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Single-Dot Optical Spectroscopy of Self-Organized Strained InGaAs Quantum Disks on (311)B-GaAs Substrate

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Single-dot photoluminescence and photoluminescence excitation spectroscopy on *true single*-dots in recently discovered strained InGaAs/AlGaAs disks spontaneously formed on GaAs-(311)B face has been undertaken. Simply by observing the luminescence under microscope and seeking dot luminescence in an energy range far from the center of the statistical distribution, well-isolated single dot luminescence and even excitation spectra could be measured. The ultra-narrow ground-state dot luminescence of Lorenzian lineshape and number of equaly distictive narrow excitated state absorption have been observed. Fully quantized zero-dimensional dot levels are thus manifested and number of specific natures expected for such system are confirmed.

Spectroscopic evidences of discrete natures of electronic level structure in artificially defined pseudo-0D semiconductor nano-structures is an ultimate proof of quantum confinement effects. Common obstacle toward 0D-spectrum analysis, however, is broadening of the spectrum due to the distribution of dots in size and/or composition.[1-4] What is essentially in need is thus a single-dot spectroscopy where a number of invidividual dot luminescences are observed of spectral linewidth much smaller than distribution width.[2-4] We have reported the results of our first trial[5] on recently discovered InGaAs disk-like embedded structure spontaneously formed via strain-driven reorganization of InGaAs/AlGaAs hetero-structure on GaAs (311)B-face.[6] By reducing the size of the optical probe, we could observe distinctive narrow luminescences from a number of *single* disks and extract their unique individual optical properties.[5] Such experiments have, however, undergone difficulties due to still rather a large number of dots within a microscopic view, and have also been spoiled by the unknown background signal overlapping with the dot luminescence. Whether such background originates from coexsisting structure or from a numerous number of dot emissions has yet been fully examined.[2,5] Following the previous experiments, we have undertaken a *true* single-dot spectroscopy. By examining statistical distribution of InGaAs disks, we have seeked a simple way to resolve single-dot luminescence from the others. Results are presented of optical assesments on single disks and discussion are presented on the present zero-dimensional system.

The samples used for this study were all grown by low-pressure metalorganic vapor phase epitaxy on GaAs (311)B-substrates, using trimethyl-gallium (TMG), -aluminum (TMA), -indium (TMI), and arsine as gas sources. Following the 100-nm $Al_{0.5}Ga_{0.5}As$ buffer layer, a thicker InGaAs layer was grown as intiating layer thereby capped by $Al_{0.5}Ga_{0.5}As$, then the thin disk InGaAs layer was grown thereafter undergone 1-3-minute growth interruption for spontaneous disk formation.[6] It should be noted that for embedding InGaAs disks, we needs no AlGaAs overgrowth, since the spontaneous rearrangement results in InGaAs islanding as well as automatic AlGaAs coverage on top through complex strain minimization and mixing/segregation of InGaAs/AlGaAs. The growth temperature was between 690°C and 750°C. A series of disk samples with different ensemble averages of size and composition were prepared simply by changing the nominal composition of InGaAs since the strain within InGaAs is a primary cause of the islanding. Well-isolated quantum disks were obtained for a nominal In content between 0.2 and 0.4, and a nominal InGaAs thickness between 2 and 6 nm, corresponding to disk diameters between 20 and 100 nm.[6] Typically 20-50 dots were found in an area of 2μm x 2 μm.

A number of sharp (typical FWHM less than 0.3 meV and the narrowest less than 60 μeV) luminescence lines with very high radiative efficiency were observed under the microscopic view of 2-μm in diameter (see Figs. 1-3).[5] Typical low-temperature micro-PL spectra taken under an excitation of 514.5-nm line of an Ar^{+}-ion laser are shown in Fig.1 for a sample of 3-nm thick $In_{0.3}Ga_{0.7}As$. Under very low excitation, only a few very narrow lines are visible (Fig.1(e)); while under increasing excitation intensity some new lines emerge and grow with excitation. All lines seem, however, to saturate their intensity under a rather a low pumping intensity; e.g. a PL line near the center of ditribution saturates already under an excitation of 54.9 nW per spot of 2-μm in diameter. At highest excitation, these lines overlap with pronounced tails and seem to converge into a broad line of Gaussian distribution.

Number and intensity of the sharp luminescence lines as in Fig.1 were found to be clearly dependent on the excitation energy: When the excitation energy, E_{laser}, exceeds the $Al_{0.5}Ga_{0.5}As$ direct gap, a markedly large number of intense lines emerge, while lower-energy excitation selects only a fraction of observable lines. When E_{laser} greater than the $Al_{0.5}Ga_{0.5}As$ (barrier) bandgap, photo created carriers can be fed into all of the low-lying dot states, thereby all the dots within the laser spot may emit light. In contrast, under an excitation below barrier energy gap, only the dots of which excited states are resonant by chance are excited, since the carrier transfer among the different dots is presumably absent.

The results presented in Fig.1 indicate that a series of the dots indeed distribute statistically in size (thickness) and/or composition, and each dot has unique optical properties. This in turn give us a way to observe a single dot: the number of the dots in the viwed area can be further decreased by looking for the dots far from the center of distribution. By this way and further by tuning the laser energy and intensity to enhance specific lines, the PL and PLE measurements were made on a few well-resolved single-dots even in a same sample wafer. Figure 2 demonstrates a result of such experiment, showing the lineshape of an isolated dot luminescence in a 3-nm thick $In_{0.35}Ga_{0.65}As$ sample (estimated disk diameter ≈ 30 nm). Plotted also is a Lorenzian fit, which reproduces the measured spectrum

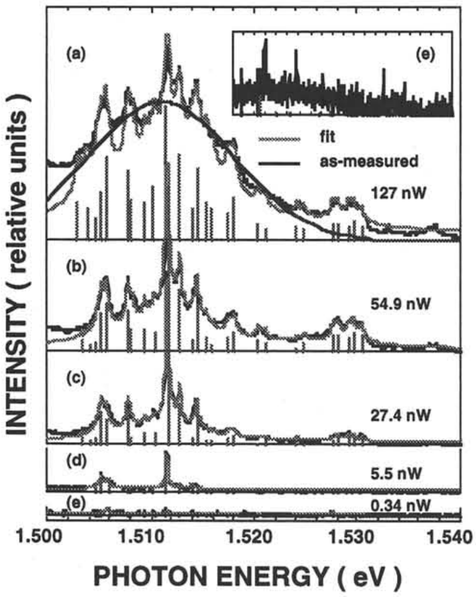

Fig.1 10-K microscope photoluminescence spectra taken on a 3-nm $In_{0.3}Ga_{0.7}As$ disk sample. Excitation by 514.5-nm line of

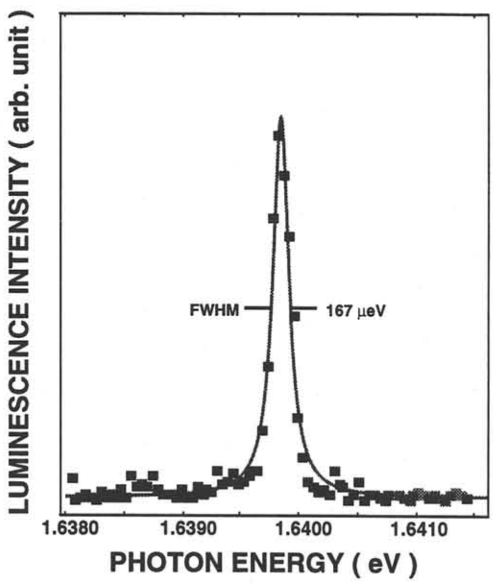

Fig.2 10-K microscope photoluminescence spectrum of a single dot in a 3-nm $In_{0.3}Ga_{0.7}As$ sample. Also shown is a Lorenzian lineshape fitting with a FWHM of 167 μeV.

quite well, as expected theoretically for *single* dot. This implies that we truely observe an isolated dot. The Lorenzian lineshape indicates that relaxation process of the dot dipole moment obeys an expronential function in time-evolution.

Having known the luminescence lineshape functional, we have undertaken lineshape fittings on all the spectra in Fig.1. Results are shown in the same figure as gray spectra with vertical lines representing the energies and intensities of the observed linese. Note that for spectra taken under very low excitation, Lorenzian lineshape was found to be a good approximation as in Fig.2. While for high excitation the lineshapes has broadened tail; simple Lorenzian lineshape unables to fit the spectral responce between each PL line in Fig.1. PL intensity and linewidth are then extracted and plotted for several PL lines as functions of pumping intensity in Fig.3(a) and Fig.3(b). The saturation of the PL intensity at rather a low excitation is well demonstrated in Fig3(a). Shown in Fig.3(b) is linewidth vs. excitaion intensity relations. Apprently the PL linewidth increases for higher excitation, proving the well-known basic physics of absorption saturation and simultaneous linebroadening. Therefore, the featureless spectral responce between PL lines, growing and letting the overall spectrum smooth for higher excitation is due to the tail broadening of each lines. Note that for some weak lines the line broadening was found to be absent.

Figure 4 shows a variety of PL and PLE spectra taken on a small region in the same sample. The center of distribution in this sample was 1.69 eV, therefore we looked for the isolated dot luminescence below this energy. Each of PLE spectrum shows a series of distinctive narrow absorption peaks. Such spectra proves undoubtedly fully-quantized discrete levels of the dots, and are in a marked contrast to the case in self-organized InAs/GaAs dots grown by molecular beam epitaxy, where PLE spectrum reveals only a series of phonon-related transitions.[7] It is also noteworthy that we observed no luminescence lines corresponding to the series of strong and sharp absorption features. This leads us to conclude that the sharp PL lines are due to the ground state transitions. Note that the second excited states, e.g. in the spectra (a) and (b), are doublets, inferring the disk is not cylindrical but of two-hold symmetry under the rotation along the growth axis. One may notices as well bunches of small features distributing over all the spectral range above the ground state, which resemble to a continuum. These features, however, seem to be composed of numbers of small peaks and dips, and they were reproducible under a number of measurements. It was also found that each

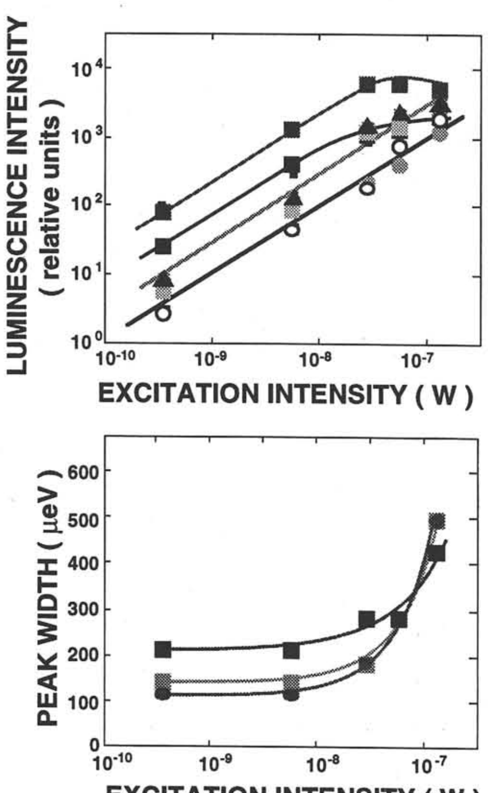

Fig.3 (a) PL intensity vs excitation intensity relations for eight different dots. (b) PL linewidth vs excitation intensity relations for the same set of dots.

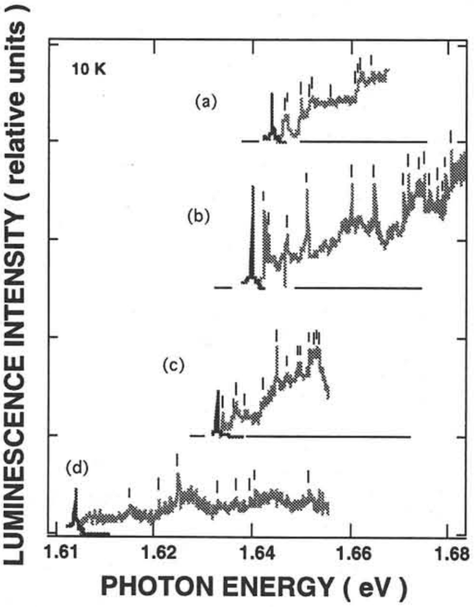

Fig.4 10-K microscope photoluminescence excitation spectrum of four single-dots. The leftmost spectrun in each dot is the ground state luminescence.

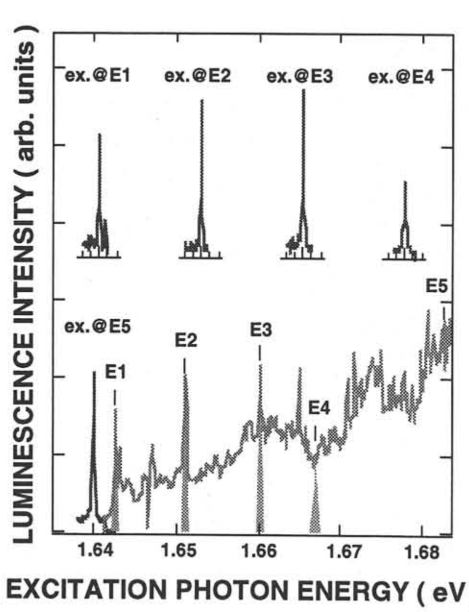

Fig.5 Bottom:PLE spectrum of the same dot (b) in Fig.3. Upper:Series of the ground state luminescences taken under different excitation energies, E1, E2, E3, E4 and E5.

of the nearby dots showed absolutely different spectral features as shown in Fig.4: None of the feature was common in different dots, i.e., no common band-states such as 2D- or 3D-band, were found as feeding states to the dots. In another word, since each disk is unique in size and composition, each set of PL and PLE spectra is unique, representing the fingerprint of the underlining disk. This is actually the reason why one could select some specific disks by optical probe. Therefore these pseudo-continuum background is attributed to the numerous number of weak transitions. Another experimental proof to the nonrelevance of the contiguous states is the following: We chose the dots (b) and excited it with different energies, E1,E2, E3, E4 and E5 (Fig.5). In all cases, none of the enhancement of the background signal was observed. Even when excitation is off-resonant with a distinct absorption peak, as in the case of E4, the observed peak show no enhanced background signal. It is therefore concluded that "pseudo-contiguous" absorption actually consist of large number of sharp absorption lines of weak intensity. Pressumablly they are to be optically prohibitted. They may gain finit optical strength due to the breakdown of the optical selection rule: the low symmetry of the system may accout this. Other reasons are still open for future studies such that undertaking polarization resolved absorption measurements, thereby revealing the nature of each transition.

The observed sharp and dintinctive emissions and absorptions as well as the ease of PL saturation are strong manifestations of fully quantized discrete levels of the zero-dimensional system. Therefore, single-dots have in fact been examined. They exhibits natural lineshape with homogeneous width. Because of their Lorenzian lineshape, the time-evolution of the relevant transition should obey a pure single exponential decay function (Fourier transformation of Lorenzian): the narrowest line observed gives a lowerbound (resolution of spectrometer) of such (phase) relaxation time, 11 psec. While excitation intensity is high, the saturation of absorption as well as the lineshape broadening occur as expected theoretically: possiblly other interactions with phonons or carriers are getting nonnegligible. Indeed the direct Fourier transformation of the observed broadened lineshape gives a multi-component decay function, proving the participation of multiple processes.

Our measurements reveal most of the characteristics expected for the single isolated dot except the following. In our InGaAs disk system the relaxation from the excited to ground states is fast enough as compared to the excited-state radiative recombination time, since we have observed no excited-state luminescence. So-called phonon bottle-neck[8,9] is therefore seems negligeble. Further study concerning the dynamics of the excited population of exciton will elucidate this problem.

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