Observation of Discrete Electronic States in a Zero-Dimensional Semiconductor Nanostructure

M. A. Reed, J. N. Randall, R. J. Aggarwal, ^(a) R. J. Matyi, T. M. Moore, and A. E. Wetsel^(b)

Central Research Laboratories, Texas Instruments Incorporated, Dallas, Texas 75265 (Received 2 October 1987)

Electronic transport through a three-dimensionally confined semiconductor quantum well ("quantum dot") has been investigated. Fine structure observed in resonant tunneling through the quantum dot corresponds to the discrete density of states of a zero-dimensional system.

PACS numbers: 73.20.Dx, 72.15.Rn, 72.70.+m, 73.40.Gk

Carrier confinement to reduced dimensions in a semiconductor was first demonstrated in GaAs-AlGaAs quantum wells by electronic¹ and optical² spectroscopy in 1974. This achievement had led to numerous important developments in basic semiconductor physics and device technology. Structures produced by ultrathin-film growth are inherently two dimensional, and thus investigations have been largely confined to heterostructures where only the carrier momentum normal to the interfaces is quantized. Recent advances in microfabrication technology $^{3-5}$ have allowed the fabrication of structures with quantum confinement to one dimension ("quantum wires")^{6,7} and have initiated intriguing investigations into one-dimensional physics, such as localization and electron-electron interaction,^{8,9} single-electron trapping,¹⁰ and universal conductance fluctuations.¹¹ It is expected that the realization of semiconductor heterostructures with quantum confinement to zero dimensions ("quantum dots") will yield equally intriguing phenomena. Attempts to observe confinement optically have been reported recently,¹²⁻¹⁶ but the spectra do not show the characteristic structure of a series of isolated peaks expected from a zero-dimensional electron-hole gas. We have therefore studied such structures by electronic transport, and in this Letter present evidence for electronic transport through a discrete spectrum of states in a nanostructure confined in all three spatial dimensions.

The approach used to produce quantum-dot nanostructures suitable for electronic transport studies was to confine resonant-tunneling heterostructures laterally with a fabrication-imposed potential.¹⁷ This approach embeds a quasibound quantum dot between two quantum-wire contacts. The initial molecular-beamepitaxial structure is a $0.5-\mu m n^+$ -GaAs contact (Si doped at 2×10^{18} cm⁻³, graded to approximately 10^{16} cm⁻³ over 200 Å, followed by a 100-Å undoped GaAs spacer layer), a 40-Å Al_{0.25}Ga_{0.75} As tunnel barrier, and a 50-Å undoped $In_xGa_{1-x}As$ quantum well. The structure was grown to be nominally symmetric about a plane through the center of the quantum well. Employing a $In_xGa_{1-x}As$ quantum well allows one to lower the quantum well states with respect to the conduction-band edge while keeping the vertical dimensions fixed; x values

studied ranged from 0 to 0.08. Large-area ($\ge 2 \mu m$ square) mesas of a typical structure (x = 0.08) fabricated by conventional means exhibited two resonant peaks: a ground state at 50 mV with a peak current density of 30 A/cm², and an excited state at 700 mV with a peak current density of 8.1×10³ A/cm², both measured at 77 K.

Electron-beam lithography defined an ensemble of AuGe/Ni/Au Ohmic metallization dots (single- or multiple-dot regions), nominally 1000-2500 Å in diameter, on the top n^+ -GaAs contact by use of a bilayer polymethylmethacrylate (PMMA) resist and liftoff. The metal-dot Ohmic contact served as an etch mask for highly anisotropic reactive-ion etching with BCl₃ as an etch gas, defining columns in the epitaxial structure. A scanning electron micrograph of a collection of these etched structures is seen in Fig. 1. To make contact to the tops of the columns, a planarizing and insulating polyimide was spun on the sample and then etched back by O₂ reactive-ion etching to expose the metal contacts on the tops of the columns. A gold contact pad was then



FIG. 1. A scanning electron micrograph of various size GaAs nanostructures containing quantum dots. The dark region on top of the column is the electron-beam defined Ohmic contact and etch mask. The horizontal bars are $0.5 \,\mu$ m.



FIG. 2. Schematic illustration of the vertical (a - a') and lateral (b - b') potentials of a column containing a quantum dot, under zero and applied bias. $\Phi(r)$ is the (radial) potential, R is the physical radius of the column, r is the radial coordinate, W is the depletion depth, Φ_T is the height of the potential determined by the Fermi-level (E_F) pinning, and $E_{c,\Gamma}$ is the Γ -point conduction-band energy.

evaporated over the top(s) of the column(s). The bottom conductive substrate provided electrical continuity.

Figure 2 schematically illustrates the lateral (radial) potential of a column containing a quantum dot, and the spectrum of three-dimensionally confined electron states under zero and applied bias. A spectrum of discrete states will give rise to a series of resonances in transmitted current as each state drops below the conductionband edge of the injection contact. To observe lateral quantization of quantum well state(s), the physical size of the structure must be sufficiently small that quantization of the lateral momenta produces energy splittings > kT. Concurrently, the lateral dimensions of the structure must be large enough that pinchoff of the column by the depletion layers formed on the sidewalls of the GaAs column does not occur. As a result of the Fermi-level pinning of the exposed GaAs surface, the conduction band bends upward (with respect to the Fermi level), and where it intersects the Fermi level determines in real space the edge of the central conduction-path core. We can express the radial potential $\Phi(r)$ in the column [for $(R-W) \le r \le R$], assumed axially symmetric, as

$$\Phi(r) = \Phi_T [1 - (R - r)/W]^2, \tag{1}$$

where r is the radial coordinate, R is the physical radius of the column, W is the depletion depth, and Φ_T is the height of the potential determined by the Fermi-level pinning. When the lateral dimension is reduced to 2W or less, the lateral potential becomes parabolic though conduction through the central conduction-path core is pinched off.

A structure that satisfies both constraints was achieved with a In_{0.08}Ga_{0.92}As quantum-well double-barrier structure with a physical (lithographic) lateral dimension of $\simeq 1000$ Å. Figure 3 shows the current-voltage characteristics of this (single) microstructure as a function of temperature. If we assume that the current density through the structure is approximately the same as in a large-area device, measurement of the peak resonant current implies a minimum (circular) conduction-path core of 130 Å for this structure; thus, a lateral parabolic potential approximation is valid. This implies a depletion depth of \simeq 430 Å at the double-barrier structure, in reasonable agreement with that expected from the known doping level (at 2×10^{18} cm⁻³, W = 220 Å) and with the realization that W will enlarge in the undoped double-barrier region. The splitting of the discrete electron levels in the quantum dot is then

$$\Delta E = (2\Phi_T/m^*)^{1/2}\hbar/R, \qquad (2)$$

where m^* is the effective mass of the electrons in the quantum well (linearly extrapolated between that of GaAs and InAs) and R is the physical radius. With a Fermi-level pinning of 0.7 eV, the states should be split



FIG. 3. Current-voltage characteristics of a single quantum-dot nanostructure as a function of temperature, showing resonant tunneling through the discrete states of the n=2 quantum well resonance. The arrows indicate the voltage positions of the discrete states for the T=1.0-K curve.

evenly by 26 meV.

Only the excited-state resonance of this structure is observed since the current expected from the groundstate resonance is at the detection limits of the test apparatus. At high temperature, the characteristic negative differential resistance of the double-barrier structure is evident. As the temperature is lowered, two effects occur. First, the resonant peak shifts slightly higher in voltage (because of the increase of the GaAs contact resistance with temperature) and decreases in current (because of the freezing out of excess leakage current). Secondly, there appears a series of peaks superimposed on the negative-differential-resistance peak. In the range 0.75-0.9 V the peaks are approximately equally spaced, with a splitting of $\simeq 50$ mV. Under the assumption that most of the bias is incrementally dropped across the double-barrier structure, the splitting of the equally spaced series is 25 meV, in excellent agreement with the value determined from the physical dimension of the structure. We believe that the structure observed here corresponds to resonant tunneling through the spectra of discrete quasibound (in the z direction) states in the quantum dot which correspond to the density of states of a three-dimensional semiconductor quantum well.

Another peak, presumably the ground state of the harmonic-oscillator potential, occurs $\approx 80 \text{ mV}$ below the equally spaced series. The origin of this anomalously large splitting is not understood, though nonparabolicity of the lateral confining potential cannot be ruled out.¹⁸

In conclusion, we have measured electronic transport through quantum well states that have been laterally confined in all three dimensions (by heterojunction barriers in one dimension and a fabrication-imposed potential in the lateral dimensions) on the nanometer scale. We have performed electrical spectroscopy in the form of resonant tunneling through the spectrum of electron states, and observe resonances that correspond to the density of states of a zero-dimensional system.

We are indebted to R. T. Bate, W. R. Frensley, J. H. Luscombe, and R. H. Silsbee for helpful discussions and analysis, and to R. K. Aldert, D. A. Schultz, P. F. Stickney, and J. R. Thomason for technical assistance. This research was supported in part by the U.S. Army Research Office and the U.S. Office of Naval Research.

^(a)Present address: Massachusetts Institute of Technology, Cambridge, MA 02139.

^(b)Present address: Harvard University, Cambridge, MA 02138.

¹L. L. Chang, L. Esaki, and R. Tsu, Appl. Phys. Lett. 24, 593 (1974).

 2 R. Dingle, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. 33, 827 (1974).

 3 A. N. Broer, W. W. Molzen, J. J. Cuomo, and N. D. Wittels, Appl. Phys. Lett. **29**, 596 (1976).

⁴R. E. Howard, P. F. Liao, W. J. Skocpol, L. D. Jackel, and H. G. Craighead, Science **221**, 117 (1983).

⁵H. G. Craighead, J. Appl. Phys. 55, 4430 (1984).

⁶H. Sakaki, Jpn. J. Appl. Phys. **19**, L735 (1980).

⁷W. J. Skocpol, L. D. Jackel, R. E. Howard, E. L. Hu, and L. A. Fetter, Physica (Utrecht) **117 & 118**, 667 (1983).

 8 R. G. Wheeler, K. K. Choi, A. Goel, R. Wisnieff, and D. E. Prober, Phys. Rev. Lett. **49**, 1674 (1982).

⁹T. J. Thornton, M. Pepper, H. Ahmed, D. Andrews, and G. J. Davies, Phys. Rev. Lett. **56**, 1198 (1986).

¹⁰K. S. Raals, W. J. Skocpol, L. D. Jackel, R. E. Howard, L. A. Fetter, R. W. Epworth, and D. M. Tennant, Phys. Rev. Lett. **52**, 228 (1984).

¹¹W. J. Sckocpol, P. M. Mankiewich, R. E. Howard, L. D. Jackel, D. M. Tennant, and A. D. Stone, Phys. Rev. Lett. **56**, 2865 (1986).

¹²M. A. Reed, R. T. Bate, K. Bradshaw, W. M. Duncan, W. R. Frensley, J. W. Lee, and H.-D. Shih, J. Vac. Sci. Technol. B 4, 358 (1986).

¹³K. Kash, A. Scherer, J. M. Worlock, H. G. Craighead, and M. C. Tamargo, Appl. Phys. Lett. **49**, 1043 (1986).

¹⁴J. Cibert, P. M. Petroff, G. J. Dolan, S. J. Pearton, A. C. Gossard, and J. H. English, Appl. Phys. Lett. **49**, 1275 (1986).

¹⁵H. Temkin, G. J. Dolan, M. B. Panish, and S. N. G. Chu, Appl. Phys. Lett. **50**, 413 (1987).

¹⁶R. L. Kubena, R. J. Joyce, J. W. Ward, H. L. Garvin, F. P. Stratton, and R. G. Brault, Appl. Phys. Lett. **50**, 1589 (1987).

¹⁷J. N. Randall, M. A. Reed, T. M. Moore, R. J. Matyi, and J. W. Lee, to be published; A. E. Wetsel, M. A. Reed, J. N. Randall, R. J. Aggarwal, R. J. Matyi, and T. M. Moore, to be published.

¹⁸M. Luban and D. L. Pursey, Phys. Rev. D 33, 431 (1986).



FIG. 1. A scanning electron micrograph of various size GaAs nanostructures containing quantum dots. The dark region on top of the column is the electron-beam defined Ohmic contact and etch mask. The horizontal bars are $0.5 \ \mu m$.



FIG. 2. Schematic illustration of the vertical (a-a') and lateral (b-b') potentials of a column containing a quantum dot, under zero and applied bias. $\Phi(r)$ is the (radial) potential, R is the physical radius of the column, r is the radial coordinate, W is the depletion depth, Φ_T is the height of the potential determined by the Fermi-level (E_F) pinning, and $E_{c,\Gamma}$ is the Γ -point conduction-band energy.