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NEAR FIELD SCANNING OPTICAL SPECTROSCOPY OF InP SINGLE QUANTUM DOTS

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We studied InP quantum dots which are prepared by strain induced self-assembly on GaAs substrates with a GaInP buffer layer using a near field scanning optical microscope operating at near liquid He bath temperatures in the collection mode. Single quantum dots are identified spatially and spectrally due to their photoluminescence spectrum. Series of luminescence lines due to single dots of different sizes are discussed in terms of dot height and width fluctuations.

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In recent years Near Field Scanning Optical Microscopy (NSOM) has proven to be a very useful tool for the investigation of quantum sized semiconductor structures because it provides optical spectroscopy with a spatial resolution on the order of the size of the quantum structures themselves [1 – 4]. Main problems limiting the spatial and spectral resolution are e.g. the penetration depth of light in semiconductors and the carrier diffusion, which are on a scale of hundreds of nanometers. The last effect is especially critical for the investigation of quantum structures in the case of non-resonant excitation of the barrier material. Both problems are especially troublesome for NSOMs operating in the illumination mode. We show that it is possible to overcome these problems using a NSOM which is operated in the collection mode.

The scanning head as illustrated in Fig. 1 was placed inside a standard variable temperature He cryostat and cooled by He gas flow. Experiments can be performed at sample temperatures near liquid He-temperature. The fiber tip used for this experiment was fabricated with a fiber pulling setup and coated with a 100 nm aluminum layer. Tip apertures below 100 nm in diameter have been realized. The scanning process is done with a piezo scanning head and controlled with a shear force feedback via laser 1, which is guided through the cryostat windows and crosses the fiber tip. Both the laserdiode and the photodiode are installed outside of the cryostat. For excitation of the carriers, an Ar^+-laser operating at 514 nm was focused by a lens system through one cryostat window at the position of the fiber tip on the sample. The excitation power can be varied up to 100 W/cm^2. The photoluminescence (PL) spectrum $I(x, y, \lambda)$ collected with the fiber tip at position $[x, y]$ is dispersed with a 0.3 m monochromator and detected by a LN_2-cooled Si-CCD- camera.

By detecting the PL with this collection mode setup only photons from carriers recombining below the tip are collected and so the spatial and spectral resolution of the

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PL signal primarily depends on the quality of the tip. Toda et al. [3] and Flack et al. [4] demonstrated investigations of quantum dot structures with a NSOM in the illumination mode. However, for this mode the diffusion of optically excited carriers limits the spatial resolution [3 – 10]. In the present setup using a colletion mode, diffusion effects are involved in the population of the dot by electron-hole pairs. The spatial resolution, however, is fully determined by the collection of the luminescence in the near field mode.

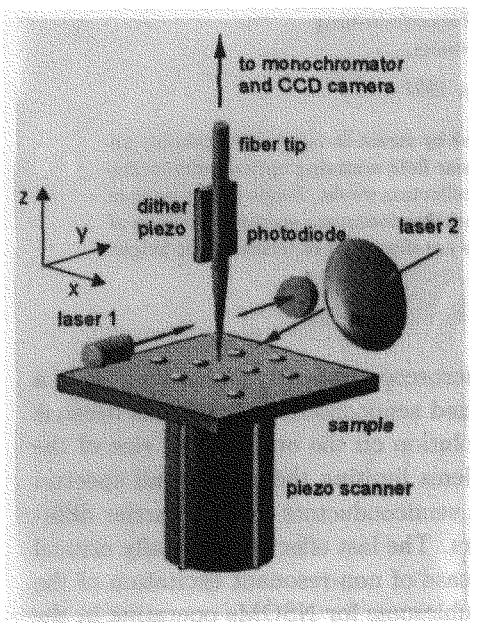

Fig.1. Experimental setup of the collection mode Near Field Optical Microscope inside a He cryostat

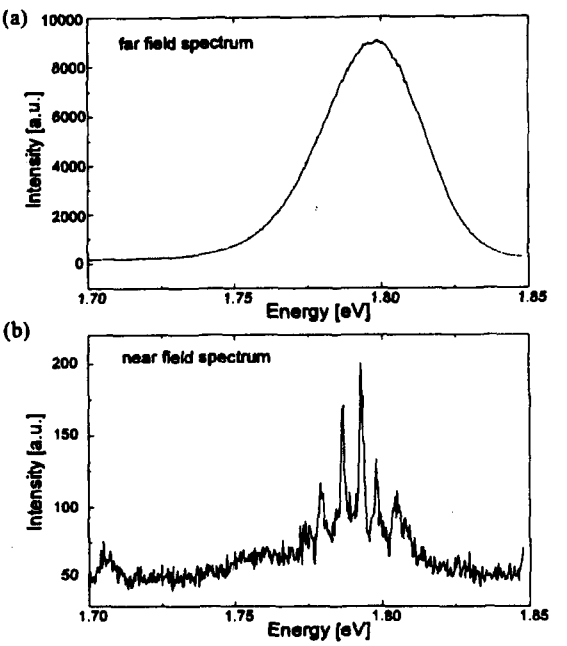

Fig.2. (*a*) Photoluminescence spectrum detected with the fiber tip at a fixed position on the sample in the far field ($z >> \lambda$). (*b*) PL spectrum in the near field regime ($z < \lambda$), $T = 10$K

The samples were grown on semi-insulating (100) GaAs substrates in a conventional MBE system using GaP as P_2 source. For further details about the P_2 source see Ref. [11]. After removing the oxide, a 400 nm thick GaAs buffer is grown under a beam equivalent pressure of $4{\cdot}10^{-6}$ Torr at a substrate temperature of 580°C. Then the deposition is interrupted to change from As to P rich growth conditions. The deposition sequence includes 200 nm $Ga_{0.46}In_{0.54}P$, followed by 3.0 monolayers (ML) InP. The InP islands are finally overgrown by a 50 nm thick $Ga_{0.46}In_{0.54}P$ layer. This layer sequence is grown at 470°C substrate temperature and under a P_2 beam equivalent pressure of $6{\cdot}10^{-6}$ Torr. The surface of the $Ga_{0.46}In_{0.54}P$ layer shows a 2x1 reconstruction using reflection high energy diffraction (RHEED). We use a growth rate of 1 ML/s for $Ga_{0.46}In_{0.54}P$ and 0.5 ML/s for InP. The composition of the $Ga_{0.46}In_{0.54}P$ layers is determined by double-crystal x-ray diffraction (DCXR). The InP quantum dots grown with this procedure have approximately a diameter of 18 nm and a height of 2.4 nm, as indicated by TEM studies [12].

To test the spectral resolution of the system PL spectra at fixed sample positions were recorded. For the far field case, that means the tip-to-sample distance z is significantly larger than the wavelength of light λ, the PL of a relatively large number of QDs is observed. Since the size of these QDs varies statistically around a mean value, a Gaussian shaped PL signal is seen in the spectrum (Fig. 2*a*). When the tip is placed in the near field regime ($z < \lambda$) sharp emission lines of single QDs with intensities well above the noise level are observed as displayed in Fig. 2*b*. The minimum spectral linewidths of the luminescence spikes are around 1 meV which was about the spectral resolution. The energy of these emission lines depends on the position $[x, y]$ at which the spectra are recorded.

Because the sample was overgrown with a 50 nm thick $Ga_{0.46}In_{0.54}P$ layer no spatial variations can be seen with AFM and STM techniques. The overgrowth layer also limits the spatial resolution for detection of the spectra to the scale of the layer thickness, since photons from carriers recombining inside the quantum dots are emitted isotropically 50 nm beneath the surface.

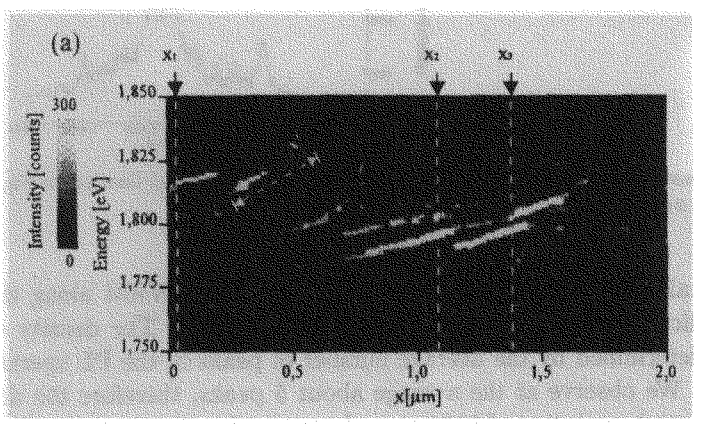

Fig.3. PL recorded in the near field regime during a linescan in x-direction at $T = 10\,\mathrm{K}$

A typical dataset is displayed in Fig. 3. The plot consists of 76 spectra, each of them was taken with an integration time of 30 s and for every spectrum the sample was moved about 26 nm along the x-direction. The intensity is scaled such that bright areas in the energy-versus-x-position image in Fig. 3 correspond to a large number of photon counts in the PL spectrum. Spectra at fixed sample positions $x = x_1, x_2, x_3$ are displayed in Fig. 4*b*.

One observes that single emission lines far above the noise level appear and disappear while the tip is scanned along the x-direction. These lines, which arise from single quantum dots, exist over distances in the range of 100 to 300 nm. The linescan in Fig. 3 is accompanied by a blue shift of the single dot emission lines of about 10 meV. If the scanning direction is reversed, we see an exactly corresponding red shift. Temperature and excitation intensity dependent measurements of the PL at single positions on this sample were done and according to these results we attribute this energy shift to local heating effects of the sample by the fiber tip, which is placed in the laser beam focus in close proximity to the sample. However other effects like the deformation of the quantum dot band structure by the strong electric field due to the close proximity of the fiber tip to the quantum dot in the laser focus could also play an important role.

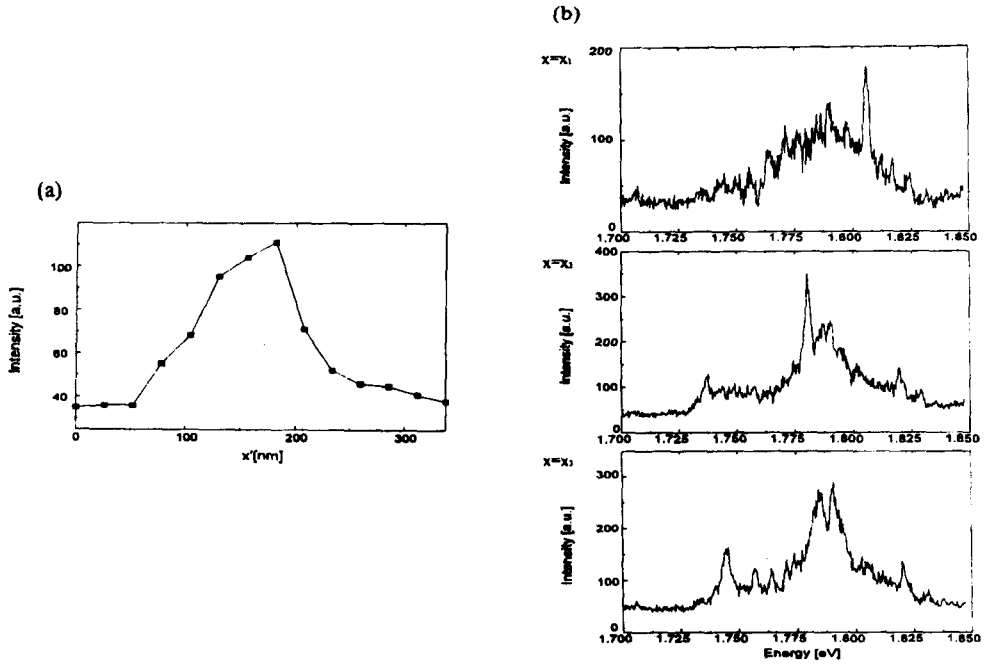

Рис.4. (*a*) Linecut across an observed dot along x' as indicated in Fig.3. In (*b*) several linecuts of Fig.3 are shown at different positions x_1, x_2, x_3

A linecut along a typical emission line is shown in Fig. 4*a*. The FWHM along the x-direction of this plot indicates a spatial resolution of about 100 nm. The density of the quantum dots can be determined by the average number of peaks in the PL spectra recorded in the near field. We observe at the average about 5 peaks, therefore the dot density is $500/\mu m^2$ for a spatial resolution of 100 nm as determined in the linescan. This value agrees very well with previous measurements done with cross sectional TEM and AFM [12].

As shown in Fig. 4*b* the near field dot emission lines are characterized by two different energy separations. On one hand side a typical spacing between single dot transition peaks on the order of 5 meV occurs frequently in the spectra. In addition, we observe also a much larger spacing of about 50 meV in many spectra (e.g. bottom and center traces in Fig. 4*b*). We attribute the luminescence lines with smaller energy separation to dots with slightly different diameters, whereas the larger spacing between the luminescence lines may be due to height variations of the approximately disk-shaped dots. By using a quantum dot model based on electron and hole effective masses of $m_e/m_0 = 0.079$ and $m_{hh}/m_0 = 0.150$ for the strained InP and conduction and valence band discontinuity of 676 meV and 70 meV, respectively, we obtain for a one monolayer height fluctuation a shift of the emission peak of about 50 meV. Within this model the smaller energy spacing corresponds to changes of the dot diameter of about 10 percent. Both values are consistent with studies of size fluctuations in self-assembled dots prior to overgrowth by TEM and AFM [12]. In addition, some of the observed single dot transition lines might be due to ground and excited state recombination in a given dot. This is also indicated by a similar spatial extent and blueshift of some of the transitions streaks in the

linescan (Fig. 3). Due to the small dot size the calculated energy spacing between allowed transitions in the ground state and the first excited states of 40 meV is larger than the observed separation of most of the peaks. This may indicate contributions of forbidden transitions in quantum dots, which could be favored e.g. by shape anisotropies of the dots or local electric fields.

In summary, we have studied single InP dot emission lines with a NSOM operated in the collection mode. Carrier diffusion and light penetration effects which limits the resolution of spatially resolved spectroscopy in illumination mode setups can be largely eliminated by operating the NSOM in the collection mode. By using the combined spatial and spectral resolution of our system we are able to distinguish dots with varying height from dots with variations of the lateral diameter.

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