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Baranov A. V., Davydov V., Fedorov A. V., Ikezawa M., Ren H.-W., Sugou S., Masumoto

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Interferometric coherence measurement of stress-induced In$_x$Ga$_{1-x}$As/GaAs quantum dots at the resonant-luminescence phonon sideband

A. V. Baranov, 1,2 V. Davydov, 1,* A. V. Fedorov, 2,3 M. Ikezawa, 4 H.-W. Ren, 1,1 S. Sugou, 1,5 and Yasuaki Masumoto 1,4

1. INTRODUCTION

Semiconductor quantum dots (QD’s) present a unique class of heterostructures possessing atomiclike discrete energy states and very narrow spectral lines due to three-dimensional quantum confinement. The homogeneous widths $\hbar \gamma_2$ about $\sim 50 \mu$eV were observed in photoluminescence (PL) spectra of single QD’s of different types at low temperature. 1–3 A longest phase relaxation time of the fundamental exciton transition $T_2 = 1/\gamma_2$ of about 1.3 ns ($\hbar \gamma_2 = 0.5 \mu$eV) has been found by a photon-echo spectroscopy for weakly confined CuCl QD’s in an NaCl matrix at 1 K (Ref. 4); $T_2$ of about 630 ps has been measured by a four-wave-mixing (FWM) technique for strongly confined In$_x$Ga$_{1-x}$As self-assembled QD’s (SAQD’s) at 7 K. Even for excited states, $T_2$ of $\sim 40$ ps has been measured by means of a wave-packet interferometry for a single QD formed by width fluctuation in a GaAs quantum well (QW) and for In$_x$Ga$_{1-x}$As QD selected from a SAQD ensemble. 7 The low-temperature long coherence of the electronic state of QD’s is very important for realization of efficient nonlinear optical processes in the QD systems and makes QD’s a hopeful object for quantum computation and coherent information processing. Nevertheless, there is a lack of data on the dephasing time of inhomogeneously broadened QD systems fabricated by epitaxial growth, although they are most hopeful to be used in the optical device engineering. Because of low QD optical density, conventional methods of studying the systems, e.g. the FWM technique, may be used only for many layers of QD’s (Ref. 8) or those in purposely prepared waveguides. 9,5 At the same time a technique of the wave-packet interferometry developed during the last few years looks very attractive for the study of the dephasing processes in the QD systems. 6,10,11

This technique is based on interference of two coherent polarization waves excited by the two coherent pulse trains at the frequency $\omega_2$ resonant with an optical transition of the studied system. Optical responses of two level system that are proportional to the square of the polarization amplitude, e.g. absorption, will possess oscillations $N(\tau) = N(0)[1 + s(\tau) \cos(\omega_2 \tau)]$ with a change of a mutual delay $\tau$ between the pulses. 6,10,11 Because of intrinsic polarization decay, an amplitude of the oscillations $s(\tau)$ decays with a time constant of $T_2$. It is possible to obtain $T_2$ of the resonant states of mesoscopic semiconductor systems by the time-integrated detection of the related signal as a function of $\tau$. The signals of the resonant Rayleigh scattering, 11 the resonant fluorescence, 12 and the luminescence from low-lying electron state populated through the higher-energy state 6 have been analyzed to get the $T_2$ values. At the same time, it is known that signals corresponding to incoherent processes involving phonons of the system, e.g. resonant Raman scattering or optical-phonon sideband of the resonantly excited luminescence, also contain information about $T_2$ of the resonant electronic states of the system 13,14 and can be analyzed in the same manner. Although the Stokes-shifted signals allow us to simplify the study of the lowest-energy state coherence, since they are not masked by coherent stray light, such experiments have not been performed yet. For the studies of the QD systems, it is necessary to consider that most of real QD systems possess essentially inhomogeneous spectrum of electronic transitions. It is expected from general considerations that the inhomogeneity should result in shortening of the $s(\tau)$ decay observed in the time-integrated experiments. However, the lack of an adequate theoretical model describing the inhomogeneity effect on the time-integrated signals makes it difficult to derive intrinsic $T_2$ of interest.

In this work, we report the interferometric coherence measurement of the lowest-energy electronic state of stress-induced In$_{0.1}$Ga$_{0.9}$As QD (SIQD) system by means of time-integrated detection of optical phonon lines in the resonant photoluminescence (RPL) spectra of QD’s at 2 K. Relatively long dephasing time is expected for SIQD’s as compared with other QD systems fabricated by epitaxial growth, e.g. self-assembled QD’s. 15 The studied SIQD’s system possesses essentially inhomogeneous spectrum of electronic transitions.
FIG. 1. Solid and dotted lines represent the PL spectra of the sample at 2 K in the region of the 1-1 and 2-2 transitions of SIQD’s and the lowest transition of QW excited by a cw Ti:sapphire laser at 1822 meV with power densities of 15 and 150 W/cm², respectively. The pairs of narrow lines are the segments of the RPL spectra of SIQD’s for different excitation energies Eₜ, at low excitation power limit. The different base levels of the segments represent the actual backgrounds of the RPL signals resulted from excitation of larger QD’s. The open circles (squares) show the integral intensities of the LO- (TO-) phonon lines for different Eₜ, i.e. excitation spectra of the lines, therefore positions of the circles (squares) are shifted with respect to the relevant segments by the phonon energies. A dashed curve is a guide for eyes. Inset shows the time dependence of the LO-line intensity under 2-ps pulse excitation. Eₜ = 1260.73 meV and E_D = 1224.13 meV.

with full width at half maximum of ~20 meV. Basing on developed theoretical model describing time-integrated signals of the phonon-assisted secondary emission of inhomogeneously broadened system, we demonstrate that intrinsic dephasing time of the SIQD electronic transitions can be determined from the interferometric experiments with spectral filtering of the phonon-assisted RPL, when independent information on the phonon lifetime is available.

II. SAMPLE AND ITS CHARACTERIZATION

SIQD’s were prepared in an In₀.₁Ga₀.₉As quantum well sandwiched between epilaxial GaAs barriers grown on a semi-insulating GaAs (001) substrate. A thickness of the In₀.₁Ga₀.₉As layer is 8 nm. The GaAs top barrier is 6.5 nm. SIQD’s were formed by local strains caused by InP stressors grown in a Stranski-Krastanow mode on the top of the whole structure. The stressors are 60 nm in diameter, and 20 nm high and the areal density is 2×10⁹ cm⁻². Details of the sample structure were published elsewhere. Electronic energy structure in the SIQD’s identified by the state-filling block of about 60 μeV dominate the spectra (Fig. 1). The spectra were measured in a near backscattering geometry by the use of a continuous-wave (cw) Ti:sapphire laser and a 1-m double monochromator with combined spectral resolution of 10 μeV. Analogous pair of lines was found when the excitation photon energies Eₜ of 1169 meV or 1345 meV were far from the transitions of SIQD’s. The lines correspond to the off-resonant Raman scattering by the TO and LO phonons of GaAs. A nonzero amplitude of the TO-phonon line comes from experimental imperfect backscattering geometry. Strong resonant enhancement of the line intensities is observed when Eₜ falls in the region of the inhomogeneously broadened 1-1 and 2-2 transitions of SIQD’s (Fig. 1). Simultaneously, the LO-phonon line becomes depolarized in contrast to the off-resonant case. The lowest-energy maximum in the excitation spectra of the lines corresponds to the 1-1 transition, whereas the higher-energy satellite is shifted by the LO- (TO-) phonon energy. The results clearly indicate that the zone center GaAs LO and TO phonons are effectively coupled with the low-energy electronic states of SIQD’s giving rise to the narrow Raman-like lines. The conclusion is correct, whether the resonant Raman scattering or phonon sideband of the RPL are responsible for the lines in the cw spectra. However, for analysis of the time-integrated signals of the lines excited by a pair of 2-ps laser pulses, that will be described below, it is important to know the origin of the lines at the pulse excitation. A conventional time-resolved experiment by the use of a 2-ps laser-pulse excitation and an infrared streak camera with time resolution of about 30 ps showed that the phonon-assisted RPL makes main contribution to the LO- (TO-) line intensity. Indeed, an integral intensity of the slow (luminescence) component of the LO- (TO-) line signal excited in resonance with 1-1 transition (inset in Fig. 1) is essentially larger (> ten times) than that of the fast (Raman) component. It follows that at 2-ps pulse excitation the lines come mainly from the phonon-assisted annihilation of directly excited electron-hole pairs when SIQD’s are excited in the region of the 1-1 transition (incoming resonance). At higher excitation energies, along with this process the annihilation of the pairs created by the optical-phonon-assisted absorption of the light can be responsible for the signal (outgoing resonance) as well as the excitation of the electron-hole pairs due to 2-2 transition followed by their relaxation to the lowest-energy states with emission of the LO(TO) phonon and consequent recombination. At the same time, a broadband luminescence background observed in the RPL spectra at analogous incident photon energies (Fig. 1), most likely, arises due to the second process involving acoustic phonons of the system.

III. INTERFEROMETRIC EXPERIMENT

In the interferometric coherence measurement (block scheme of the experimental setup is shown in Fig. 2), a modified Michelson interferometer has been used to form two coherent pulse trains with the same polarization and intensity separated by variable time delay τ. The pulses were generated by a ~2-ps mode-locked tunable Ti:sapphire laser at a 82-MHz repetition rate. The pulse trains emerging from
the interferometer illuminated the sample immersed in superfluid helium with the power density below the state-filling level. The luminescence was fed to a 1-m double monochromator (M1) equipped with an infrared photomultiplier. The monochromator served as a spectral filter that transmitted the luminescence at \( \hbar \omega_D \) with bandpass \( \hbar \Gamma_D \) determined by the slit width W/2. We used a continuously scanning interferometer instead of actively stabilized one commonly utilized in the coherent control experiments.\(^6,\)\(^8,\)\(^10,\)\(^11\) Then, the mutual time delay \( \tau \) is slowly varied in time \( t \) by the relation \( \tau = 2v t / c \), where \( v \) is the scanning speed of an interferometer retroreflector, \( c \) is the light speed, and the time-integrated luminescence signal \( N(\tau) \) is described by

\[
N(\tau)/N(0) = 1 - s(\tau) \cos(\omega' \tau) = 1 - s(\tau) \cos(\omega^0 t),
\]

where \( \omega' = 2 \omega v / c \). The minus sign comes from the odd number of reflections of this beam off the interferometer’s beamsplitters.

The detected signals \( N(\tau) \) oscillate with low frequency \( f = \omega' / 2\pi \) of about several tens hertz that allows one to use a lock-in technique in order to directly record the envelope of the oscillations \( s(\tau) \) containing information about \( T_2 \) with improved signal-to-noise ratio. For example, \( f = 50 \) Hz corresponds to \( \omega = \omega_L = 1.946 \times 10^{15} \) s\(^{-1} \) (\( E_L = 1277 \) meV) at experimentally used \( v = 2.4 \times 10^{-3} \) cm/s. Continuously scanning the interferometer, we took the data triggered by the ac component of the signal of the photodiode (PD1) placed behind the output slit of an auxiliary monochromator (M2). It extracted a narrow spectral component at the energy of \( \hbar \omega_L = \hbar \omega_D \) corresponding to the spectral maximum of another laser-pulse beam emerging from the interferometer. The coherence length of the component was found to be \( \approx 8 \) cm that enabled us to measure \( \tau \) in time scale up to 200 ps. The PD1 signal \( N_s(t) = \cos(\omega' t) \) served as a reference to the two lock-in amplifiers (LIA’s). One of them (LIA2) collected the luminescence signal \( N(\tau) \), and the second one (LIA1) was used for simultaneous recording of the pulse autocorrelation \( a(\tau) \) utilizing the signal from the second photodiode (PD2) directly illuminated by the light beam from the interferometer. The amplitudes of the corresponding LIA outputs are proportional to \( s(\tau) \) and \( a(\tau) \). The high stability of the interferometer and the scanning velocity \( v \) allowed us to obtain the good signal-to-noise ratio with an accumulation time constant up to several seconds (the uncontrolled jitter of the relative phase between the excitation pulses was determined to be within 0.2-rad rms) and measure the \( \omega_L \) values with accuracy of 0.5%.

IV. RESULTS AND DISCUSSION

In Fig. 3 the envelopes \( s(\tau) \) of the time-integrated signals of the LO-phonon-assisted RPL for different values of the spectral selection bandpass \( W \) (in energy units). The mean photon energy of the incident pulses, \( E_L = 1260 \) meV is in the region of the 1-1 transition of SIQD’s and the detection energy \( E_D = E_L - \hbar \Omega_{LO} = 1233.4 \) meV corresponds to the maximum of the LO-phonon band. \( a(\tau) \) is the laser-pulse autocorrelation. Full spectral width of the LO-phonon line, \( 2\hbar \Gamma_{PH} = 120 \) \( \mu \)eV.
transition (see Fig. 1). For the elastically scattered light, the decrease of $W$ resulted in the continuous increase of the width of $s(\tau)$ due to interference of two coherent pulses passed through the monochromator, as it was expected. For the luminescence base, the $s(\tau)$ was found to be independent of $W$ and to agree with $a(t_D)$ within experimental errors. Such features of the signal are typical for the luminescence from an optical transition populated through photoexcited higher-energy states with continuous spectrum, e.g., at excitation in the free-carrier continuum of the bulk semiconductor and detection of the exciton emission.\textsuperscript{12} In our case it is, most likely, caused by a large inhomogeneous distribution of energy separation between the lowest and higher exciton levels in the SIQD system analogously reported for the self-assembled InAs QD’s.\textsuperscript{22} Then, though the detected luminescence comes from the lowest-energy states selected by the filter, these states are populated from the higher-energy quasistatic continuum state through intraband relaxation mediated by acoustic phonons with continuous energy spectrum. Thus, $W$ dependence of $s(\tau)$ for the phonon-assisted RPL signal quite differs from those for other signals.

In order to explain the unusual $W$ dependence of the phonon-assisted RPL signal and to determine what information on relaxation constants of the SIQD’s can be obtained from the signal analysis, we carried out theoretical analysis of a spontaneous RPL of InGaAs.\textsuperscript{23} The spectral-selected time-integrated signal excited by two coherent pulse trains has been calculated for an inhomogeneously broadened system with two electronic states by the use of perturbation theory for the generalized master equation.\textsuperscript{24} Electron-phonon interaction was considered in the adiabatic approximation. One phonon mode with fixed frequency was taken into account since, accordingly to our experimental data, the zone-center GaAs optical phonons contribute mainly to the interaction. The inhomogeneity width was supposed to be much broader than the spectral width of the laser pulses, which is in turn much more than the relaxation rate constants of the system quantum states taken into consideration and smaller than frequencies of the optical phonons. These assumptions hold for the studied SIQD system.

Then the signal of the secondary radiation for incoming resonance averaged over the transition frequency is determined by

$$N(\tau) = \frac{8\pi\beta}{\gamma_1} \sigma^2 \Gamma_D^2 F(\Delta_{0L}) \left[ \frac{1}{\Gamma_D(\sigma - \Gamma_D)^2(\sigma + \Gamma_D)^2} \right. \times \{1 + K(\tau)e^{-\Gamma_D\tau}\cos[(\omega_D + \Omega)\tau]\}$$

$$- \frac{1}{\sigma(\sigma - \Gamma_D)^2(\sigma + \Gamma_D)^2} \{1 + K(\tau)e^{-\sigma\tau}\cos(\omega_L\tau)\}$$

$$- \frac{1}{2\sigma^3(\sigma - \Gamma_D)(\sigma + \Gamma_D)} \{1 + K(\tau)(1 + \sigma\tau)e^{-\sigma\tau}\cos(\omega_L\tau)\} \Big] .$$

If $\Gamma_D \gg \sigma$, then

$$N(\tau) = \frac{4\pi\beta}{\gamma_1} \frac{\Gamma_D}{\sigma^2} F(\Delta_{0L}) \{1 + K(\tau)(1 + \sigma\tau)e^{-\sigma\tau}\cos(\omega_L\tau)\} .$$

Since $\sigma \gg 2\gamma_2 + \gamma_{ph}/2$ by assumption, $K(\tau) \approx 1$ and $s(\tau) = (1 + \sigma\tau)e^{-\sigma\tau}$, i.e., it coincides with the laser-pulse autocorrelation $a(t)$. It is easy to show that the conclusion is fulfilled for any shape of the pulses.

If $\Gamma_D \ll \sigma$, then

$$N(\tau) = \frac{8\pi\beta}{\gamma_1} \frac{\Gamma_D}{\sigma^2} F(\Delta_{0L}) \times \left[ 1 + K(\tau)e^{-\Gamma_D\tau}\cos[(\omega_D + \Omega)\tau] \right] .$$

Importantly the decay of the signal is independent of the laser-pulse shape and the cosine amplitude $s(\tau) = K(\tau)e^{-\Gamma_D\tau}$ [see Eq. (1)]. Moreover, the decay rate of $s(\tau)$ monotonically decreases with $\Gamma_D$ and $s(\tau)$ tends to $K(\tau)$ at $\Gamma_D \ll 2\gamma_2 + \gamma_{ph}/2$ that holds for any kind of spectral filter. Just in this limit, direct information about relaxation constants of interest can be obtained from the $s(\tau)$ analysis. Since the signal does not depend on the pulse shape, it is reasonable to assign it to a free-polarization one\textsuperscript{25} that consists of two components in accordance with Eq. (3)\textsuperscript{26} First of them decaying with $2\gamma_2 + \gamma_{ph}/2$ and proportional to the pure dephasing $\gamma_2$ can be attributed to the phonon-assisted RPL. Such definition is in accordance with the phonon-assisted RPL of inhomogeneously broadened system at monochromatic cw excitation, where a spectral width (HWHM) of the related line is $2\gamma_2 + \gamma_{ph}/2$ and amplitude is proportional to $\gamma_2$. It is the term that contains information about the intrinsic dephasing constant $\gamma_2$ of the resonant transitions. Second
term decays with $\gamma_{ph}/2$ and could be assigned to the resonant Raman scattering. When $\gamma_2 \gg \gamma_1/2$, the luminescence term dominates the time-integrated signal that decays with $2 \gamma_2 + \gamma_{ph}/2$, i.e., it contains information on the coherence decay of the resonant electronic transitions.

Let us consider a correspondence between the limitation of the model and our experimental data. The direct time-resolved experiment at picosecond excitation on the SIQD system shows that the luminescence dominates the Stokes-shifted signal. The condition of exact resonance $\delta_D = \omega_L - \omega_D - \Omega = 0$ is fulfilled with accuracy close to $\Gamma_D$. The observed $W$, or $\Gamma_D$ dependence of $s(\tau)$ is qualitatively evident from Eqs. (5) and (6): $s(\tau)$ tends to $a(\tau)$ and $K(\tau)$ in the limits of broad and narrow spectral bandpasses, correspondingly. Although the values of $\gamma_2$ and $\gamma_{ph}$ are a priori unknown for the SIQD's as well as the exact value of $\Gamma_D$, it is reasonable to think that at saturation condition found experimentally for $W$ smaller than about 0.1 meV the inequality of $\Gamma_D \ll 2 \gamma_2 + \gamma_{ph}/2$ approximately holds. Physical meaning of the spectral selection in the interferometric experiment is analogous to that in conventional cw experiments, where bandpass of the filter should be smaller than spectral width of the measured line in order to get a correct value of the width. As a result, the spectral-selected time-integrated signal is described by a single exponential decay

$$s(\tau) = e^{-(2 \gamma_2 + \gamma_{ph}/2) \tau}.$$  

Therefore, the inhomogeneous broadening of the electronic transitions gives rise to the $\gamma_{ph}$ term in decay of $s(\tau)$ and $\gamma_2$ can be determined if $\gamma_{ph}$ is known from independent measurements, e.g., from the off-resonant Raman-scattering experiment.

The wave-packet interferometry experiment on a single QD controlled the coherent status and measured $\gamma_2$ for the excited but not for the exciton ground state of QD. At the same time the analysis of the signal of the phonon-assisted RPL under resonant excitation of the lowest-energy transition allows to determine dephasing constant for the exciton ground state of QD. For the SIQD system, it corresponds to excitation at the low-energy side of the related PL band (1-1 transition). We measured $s(\tau)$ for the LO line at $E_L$ in this region ($E_L = 1234, 1244, 1260, 1270$ meV) at spectral bandpass corresponding to the saturation limit. Typical $s(\tau)$ for $E_L = 1234$ meV is shown in Fig. 4(a). It was found that $s(\tau)$ were well described by the monoeXponential decay. Fitting of the curves by the use of Eq. (7) allows us to get the $\gamma = 2 \gamma_2 + \gamma_{ph}/2$ values. As is seen in Fig. 4(b), $h\gamma$ is practically constant of about 60 $\mu$eV with $h\gamma = 55 \pm 5$ $\mu$eV for the lowest incident photon energy of 1234 meV. A set of precise independent off-resonant Raman measurements of the GaAs LO-phonon linewidth performed on the same sample at 2 K by the use of a 1064.2-nm cw radiation of a Nd$^{3+}$:YVO$_4$ laser gave us the $h\gamma_{ph}$ value of 85$\pm$10 $\mu$eV. The value was found by a standard deconvolution of LO-phonon signal with Lorentzian LO-phonon line and Gaussian apparatus response measured in the same experiment by the use of the incident light. Then we can determine the $h\gamma_2$ value for the lowest-energy excitonic transition as $h\gamma_2 = h(\gamma - \gamma_{ph}/2)/2 \approx 6.3 \pm 1.5$ $\mu$eV that corresponds to the dephasing time value $T_2$ of 100$\pm$25 ps.

At higher $E_L$ (in the overlapping region of the 1-1 and 2-2 transitions and in the region of the 2-2 transition) the $s(\tau)$ time traces become narrower [Fig. 4(a)]. A spike related to the thermalized luminescence background and well described by $a(\tau)$ appears on the top of the curves, which indicates that processes involving electronic states different from lowest one additively contribute to the signals. Analysis of the signal becomes much complicated because of unknown contribution of other processes involving optical phonons. In this case, the signal $s(\tau)$ cannot be described by the above-mentioned simple model. Most likely, the shortening of the $s(\tau)$ decay is caused by an increasing contribution of the next excited state with higher $\gamma_2$ to the signal owing to overlapping of the 1-1 and 2-2 transition energies for different SIQD’s.

The low temperature $T_2$ of the lowest exciton state in In$_{0.4}$Ga$_{0.6}$As SIQD’s is longer than that of the HH exciton in single GaAs QW by an order of magnitude$^{11,27}$ and than $T_2$ of the excited states in single QD.$^{5,7}$ However, it is shorter compared with highest known $T_2$ in CuCl QD’s (Ref. 4) and

![Image](https://example.com/image.png)
In conclusion, we show that dephasing time of lowest electronic transition of QD’s in inhomogeneously broadened ensemble can be determined from analysis of time-integrated signal of optical-phonon-assisted RPL excited by two coherent pulse trains. Spectrally selected phonon-assisted RPL signals as a function of mutual delay between the pulses were analyzed in the framework of a theoretical model developed for the inhomogeneously broadened system with two electronic states and adiabatic electron-phonon interaction. It has been shown that spectral selection of the luminescence is necessary condition in order to get information on dephasing constant of the electronic transition in inhomogeneously broadened systems and an optical-phonon dephasing rate additively contributes to decay of the signal. The interferometric measurements have been carried out for In$_{0.1}$Ga$_{0.9}$As/GaAs SIQD system by the use of 2-ps pulse trains. Characteristic property of studied SIQD’s was found to be coupling of their low-energy electronic excitations with optical phonons of GaAs barriers or substrate that gives rise to analyzed phonon sideband of resonant luminescence under cw and pulse excitation. Dephasing rate of the lowest exciton state of the studied SIQD’s of 6.3±1.5 µeV (T$_2$=100±25 ps) has been found from the interferometric data by the use of appropriate fitting procedure and independent off-resonant Raman data on dephasing rate of zone-center bulk GaAs optical phonon.

The time-integrated signals of the phonon-assisted RPL can be used for the lowest-state coherence measurements of any inhomogeneously broadened systems with the narrow optical-phonon resonances in their RPL spectra. Importantly these data and analogous data obtained for phononless RPL signal allow one to get complete information on electron and phonon dephasing of the systems. It is worth noting that in the absence of the inhomogeneous broadening, e.g. for single QD, the time-integrated signals of the phonon-assisted RPL give information exclusively about $\gamma_2$.

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\*Permanent address: S.I. Vavilov State Optical Institute, 199034 St. Petersburg, Russia.
\*Present address: Space Vacuum Epitaxy Center, University of Houston, Houston, TX 77204-5507.

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It should be noted that the signal is caused by spontaneous radiation and qualitatively different from that of free polarization well known in spectroscopy of transient coherent processes.


