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Exciton coherence in clean single InP/InAsP/InP nanowire quantum dots emitting in infra-red measured by Fourier spectroscopy

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Abstract. We report optical properties of InP/InAsP/InP nanowire quantum dots and single-photon Fourier spectroscopy of an exciton in a single InAsP quantum dot embedded in an InP nanowire. The coherent length of the time-averaged emission originating from the single InAsP QD was measured by a Mach-Zehnder interferometer inserted in the photoluminescence path. Effects of fluctuations in surrounding excess charges trapped in the InP nanowire were investigated by excitation power and energy dependencies.

Single-photon and/or entangled-photon pair sources based on solid-state device have drawn increasing attention, which are key to quantum information networks [1]. Emissions from 0D-exciton complexes localized in semiconductor quantum dots (QDs) are one of the most promising candidates for single-photon and/or entangled-photon pair sources [2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. Recently, research on semiconductor QD embedded in epitaxially grown nanowire (NW) structures has been a subject of interest in practical nano-scale device applications, such as photon sources, single-electron storage, and quantum logic circuit. This 0D+1D combined structures have been expected to improve the controllability of emission energy corresponding to size, shape anisotropy, and position of QD. In addition, epitaxially grown NWs can remediate fast dephasing of carriers in NW structures fabricated by top-down techniques due to the reduction of surface traps induced by defects and/or disorders. Besides, 0D excitons confined in a semiconductor QD can interact with its environment through carrier-phonon, carrier-carrier, and electron-nuclei interactions. The coherence length is an important parameter for the characterization of the single-photon source because decoherence (dephasing) of single-photon source sets up the bit error in the demodulation at the receiver [13]. In this study we report the optical properties of InAsP QDs embedded in InP NWs (hereafter called NW-QDs) and discuss dephasing of exciton confined in the InAs QD. Excitation power and energy dependencies of coherence times of a single NW-QD emitting in infra-red are measured by single self-photon Fourier spectroscopy.

The sample used here is InP/InAsP/InP NW-QDs grown by selective-area metalorganic vapor phase epitaxy. The lateral diameter of QD was \(~100\) nm corresponding to that of NW (Fig. 1(a)). The detailed growth conditions and the characteristics are seen in Ref. [14]. The single NW-QD photoluminescence
(PL) spectroscopy was performed by standard micro-PL measurements in the far field as shown in Fig. 1(b). Excitation laser beam travelling along NW growth direction was focused on the sample surface by a microscope objective. The QD and NW emissions collected by the same objective lens were dispersed by a double grating spectrometer \( f = 1.0 \text{ m} \) and were detected with a liquid-nitrogen-cooled InGaAs photodiode array. The typical exposure time was 1 s to obtain one spectrum with a high signal-to-noise ratio.

Figure 1. (a) SEM image of NW-QD. (b) Schematic of the experimental setup: single NW-QD-PL passing through a Mach-Zehnder interferometer (MZI). The radiative recombination decay was obtained by streak camera. FM: flipper-mounted mirror. PS: phase shifter. NPBS: non-polarized beam splitter cube.

Figure 2(a) shows a time-integrated PL spectrum of a single NW-QD at 5 K. The excitation has been carried out with He-Ne laser above InP NW bandgap energy. At low excitation intensity, the sharp peak centered around 1.023282 eV (FWHM of 46 \( \mu \text{eV} \)) for QD is observed. This sharp emission shows no significant fine structure splitting induced by an anisotropic exchange interaction [15, 16, 17]. Figure 2(b) shows excitation power dependence of PL intensity (red solid circles) and emission energy (blue solid triangles) of QD. With increasing excitation power density, the PL intensity linearly increased and saturated. Before the saturation of PL intensity, the emission energy is slightly shifted (\( \sim 200 \mu \text{eV} \)). This energy-shift is attributed to the Stark effect due to the internal electric field induced by the dipoles at the wurtzite and zinc-blende heterointerfaces in the InP NW [18].

We have performed a time resolved PL (TR-PL) measurement using pulsed-Ti:sapphire laser excitation at a repetition rate of 76 MHz. Streak cameras (Hamamatsu: C9510-NIR and C5683) were used to obtain TR-PLs of both structures. Figure 2(c) shows transient PL signals of single InP NW (blue solid triangles) and InAsP QD (red solid circles). The observed TR-PL of InP NW proves to consist of at least two decay components: InP substrate (fast decay components) and InP NW (slow decay components). From the streak camera image overlapping of NW and substrate emissions (inset of Fig. 2(c)), the remaining PL signals of InP NW were observed before zero delay time (marked by circle), corresponding to type-II recombination [18]. In what follows we focus on the QD emission with recombination lifetime of \( \sim 2 \) ns obtained by fitting to a single exponential function (red solid curve).

A Mach-Zehnder interferometer inserted in the PL path was used to apply a first-order correlation measurement on single NW-QD exciton emissions; this is a type of time-domain spectroscopy called single-photon Fourier spectroscopy and first demonstrated by Kammerer et al [19]. Fourier spectroscopy is an interesting method to explore the spectral dynamics of a single transition by both high temporal and high spectral resolutions with very low photon losses [19, 20, 21, 22, 23, 24]. The PL passing through the interferometer was sent to the double-grating spectrometer equipped with an InGaAs photodiode array. Here two types of excitation sources were used: a cw-Ti:sapphire laser (925 nm) below nanowire bandgap excitation and He-Ne laser for barrier excitation. Rotating a thin glass plate set in one of the interferometer arm gave a fine tuning of the relative phase (PS in Fig. 1(b)), \( E_0 \tau_1 / \hbar \),
between the two arms. Short-period fringe evolution of single NW-QD exciton PL emission is shown in inset of Fig. 2(a) at \( \tau = 10 \) ps. The fringe evolution as a function of the delay \( \tau \) are given by

\[
I(\tau) = I_0[1 + V(\tau) \cos(E_0 \tau_i/\hbar + \theta)],
\]

where \( E_0, I_0, V(\tau), \) and \( \theta \) are the central detection energy (\( \sim 1.023282 \) eV), the averaged signal intensity, the visibility contrast given by the modulus of the Fourier transform of the intensity spectrum, and the phase, respectively. By varying the time delay \( \tau \), we recorded interference fringes of single-photon events. Since the visibility contrast decays with increasing delay time \( \tau \) between two arms, we can obtain the coherence time by measuring the function \( V(\tau) \). Figure 3(a) shows the visibility plot of the single QD-PL interferogram as a function of the path-length difference between the two arms of the interferometer. The visibility of interference fringes decays with an almost simple exponential function. With decreasing excitation power, the coherence time increases up to \( \sim 30 \) ps as shown in Fig. 3(b). This suggests that the dephasing induced by the environmental charge fluctuations were sufficiently suppressed under weak excitation. The prolonged dephasing time corresponds to the energy-shift, \( \Delta E \), of the QD exciton emission as shown in the right axis of Fig. 3(b), which is corresponding to the number of surrounding excess charges in NW. In fact, we obtained the same coherence time in the excitation power of \( 80 \) \( \mu \)W with below InP nanowire bandgap energy (using cw-Ti:sapphire laser) and FWHM of PL peak deduced by coherence time, \( 2\hbar/T_2 = 44 \) \( \mu \)eV.

**Figure 2.** (a) PL spectrum of a single QD under He-Ne laser excitation (0.2 \( \mu \)W) at 5 K. (b) Solid (red) circles and solid (blue) triangles indicate the PL peak intensity and PL peak emission energy as a function of excitation power, respectively. (c) Time-resolved PL signals: single InAsP QD (solid (red) circles), InP nanowire (solid (blue) triangles), and InP substrate (open circles). Inset: streak camera image of a single NW exciton emission.

**Figure 3.** (a) Visibility plot of the single-dot PL. Inset: Short-period fringe evolution of a single-dot PL at delay time of 10 ns. (b) Excitation power \( (P) \) dependence of \( T_2 \) (solid (red) circles) with He-Ne laser excitation. Solid (blue) circle indicates \( T_2 \) with cw-TIS (925 nm) excitation at \( P=80 \) \( \mu \)W. PL peak energy, \( \Delta E = E(P) − E(2 \mu W) \), (open triangles) are replotted. Inset: PL spectra with \( P \) of 0.25 \( \mu \)W (solid line) and 0.9 \( \mu \)W (dashed line).
We report the single-photon Fourier spectroscopy of exciton in a single NW-QD. With decreasing excitation power and energy, the effects of fluctuations induced by the environmental excess charges are suppressed. These experimental facts suggest that the emissions originating from InAsP QDs embedded in the standing InP nanowires are promising candidates for single-photon source at a 1.2 micron-meter wavelength.

References