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Manipulating Ge quantum dots on ultrathin Si_xGe_{1-x} oxide films using scanning tunneling microscope tips

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Abstract

Germanium quantum dots (QDs) were extracted from ultrathin Si_xGe_{1-x} oxide films using scanning tunneling microscope (STM) tips. The extraction was most efficiently performed at a positive sample bias voltage of +5.0 V. The tunneling current dependence of the extraction efficiency was explained by the electric field evaporation transfer mechanism for positive Ge ions from QDs to STM tips. Ge QDs (~7 nm) were formed and isolated spatially by extracting the surrounding Ge QDs with an ultrahigh density of $>10^{12}$ cm^{-2}. Scanning tunneling spectroscopy of the spatially-isolated QDs revealed that QDs with an ultrahigh density are electrically-isolated from the adjacent dots.

Keywords: Molecular beam epitaxy; Scanning tunneling microscope; Scanning tunneling spectroscopy; Field evaporation, nanopatterning; Germanium; Silicon

1. Introduction

Self-assembled quantum dots (QDs) of group IV semiconductors have been intensively studied for their quantum effects [1–4]. Artificial structures formed by self-assembled QDs have drawn much attention because they may contain new material properties. One method of arranging self-assembled QDs relies on the use of scanning tunneling microscope (STM). The atomic or molecular movements caused by STM manipulation are induced by several effects, such as the physical or chemical interaction between STM tips and the surface atoms [5], the strong electrical field effect induced by the tips [6,7], and the electronic or vibrational excitation effects induced by carrier injection from the STM tips [8–12]. Using these effects, various nanostructures (such as Si islands) were formed on the surfaces through field-induced diffusion [6,7]. The tip–induced electronic or vibrational excitation effect enhanced H atom desorption from H-terminated surfaces to form 1 nm wide bare Si lines [8].

Extracting self-assembled Ge QDs from ultrathin SiO_2 films using STM tips was performed by electron beam (EB) irradiation with a scanning electron microscope in local surface areas under the STM tips [13]. Manipulation using only STM tips without EB irradiation, which is a more valuable technique due to its availability, was difficult because Ge QDs are strongly bonded to the SiO_2 films. On the other hand, for Ge QDs on Si_xGe_{1-x} oxide films, the bonds between Ge QDs and the oxide films are presumably weaker because Si_xGe_{1-x} oxide films composed of both Si and Ge oxides are defective [14]. In this paper, we extracted Ge QDs from ultrathin Si_xGe_{1-x} oxide films using only STM tips. The extraction was caused by the field evaporation transfer for positive Ge ions from QDs to STM tips.

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We conducted scanning tunneling spectroscopy (STS) of the spatially-isolated Ge QDs by extracting the surrounding QDs with an ultrahigh density of 10^{12} cm^{-2}, and found that the ultrahigh density QDs are electrically-isolated from the adjacent dots.

2. Experimental

Samples cut from an n-type Si(1 1 1) wafer with a resistivity of 0.1–100 Ω cm were introduced into an ultrahigh-vacuum (UHV) chamber at a base pressure of $\sim 1 \times 10^{-8}$ Pa. The chamber was equipped with an STM and a reflection high-energy electron diffraction (RHEED) apparatus. Germanium with a thickness of 3 bilayers (BL) was deposited on Si(1 1 1)–(7 × 7) surfaces (pre-cleaned by flashing at 1250 °C) at a rate of ∼0.5 BL/min at 600 °C using a Knudsen cell. Two-dimensional Ge wetting layers were formed without three-dimensional islands [14]. Intermixing between Si and Ge atoms in a Ge wetting layer grown on Si(1 1 1) surfaces [15,16] is a controversial issue, and the composition of Si and Ge has yet to be elucidated. The Ge wetting layers were oxidized at 500 °C for 10 min at a molecular oxygen gas pressure of 2×10^{-4} Pa to form Si_xGe_{1-x} oxide films on Si(1 1 1). The Si_xGe_{1-x} oxide films consisted of both Si oxide (∼0.34 nm) and Ge oxide (∼0.11 nm); these details were described in a previous paper [14]. Germanium with 2–3.5 BL was deposited onto such ultrathin Si_xGe_{1-x} oxide films at a rate of ∼0.5 BL/min at 330–500 °C, to form Ge QDs smaller than 10 nm. RHEED observations showed diffraction patterns indicative of epitaxial growth without Debye ring pattern, indicating that most of the Ge QDs were epitaxially grown on the oxide films. The QDs contacted with the Ge wetting layers through voids in the oxide films [14]. STM imaging for surface observation was performed at a sample bias voltage of $V_S = +2$ V, and a tunneling current of $I_T =$ 30 pA, at room temperature using W tips, under which condition STM imaging did not change the surface topography. We manipulated Ge QDs using STM tips at various V_S and I_T ranging from −7 to +7 V and 30 pA to 1 nA, respectively.

3. Results and discussion

The STM image in Fig. 1(a) was obtained after scanning the area in the square at $V_S = +4.0$ V and $I_T = 0.3$ nA for ∼5 nm Ge QD samples with a density of $\sim 10^{12}$ cm^{-2}. The ball-shaped bright contrast seen in Fig. 1(a) is indicative of Ge QDs, while the dark area inside the square indicates the removal of the Ge QDs through STM scanning. The dark area outside the square (i.e., outside the scanning area) reveals that Ge QDs were extracted several nm away from the position just beneath the STM tip. To extract a single Ge QD, we moved the STM tip above a specific Ge QD under non-destructive imaging conditions ($V_S = +2$ V and $I_T = 30$ pA) and put the STM tip just above the Ge QD for 10 s at $V_S = +5.0$ V and $I_T = 0.1$ nA to extract the Ge QD. Fig. 1(b) and (c) show the STM images before and after extracting the target Ge QD (indicated by arrows), demonstrating that part or all of the target Ge QD was removed. Fig. 1(d) shows the letter "I" formed by dot extraction using this technique.

We investigated the dependence of the extraction efficiency on the sample bias voltage at $I_T = 0.1$ nA, as shown in Fig. 2(a). We defined the extraction efficiency of Ge QDs as the number of Ge QDs extracted during one scan (at a scanning speed is 2.7 μm/s), divided by the total number of Ge QDs in the scanning area (70 × 70 nm^2) before the extraction. On the whole, the extraction efficiency for the positive sample bias voltage is higher than that for the negative sample bias voltage. On the side of the positive sample bias voltage, the peak near $V_S = +5.0$ V was quite pronounced. We also investigated the tunneling current dependence of the extraction efficiency at the off-peak $V_S = +4.0$ V, which shows a monotonic increase for the extraction efficiency in Fig. 2(b).

For the ultrahigh density QDs (>10^{12} cm^{-2}), whether the STS results of QDs include dot–dot interactions or not is ambiguous. To clarify this, we isolated a Ge QD spatially by extracting the surrounding Ge QDs with ultrahigh density. STM images before and after isolation are shown in Fig. 3(a) and (b). In the STM image of Fig. 3(c), wide-field image of Fig. 3(b), the Ge QD shown by the arrow

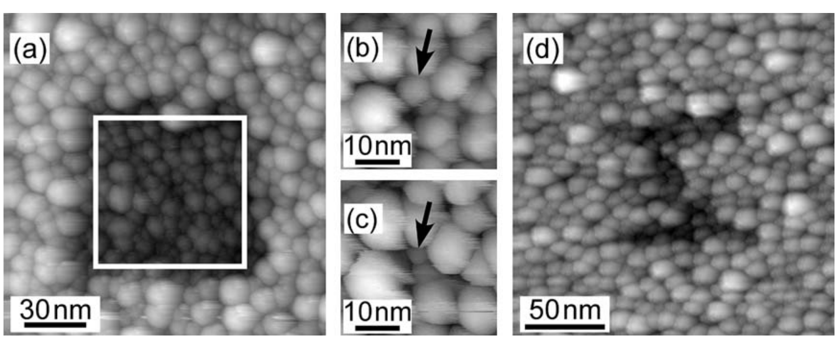

Fig. 1. (a) STM image of Ge QDs on Si_xGe_{1-x} oxide films formed by 2.2 BL Ge deposition at 380 °C, after scanning the area in square at $V_S = +4.0$ V at $I_T = 0.3$ nA. STM images (b) before and (c) after extracting the target Ge QD, indicated by arrows at $V_S = +5.0$ V and $I_T = 0.1$ nA for 10 s, where the QDs were formed by 3.5 BL Ge deposition at 330 °C. STM imaging conditions are $V_S = +2.0$ V at $I_T = 30$ pA. (d) Letter "I" formed by dot extraction.