Single-photon Fourier spectroscopy of excitons and biexcitons in single quantum dots

V. Zwiller,* T. Aichele, and O. Benson
Nano-Optics, Physics Department, Humboldt University, D-10117 Berlin, Germany
(Received 25 February 2003; revised manuscript received 29 December 2003; published 12 April 2004)

We report on the direct measurement of the linewidth of the exciton and biexciton emission in single self-assembled InP quantum dots. The measurements were performed using a Michelson interferometer combined with a Hanbury-Brown and Twiss correlator. The setup allowed to demonstrate single-photon emission from single dots by detecting antibunching and to determine the coherence length of single photons emitted from the very same dots. A charge-coupled device camera used as detector in the Michelson interferometer allowed to determine the emission linewidth of many dots in parallel under identical experimental conditions at different temperatures.

DOI: 10.1103/PhysRevB.69.165307 PACS number(s): 78.67.Hc, 78.67.−n, 42.50.Ar, 78.55.−m

The recent advances in the field of single-photon generation using single quantum dots[1–6] provide a key element for novel experiments in quantum optics such as quantum cryptography[7,8] and quantum computing.[9,10] In these experiments, single-photon emission is usually demonstrated by detecting antibunching in a photon correlation measurement in a Hanbury-Brown and Twiss arrangement. An important step forward from incoherent single-photon emission and detection is the observation of coherent effects such as single-photon interference.[11] Here, a critical parameter is the coherence length or the linewidth of the single-photon wave packets. Linewidth measurements of single-emission lines from semiconductor quantum dots[12] have been performed using high-resolution spectroscopy,[13,14] four-wave mixing,[15,16] and coherent spectroscopy in the time domain.[17,18] Another technique which provides direct information about an emitter's line shape, linewidth, and coherence length is Fourier spectroscopy. Fourier spectroscopy is a superior method for single-photon spectroscopy since it combines high precision and a possible robust setup with very low photon losses. First attempts to establish this method for single quantum dot spectroscopy have been reported.[19] In this paper we report direct linewidth measurements performed on single self-assembled InP quantum dots using Fourier spectroscopy with a Michelson interferometer. We also measured the photon correlation of the very same emission line in order to prove single-photon emission. From a more fundamental point of view, our setup combines the measurement of wavelike and particlelike features of single photons emitted from a single quantum dot. We determine the linewidth of the exciton and biexciton emission of several dots in parallel at different temperatures.

The sample was grown by metal-organic vapor phase epitaxy and consists of a low density of InP quantum dots imbedded in GaInP. These dots can emit single photons at an ideal wavelength (~690 nm) for silicon detectors.[6] When imaging through a narrow bandpass filter, the dots are spaced by an average of several microns, making it easy to select single dots or to image several dots simultaneously. An aluminum mirror was positioned 100 nm below the dots to increase the extraction efficiency. All measurements were done in a continuous flow liquid-helium cryostat. For pulsed excitation, the output of a Ti:sapphire laser emitting 150 fs pulses with a repetition rate of 76 MHz was frequency doubled to generate pulses with a wavelength of 450 nm. For continuous-wave excitation a HeNe laser (632.8 nm) was used. The luminescence was collected with a 0.75 numerical aperture objective, spatially filtered with a removable pinhole and filtered through a narrow bandpass filter [1.2 nm full width at half maximum (FWHM) of white light transmission]. A unique feature of our setup is that the signal could either be sent to a Michelson interferometer to perform Fourier spectroscopy or to a Hanbury-Brown and Twiss (HBT) correlator for correlation measurements. In the HBT setup, avalanche photodiodes (APDs) detect single photons and their signal is used to start and stop a time to amplitude converter. The time resolution of the system was measured to be 800 ps. Lifetime measurements could be obtained by measuring the correlation between the signal from an APD in the HBT and from a photodiode on the laser. The Michelson interferometer consists of one arm with a fixed mirror and another arm with a mirror mounted on a piezotranslator with a range of 70 μm mounted on a mechanical translator with a 5 mm range. A charge-coupled device (CCD) was used to record the intensity at the Michelson interferometer output. A schematic of the setup is shown in Fig. 1(a). By removing the pinhole, several dots could be studied in parallel as shown in Fig. 1(b). Conventional imaging of the sample was done by blocking one arm of the Michelson interferometer.

Fourier spectroscopy with the Michelson interferometer was done by recording a film with the CCD while the mirror, i.e., the arm length of one interferometer arm, was scanned. The integration time for each frame was 500 ms. This enabled us to perform single-dot measurements in parallel (typically about ten dots). Long scans (70 μm) were performed to measure the exact wavelength and line shapes and short scans were done at widely different distances over 5 mm to measure the visibility defined as $V=(I_{max}-I_{min})/(I_{max}+I_{min})$ where I_{max} and I_{min} are the maximum and minimum measured dot emission intensities, respectively. The dependence of the intensity on the interferometer armlength was then extracted by measuring the recorded intensity of a single dot through all the frames, resulting in a tracelike one shown in Fig. 1(c). The background was subtracted from the signal by measuring the noise level on a nearby region of the CCD with the same area where no dot

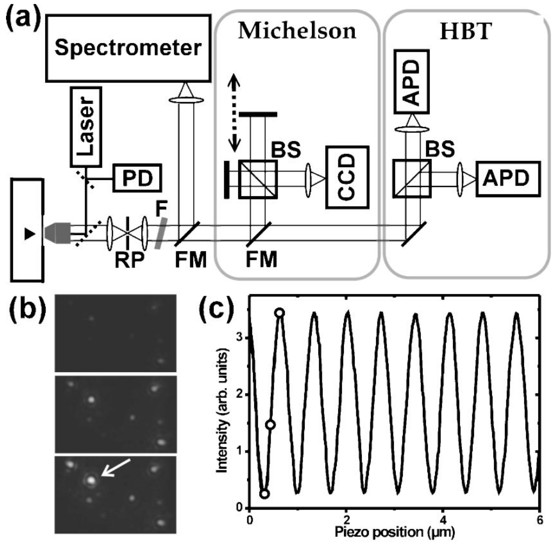

FIG. 1. (a) Experimental setup. Flippable mirrors (FM) are used to direct the luminescence either to a spectrometer, to a Michelson interferometer, or to a Hanbury-Brown and Twiss correlator. The later two are built around nonpolarizing beam splitters (BS). A removable pinhole (RP) can be used to select a single dot. A narrow bandpass filter (F) is tilted to transmit single spectral lines. (b) Images taken through the Michelson interferometer showing several dots with varying intensity as the mirror is scanned. (c) Single-dot photoluminescence intensity for the dot marked by an arrow in (b) as a function of mirror position. The circles indicate the three positions where the images were taken.

was detected. A sinusoidal fit was then used to extract the visibility value for each measurement. This enables us to extract the emission linewidth directly without making any assumption.

The power dependence of the lines was used to identify the exciton and biexciton emission lines. The exciton emission intensity shows a linear dependence on the excitation power while the biexciton line intensity follows a quadratic power dependence. Cross correlations between the exciton and biexciton emission[20–22] have also been measured on this sample to identify the emission lines, and these results will be reported elsewhere.[23] Typical single-dot spectra are shown in Fig. 2(a). The top two spectra were taken by filtering the exciton and biexciton emission while the bottom spectrum was taken without any filter. All spectra were obtained with 300 ms of integration time. A time-resolved measurement done on the exciton emission at a temperature of 10 K is shown in Fig. 2(b); an exponential fit reveals a radiative lifetime of 1.2 ns$\pm$0.4 ns for the exciton emission in agreement with previous time-resolved studies on single InP dots.[24] Fourier spectroscopy was then performed individually on each line by filtering with the narrow bandpass filter. Before each interferometry measurement, photon correlations were measured to confirm the antibunched nature of the quantum dot emission.

Figure 2(c) shows the visibility of the exciton emission measured over a 5 mm scan at a temperature of 6 K. The inset shows photon correlations measured on the filtered exciton emission under the same conditions and on the same dot with 444 s of integration time. The normalized area of the peak at zero time delay is far below 0.5 which demonstrates predominant single-photon emission.[6] The solid line in Fig. 2(c) is a fit of the form $1/[\exp(t/\tau)+\exp(-t/\tau)]$

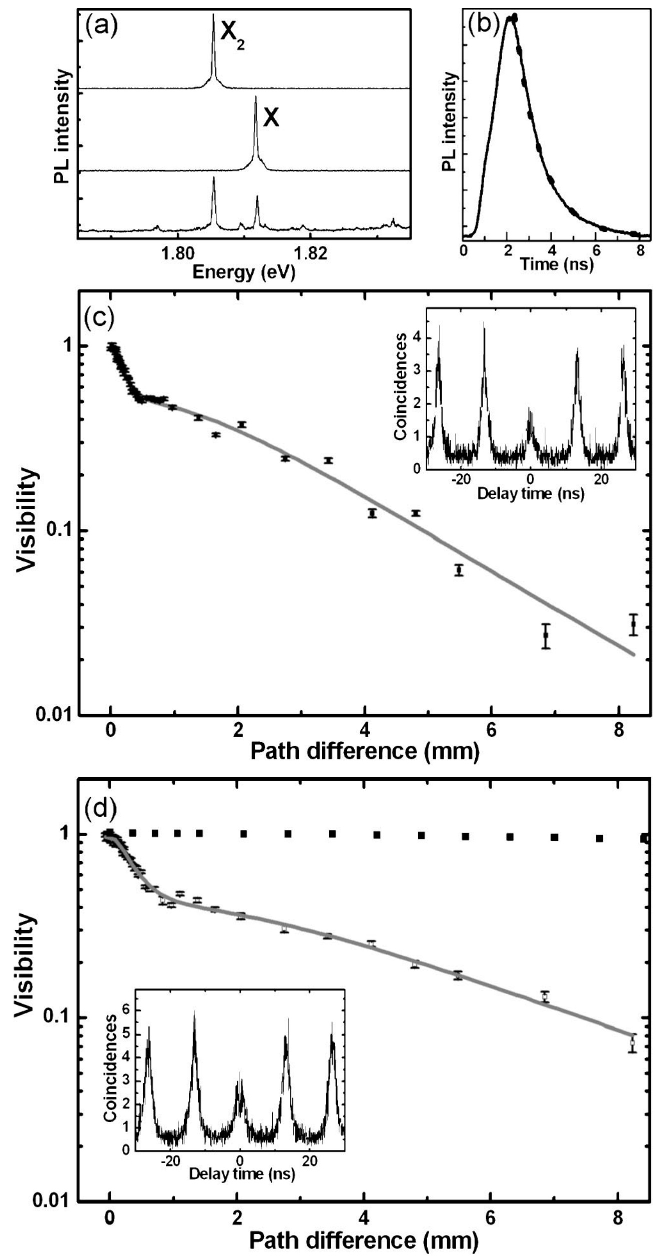

FIG. 2. (a) Single quantum dot spectra, the exciton (X) and biexciton (X_2) emission lines, are indicated. The top two spectra are taken through a narrow bandpass filter centered on the exciton and biexciton emission. The bottom spectrum was taken on the same dot without any filtering. (b) Time-resolved measurement of the exciton emission. The dotted gray line is an exponential fit. (c) Interference visibility for the exciton emission and for the biexciton emission (log scales). (d) The squares represent visibility measurements done on a continuous wave laser. The insets show correlation measurements done under the same conditions. All measurements were done at 6 K.

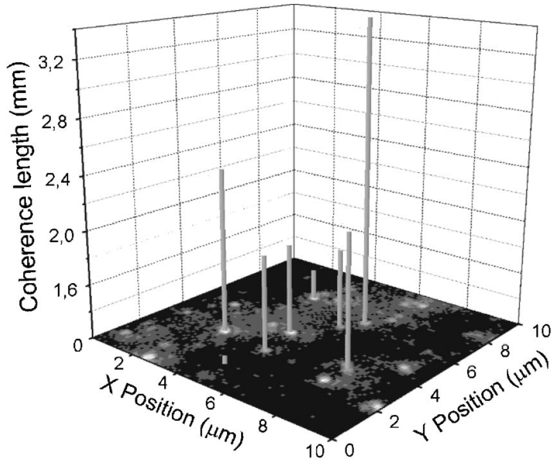

FIG. 3. Result of a parallel coherence length measurement performed on several dots at a temperature of 6 K. The bars indicate the coherence length of the single photons emitted from each corresponding dot. The x-y plane displays a CCD camera image.

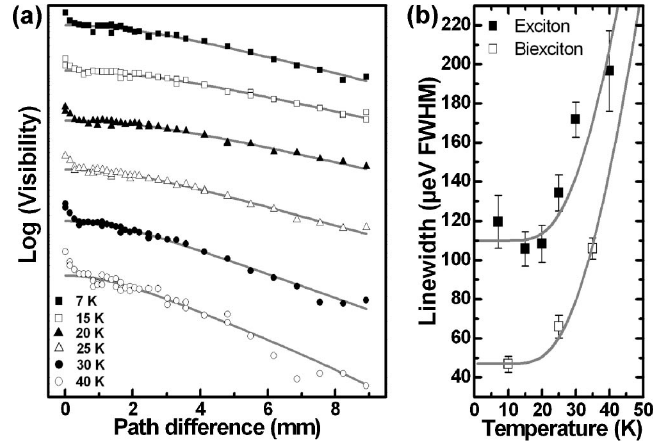

FIG. 4. (a) Temperature dependence of the visibility of the exciton emission. An offset was added to each trace for clarity. (b) Exciton and biexciton (from another dot) emission linewidth as a function of temperature.

expected for the case of a Lorentzian spectral lineshape. There are two regimes characterized by two different exponential slopes. Similar to measurements on InAs self-assembled quantum dots[16] this behavior corresponds to a narrow emission line (zero-phonon line) on top of broader shoulders. In our measurements, the shortest coherence length or the broadest width of the shoulder in the corresponding spectrum is set by our narrow bandpass filter. We checked experimentally that the emission of a white light source filtered by a narrow bandpass filter of 1.2 nm FWHM resulted in a coherence length of 0.3 mm which is in good agreement with the observed short coherence length. Emission of the wetting layer and barrier material predominantly contribute to this incoherent background in our experiment. In the following we concentrate on the coherence length and linewidth of the zero-phonon line.

The extracted coherence length from our fit to the zero-phonon line for the exciton emission is 2.1 mm, yielding an exciton emission linewidth of 186 μeV (FWHM) for a Lorentzian line shape. Figure 2(d) shows the visibility of the biexciton emission measured on the same dot under the same conditions. Here again, the inset shows a photon correlation measurement demonstrating single-photon emission from the biexciton recombination with an integration time of 666 s. The coherence length of the biexciton is 3.5 mm which corresponds to a linewidth of 112 μeV.

We want to stress here that our method allows to clearly identify emission lines from single quantum dots by studying the single-photon emission. Additionally we are able to perform measurements in parallel on several dots in a given area under exactly the same experimental conditions. We could thus determine a typical width of the distribution of the linewidth of single quantum dots. In order to demonstrate this, we have performed similar measurements over a dozen of dots. The results of a parallel coherence length measurement on the emission from eight dots are shown in Fig. 3 where the integration time for each frame was 150 ms. The visibilities of these eight dots were extracted from one mirror scan. Variations in the single-photon coherence lengths between 1.2 and 3.2 mm are observed. Because these different results are obtained simultaneously at the same energy under the same excitation conditions, the variations in coherence length reveal intrinsic differences in the dephasing mechanisms from quantum dot to quantum dot.

In our sample we typically obtained linewidths which were up to two orders of magnitude broader than what is expected from the measured radiative lifetime of 1.2 ns. This shows that decoherence processes are at work in these quantum dots even at low temperatures. Similar measurements done under HeNe excitation to avoid exciting the barrier material yielded similar results. This indicates that the dephasing mechanisms in these quantum dots do not originate from an interaction with excitons in the barrier material. It has been speculated that the linewidth broadening in InP dots is due to charging effects and fluctuating charge traps in the barrier material.[25] Although we could not detect blinking or spectral diffusion on a time scale longer than 10 msec on the dots we investigated, we could not exclude that these effects are present on a much faster time scale leading to the observed linewidths.

The temperature dependence of the exciton emission coherence length is shown in Fig. 4(a) for a typical quantum dot measured with 100 ms of integration per frame. The measurements were performed on the same dot from 7 K up to 40 K and show an increasing linewidth with increasing temperature attributed to increasing thermal phonon population. The extracted temperature-dependent linewidth for the exciton and biexciton emission is presented in Fig. 4(b). We also plotted the measured results for the biexciton emission from another dot. For exciton and biexciton emission from the same dot we typically found similar linewidths, as discussed for the results displayed in Figs. 2(c) and 2(d). The fits in Fig. 4(b) are based on an exponential function with an activation energy of 11.5 meV for both fits. This value is in good agreement with measurements of the transverseacoustic phonon energy in InP dots embedded in GaInP.[26]

We have measured the linewidth of the exciton and biexciton emission from several single InP quantum dots in parallel. A typical coherence length between 2 and 5 mm corresponding to linewidths between 50 μeV and 250 μeV was found at 7 K. In our setup, we used Fourier spectroscopy together with photon correlation measurements. This demonstrates the possibility of combining two very sensitive spectroscopic methods, namely, Fourier spectroscopy and intensity correlation measurements at the single-photon level.

We gratefully thank Werner Seifert for providing the sample. This work was supported by the Deutsche Forschungsgemeinschaft DFG, Grant No. BE22241. V.Z. acknowledges financial support from the European Union Marie Curie program.

*Present address: Quantum Photonics, ETH, CH-8093 Zürich, Switzerland. Electronic address: zwiller@phys.ethz.ch

[1] P. Michler, A. Imamoglu, M.D. Mason, P.J. Carson, G.F. Strouse, and S.K. Buratto, Nature (London) **406**, 968 (2000).
[2] P. Michler, A. Kiraz, C. Becher, W.V. Schoenfeld, P.M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Science **290**, 2282 (2000).
[3] C. Santori, M. Pelton, G. Solomon, Y. Dale, and Y. Yamamoto, Phys. Rev. Lett. **86**, 1502 (2001).
[4] V. Zwiller, H. Blom, P. Jonsson, N. Panev, S. Jeppesen, T. Tsegaye, E. Goobar, M.-E. Pistol, L. Samuelson, and G. Björk, Appl. Phys. Lett. **78**, 2476 (2001).
[5] C. Becher, A. Kiraz, P. Michler, A. Imamoglu, W.V. Schoenfeld, P.M. Petroff, L. Zhang, and E. Hu, Phys. Rev. B **63**, 121312 (2001).
[6] V. Zwiller, T. Aichele, W. Seifert, J. Persson, and O. Benson, Appl. Phys. Lett. (to be published).
[7] N. Gisin, G. Ribordy, W. Tittel, and H. Zbinden, Rev. Mod. Phys. **74**, 145 (2002).
[8] E. Waks, K. Inoue, C. Santori, D. Fattal, J. Vukovic, G. Solomon, and Y. Yamamoto, Nature (London) **420**, 762 (2002).
[9] E. Knill, R. Laflamme, and G.J. Milburn, Nature (London) **409**, 46 (2001).
[10] Special issue of Superlattices Microstruct. **31**, 2 (2002).
[11] C. Santori, D. Fattal, J. Vucković, G.S. Solomon, and Y. Yamamoto, Nature (London) **419**, 594 (2002).
[12] Y. Masumoto, J. Lumin. **100**, 191 (2002).
[13] D. Gammon, E.S. Snow, B.V. Shanabrook, D.S. Katzer, and D. Park, Science **273**, 87 (1996).
[14] M. Bayer and A. Forchel, Phys. Rev. B **65**, 041308 (2002).
[15] D. Birkedal, K. Leosson, and J.M. Hvam, Phys. Rev. Lett. **87**, 227401 (2001).
[16] P. Borri, W. Langbein, S. Schneider, U. Woggon, R.L. Sellin, D. Ouyang, and D. Bimberg, Phys. Rev. Lett. **87**, 157401 (2001).
[17] N.H. Bonadeo, J. Erland, D. Gammon, D. Park, D.S. Katzer, and D.G. Steel, Science **282**, 1473 (1998).
[18] Y. Toda, T. Sugimoto, M. Nishioka, and Y. Arakawa, Appl. Phys. Lett. **76**, 3887 (2000).
[19] C. Kammerer, G. Cassabois, C. Voisin, M. Perrin, C. Delalande, Ph. Roussignol, and J.M. Gérard, Appl. Phys. Lett. **81**, 2737 (2002).
[20] E. Moreau, I. Robert, L. Manin, V. Thierry-Mieg, J.M. Gérard, and I. Abram, Phys. Rev. Lett. **87**, 183601 (2001).
[21] A. Kiraz, S. Falth, C. Becher, B. Gayral, W.V. Schoenfeld, P.M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Phys. Rev. B **65**, 161303 (2002).
[22] C. Santori, D. Fattal, M. Pelton, G.S. Solomon, and Y. Yamamoto, Phys. Rev. B **66**, 045308 (2002).
[23] T. Aichele *et al.* (submitted).
[24] V. Zwiller, M.-E. Pistol, D. Hessman, R. Cederström, W. Seifert, and L. Samuelson, Phys. Rev. B **59**, 5021 (1999).
[25] P.G. Blome, M. Wenderoth, M. Hubner, R.G. Ulbrich, J. Porsche, and F. Scholz, Phys. Rev. B **61**, 8382 (2000).
[26] I.V. Ignatiev, I. Kozin, V.G. Davydov, S.V. Nair, J.-S. Lee, H.-W. Ren, S. Sugou, and Y. Masumoto, Phys. Rev. B **63**, 075316 (2001).