Defense Technical Information Center
Compilation Part Notice

ADP013106

TITLE: Observation of Quantum Beats in Photoluminescence of
Self-Assembled Quantum Dots

DISTRIBUTION: Approved for public release, distribution unlimited
Availability: Hard copy only.

This paper is part of the following report:

TITLE: Nanostructures: Physics and Technology International Symposium
[8th] Held in St. Petersburg, Russia on June 19-23, 2000 Proceedings

To order the complete compilation report, use: ADA407315

The component part is provided here to allow users access to individually authored sections
of proceedings, annals, symposia, etc. However, the component should be considered within
the context of the overall compilation report and not as a stand-alone technical report.

The following component part numbers comprise the compilation report:
ADP013002 thru ADP013146

Observation of quantum beats in photoluminescence of self-assembled quantum dots

V. Davydov†§, A. V. Fedorov£§, I. V. Ignatiev†‡, S. V. Nair†, H.-W. Ren†, M. Sugisaki†, S. Sugou† and Y. Masumoto†¶

† Single Quantum Dot Project, ERATO, JST, Tsukuba 300–2635, Japan
‡ Institute of Physics, St Petersburg State University, St Petersburg 198904, Russia
§ Vavilov State Optical Institute, St Petersburg, 190034, Russia
¶ Institute of Physics, University of Tsukuba, Tsukuba 305–8571, Japan
£ Venture Business Laboratory, University of Tsukuba, Tsukuba 305–8571, Japan

Abstract. The photoluminescence (PL) kinetics of heterostructures with InP self-assembled quantum dots were studied under quasiresonant excitation in presence of external electric field. An oscillation behavior of the PL kinetics is observed at small applied bias. Period of the oscillation is about 30 ps and decay time is 20 to 50 ps depending on spectral point. We studied the PL kinetics in magnetic and electric fields. All set of the data gives a clear evidence that the oscillations are caused by the quantum beats of the optical transitions. A possible model of the observed phenomenon is discussed.

A fine structure of the energy states of self-assembled quantum dots (QDs) is difficult to study due to the large inhomogeneous broadening. Some attempts of application of usual spectroscopy technique to a single quantum dot meet the serious problems because of weak signal and poor reproducibility. A study of fine structure in time domain by technique like pump-probe or four-wave mixing is also strongly restricted for a single layer of QD's. Here we propose a different way to study the fine structure. We discovered a new phenomenon of quantum beats (QB's) that are observed in the photoluminescence (PL) kinetics of QD's in presence of external electric field. This phenomenon opens up wide possibilities to study the fine energy structure of self-assembled QD's.

We studied a heterostructure with one layer of InP self-assembled QD's embedded between $Ga_{0.5}In_{0.5}P$ barrier layers. The areal density of the QD's is about 10^{10} cm^{-2}. Average base diameter is about 30 nm and the height is about 5 nm. The sample was grown by gas source molecular beam epitaxy on an n^+ GaAs substrate. The total thickness of the undoped epitaxial layer is 500 nm. The sample was provided with a semi-transparent gold Shottky contact (thickness ≈20 nm) on the top surface and an ohmic contact on the back surface.

The PL of the sample was excited selectively by a mode-locked Ti:sapphire laser whose wavelength was tuned within or slightly above the PL band of the QD's (hereafter referred to as quasiresonant excitation). The pulse duration was about 1 ps, and the pulse repetition rate was 82 MHz. To prevent the creation of more than one electron-hole pair in a QD, we used a fairly low pump power density of about 100 W/cm^2. The PL kinetics were studied using a 0.25 m double subtractive dispersion monochromator and a streak-camera. The time resolution of the setup is about 6 ps. The PL spectra were measured also using a continuous wave (cw) excitation, a 1 m double monochromator and a photon counting system. The samle was immersed in the superfluid helium at a temperature below 2 K.
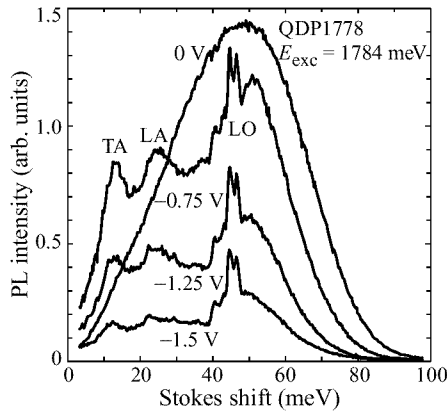
Fig. 1. The PL spectra of InP QD's at the various applied bias indicated near each spectrum. The phonon resonances discussed in the text are marked by "TA", "LA" and "LO".
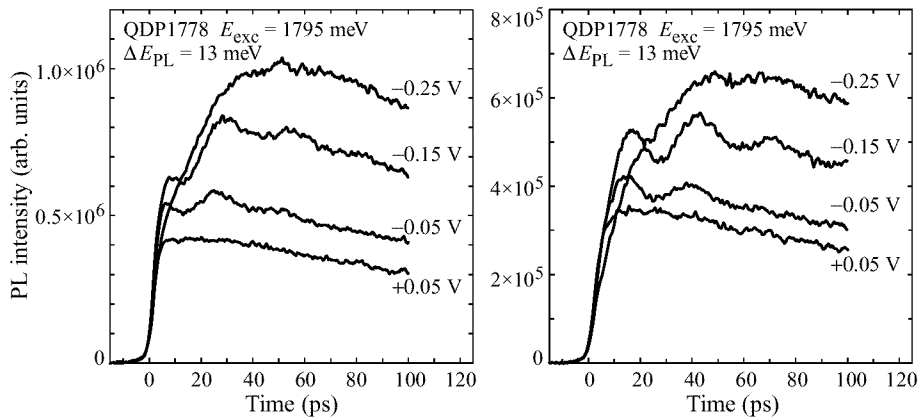

Fig. 2. The PL kinetics measured at TA resonance in parallel (left panel) and orthogonal (right panel) polarizations at the various applied bias indicated near each curve.

The PL spectra of the InP QD's measured under quasiresonant excitation at a few different negative biases applied to the sample surface are shown in Fig. 1. At zero bias, the PL spectrum has a smooth profile. When negative bias is applied, the PL is suppressed and the different resonances appear. Most prominent of them are shifted from excitation line by the transverse acoustic, longitudinal acoustic and longitudinal optic phonon energies of InP crystal. These resonances are marked in Fig. 1 by "TA", "LA" and "LO", respectively. The details of the behavior of the PL in electric field is discussed elsewhere [ ].

The PL kinetics within the first 100 ps at the TA resonance are shown in Fig. 2. They were measured under the linearly polarized excitation and the PL detection in two linear polarizations that are parallel and orthogonal to the laser beam polarization. At zero or positive bias, the PL kinetics have a fast rising part and slow decay without any features.

However at small negative bias, an oscillation type behavior of PL kinetics is clearly seen. These oscillations have exactly opposite phase in two polarizations. No oscillations are observed when nonpolarized PL is detected. Oscillations are observed for any orientation of the laser beam polarization relative to the crystallographic axes of the sample. Difference between PL kinetics measured in the parallel and orthogonal polarizations can
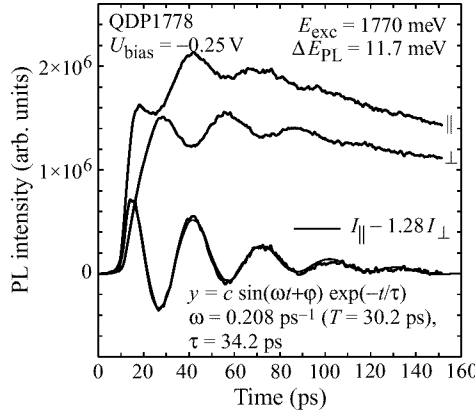
Fig. 3. The PL kinetics measured at TA resonance in parallel and orthogonal polarizations at $U_{bias} = -0.25$ V (top curves) and a difference between two curves and a fit (dashed line).

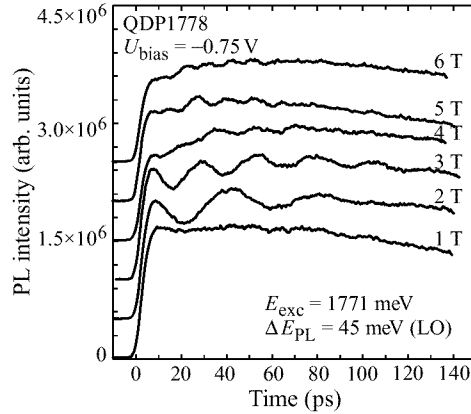**Fig. 4.** The PL kinetics measured at TA resonance in parallel polarizations at $U_{bias} = -0.75$ V and different values of magnetic field indicated near each curve. The curves are shifted for clarity.

be well fitted by a simple expression $y = \sin(\omega t + \phi) \exp(-t/\tau)$ as is shown in Fig. 3.

The oscillations in PL kinetics are observed in the whole spectral region between excitation line and LA resonance and also at LO+TA spectral point with Stokes shift 55 meV. However the oscillations are not observed at LO resonance. Period of oscillations T is about 30 ps and decay time τ is 20 to 50 ps depending on spectral point. We observed also the oscillation in PL kinetics of another sample containing slightly large InP QD's (base diameter is about 40 nm). Period of oscillations is about 40 ps in this case.

To study the physical nature of the observed phenomenon, we measured the PL kinetics in magnetic field. We found that the magnetic field destroys the regular oscillations at the value of magnetic field of about 1.5 T. At the same time, new oscillations appear that depend strongly on magnetic field. An example of these oscillations are shown in Fig. 4. The oscillation are observable for any negative bias and any spectral points except the region between LA and LO resonances.

Presented data give a clear evidence that the observed oscillations in PL kinetics are caused by quantum beats of the coherently excited states. Period of oscillations of about

30 ps corresponds to the energy separation between excited states of about 100 μeV. Decay time τ is determined by the time of coherency T_2. An important and new result is that the coherency is kept after relaxation of excitation to the luminescent state by a single phonon emission.

We do not know yet the exact physical nature of the quantum states responsible for the observed phenomenon. The oscillations observed without magnetic field (see Figs. 2 and 3) could be explained as follows. Laser pulses excite coherently two optical transitions. One of them is the phonon assisted absorption when the excitation creates an electron and a hole in their ground states, and a phonon. Probability of this transition is low due to small electron-phonon interaction in InP QD's. Another optical transition is the electronic transition between the excited electron and/or hole states. Because of quasiresonant excitation used, the excitation probably creates a hole in the excited state and an electron in the ground state. Probability of this transition should be low due to selection rules. However an external electric field may change this probability. So the probability of both transition may be equal at some bias. In this case, we may expect the quantum beats of these transitions if some splitting of the electronic and the resonant vibronic states due to their interaction takes place. In the framework of this model, the energy splitting of about 100 μeV estimated from the quantum beats reflects the interaction strength with acoustic phonons in the QD's. The interaction with LO is presumably much stronger therefore the oscillation at LO resonance is probably much faster and is not seen due to limited time resolution of 6 ps.

Open question in this simple model is that why interaction of a discrete electronic energy level with a continuous spectrum of vibronic states creates two quasidiscrete states.

The quantum beats observed in magnetic field are caused probably by the splitting of the electron and hole ground states. A coherence is transferred from excited states to the ground states by a single phonon assisted relaxation in that QD's where this relaxation is possible. That is why the quantum beats are not observed in the spectral points corresponding to the phonon bandgap between LA and LO phonons.

In conclusion, a new phenomenon of quantum beats in the PL of self-assembled InP QD's is observed. This phenomenon opens up wide possibilities to study the fine energy structure of the QD's.

References

[1] I. E. Kozin, I. V. Ignatiev, H.-W. Ren, S. Sugou and Y. Masumoto, *J. Lumin.*, (2000), at press;

[2] I. V. Ignatiev *et al.*, *Phys. Rev. B* at press.