

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

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Dephasing of the lowest-energy electronic transition of $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}/\text{GaAs}$ stress-induced quantum dots in inhomogeneously broadened system has been investigated by the interferometric double pulse excitation and time-integrated detection of optical-phonon sideband in their resonant-photoluminescence spectra. It was found that combination of the narrow phonon resonance and spectral filtering of the signals allow us to determine the dephasing time in inhomogeneously broadened ensemble of quantum dots. The dephasing time of about 100 ps at 2 K shows that a pure dephasing is still essential in the studied system, most likely, as a result of coupling of electronic excitations in quantum dots with optical phonons of surroundings via deformation potential.

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I. INTRODUCTION

Semiconductor quantum dots (QD's) present a unique class of heterostructures possessing atomlike discrete energy states and very narrow spectral lines due to three-dimensional quantum confinement. The homogeneous widths $\hbar\gamma_2$ about $\sim 50 \mu\text{eV}$ 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 ($\hbar\gamma_2 \approx 0.5 \mu\text{eV}$) 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 $\text{In}_x\text{Ga}_{1-x}\text{As}$ self-assembled QD's (SAQD's) at 7 K. Even for excited states, T_2 of ~ 40 ps has been measured by means of a wave-packet interferometry for a single QD formed by width fluctuation in a GaAs quantum well⁶ (QW) and for $\text{In}_x\text{Ga}_{1-x}\text{As}$ 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 ω_L 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 $N(\tau)=N(0)[1+s(\tau)\cos(\omega_L\tau)]$ with a change of a mutual delay τ 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 τ . 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 state⁶ 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 $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ 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

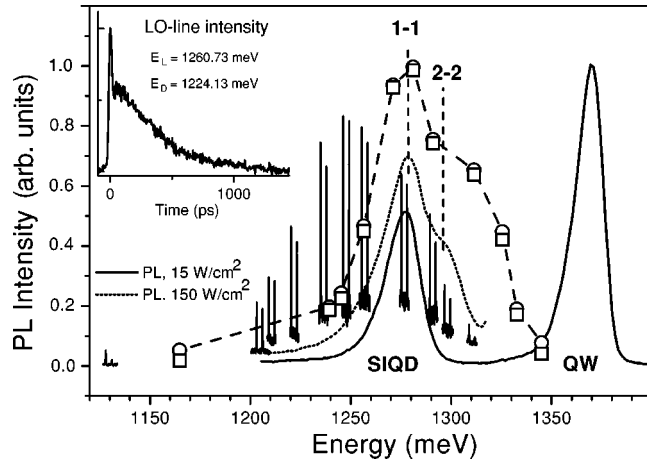


FIG. 1. Solid and dotted lines represent the PL spectra of the sample at 2 K in the region of the 1-1 and 2-2 transitions of SIQD's and the lowest transition of QW excited by a cw Ti:sapphire laser at 1822 meV with power densities of 15 and 150 W/cm², respectively. The pairs of narrow lines are the segments of the RPL spectra of SIQD's for different excitation energies E_L 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 E_L , 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. $E_L = 1260.73$ 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 In_{0.1}Ga_{0.9}As quantum well sandwiched between epitaxial GaAs barriers grown on a semi-insulating GaAs (001) substrate. A thickness of the In_xGa_{1-x}As 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 2×10^9 cm⁻². Details of the sample structure were published elsewhere.¹⁶ Electronic energy structure in the SIQD's identified by the state-filling experiment at 2 K is typical for similar QD's.¹⁷ The luminescence bands corresponding to the 1-1 transition (at 1277 meV) and the 2-2 transition (at 1296 meV) are shown in Fig. 1. A distinctive feature of the RPL spectra excited in the region of the low-energy transitions of SIQD's at 2 K is that pairs of narrow lines with the Stokes shifts of 33.60 meV and 36.55 meV, and half width at half maximum (HWHM)

($\hbar\Gamma_{PH}$) of about 60 μ eV 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 μ eV. Analogous pair of lines was found when the excitation photon energies E_L 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.¹⁸ A nonzero amplitude of the TO-phonon line comes from experimental imperfect backscattering geometry. Strong resonant enhancement of the line intensities is observed when E_L 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.¹⁹ 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

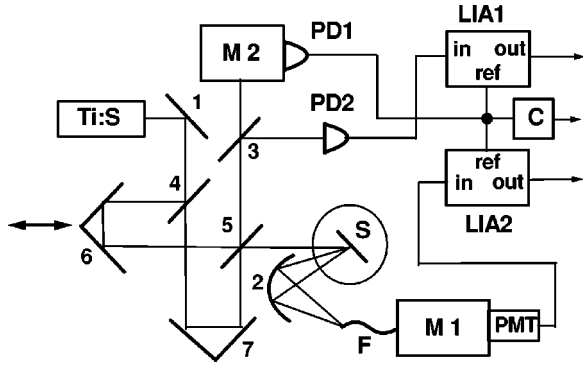


FIG. 2. Experimental setup: Ti:S represents picosecond mode-locked Ti:sapphire laser; M represents monochromators; PD represents photodiodes; S is the sample in the cryostat; F is the fiber bundle; PMT represents photomultiplier; LIA represents lock-in amplifiers; C represents counter. 1,2 are the mirrors; 3–7 are the elements of the interferometer.

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.^{6,10,11} Then, the mutual time delay τ is slowly varied in time t by the relation $\tau = 2vt/c$, 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

$$N(\tau)/N(0) = 1 - s(\tau)\cos(\omega\tau) = 1 - s(\tau)\cos(\omega't), \quad (1)$$

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 $\omega = \omega_L = 1.946 \times 10^{15} \text{ s}^{-1}$ ($E_L = 1277$ meV) at experimentally used v of 2.4×10^{-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 $\hbar\omega_r = \hbar\omega_L$ corresponding to the spectral maximum of another laser-pulse beam emerging from the interferometer. The coherence length of the component was found to be ~ 8 cm that enabled us to measure τ in time scale up to 200 ps. The PD1 signal $N_r(t) \sim \cos(\omega't)$ 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

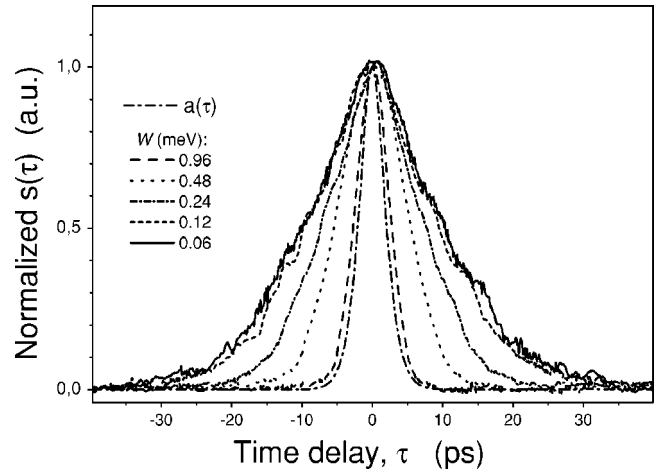


FIG. 3. Envelopes $s(\tau)$ of the time-integrated signals of the LO-phonon-assisted RPL for different values of the spectral selection bandpass W (in energy units). The mean photon energy of the incident pulses, $E_L = 1260$ meV is in the region of the 1-1 transition of SIQD's and the detection energy $E_D = E_L - \hbar\Omega_{LO} = 1233.4$ 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, $2\hbar\Gamma_{PH} = 120 \mu\text{eV}$.

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 ω_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 are shown 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 maximum of the LO-phonon band. It was initially found that $N(\tau)$ oscillates with frequency of ω_L that is a result of the interference in the absorption channel.^{10,12,20} As seen from Fig. 3, the time trace of $s(\tau)$ dramatically depends on W . With the decrease of W down to a value of about $2\hbar\Gamma_{PH}$, the $s(\tau)$ width monotonically increases, and does not depend on bandpass below this limit (Fig. 3). Analogous measurements for the TO-phonon line show the same results. Keeping in mind a different bandpass dependence of the resonantly excited coherent and incoherent secondary radiation,^{11,12,21} we have studied the effect of the spectral filtering on the time trace of $s(\tau)$ for other excitation-detection conditions: detection of the laser light elastically scattered from the sample, $E_D = E_L$, and detection of the broad luminescence base in the vicinity ($\varepsilon = \pm 3$ meV) of the LO- (TO-) phonon lines ($E_L = 1280$ meV, $E_D = E_L - \hbar\Omega_{LO(TO)} + \varepsilon$). The luminescence background is observed at E_L above the energy of the 1-1

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.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.²² Then, though the detected luminescence comes from the lowest-energy states selected by the filter, these states are populated from the higher-energy quasicontinuum 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 phonon-assisted secondary radiation.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.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

$$N(\tau) = \frac{8\pi\beta}{\gamma_1} \sigma^2 \Gamma_D^2 F(\Delta_{0L}) \times \int_{-\infty}^{\infty} dx \frac{1}{x^2 + \Gamma_D^2} \frac{1 + K(\tau) \cos[(x + \omega_D + \Omega)\tau]}{[(x - \delta_D)^2 + \sigma^2]^2}, \quad (2)$$

where

$$K(\tau) = \frac{\hat{\gamma}_2}{\gamma_2} e^{-(2\gamma_2 + \gamma_{ph}/2)\tau} + \frac{\gamma_1}{2\gamma_2} e^{-\gamma_{ph}\tau/2}, \quad (3)$$

$\Delta_{0L} = \omega_0 - \omega_L$, $\delta_D = \omega_L - \omega_D - \Omega$, ω_0 is the central frequency of distribution $F(\Delta_{0L})$ for electronic transitions in the inhomogeneous system, ω_L is the carrier frequency of

laser pulses with spectral width σ , ω_D and Γ_D are the frequency and bandpass of the spectral filter, β is the constant, Ω is the optical-phonon frequency, $\gamma_2 = \gamma_1/2 + \hat{\gamma}_2$, γ_2 and $\hat{\gamma}_2$ are the coherence loss rate constants due to the full and pure phase disruptions, and γ_1 and γ_{ph} are the inverse lifetimes of electronic population and phonon. The expression has been derived for exponential laser pulses ($\sim \exp[-\sigma|t|]$) and a Fabry-Perot spectral filter.

In the case of exact resonance $\delta_D = 0$,

$$N(\tau) = \frac{8\pi\beta}{\gamma_1} \sigma^2 \Gamma_D^2 F(\Delta_{0L}) \left[\frac{1}{\Gamma_D(\sigma - \Gamma_D)^2(\sigma + \Gamma_D)^2} \times \{1 + K(\tau)e^{-\Gamma_D\tau} \cos[(\omega_D + \Omega)\tau]\} - \frac{1}{\sigma(\sigma - \Gamma_D)^2(\sigma + \Gamma_D)^2} \{1 + K(\tau)e^{-\sigma\tau} \cos\omega_L\tau\} - \frac{1}{2\sigma^3(\sigma - \Gamma_D)(\sigma + \Gamma_D)} \{1 + K(\tau)(1 + \sigma\tau)e^{-\sigma\tau} \cos\omega_L\tau\} \right]. \quad (4)$$

If $\Gamma_D \gg \sigma$, then

$$N(\tau) = \frac{4\pi\beta}{\gamma_1} \frac{1}{\sigma} F(\Delta_{0L}) \{1 + K(\tau)(1 + \sigma\tau)e^{-\sigma\tau} \cos\omega_L\tau\}. \quad (5)$$

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

If $\Gamma_D \ll \sigma$, then

$$N(\tau) = \frac{8\pi\beta}{\gamma_1} \frac{\Gamma_D}{\sigma^2} F(\Delta_{0L}) \times [1 + K(\tau)e^{-\Gamma_D\tau} \cos[(\omega_D + \Omega)\tau]]. \quad (6)$$

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 Γ_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 one²⁵ that consists of two components in accordance with Eq. (3).²⁶ First of them decaying with $2\gamma_2 + \gamma_{ph}/2$ and proportional to the pure dephasing $\hat{\gamma}_2$ 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 $\hat{\gamma}_2$. It is the term that contains information about the intrinsic dephasing constant γ_2 of the resonant transitions. Second

term decays with $\gamma_{ph}/2$ and could be assigned to the resonant Raman scattering. When $\hat{\gamma}_2 \gg \gamma_1/2$, 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 Γ_D . The observed W , or Γ_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 γ_2 and γ_{ph} are *a priori* unknown for the SIQD's as well as the exact value of Γ_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

$$s(\tau) = e^{-(2\gamma_2 + \gamma_{ph}/2)\tau}. \quad (7)$$

Therefore, the inhomogeneous broadening of the electronic transitions gives rise to the γ_{ph} term in decay of $s(\tau)$ and γ_2 can be determined if γ_{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 γ_2 for the excited but not for the exciton ground state of QD.⁶ 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 ($E_L = 1234, 1244, 1260,$ and 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), $\hbar\gamma$ is practically constant of about $60 \mu\text{eV}$ with $\hbar\gamma = 55 \pm 5 \mu\text{eV}$ 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 $\text{Nd}^{+3}:\text{YVO}_4$ laser gave us the $\hbar\gamma_{ph}$ value of $85 \pm 10 \mu\text{eV}$. 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 $\hbar\gamma_2$ value for the lowest-

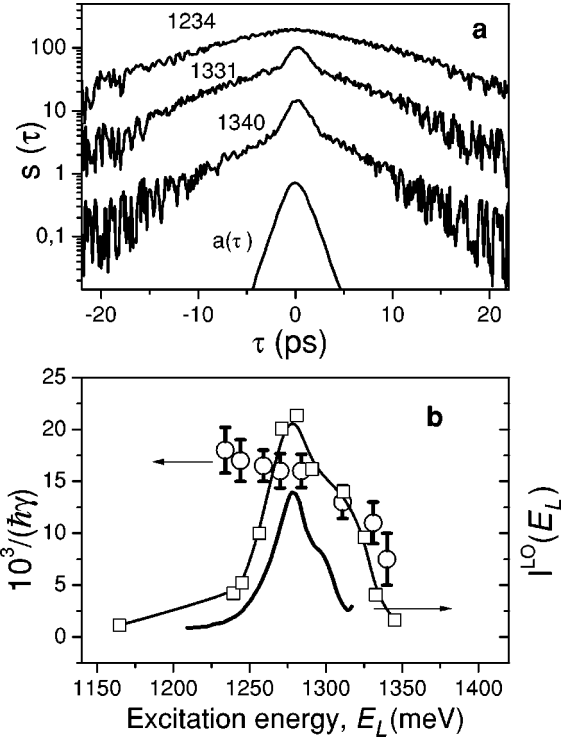


FIG. 4. (a) A semilogarithmic plot of the LO-line signal $s(\tau)$ for several E_L (shown in a unit of meV). The curves are vertically displaced for clarity. Narrow spikes on the top of some curves are well described by $a(\tau)$ and are related to the broad luminescence base. (b) Open circles correspond to the γ values determined from monoexponential fitting of $s(\tau)$ for different E_L ($\hbar\gamma$ is shown in a unit of μeV). Note, only for $E_L \leq 1270$ meV the γ values are described by $2\gamma_2 + \gamma_{ph}/2$, see the text. Relevant portions of the PL spectra of SIQD's (solid line) and the excitation spectrum of the LO-phonon line, $I^{LO}(E_L)$ (open squares, dashed line) are shown for comparison.

energy excitonic transition as $\hbar\gamma_2 = \hbar(\gamma - \gamma_{ph}/2)/2 \approx 6.3 \pm 1.5 \mu\text{eV}$ that corresponds to the dephasing time value T_2 of 100 ± 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 γ_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 $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}$ 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.^{6,7} However, it is shorter compared with highest known T_2 in CuCl QD's (Ref. 4) and

$\text{In}_x\text{Ga}_{1-x}\text{As}$ SAQD's.⁵ Comparison with recombination lifetime of the lowest exciton state (T_1 is about 300 ps, $\hbar\gamma_1 = 2.2 \mu\text{eV}$) found in direct time-resolved experiment shows that pure dephasing processes ($\hbar\hat{\gamma}_2 = 5.2 \mu\text{eV}$) are essential for the lowest-energy transition in the studied SIQD's even at $T=2$ K. The fact is in a qualitative accord with the FWM data on III-V semiconductor SAQD's at $\sim 5-7$ K where the pure dephasing either governs the total dephasing rate of the exciton fundamental transition⁸ or its rate is comparable with the depopulation one.⁵ A quantitative discrepancy between our and cited data as well as mechanisms governing the pure dephasing in the QD's systems is still subject for discussion. A comparative analysis of temperature dependences of depopulation and dephasing rates is normally used for clarification of dephasing processes in QD's usually treated in the framework of theory of crystal defects.^{28,29} The simplest case is two-level systems with radiative limited depopulation rate and pure dephasing rate caused by an elastic phonon scattering. The optical-phonon contribution is negligible at temperatures up to few tens of Kelvin due to the reduction of the phonon population.³⁰ Then acoustic phonons with continuous energy spectrum govern the pure dephasing rate resulting in its T^7 dependence.³¹ A presence of the exciton fine structure with characteristic energy gap $\Delta\varepsilon$, that is an inherent property of most QD systems, causes qualitatively other temperature dependence: in addition to the elastic phonon scattering, transitions between levels of the structure with emission and absorption of the acoustic phonons will contribute to γ_2 via both the γ_1 and $\hat{\gamma}_2$ temperature dependences. In high-temperature limit $\Delta\varepsilon < k_B T$, γ_1 and $\hat{\gamma}_2$ will be proportional to T and T^2 , respectively. In opposite limit γ_1 is temperature independent for transitions with emission of the phonons and $\gamma_1 \sim \exp(-\Delta\varepsilon/k_B T)$ for transitions with absorption of the phonons, while $\hat{\gamma}_2 \sim \exp(-\Delta\varepsilon/k_B T)$ for both absorption and emission. Just this process was concluded to be responsible for the low-temperature total dephasing rate in $\text{In}_x\text{Ga}_{1-x}\text{As}$ SAQD's,⁵ where the linear temperature dependence of γ_2 has been observed in the range of 5–40 K. The high-temperature limit holds for this type of QD's where $\Delta\varepsilon$ are in the range of few hundred μeV .³² The same linear dependence has been observed for $\text{In}_x\text{Al}_{1-x}\text{GaAs}$ SAQD's.⁸ It follows that main contribution to the temperature dependence of γ_2 comes from that of γ_1 . At the same time, physics of pure dephasing remains not quite clear since predicted dependence of $\hat{\gamma}_2 = aT^2 + bT^7$ was not detected despite the fact that measured values of $\hat{\gamma}_2$ were comparable with or more than those of γ_1 . In studied $\text{In}_x\text{Ga}_{1-x}\text{As}$ SIQD's relative contribution of $\hat{\gamma}_2$ to total dephasing rate is even greater than that found in Ref. 8 and we suppose that the γ_2 temperature-dependence measurements performed for this system will allow one to look inside the physics of the low-temperature pure dephasing in QD's.

V. CONCLUSION

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 $\text{In}_{0.1}\text{Ga}_{0.9}\text{As}/\text{GaAs}$ 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 $6.3 \pm 1.5 \mu\text{eV}$ ($T_2 = 100 \pm 25$ 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 γ_2 .

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