Fabrication of closely spaced quantum dot diodes

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Quantum dot devices have been proposed as the basic structure for an integrated circuit technology with extremely high functional density [R. T. Bate, Sci. Am. 258, 96 (1988)]. Electronic transport has been reported through quantum dot diodes which are resonant tunneling devices with lateral dimensions small enough to produce zero-dimensional (0D) confinement effects [M. A. Reed, J. N. Randall, R. J. Aggarwal, R. J. Matyi, T. M. Moore, and A. E. Wetsel, Phys. Rev. Lett. 60, 535 (1988); G. Faini, A. Ramdane, F. Mollot, and H. Launois, (to be published) NATO Workshop Spain 1990; S. Tarucha, Y. Hirayama, T. Saku, and T. Kimura, Phys. Rev. B 41, 5459 (1990)]. We seek improvements in the switching characteristics of these 0D tunneling devices as well as processing techniques to fabricate them very close together in an attempt to study field coupling between dots. We will discuss the processing issues involved with fabricating very closely spaced quantum dot diodes. We are building pairs of quantum dot diodes which are approximately 1500 ångstroms in diameter and spaced a few hundred ångstroms apart. e-beam lithography is used to define the dot patterns. The use of nonalloyed InGaAs contacts, which is essential to providing ohmic-type contacts of this small diameter, [J. N. Randall, C. H. Yang, Y. C. Kao, and T. M. Moore, J. Vac. Sci. Technol. B 7, 2007 (1989); P. Gueret, P. Buchmann, K. Daetwyler, and P. Vettiger, Appl. Phys. Lett. 55, 1735 (1989)], presents complications for the reactive ion etching of these structures. The task of independently contacting two very small and closely spaced contacts requires high resolution lithography and accurate re-alignment techniques. The e-beam lithography techniques used to accomplish this task will be presented. Finally some transport measurements through isolated quantum dot diodes will be presented.

I. INTRODUCTION

The search for a successor to the current integrated circuit (IC) technology has led us to investigate quantum effect devices which employ resonant tunneling. Resonant tunneling involves electronic transport which is far from equilibrium and has demonstrated gain and room temperature operation.\textsuperscript{1,2} Other devices employing quantum interference effects have been proposed.\textsuperscript{3,4} However, these types of devices operate in the near equilibrium transport regime and are probably restricted to cryogenic operation. Our interest in maximizing functional density has led to "zero-dimensional" quantum dot devices. It is unlikely that quantum dot devices will operate satisfactorily in conventional circuit architectures; therefore, investigations of quantum-compatible circuit architectures are being carried out. We are considering cellular automata architectures because they involve only short range interconnections which may be synergistic with quantum coupling mechanisms between closely spaced quantum dots. The type of cellular automata architecture envisioned for quantum coupled devices is a two-dimensional array where data is introduced and retrieved from the periphery of the arrays where only nearest-neighbor and possibly next-nearest-neighbor interaction between devices would be allowed. The functionality of the circuit is determined by the interaction rules for the devices. There are many possible interaction rules for devices in such an array, most of which do not lead interesting interaction. We are searching for interaction rules which satisfy the dual constraints of producing useful computation and of being realizable with some form of quantum coupling. This paper will primarily discuss the efforts to build structures which will be used to measure coupling between closely spaced vertical tunneling quantum dot devices.

II. COUPLING BETWEEN QUANTUM DOTS

There are several possible mechanisms for coupling between quantum dots, such as wave function overlap, charge exchange via tunneling, energy exchange via phonons or photons, and electromagnetic field interaction.

A. Wave function overlap and tunneling

Wave function overlap and tunneling between dots are very short range effects which may be exploited only with distances between dots of approximately 100 Å or less. Dot-to-dot resonant tunneling is being explored by a triple barrier quantum dot diode. In this structure there is electron tunneling from one dot to another and efforts to understand the coupling between the dots are underway.\textsuperscript{5} However, in these structures the dots are stacked vertically and the charge transport is also in the vertical direction. Some form of lateral coupling will be required to be compatible with a circuit consisting of a two dimensional lateral array. Coupling mechanisms in lateral arrays of quantum dots has been explored in systems which use depletion confinement.\textsuperscript{6-9} However, these structures either operate near equilibrium or use depletion regions as tunnel barriers which will limit their
operation to cryogenic temperatures. While lateral heterojunction tunneling structures can, in principle, be fabricated, there are significant advances in processing technology which must be made before this will be possible.

B. Optical coupling

Electrons in quantum dots will be able to absorb and emit phonons and photons by changing energy states in the dots. Furthermore, the quantization in the dots will assure that only specific frequencies of phonons or photons may carry energy to or from a particular dot. However, it may be difficult to restrict this type of coupling to nearest neighbor interaction as is required for a CA architecture. On the other hand, optical coupling may be an excellent method of distributing clock signals to a synchronous CA array of resonant tunneling quantum dot devices.

C. Field coupling

Although the electronic transport through quantum dot diodes is accomplished by electron tunneling current, the energy of the quantum dot is most strongly affected by electrostatic field coupling from the emitter and collector regions above and below the dot. Consider a quantum dot diode (or any resonant tunneling diode) where the thickness of the barriers is increased so that the tunneling current is vanishingly small. Despite the lack of tunneling current, the potential of the quantum states is still a function of the potentials applied to the contacts.

Consider a quantum dot with contacts above and below which are separated from the dot by barriers thin enough to allow tunneling. In addition consider that there are contacts to the left and right of the dot where there are barriers thick enough to prevent tunneling. The potentials of the quantum states in the dot are a function of the potential applied to each of its four contacts. If the vertical contacts are biased correctly there will be vertical resonant tunneling through the dot. However, the horizontal contacts may be used to modulate the tunneling current. While this structure would be difficult to fabricate it does illustrate the point that vertical resonant tunneling may be effected by horizontal electric fields from nearby potentials.

The structure we will use for examining lateral field coupling between quantum dots consists of two independently contacted, closely spaced, vertical tunneling quantum dot diodes. As a quantum dot diode goes into and out of resonance, the charge distribution in the diode will change and will therefore change the potential distribution in the diode. This change in potential will affect the potential of the quantum dot in the neighboring device. By biasing one device near resonance and varying the bias on the other, any effect on the potential of the dot with fixed bias should result in a modulation of the tunneling current. These measurements will have to be accomplished with considerable care and must consider all possible parasitic effects.

III. FABRICATION OF CLOSELY SPACED QDDS

The principal aim of this paper is to describe the fabrication techniques which have been developed to construct a pair of very closely spaced and separately contacted quantum dot diodes. There are several critical steps.

A. e-beam definition of closely spaced dots

The first step in fabricating closely spaced quantum dot diodes is to pattern two very closely spaced dots using electron beam (e-beam) lithography. The experiments described here were accomplished with a commercially available e-beam lithography system, a Philips EBPG-4HR.

In our initial experiments quantum dot diodes of approximately 1500 Å diameter were chosen as a compromise between devices small enough to show lateral quantization effects and the diminishing yield of smaller devices. Because of the proximity effect in e-beam lithography, the exposure of two closely spaced dots is not as simple as using the same exposure conditions used to pattern isolated dots. It was desirable to space the dots as closely as possible. Toward that goal, an experiment which placed dots at varying distances apart was conducted. After exposure and development, approximately 1000 Å of metal was vacuum evaporated and "lifted off" by spraying with acetone. Figure 1 is a scanning electron micrograph of dots where the distance between dots was incrementally varied in 100 Å steps. At the bottom of the micrograph the dots were close enough that the proximity effect caused them to merge. At the top the dots are clearly distinct. In between there is a region where the dots are separated by only a few hundred angstroms.

After molecular beam epitaxial growth of the resonant tunneling heterostructure, the fabrication of closely spaced quantum dot diodes begins with an optical exposure to define an alignment mark pattern. Each die includes one optical and several e-beam alignment marks which were formed in gold by a lift-off process. The dot pattern was exposed by e-beam lithography with a 50 kV beam, a 150 Å spot size, and a 50 Å beam step size. The resist was 950 K molecular.

![Figure 1: Scanning electron micrograph of lifted-off gold dots in closely spaced pairs. Spacing between dots increases in 100 Å increments from the bottom toward the top.](image)
weight polymethylmethacrylate (PMMA). Metal was evaporated and lifted off to form the dots. All dots were exposed to be nominally 1500Å in diameter. Six pairs of dots were exposed with varying spacing from dot edge to dot edge. The smallest spacing we attempted was 200Å. With exposure conditions used, dots spaced close together often merge. At the widest spacing which was 1000Å, separate dots were assured.

B. Reactive ion etching

The closely spaced dot pattern must be transferred as faithfully as possible to the molecular beam epitaxy grown heterostructure. For this purpose reactive ion etching with BCl₃ as an etch gas was employed. The process has been optimized to achieve highly anisotropic etching. Our etch conditions include a flow of 30 SCCM of BCl₃, an operating pressure of 30 mTorr, and sufficient rf-power (13.58MHz) to develop a plasma self-bias potential of 300 V. Figure 2 is a scanning electron micrograph of the structure resulting from reactive ion etching a series of closely spaced dots where the spacing between the dots was varied. As can be seen in the micrographs, it is possible to transfer the closely spaced dot pattern into GaAs.

As mentioned below, a thin InGaAs cap is used for making ultra-small ohmic contacts. The InGaAs etches considerably slower than GaAs resulting in an induction time of several minutes. While other reactive ion etching processes have been developed which etch InGaAs more easily than our BCl₃ process, our best results (with respect to anisotropy) have been obtained with the BCl₃ process by simply adjusting the etching time to account for the slower etch rate through the InGaAs cap.

C. Planarization with polyimide

In order to make contact to the top of the quantum dot diodes, a planarizing layer of polyimide is used which entirely covers the quantum dot devices. Reactive ion etching with oxygen is used to etch-back the polyimide layer until the tops of the diodes are uncovered. This is the same procedure which we use for isolated quantum dot diodes.

D. Independently contacting dots

Making separate electrical contact to two closely spaced diodes involves both producing ultrasmall ohmic contacts and high resolution lithography with alignment accuracy of a few hundred angstroms to attach metal lines to the dot contacts. It has been previously demonstrated that ohmic contacts down to 1500Å diameter can be reliably formed by grading from GaAs to In₀,₄₅Ga₀,₅₅As over a few hundred Angstroms and capping with a thin layer of In₀,₄₅Ga₀,₅₅As. In those experiments a metal contact was formed by e-beam lithography and subsequent lift-off. A silicon nitride layer was deposited over this structure and vias of the same diameter, carefully aligned to the first exposure, were patterned with e-beam lithography.

In work reported here good results were obtained with the previously unsuccessful and more straightforward approach of simply patterning and etching an ultrasmall via in a silicon nitride layer down to the semiconductor surface. Metal deposited over this surface forms a contact to the semiconductor at the bottom of the via. While we used a conventional ohmic metalization of AuGe-Ni-Au in these experiments, the previous study demonstrated that Ti-Au would also form good quality ohmics on the InGaAs capped material. Vias patterned nearby but not covered with metal were measured by imaging with a scanning electron microscope (SEM). Reactive ion etching the exposed semiconductor down through the via prior to SEM examination improved the contrast of the via image.

Ohmic contacts with variety of contact sizes from 1μm diameter down to 700Å diameter were successfully formed. Although some ohmic contacts were formed on the InGaAs capped samples without any annealing, the yield and contact resistance were significantly improved with a several minute anneal at 430°C. This improvement with annealing is similar to the previous small contact experiment results. Even after annealing there was considerable scatter in the contact resistance but the yield (percentage of ohmic versus Schottky) of even the smallest contacts was over 80%.

While these experiments of small ohmic contacts to planar surfaces capped with InGaAs have demonstrated good yields. The results on contacts atop the small diameter cylinders of quantum dot diodes have not been as successful. While the InGaAs cap has improved the yield of ohmic contacts, it is still relatively poor. Further investigations into this problem are underway.

After patterning and etching the dots and planarizing and etching back the polyimide layer there remains the challenging task of independently connecting to two metal contacts each only 1500Å in diameter and separated by only a few hundred angstroms.

The metal leads that contact the closely spaced dots are patterned by e-beam lithography. This layer is aligned to a different alignment mark on the die than was used to expose the dots. The metal leads were lifted off. An optically defined
metal pattern formed bondpads which connected to the e-beam defined metal leads.

IV. PRELIMINARY RESULTS

Initial electrical tests revealed that some quantum dot diodes were not connected. This could be attributed to misalignment and/or step coverage problems resulting from a rough polyimide surface. Some quantum dot pairs were shorted together either by dots merging or by the metal leads bridging the dots.

Some dot pairs were successfully contacted individually but were Schottky rather than ohmic contacts and did not show resonant tunneling transport. On a few percent of the dies, however, there were dot pairs which were independently contacted where each showed resonant tunneling and were not shorted together. Some of these successfully contacted pairs are among the most closely spaced devices and the spacing between devices should be on the order of 200 Å. Figure 3 is an SEM of a dot pair that has been successfully contacted. The polyimide layer has been etched away to reveal the dot structures.

The careful measurements to search for coupling between the dots has only recently commenced with no results to report at present. We will not be surprised if strong coupling between these particular devices is not discovered. These particular quantum dot diodes are double barrier, single dot designs. The difference in current through these devices between resonant and nonresonant conditions is only a few percent. In terms of peak to valley current ratio, the best result to date with a single dot diode is 1.04:1. The modest change in current through the device suggests a small change in charge distribution and limited means affecting the quantum states of dots in the neighboring diode. One must keep in mind that attempts to field couple between the diodes must contend with the screening from surface states on the surface of the diodes. Measurements will be reported at a later date. The work reported here is primarily intended to develop fabrication techniques and demonstrate the ability to build closely spaced quantum dot diodes.

In an effort to improve the peak to valley of quantum dot diodes we have been experimenting with triple-barrier, double-dot diodes. This type of structure has demonstrated better peak to valley ratios than single dot diodes. We speculate that this is because the dot-to-dot tunneling provides sharper resonances. A double dot diode has recently been reported which demonstrates negative differential resistance at room temperature. A new result is a double dot diode with a peak to valley current ratio of 1.5:1 was taken at 90 K. The I-V characteristic of this device is depicted in Fig. 4. This is the highest peak to valley ratio reported to date for a quantum dot diode. We feel other improvements in the design and materials of quantum dot diodes will permit significantly improved performance results in the near future. Improved characteristics of the isolated devices should significantly enhance the possibility of lateral coupling between closely spaced quantum dot diodes.

V. SUMMARY

The fabrication of quantum dot diodes which are closely spaced and independently contacted has been described. With such structures the lateral electrostatic coupling between pairs of vertical tunneling quantum dot diodes will be explored. The principle fabrication problem left to solve is improving yields of ohmic contacts to the tops of the quantum dot diodes. The strength of coupling between dots will depend on the performance of the individual quantum dot diodes. The triple-barrier double-dot diode structure has shown significantly better performance than single dot devices. A double-dot diode with a peak to valley current ratio of 1.5:1 has been reported.
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