floating-zone method. Some of these crystals were used as reference samples for systematic doping studies using Fe, Zn, and Ni [G. D. Gu *et al.*, *J. Cryst. Growth* **130**, 325 (1993); *ibid.* **137**, 472 (1994); D.-S. Jeon *et al.*, *Physica C* **253**, 102 (1995)]. The data presented here were obtained from a sample with T_c = 88 K, near optimal in the $Bi_2Sr_2CaCu_2O_{8+\delta}$ system. The results were reproduced in three samples from the same growth batch by Gu and colleagues. The spectra from these samples show a systematic set of subtle but important differences from other samples that we have used before [D. B. Mitzi *et al.*, *Phys. Rev. B* **41**, 6564 (1990)]. The details of these differences, their relation to the effects reported here, and the implications will be published elsewhere.
5. D. S. Dessau *et al.*, *Phys. Rev. Lett.* **71**, 2781 (1993).
6. D. S. Dessau *et al.*, *ibid.* **66**, 2160 (1991); Y. Hwu *et al.*, *ibid.* **67**, 2573 (1991); M. Norman *et al.*, *ibid.* **79**, 3506 (1997).
7. M. Randeria *et al.*, *ibid.* **74**, 4951 (1995).
8. H. Ibach and H. Luth, *Solid-State Physics* (Springer, New York, 1995).
9. For an overview, see R. B. Laughlin, *J. Phys. Chem. Solids* **56**, 1627 (1995); A. Moreo *et al.*, *ibid.*, p. 1645; R. Preuss *et al.*, *ibid.*, p. 1659; Y. Otta *et al.*, *ibid.*, p. 1741.
10. R. B. Laughlin, *Phys. Rev. Lett.* **79**, 1726 (1997).
11. V. J. Emery, S. A. Kivelson, O. Zachar, *Phys. Rev. B* **56**, 6120 (1997); *Physica C* **282–287**, 174 (1997).
12. B. G. Levi, *Phys. Today* **49** (no. 6), 17 (1996).
13. J. M. Harris *et al.*, *Phys. Rev. B* **54**, R15665 (1996).
14. D. S. Marshall *et al.*, *Phys. Rev. Lett.* **76**, 4841 (1996); A. G. Loeser *et al.*, *Science* **273**, 325 (1996); H. Ding *et al.*, *Nature* **382**, 51 (1996).
15. J. M. Tranquada *et al.*, *Nature* **375**, 561 (1995).
16. S. W. Cheong *et al.*, *Phys. Rev. Lett.* **67**, 1791 (1991); T. E. Mason *et al.*, *ibid.* **68**, 1414 (1992); T. R. Thurston *et al.*, *Phys. Rev. B* **46**, 9128 (1992).
17. K. Yamada *et al.*, in preparation.
18. H. J. Schultz, *J. Phys. (Paris)* **50**, 2833 (1989).
19. D. Poilblanc and T. M. Rice, *Phys. Rev. B* **39**, 9749 (1989); J. Zaanen and O. Gunnarson, *ibid.* **40**, 7391 (1989); S. A. Kivelson and V. J. Emery, *Physica C* **235–240**, 189 (1994).
20. S. R. White and D. J. Scalapino, cond-mat/9801274, Los Alamos e-Print Archive at xxx.lanl.gov (1998).
21. W. A. Groen, D. M. de Leeuw, L. F. Feiner, *Physica C* **165**, 55 (1990).
22. M. V. Zimmermann *et al.*, *Europhys. Lett.*, in press.
23. A. Bianconi *et al.*, *Phys. Rev. Lett.* **76**, 3412 (1996).
24. X.-G. Wen and P. A. Lee, cond-mat/9709108, Los Alamos e-Print Archive at xxx.lanl.gov (1997).
25. G. A. Sawatzky, *Nature* **342**, 480 (1989); P. W. Anderson, *Phys. Rev. Lett.* **67**, 2569 (1991).
26. Although ARPES only measures the single-particle spectral function, this connection is empirically justified because its strength correlates with T_c (*27*) and T_c scales with superfluid density (*29*) in the underdoped regime.
27. P. J. White *et al.*, *Phys. Rev. B* **54**, R15669 (1996).
28. B. O. Wells *et al.*, *Phys. Rev. Lett.* **74**, 964 (1995).
29. Y. J. Uemura, in *Proceedings of the CCAST Symposium on High-T_c Superconductivity and C60 Family, Beijing, 1994*, S. Feng and H. C. Ren, Eds. (Gordon & Breach, New York, 1995), pp. 113–142.
30. We acknowledge S. A. Kivelson, S.-C. Zhang, R. B. Laughlin, A. Puchkov, Z. Hasan, P. A. Lee, A. J. Millis, P. Coleman, and G. A. Sawatzky for helpful discussions. Photoemission experiments were performed at the Stanford Synchrotron Radiation Laboratory, which is operated by the U.S. Department of Energy's Office of Basic Energy Science, Division of Chemical Sciences. The Office's Division of Material Science provided support for this research.

18 December 1997; accepted 20 February 1998

Optical Studies of Individual InAs Quantum Dots in GaAs: Few-Particle Effects

L. Landin, M. S. Miller,* M.-E. Pistol,† C. E. Pryor, L. Samuelson

Optical emission from individual strained indium arsenide (InAs) islands buried in gallium arsenide (GaAs) was studied. At low excitation power density, the spectra from these quantum dots consist of a single line. At higher excitation power density, additional emission lines appeared at both higher and lower energies, separated from the main line by about 1 millielectron volt. At even higher excitation power density, this set of lines was replaced by a broad emission peaking below the original line. The splittings were an order of magnitude smaller than the lowest single-electron or single-hole excited state energies, indicating that the fine structure results from few-particle interactions in the dot. Calculations of few-particle effects give splittings of the observed magnitude.

Semiconductor quantum dots have been the subject of intense research in recent years. These heterostructures consist of nanometer-scale islands of one type of semiconductor either embedded in a different semiconductor or free-standing on a suitable substrate. The materials are chosen such that electrons and holes are confined to the island, resulting in a discrete spectrum for the confined charges. The spectrum of a quantum dot provides information about its internal structure, similar to the spectrum of an atom or a molecule. Unlike atoms and molecules, however, quantum dots suffer from unavoidable variation in their size, and hence, their energy levels. Previous measurements most often averaged over many dots, making it difficult to disentangle the features related to single-dot physics from those arising from dot-to-dot variation.

Recent studies of individual semiconductor quantum dots showed departures from a single-particle description of quantum dots, using magnetoconductance measurements (*1*) and differential conductance measurements (*2*). In these studies, the quantum dots were filled with only one type of charge carrier (electrons), leading to strong Coulomb effects. Studies of photon emission from localized sites arising from interface fluctuations in quantum wells showed the existence of excitons (*3*, *4*) and bi-excitons (*4*), which consist of two different types of charge carriers, electrons and holes. Tri-excitons have been observed in the CuCl system (*5*). Even studies of individual pairs of quantum dots have been perfomed, showing clear effects of coupling between the quantum dots (*6*). We expect that studies of individual quantum dots will become increasingly important in the near future, similar to the situation with single-molecule studies (*7*). The relative ease of positioning of quantum dots, in comparison with single atoms (for which atomic traps are needed), will faciliate experiments in quantum computing (*8*), to use one example. In such experiments, it will be necessary to control the electronic states of the quantum dots using, for example, fast pulsed lasers.

One extensively studied quantum dot system is InAs islands embedded in GaAs. Indium arsenide has a 7.2% larger lattice constant than GaAs, and only a thin layer of InAs can be grown on a GaAs surface before the film breaks up into small islands on a thin InAs wetting layer (*9*). When embedded in GaAs, the InAs islands are small enough to confine the electronic states strongly in all three dimensions, making good quantum dots with low-temperature luminescence energies between 1.0 and 1.4 eV. The wetting layer behaves as a thin quantum well. Several groups have formed such islands on flat substrates, using molecular beam epitaxy (*10–12*), metal-organic chemical vapor deposition (*13*, *14*), and chemical beam epitaxy (*15*). The optical emission spectra of individual dots are of great interest, because the spectra characterize the electronic structure and thus determine the properties available for optical or electrical device applications. Recently, optical emission spectra for individual InP quantum dots in barriers of GaInP were reported to have multiple emission lines for each InP dot even at low excitation power density (*16*). The InP dot spectral features were in good agreement with detailed electronic structure calculations (*17*). Spectra with narrow emission lines from small numbers of InAs dots in GaAs (*12*, *18*, *19*) have

Department of Solid State Physics, Lund University, Box 118, S-221 00 Lund, Sweden.

*Present address: Department of Electrical Engineering, University of Virginia, Thornton Hall, Charlottesville, VA 22903–2442, USA.
†To whom correspondence should be addressed. E-mail: mats-erik.pistol@ftf.lth.se

also been reported. The narrow lines are compelling evidence for three-dimensional confinement in the quantum dots. However, the forests of lines in the reported InAs spectra prevented determination of the number of emission lines per dot. The higher surface density of InAs islands compared to InP islands has so far prevented the study of emission spectra of individual InAs islands.

We investigated a large number of individual InAs quantum dots (about 50) on special low-density samples. The evolution of the optical emission spectra with increasing optical pumping first shows one line per island, followed by multiplets of lines, and finally a broad emission from each dot. Comparison with calculations demonstrates that the multiplets and broad emission must be due to few- and many-body interactions. We propose that the multiple peaks at intermediate excitation power density are a state-filling effect in which the dot is filled with more than two carriers (that is, more than one electron and one hole).

The sample used in our experiment was grown by chemical beam epitaxy. Regions of the sample were patterned with etched grooves and holes for selective island placement, and most of the islands grew in patterned areas (*20*). Photoluminescence measurements were made in regions with low island density away from the patterned features. The InAs islands were capped with a 20-nm layer of GaAs. Growth and processing details are reported elsewhere (*15*, *20*). Although there is presently no consensus on InAs island shape, high-resolution atomic force micrographs of similar islands from our group suggest a truncated pyramidal shape with a height of ~4 nm and a diameter of ~20 nm at the island base (*21*). Such shapes are strongly reminiscent of the shapes of InP, which are larger by a factor of 2 to 4 and, hence, are easier to resolve (*22*). On the basis of similarities of the present InAs island growth conditions and resulting luminescence energies with those reported by other groups, we believe that the islands we investigated are not unusual.

All our experiments were carried out at 7 K. For excitation, we used a frequency-doubled yttrium-aluminum-garnet laser emitting at 532-nm wavelength. The diameter of the laser spot was kept to about 50 μm, and the excitation power density was varied between 0.5 and 160 W/cm^2. The emitted light from the sample was collected by a microscope and either dispersed through a monochromator or analyzed and imaged through a band-pass interference filter. In both cases, we used a camera equipped with a charge-coupled device (CCD) for detection. When we used the monochromator, the image of the sample was focused through a narrow slit with the slit oriented parallel to the grating rulings. Thus, the image on the CCD was spectrally resolved in one dimension and spatially resolved in the other (Fig. 1). The monochromator spectral resolution was typically 0.1 nm. In this way, many dots could be simultaneously measured with their respective spectra still separated.

At low excitation power density (1.6 W/cm^2 or less), the emission spectra consisted of a single line (Fig. 2, A through D). The single narrow emission line is evidence of single dot emission. At higher excitation power density (around 5 W/cm^2), we saw a second line (labeled b in Fig. 2) at about 1 meV higher energy than the main line. A further increase in excitation power density results in additional lines at lower energy separated by about 1 meV from each other. Finally, at the highest excitation power density we used (160 W/cm^2), we observed a continuum-like emission. This broad emission peaked at an energy below that of the initial line. We found these behaviors to be shared by most of the quantum dots that we investigated. In Fig. 2, we also show the luminescence peak from the wetting layer at 1.44 eV. The wetting layer peak begins to show band-filling effects at pump intensities near 160 W/cm^2, that is, at the same excitation power density at which we observed the continuum-like emission from the quantum dots.

An increasing number of spectral lines at larger excitation densities are usually associated with state filling. At high excitation

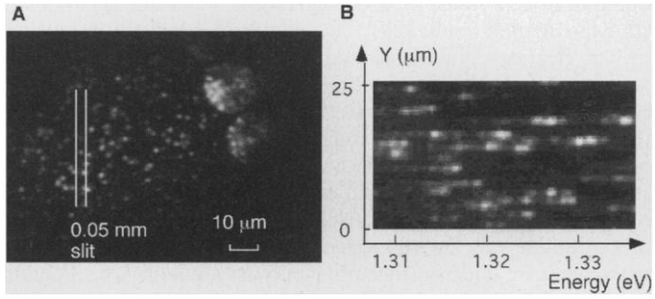

Fig. 1. A micrograph of the emission from the sample (**A**), showing emission from individual quantum dots. The emission wavelength is 960 nm, and the band-pass of the interference filter is 10 nm. In (A), we also show the size of a 0.05-mm slit in our optical setup. (**B**) A wavelength-dispersed image. The luminescing area is defined by the extension of the laser spot. The 20-μm diameter circular regions above and to the right of the center are concentric circular trenches for which island luminescence energy was found to vary as a function of position around the circles (*27*). Away from the patterns, the density of dots is very low, and the luminescence from isolated dots can be seen. The surface density of individual luminescing dots near the center of the image is ~0.03/μm^2, as observed with this filter. This value agrees with the density measured by atomic force microscopy on an otherwise similar, but uncapped, sample (*20*). This density is significantly lower than the typical reported densities of between 10^9 and 10^{11} cm^{-2}.

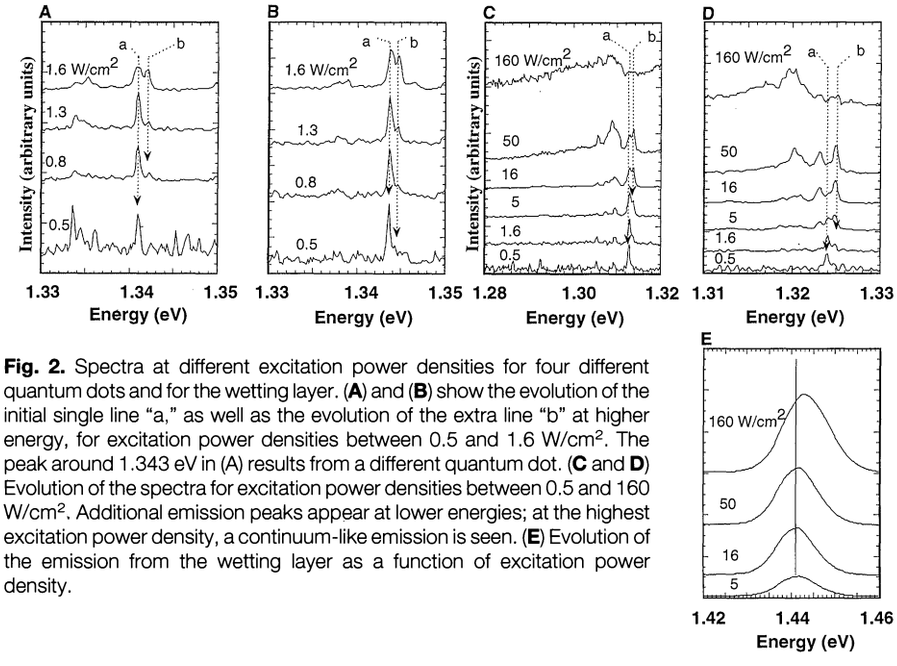

Fig. 2. Spectra at different excitation power densities for four different quantum dots and for the wetting layer. (**A**) and (**B**) show the evolution of the initial single line "a," as well as the evolution of the extra line "b" at higher energy, for excitation power densities between 0.5 and 1.6 W/cm^2. The peak around 1.343 eV in (A) results from a different quantum dot. (**C** and **D**) Evolution of the spectra for excitation power densities between 0.5 and 160 W/cm^2. Additional emission peaks appear at lower energies; at the highest excitation power density, a continuum-like emission is seen. (**E**) Evolution of the emission from the wetting layer as a function of excitation power density.

density, states fill up and emission is seen from recombination involving excited states. The challenge presented by the spectra in Figs. 2 and 3 is to understand how state filling can produce lines below the initial line, and why the additional lines are so closely spaced. To address this point, we calculated the electronic energy levels in these quantum dots, using an eight-band **k.p** theory in the envelope function approximation. The model includes strain, a realistic shape of the quantum dots, and the piezoelectric potential. More extensive calculation results are presented in detail elsewhere (*23*). The calculation was done for a dot with a height of 4.5 nm, which reproduces the observed transition energies best. We find that there is only one confined-state energy for the electrons, and that for the holes, the first excited state is 25 meV away from the ground state, in good agreement with experimental results (*24*). These results rule out electron- or hole-excited states as the source of the observed extra emission peaks. Because of time reversal invariance, the confined states in the dot are doubly degenerate. Therefore, the dot can be filled with two electrons and two holes without occupying any of the single-particle excited states. Because we are interested only in states that can undergo recombination to produce a photon, there are four possibilities involving the single-particle ground states: e + h $\rightarrow$ photon, e + 2h $\rightarrow$ h + photon, 2e + h $\rightarrow$ e + photon, and 2e + 2h $\rightarrow$ e + h + photon, where e and h each stand for a single electron or hole, respectively. For noninteracting particles, all four of the resulting photons would have the same energy. However, the Coulomb interaction changes the energies of the multiparticle states such that they are no longer the sum of single particle energies. The relative shifts can be positive or negative, depending on details of the well shape (*25*). Hence, four distinct lines should be seen, and the number of observed lines should depend on the excitation power.

We determined the photon energies by computing the multiparticle energies in the self-consistent Hartree approximation. The resulting shifts with respect to a single recombining electron-hole pair (e + h) are –1.6 meV for one additional electron (2e + h), +4.3 meV for an additional hole (e + 2h), and +2.6 meV for an additional exciton (2e + 2h). The observed spectra do, indeed, have two lines above the original and one at lower energy, in agreement with our calculation. The Hartree approximation includes neither exchange nor correlation. These effects will certainly alter the shifts and may also cause additional splittings. However, the Hartree approximation determines the magnitude of the splittings and provides an existence proof for a negative shift. Our model predicts four lines within a few millielectron volts that fill states both above and below the single line seen at low power density. For some systems, polariton effects can contribute a splitting to the fine structure, because two dots are within a wavelength of light from one another and emit at the same energy (*26*). Polariton effects should be negligible for our sample with low density of islands, because within a given 1-meV range of energy, the dot-to-dot separation is much larger than the wavelength of the emitted light. The continuous emission from the quantum dots appears simultaneously with the occurrence of band-filling in the wetting layer (Fig. 2). We attribute the continous emission from the quantum dots to an interaction between the band-filled wetting layer and the state-filled dot.

It is interesting to compare our results with results obtained on single InP quantum dots (*16*). In the InP system, several emission lines were observed, even at the lowest excitation power density used. The observed splittings were at energies that correspond to the calculated valence band splittings (*17*). In light of our results, which show a pronounced excitation power density dependence, we confirm that the fine structure, obtained at low excitation power density, of the InP dots is indeed a result of the valence band splittings.

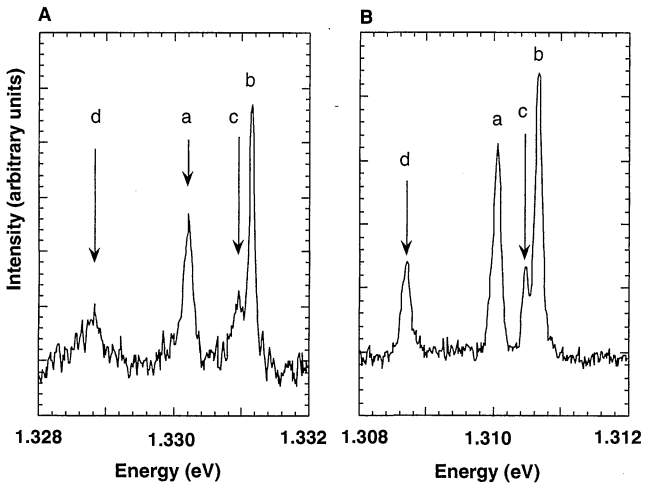

Fig. 3. High-resolution spectra of quantum dots showing fine structure, generated with an intermediate excitation power density (16 W/cm^2). Four peaks are shown for each island and are labeled "a" to "d," where a is the original single peak, b is the same as in Fig. 2, c is the shoulder peak just below b, and d is the low-energy peak. For these two islands, the lines a and d have the same splitting of ~1.4 meV with respect to one another, and the lines b and c have the same splitting of 0.25 meV. The a–d and b–c pair have different relative spacings between the islands, where the a–b splitting of the higher-energy island is 1.00 meV and of the lower-energy island is 0.65 meV.

REFERENCES AND NOTES

1. D. R. Stewart, D. Sprinzak, C. M. Marcus, C. I. Duruöz, J. S. Harris Jr., *Science* **278**, 1784 (1997).
2. L. P. Kouwenhoven *et al.*, *ibid.*, p. 1788.
3. D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, D. Park, *ibid.* **273**, 87 (1996).
4. K. Brunner, G. Abstreiter, G. Böhm, G. Tränkle, G. Weimann, *Phys. Rev. Lett.* **73**, 1138 (1994).
5. M. Ikezawa, Y. Masumoto, T. Takagahara, S. V. Nair, *ibid.* **79**, 3522 (1997).
6. G. Schedelbeck, W. Wegscheider, M. Bichler, G. Abstreiter, *Science* **278**, 1792 (1997).
7. R. F. Service, *Science* **276**, 1027 (1997).
8. E. Knill, R. Laflamme, W. H. Zurek, *ibid.* **279**, 342 (1997).
9. L. Goldstein, F. Glas, J. Y. Marzin, M. N. Charasse, G. L. Roux, *Appl. Phys. Lett.* **47**, 1099 (1985).
10. D. Leonard, M. Krishnamurthy, C. M. Reaves, S. P. DenBaars, P. M. Petroff, *ibid.* **63**, 3202 (1993).
11. S. Guha, A. Madhukar, K. C. Rajkumar, *ibid.* **57**, 2110 (1990).
12. J.-Y. Marzin, J.-M. Gerard, A. Israel, D. Barrier, G. Bastard, *Phys. Rev. Lett.* **73**, 716 (1994).
13. S. P. DenBaars, C. M. Reaves, V. Bressler-Hill, S. Varma, H. Weinberg, *J. Cryst. Growth* **145**, 721 (1994).
14. N. Carlsson *et al.*, *Appl. Phys. Lett.* **65**, 3093 (1994).
15. M. S. Miller *et al.*, *Solid State Electron.* **40**, 609 (1996).
16. D. Hessman, P. Castrillo, M.-E. Pistol, C, Pryor, L. Samuelson, *Appl. Phys. Lett.* **69**, 749 (1996).
17. C. Pryor, M.-E. Pistol, L. Samuelson, *Phys. Rev. B* **56**, 10404 (1997).
18. M. Grundmann *et al.*, *Phys. Rev. Lett.* **74**, 4043 (1995).
19. R. Leon, P. M. Petroff, D. Leonard, S. Fafard, *Science* **267**, 1966 (1995).
20. S. Jeppesen *et al.*, *Appl. Phys. Lett.* **68**, 2228 (1996).
21. T. Junno and M. S. Miller, unpublished data.
22. M.-E. Pistol *et al.*, *Proceedings of the 23rd International Conference on the Physics of Semiconductors*, Berlin, 21 to 26 July 1996, M. Scheffler and R. Zimmermann, Eds. (World Scientific, Singapore, 1996), p. 1317.
23. C. E. Pryor, http://xxx.lanl.gov/abs/cond-mat/9710304
24. K. H. Schmidt, G. Medeiro-Ribeiro, M. Oestereich, P. M. Petroff, G. H. Döhler, *Phys. Rev. B* **54**, 11346 (1996).
25. Ph. Lelong and G. Bastard, *Solid State Commun.* **98**, 819 (1996).
26. D. S. Citrin, *Opt. Lett.* **20**, 901 (1995).
27. M. S. Miller *et al.*, *J. Cryst. Growth* **175/176**, 747 (1997).

28. We thank S. Jeppesen, B. Kowalski, and I. Maximov for the growth and processing of the sample. We also thank D. Citrin and B. Kowalski for clarifying discussions. This work was performed within the Nanometer Structure Consortium in Lund, Sweden, and was supported by grants from the Swedish Natural Science Research Council (NFR), the Swedish National Board for Industrial and Technical Development (NUTEK), the Swedish Research Council for Engineering Sciences (TFR), and the Swedish Foundation for Strategic Research (SSF).

1 December 1997; accepted 19 February 1998