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Two-pulse coherent population of quantum states in inhomogeneous ensemble detected by the phonon-assisted resonant luminescence

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Abstract. The novel technique of coherent control of quantum states in semiconductor nanostructures was applied to the inhomogeneously broadened ensemble of quantum dots. By taking an advantage of the interaction between electronic states in quantum dots and phonons in the host matrix the inhomogeneous broadening was effectively eliminated. The coherence decay rate was measured.

Introduction

Recent development of ultra-short pulse tunable lasers has opened numerous opportunities in time-domain spectroscopy of quantum systems. Typical example of the great value of this approach in the field of semiconductor quantum dots (QDs) was published recently [1]. That work took advantage of the narrow optical transitions in the single QD selected from ones formed by the natural width fluctuations of the quantum well.

The present work is devoted to testing a different approach based on the spectral narrowing of the response of the inhomogeneously broadened system containing a large number of QSs. The core idea of this technique is to utilize the intrinsic narrow resonances in the III–V semiconductor heterostructures — optical phonons of the host matrix crystal [2]. These resonances can tightly bound the energy positions of levels excited by laser and those whose luminescence is detected.
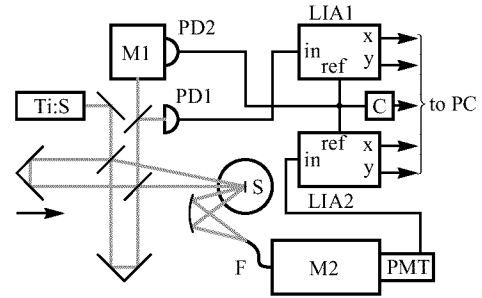
1. Experimental

1.1. Sample

The sample studied was grown by the gas source molecular beam epitaxy on the GaAs substrate and contains InGaAs quantum well sandwiched between epitaxial GaAs barriers. QDs are formed in the concentrations of local strains caused by the InP stressors grown in Stranskii–Krastanow mode on top of the whole structure. Details of the sample structure were published elsewhere [3].

It is the property of that kind of structures, that longitudinal (LO) and transversal (TO) optical phonons of the GaAs possess a narrow spectral width of the order of <1 meV and, on the other hand, are strongly coupled to the electronic states of stress-induces quantum dots. This coupling results in strong quasi-Raman lines in the resonant photoluminescence
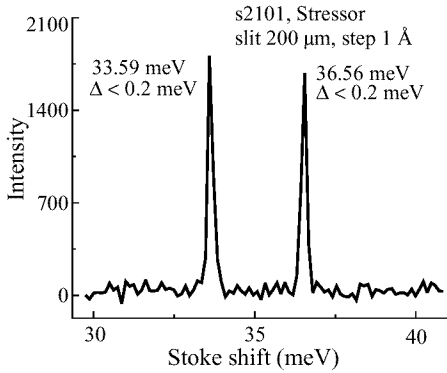
Fig. 1. Resonant photoluminescence spectrum of the stress-induced InAs quantum dots. Positions and widths of the TO (peak with lower energy) and LO (higher energy peak) phonons are shown.

Fig. 2. Experimental setup. Notation: Ti:S — laser, LIA — lock-in amplifiers, PD — photodiodes, M — monochromators, S — sample in cryostat, PMT — photomultiplier, F — fiber bundle, C — counter, PC — computer.

(RPL) spectra. An example of such a spectrum is shown in Fig. 1. The similar picture is observed in the photoluminescence excitation spectra.

1.2. Setup

The core parts of the experimental setup (schematically shown in Fig. 2) are the mode-locked tunable Ti:sapphire laser, modified Michelson interferometer, 1 m double monochromator and acquisition electronics.

Time sweep was implemented by precise mechanical displacement of one of the interferometer's retroreflectors by means of piezo microstepper motor. Both beams of the laser light emerging from the interferometer consist of the pairs of coherent pulses separated by the time delay Δt up to few hundreds picoseconds.

To obtain the exact measure of the Δt, we extract single narrow spectral component from one of this light beams by use of the auxiliarry "reference" monochromator (M1) tuned to a wavelength λ_R somewhere within the spectrum of the laser pulses. The mean intensity I_R of this component is detected by the photodiode (PD2) placed behind the output slit and can be described by an equation

$$ I_R = I_{R_0} \left[1 + r(\Delta t) \cos \left(2\pi \frac{c}{\lambda_R} \Delta t \right) \right], \tag{1} $$

where I_{R_0} is a constant proportional to the laser power at the wavelength λ_R and combined efficiency of the monochromator and photodiode, and the slowly changing envelope r is determined by the finite bandwidth of the auxiliarry monochromator and minor misalignments of the interferometer during the scan. In our experiments the value $r(0)$ is almost 100% and gradually falls to about 30% at the $\Delta t \approx 200$ ps.

The small part of the same beam was split off by the beamsplitter and detected by the broadband photodetector (PD1), that is plain photodiode without any spectral filters (except for the laser spectrum itself, of course). The signal from this photodetector I_A obeys the formula

$$ I_A = I_{A_0} \left[1 + a(\Delta t) \cos \left(2\pi \frac{c}{\lambda_0} \Delta t \right) \right]. \tag{2} $$

This formula is quite similar to the Eqn. 1, however it's terms bear a different physical sense. The λ_0 term is the mean wavelength of the laser pulse (which coincides with the wavelength of the spectral maximum for typical symmetric pulses of Ti:sapphire laser), and the envelope $a(\Delta t)$ describes an amplitude of the autocorrelation function of light *electric field*. For the transform-limited pulses, this term is equal to the square root of the light *intensity* autocorrelation function usually obtained by the autocorrelators with nonlinear crystals.

The remaining second laser beam is routed to illuminate the sample (S) immersed in the superfluid helium inside the cryostat. Because this beam has already passed the interferometer, both pulses are filling exactly the same spatial mode, and their mutual coherence does not suffer from the quality of the routing optics, cryostat windows and sample surface. The luminescence of the sample is collected by the spherical mirror and fed to the double monochromator (M2) through the fiber bundle (F). Monochromator is equipped with the cooled infrared photomultiplier tube (PMT) and tuned to the wavelength λ_d such, that energy of photon of this wavelength is 1 LO phonon lower than the mean energy of laser photons. It effectively detects the luminescence excited at the wavelength

$$\lambda_e = 1 \left(\frac{1}{\lambda_d} + \frac{\Omega_{\text{LO}}}{2\pi c} \right)^{-1}.$$

The intensity of luminescence light detected by the PMT is described by the already familiar formula

$$I_L = I_{L0} \left[1 - l(\Delta t) \cos \left(2\pi \frac{c}{\lambda_e} \Delta t \right) \right], \tag{3}$$

where both the envelope $l(\Delta t)$ and the effective detection wavelength λ_e are determined by the *combined* properties of the sample and monocromator together. Knowing the properties of monochromator (or just taking a limit of series of measurements with different slit widths) we can obtain the desired dephasing time from the decay of the envelope l. The minus sign comes from the odd number of reflections of this beam off the interferometer's beamsplitters.

Data acquisition while continuously scanning the intrferometer is triggered by the AC component of the signal (1), which also serves as a reference to the two lock-in amplifiers (LIA 1 and 2). One of them collects luminescence (3) from the PMT, and another is used for simultaneous recording of the pulse autocorrelation (2). Because all of the three wavelengths in this formulae are chosen to almost coincide, the input signals of the LIA are oscillating at the frequency very close to the reference which allows us to use long accumulation time constants in order to improve signal to noise ratio. In this case the good estimations for the envelope functions a and l can be obtained just from the amplitude $\sqrt{x^2 + y^2}$ of the corresponding LIA output.

2. Results and discussion

In Fig. 3 several of traces $l(\Delta t)$ are presented taken with the different values of the slit width of monochromator. Also the autocorrelation $a(\Delta t)$ was recorded.

It can be seen from this data, that in the limit of the narrow slit the envelope takes a simple form of

$$l(\Delta t) = \exp \left(- \left| \frac{\Delta t}{\tau} \right| \right), \tag{4}$$

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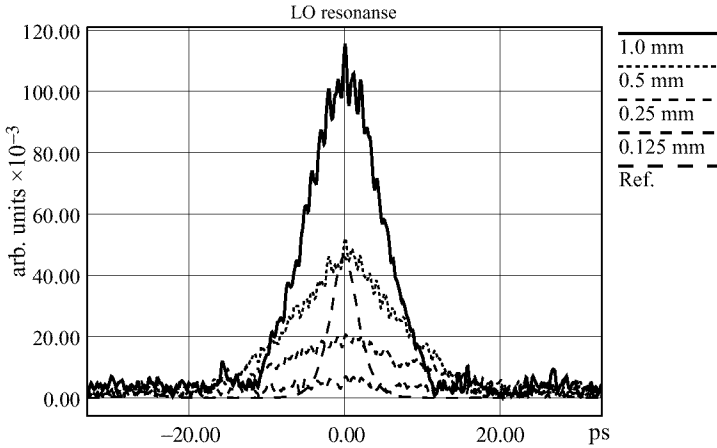
Fig. 3. Amplitudes of the coherent component seen in the luminescence of the sample and interference of the exciting light. The magnitude of latter was arbitrarily scaled to fit into the graph.

where the only parameter τ can be determined from the fit (with some small corrections for the finite width of a) and appears to be approximately 20 picoseconds, which is fairly close to the radiative lifetime of this kind of semiconductor structures. Note that the form (4) implies Lorentzian shape of the studied spectral transition, which is typical for the homogeneous broadening.

3. Conclusion

The strong interaction between the optical transitions and bulk crystal phonons in the III–V heterostructures provides a ways to perform fluorescence line narrowing spectroscopy on this systems. Using of the Ti:sapphire mode-locked laser allows to do measurements directly in the time domain. We utilize both of this features for study of the dephasing time in quantum dots.

References

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