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Apparent microcavity effect in the near-field photoluminescence of a single quantum dot

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Abstract. We present low-temperature near-field scanning optical microscopy (NSOM) measurements of self-organized InP quantum dots (QD) embedded in a $3\lambda/2$ GaInP layer. We observed an anomalously strong increase of the emission energy (~270 meV) of a single QD by changing the tip-to-surface distance over ~100 nm in the near-field region. The effect indicates formation of a near-field microcavity having extremely high photon-exciton mixing.

Introduction

Near-field optical scanning microscopy (NSOM) allows one to extend spatial resolution of optical experiments far beyond the light diffraction limit. The method opens new possibilities to study photon-matter interaction and allows the manipulation of the non-radiative components of electromagnetic waves. In the present paper we report an apparent microcavity behavior of the NSOM spectra of a $3\lambda/2$ GaInP layer containing InP QDs. The near-field microcavity effect was manifested by anomalously strong dependence of the emission energy of a single QD on the tip-to-surface distance.

1. Experiment

InP QDs were grown at 700°C by low pressure MOVPE on exactly oriented GaAs (100) substrates. Growth was started with the deposition of a 350 nm-thick $Ga_{0.5}In_{0.5}P$ lower barrier followed by nominally 3 monolayers of InP island deposition. After 5 s growth interruption the islands were overgrown by a 35 nm-thick upper $Ga_{0.5}In_{0.5}P$. The plan-view transmission electron microscopy measurements (see Fig. 1(a)) clearly reveal strain-induced contrast characteristic of QDs and point to a typical dot base ~80 nm and density $\sim 2 \times 10^9$ cm^{-2}. The thickness of the GaInP layer was equal $\sim 3\lambda/2$ for the QD emission wavelength ~730 nm.

The low-temperature near-field scanning optical microscope (NSOM) was built using a cryogenic positioning system CryoSXM by Topac, Inc. The NSOM photo-luminescence (PL) spectra were taken in collection-illumination mode at 10 K under excitation of 514.5 nm line with a power 5 μW. We use tapered fiber tips with a diameter ~0.25 μm and with a thickness of the Al coating of 20 (tips of type I) and 60 nm (tips of type II).

2. Results and discussion

2.1. Spatially resolved near-field PL spectra of InP QDs

Using the *tips of type I* we measured the PL of our structure in a standard NSOM regime (Fig. 1(b) and (c)). In this regime the intensity of the emission spectra gradually increases

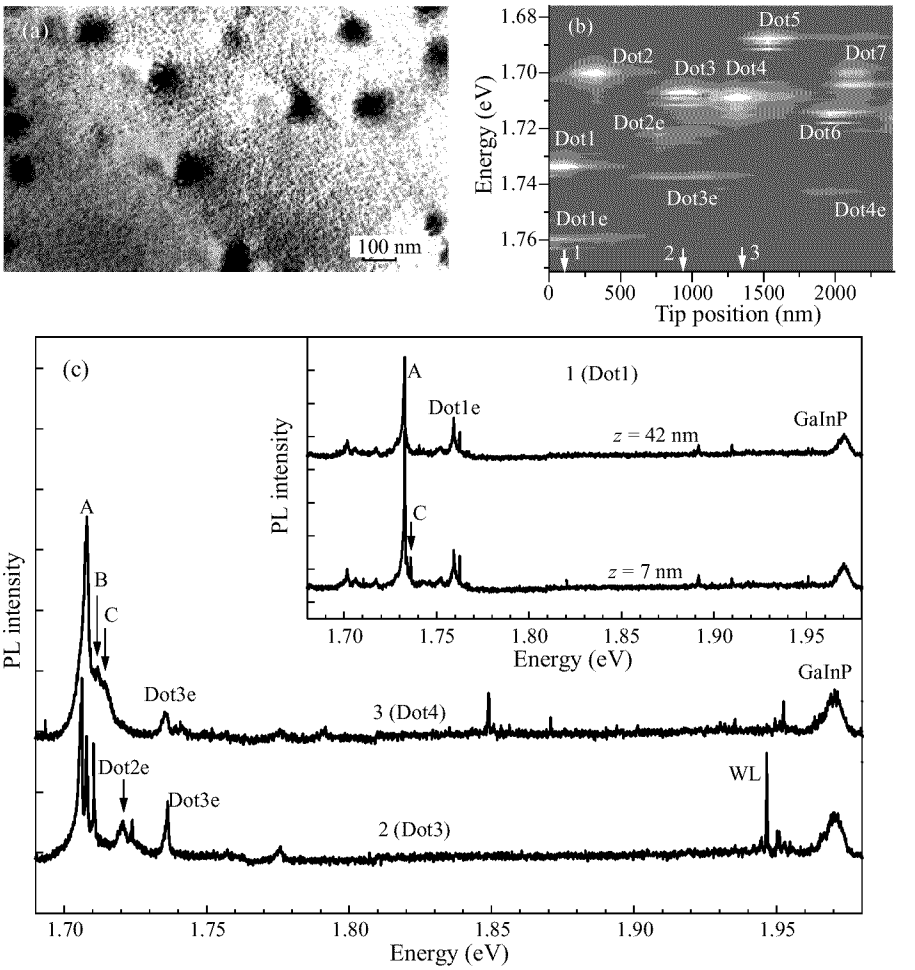

Fig. 1. (a) (upper left) plan-view TEM image of InP QDs; (b) (upper right) spatial evolution of the low-temperature near-field PL spectra InP QDs. Arrows show tip position for spectra presented in (c); (c) (under) low-temperature near-field PL spectra for the tip located on individual InP QDs. Insert shows spectra taken for two different distances from the surface.

when the tip-to-sample surface distance (z) decreases until the tip reaches the sample surface. No change in the energy position of the QDs emission lines are observed in this case (see insert in Fig. 1(c)).

Figure 1(b) shows spatially and spectrally resolved near-field PL intensity obtained in this regime, allowing the resolution of individual QDs (Dot1-Dot7, Dot1e-Dot4e). The QD density estimated from these data is $\sim 2 \times 10^9$ cm^{-2}, which agrees well with TEM data (Fig. 1(a)). In Fig. 1(c) we present the near-field emission spectra taken in lateral positions in which the tip was located on Dot2, Dot3 and Dot4. We can see that the typical emission spectra of a single InP QD consists of a triplet multiexcitonic manifold [1, 2] having energy 1.69–1.76 eV. It has an energy splitting of 1–6 meV and a halfwidth (γ) of the components, conditionally denoted as A, B, C, of 0.2–5 meV. We attribute the difference in the energy splitting and the line halfwidth to the charging of the QDs due to the presence of an impurity

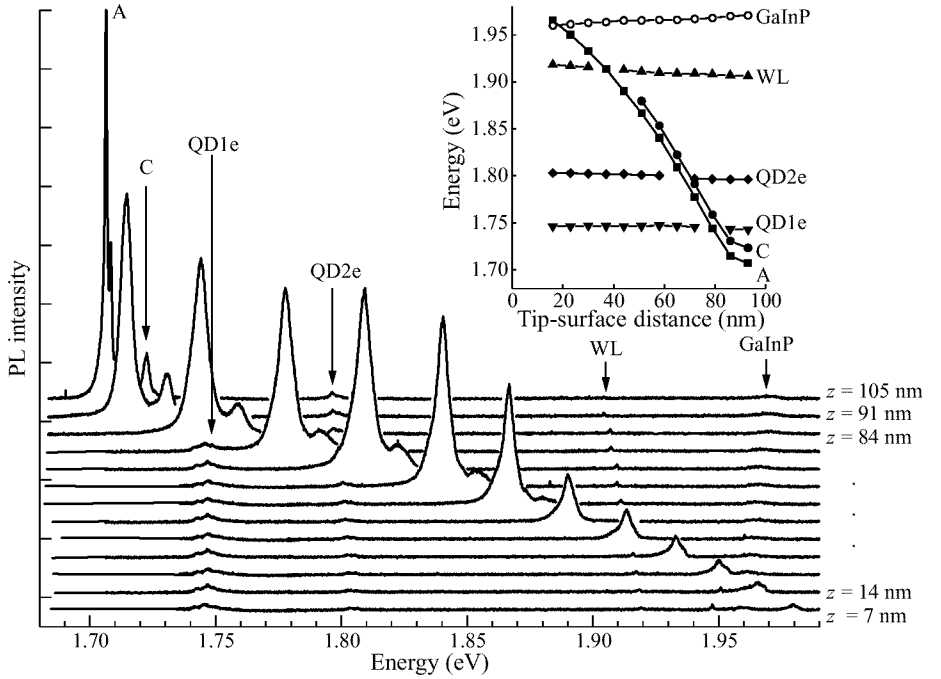

Fig. 2. Evolution of the low-temperature near-field PL spectra of InP QD under approach of the type II tip to the surface. Insert shows the shift of the emission lines as a function of tip-to-surface position.

inside the dot or in the nearby region.

In addition to the lines related to the spatially selected (central) QD, the spectra contain an emission band from the GaInP matrix at ~1.97 eV (γ = 15 meV), ultranarrow lines ($\gamma \leq 0.2$ meV) from a wetting layer (WL) centered at 1.95 eV, and very weak multiexcitonic manifolds from the QDs located close to the tip edge (Dot1e, Dot2e, and Dot3e).

2.2. *Strong shift of the QD emission line in the near-field*

For the *tips of type II* the standard NSOM regime described before takes place for our samples only for z values higher than a certain critical value of $z_0 \sim 100$ nm. For the lower tip-to surface distances the energy of the multiexciton manifold of the central QD strongly increases as z decreases.

The PL spectra taken at different tip vertical positions below z_0 are presented in Fig. 2. We can see that line A, related to the main emission component of the central QD for $z > z_0$, shifts approximately 270 meV in the range $z = 7$ nm to z_0 and when the tip approaches the surface it has energy *higher* than that of GaInP matrix. In this detuning region the manifold components broaden and their shape changes. Simultaneously the intensity is decreased as z goes to 0. From spectra in Fig. 2 we can see that much smaller energy shifts (1–5 meV) take place for the weak emission lines QD1e, QD2e, WL and GaInP. For the QD1e, QD2e and WL lines the energy increases with z decreasing, while for the GaInP line the energy decreases.

2.3. Discussion

The observed behavior of the central QD emission for $z < z_0$ is similar to Fabry–Perot detuning in planar microcavities [3, 4]. As the detuning effect is absent for tips of type I, we can suppose that in our experiments the microcavity is formed by the GaAs/GaInP interface, $3\lambda/2$ GaInP layer and an "aperture" created by the subwavelength ring of the Al coating of the NSOM tip. The strongly shifted emission line thus can be interpreted as a resonant photon mode of this cavity. However, as a QD has a discrete energy spectrum, the continuous detuning of the cavity emission in the range where no spontaneous photons are emitted (Fig. 1(c)) suggests a strong coupling of the central QD exciton with a resonant cavity photon [5]. The resonant cavity photon also couples with the excitons localized in the edge QDs, WL and GaInP which is evident from the energy shifts of the corresponding lines, but here the coupling is much smaller. In NSOM emission spectra we did not observe any evidence of the Rabi splitting for the central QD, which may be due to inhibiting of the emission of the lower polariton branch, or even due to quite a different mechanism of QD emission detuning. We expect to detect the Rabi splitting in NSOM reflectivity experiments which are now in progress.

In conclusion our experiments reveal an apparent microcavity effect in the near-field spectra of a single QD embedded in $3\lambda/2$ layer. The strong detuning of the QD emission energy observed opens new possibilities for photonic device engineering.

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References

[1] E. Dekel, D. Gershoni, E. Ehrenfreund, D. Spector, J. M. Garcia and P. M. Petroff, *Phys. Rev. Lett.* **80**, 4991 (1998).

[2] D. Hessman, P. Castrillo, M.-E. Pistol, C. Pryor and L. Samuelson, *Appl. Phys. Lett.* **69**, 749 (1996).

[3] P. Royo, R. P. Stanley, R. Houdre, M. Ilegems, M. Moser, R. Hovel, H. P. Schweizer and K. H. Gulden, *Appl. Phys. Lett.* **75**, 4052 (1999).

[4] M. C. Larson and J. S. Harris, Jr, *Appl. Phys. Lett.* **68**, 891 (1996).

[5] *Confined Excitons and Photons: New Physics and Devices*, eds E. Burstein and C. Weisbuch (Plenum, New York, 1995).