nu_p < 0.01$ cm^{-1} also by means of the pulse oscillograms. The laser radiation entered the experimental setup (Fig.1) through a passive decoupler—a polarizer 8 and a Fresnel rhombus 7. The setup was based on a two-beam Michelson interferometer with independent inversion of the wave front in each of the arms. The light beam was split with a semitransparent mirror 6 into two beams of equal intensity, which were guided by 100% mirrors 3, 4, and 5 into the SBS cell 1. The radiation was focused into the cell by lens 2 of focal length 20 cm, and the beams inside the cell did not

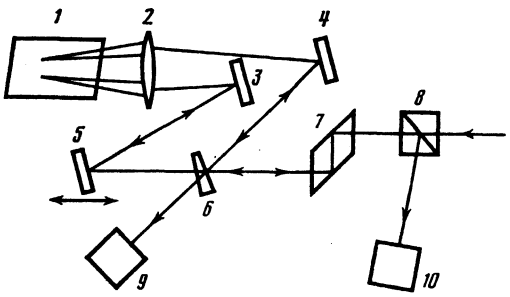

FIG. 1. Diagram of experimental setup.

overlap, so that the initial phases of the scattered waves in the two arms of the interferometer varied from pulse to pulse in independent and random fashion. The reflected light beams with inverted wave fronts interfered next on the surface of the semitransparent mirror 6. Depending on the initial phase difference and on the fluctuations of the Stokes signal, the total energy of the scattered radiation was received during the pulse time by calorimeters 9 and 10. The quality of the wave-front inversion was verified by a standard autocalibration procedure using a mirror wedge. At the 3- to 4-fold excess above the SBS threshold at which the measurements were performed in the present study, the directivity patterns of the single-mode pump beam and of the scattered radiation coincided. Displacement of mirror 5 with suitable adjustment of mirror 3 made it possible to vary the difference ΔL between the lengths of the interferometer arms from zero to several dozen centimeters.

3. PROCEDURE FOR STATISTICAL ANALYSIS OF THE EXPERIMENTAL DATA

The fraction of the total scattered-radiation energy that entered as the result of interference into one of the two recording channels is equal, on the basis of (1) and (4), to

$$\frac{K_1}{K_1+K_2} \approx \frac{1}{2} + V(\tau)\frac{1}{2T}\int_{-T/2}^{T/2} \cos\Delta\varphi(t)\,dt. \qquad (5)$$

Here K_1 and K_2 are the SBS signal energies measured by calorimeters 9 and 10; T is the duration of the Stokes pulse; $\Delta\varphi(t) = \varphi_1(t) - \varphi_2(t)$ is the random phase difference between the scattered waves; $V(\tau)$ is the pump correlation function; τ is the path-time difference corresponding to the difference ΔL between the lengths of the interferometer arms. Expression (5) was obtained under the assumption that the pump spectrum is symmetrical about the central frequency, $\tau \ll T$ and $\tau_p \ll \tau_F$.

At $\tau = 0$ the correlation function $V(0) = 1$ and expression (5) coincides with the expression for monochromatic pumping. This means that at a zero path difference it is possible to use the statistical method of estimating the characteristic time τ_F of dephasing of the Stokes wave, even in the case of broad-band pumping, from the shape of the distribution function of the random quantity $K_1/(K_1+K_2)$ constructed on the basis of a large number of tests. For a comparison of the experimental results with the theory, Fig. 2 shows the distribution functions plotted in accordance with

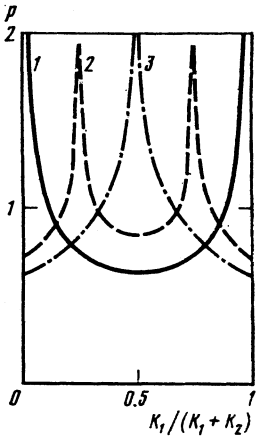

FIG. 2. Theoretical distributions of the probability density of the normalized energy of a Stokes pulse in one of the recording channels for different models of the behavior of $\Delta\varphi(t)$ during the pulse: 1) $\Delta\varphi = \text{const}$; 2) one dephasing of $\Delta\varphi$ at the end of the pulse; 3) one dephasing of $\Delta\varphi$ at the center of the pulse.

Eq. (5), using the model of jumplike change of the phase difference by a random amount uniformly distributed between zero and 2π, at a random initial phase difference and at a rectangular shape of the Stokes pulse. If the dephasing $\Delta\varphi$ takes place after a time $\tau_{dp} > T/2$ after the start of the pump, the right-hand maximum of curve 2 is located at

$$[K_1/(K_1+K_2)]_{p=\max} = \tau_{dp}/T. \tag{6}$$

At $\tau_{dp} = T_2$, curve 2 goes over into curve 3 (the peaks merge). Assuming that $\tau_F \approx \tau_{dp}$, we can estimate this quantity by using the experimentally obtained distribution of the probability $p[K_1/(K_1+K_2)]$ and relation (6). We note that when the number of dephasings during the time of the pulse increases, the scatter of $K_1/(K_1+K_2)$ decreases—the distribution tends to a Gaussian with a maximum at $K_1/(K_1+K_2) = 0.5$ and a mean squared deviation $1/2(k+1)$, where k is the number of dephasings per pulse. In our experiments, however, the pulse duration was chosen such that in the substances investigated not more than one dephasing took place per pulse.

At nonzero path difference ($\tau \neq 0$) the range of the possible realizations of the quantity $K_1/(K_1+K_2)$ is restricted, as seen from (5), to the values $(1/2)[1-V(\tau)]$ and $(1/2)[1+V(\tau)]$. Thus, by measuring the width of the experimentally obtained statistical distributions of this quantity at different τ we can determine the pump correlation function

$$V(\tau) = \max\left(\frac{K_1}{K_1+K_2}\right) - \min\left(\frac{K_1}{K_1+K_2}\right). \tag{7}$$

The functions $p[K_1/(K_1+K_2)]$ were plotted in the experiment the basis of histograms of the distribution of $K_1/(K_1+K_2)$, which were obtained by breaking up the range (0–1) of this quantity into 20 equal intervals and counting the number of realizations that land in each of these intervals. We estimate first the number of tests N necessary to construct the distribution function by this procedure. If n_i is the number of realizations of $K_1/(K_1+K_2)$ in the ith interval, then in accordance with the de Moivre–Laplace theorem the mean value over the given interval of the sought probability density is

$$p_i = \lim_{N\to\infty} (20n_i/N), \tag{8}$$

with the mean squared deviation of the quantity $20n_i/N$ from p_i at large N amounting to

$$\sigma_i \approx (20p_i/N)^{1/2}. \tag{9}$$

Preliminary experiments have shown that the peaks of the probability distribution function $p[K_1/(K_1+K_2)]$, whose position determines in accordance with (6) the characteristic dephasing time, are not as clearly pronounced in a real case as in the idealized theoretical model (see Fig. 2), and to determine them reliably it is necessary to plot the histogram with accuracy not worse than 15%. It can be seen from (9) that to attain this accuracy at $p_i \sim 1$ it is necessary to make not less than 1000 tests.

The experimental conditions have made it possible to perform runs of up to 500 measurements. However, owing to the symmetry of the interferometer arms, and consequently to the *a priori* symmetry of the sought distribution function, it became possible to sum the corresponding quantities n_i in those histogram columns which are symmetrical about the point $K_1/(K_1+K_2) = 0.5$. This operation is equivalent to an effective doubling of N, thereby decreasing the error in the determination of p_i by a factor $\sqrt{2}$. The balance of the interferometer arms was attested to by the fact that in the different runs the number of realizations each of the halves of the distribution amounted to 250 at the mathematically expected mean deviation $0.5N^{1/2} \approx 11$ from this quantity.

4. DISCUSSION OF RESULTS

To confirm the procedure proposed by us we verify first that the values of τ_F obtained in monochromatic excitation agree with the theoretical estimates.

The change of the characteristic time of the fluctuations radiation in the course of the amplification is due to deformation of the spectrum. In particular, it is easy to show that in the case of exponential amplification, in a monochromatic pumping field with a growth rate $\Gamma \gg 1$, the Lorentzian spectrum of the spontaneous Stokes component, with half-width Ω at half-maximum, is transformed into a Gaussian spectrum with a variance

$$\Delta^2 = \Omega^2/2\Gamma. \tag{10}$$

The corresponding correlation function for spontaneous Stokes components is performed:

$$R_{sp}(\tau) = \exp(-\Omega|\tau|), \tag{11}$$

and for the amplified radiation

$$R_{amp}(\tau) = \exp(-\Delta^2\tau^2/2). \tag{12}$$

We use the definition given in Ref. 12 for the correlation time:

$$\tau_c = 2\int_0^\infty R^2(\tau)\,d\tau. \tag{13}$$

According to this formula we obtain from (11)

$$\tau_c^{sp} = T_2 = \Omega^{-1}, \tag{14}$$

and from (12), inasmuch as $E_s(t) \propto F(t)$ for monochromatic pumping,

$$\tau_c^{amp} = \tau_F = \pi^{1/2}/\Delta. \quad (15)$$

From (10), (14), and (15) follows a relation between the lifetime T_2 of the acoustic oscillations in the medium and the characteristic time of the scattered-radiation fluctuations:

$$\tau_F = (2\pi\Gamma)^{1/2} T_2. \quad (16)$$

Under condition typical of stimulated emission of light we have $\Gamma \approx 25$. Consequently

$$\tau_F \approx 13 T_2. \quad (17)$$

The results of an investigation of spontaneous Brillouin scattering[13] give, when recalculated for the neodymium-laser wavelength, values $T_2 = 1$ nsec for carbon tetrachloride and $T_2 = 2$ nsec for benzene. Accordingly we obtain from (17) for these active media $\tau_F \approx 13$ and $\tau_F \approx 26$ nsec, respectively.

Let us compare these calculated correlation times of the Stokes component of stimulated Brillouin scattering with the characteristic dephasing times obtained in the present paper for monochromatic excitation ($\Delta\nu_p \ll \Delta\nu_l$). Figure 3 shows histograms constructed by the procedure described above, using monochromatic pumping. The smooth envelopes of these histograms, with allowance for the fact that in accordance with (8) we have $p_i \approx n_i/50$, yield the probability distribution functions $p[K_1/(K_1 + K_2)]$ shown in Figs. 4a and 4d. For carbon tetrachloride the distribution in Fig. 4d, according to (6) and with allowance for the fact that the duration of the Stokes pulse was 20 nsec, yields $\tau_F \approx 15$ nsec. As for the distribution obtained for benzene (see Fig. 4a), it corresponds to the case $\tau_F > 20$ nsec, and a certain decrease of the probability on the edges of the distribution, compared with the theoretical ones (curve 1 of Fig. 2) is due to the presence in each of the interferometer arms, besides the inverted radiation, of a small value of the noise component, which does not participate in the interference and is equally distributed among the registration channels. Thus, in the case of monochromatic pumping the results obtained from an analysis of statistical distributions shown in Fig. 4a and

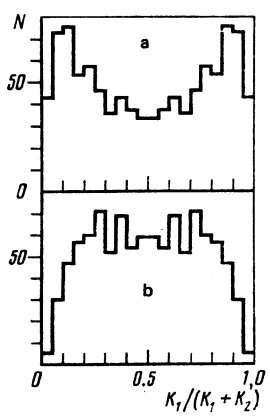

FIG. 3. Symmetrized histograms of the distribution of the number of realization of the normalized energy of a Stokes pulse in one of the recording channels at an effective number $N = 1000$ tests and $\Delta\nu_p \approx 0.001$ cm^{-1}. Active medium: a) benzene; b) carbon tetrachloride.

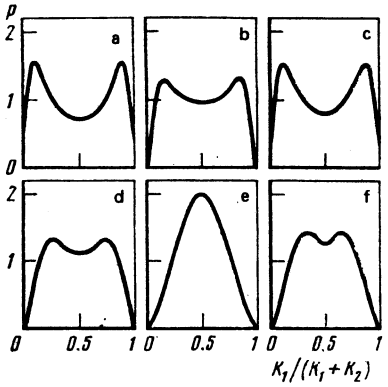

FIG. 4. Experimental distributions of the probability density of the normalized energy of a Stokes pulse in one of the recording channels at different relations between $\Delta\nu_p$ and $\Delta\nu_l$. Active medium: a, b, c) benzene ($T_2 \approx 2$ nsec, $\Delta\nu_l \approx 0.005$ cm^{-1}); d, e, f) carbon tetrachloride ($T_2 \approx 1$ nsec; $\Delta\nu_l \approx 0.01$ cm^{-1}). Width $\Delta\nu_p$ of pump spectrum: 0.001 cm^{-1} (a, d); 0.01 cm^{-1} (b); 0.03 cm^{-1} (e); 0.3 cm^{-1} (c, f).

4d are in good agreement with the theoretically calculated values of τ_F. This gives grounds for using this procedure to estimate also at other widths of the pump spectrum the characteristic time of the variation of the function $F(t)$, which determines the difference between the temporal structure of the Stokes signal from the temporal structure of the exciting radiation.

In the case of broad-band pumping ($\Delta\nu_p \gg \Delta\nu_l$), as follows from the probability distributions shown in Fig. 4c and 4f, we have $\tau_F > 20$ nsec for benzene and $\tau_F \approx 13$ nsec for carbon tetrachloride, i.e., the values of τ_F differ little from those obtained under monochromatic excitation; this agrees with D'yakov's theoretical results.[7]

Figures 4b and 4e show the distributions obtained at $\Delta\nu_p \sim \Delta\nu_l$. As noted above, the previously employed methods of investigating the temporal structure of Stokes radiation are not suitable in the indicated intermediate region of pump-spectrum widths, so that the measurements performed here are of particular interest. The results of these

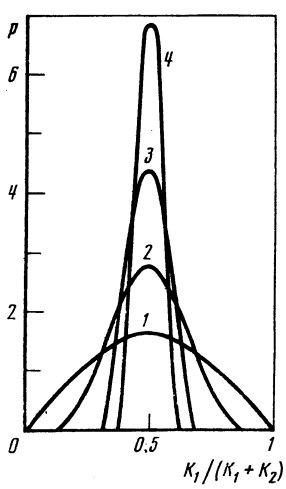

FIG. 5. Probability-density distributions of normalized energy of Stokes pulse in one of the recording channels at various difference between the interferometer arm lengths: 1) $\Delta L = 0$; 2) $\Delta L = 4$ cm; 3) $\Delta L = 9$ cm; 4) $\Delta L = 16$ cm.

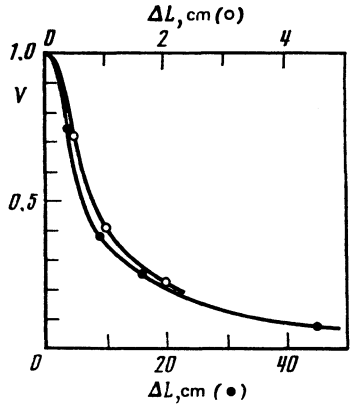

FIG. 6. Pump correlation function $V(\tau)\equiv V(\Delta L/c)$ at different spectrum widths: ●) $\Delta\nu_p\approx 0.03$ cm^{-1}; ○) $\Delta\nu_p\approx 0.3$ cm^{-1}.

measurements yield $\tau_F\approx 17$ nsec for benzene and $\tau_F\approx 10$ nsec for carbon tetrachloride. Thus, at $\Delta\nu_p\sim\Delta\nu_l$ the characteristic time of the fluctuations of the Stokes radiation is approximately 1.5 times smaller than in cases of monochromatic and broad-band pumping.

The decrease of the characteristic time τ_F at $\Delta\nu_p\approx\Delta\nu_l$ can be explained by starting from Eqs. (2) and (3), namely, in monochromatic and in broad-band pumping the quantity $\xi(t,\theta)$ in the argument of the Bessel function is equal to zero, while at $\Delta\nu_p\sim\Delta\nu_l$ the characteristic time of the fluctuations of the alternating part of the pump intensity $I_p(t)$ becomes comparable with the integration region 0—θ in (3). This leads to the appearance of an additional dependence of the function $F(t)$ on the time and, as a consequence, to a decrease of the characteristic time τ_F.

The derivation of the pump correlation function with the aid of the statistical procedure described above is illustrated in Figs. 5 and 6. Figure 5 shows the distribution obtained in the case of scattering of pump radiation of spectral width $\Delta\nu_p\approx 0.03$ cm^{-1} in carbon tetrachloride. On the basis of these curves, and also on the basis of the distribution (not shown here) at $\Delta L = 45$ cm, in accord with Eq. (7), we determined the correlation functions shown in Fig. 6 by the black circles. The correlations functions at $\Delta\nu_p\approx 0.3$ cm^{-1}, shown in Fig. 6 by the light circles, were determined in similar fashion. In the latter case, while the distribution became narrower with increasing ΔL, its "double-hump" form (see Fig. 4f) was preserved, in agreement with formula (5) obtained by us.

The correlation-function plots drawn in Fig. 6 through the experimental points are close to exponential. They correspond to Lorentzian spectra with widths 0.03 and 0.3 cm^{-1} at half-maximum, in agreement with the spectroscopic measurements.

We note that since $\tau_p \ll \tau_F$ for nonmonochromatic pumping, the correlation functions obtained characterize in accordance with (1) both the pump and the Stokes radiation.

5. CONCLUSION

The statistical procedure developed by us, based on the use of an interferometer with mirrors that invert the wave front, have made it possible to investigate in a unified manner the characteristic Stokes-radiation fluctuations due both to the reproduction of the fluctuations of the exciting radiation and to the finite lifetimes of the acoustic phonons from which the stimulated scattering takes place. Investigations of the temporal structure of the scattered radiation were carried out, for the first time ever, by a unified procedure at the different widths of the pump spectrum. In a region close to the linewidth of the spontaneous scattering, a decrease was observed in the characteristic dephasing time compared with the cases of monochromatic and broad-band excitations.

We note that the use of a periodic-pulsed laser in conjunction with an automated system for recording and reducing the information will make it possible in the future to simplify substantially the performance of similar investigations and to increase their accuracy.

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