Ge–Si intermixing in Ge quantum dots on Si

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Abstract

We have provided direct evidence for the presence of considerable Si–Ge intermixing in strained and unstrained Ge quantum dots deposited on Si(001) and Si(111). The local structure around Ge was probed by using Ge K-edge X-ray absorption spectroscopy; complementary evidence for intermixing was provided by AFM and STM studies. These results implied that the strain energy in the dots was reduced by Si atoms diffusing into the dots, resulting in a modified form of Stranski–Krastanov growth. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The properties and structure of self-organized semiconductor quantum dots have been the subject of considerable recent interest, due to the possibility of tailoring the electronic structure. Since it is the underlying atomic structure which determines the electronic properties, its characterization is of paramount importance. Recently, there have been many reports indicating that a hitherto neglected phenomenon, Si–Ge intermixing, plays an important role in the Stranski–Krastanow growth of Ge dots on Si [1–4]. In the present paper, we review our X-ray absorption (XAFS) results and provide complementary evidence obtained by atomic force microscopy (AFM) and by scanning tunneling microscopy (STM).

2. Sample growth

Two sets of samples were deposited on Si(001) and Si(111), by ultra high vacuum chemical vapor deposition (UHV-CVD) and molecular beam epitaxy (MBE), respectively.

UHV-CVD growth was obtained in a reactor whose base pressure was in the low 10−10 torr range. The samples were deposited using germane at a pressure $P_{\text{dep}} = 0.5$ mtorr without a carrier gas. The morphological characterization of the samples was performed by means of an ambient Park Scientific Instruments CP AFM operating in contact mode [5,6]. Six samples were studied, with an equivalent thickness ranging from 5.8 to 38 nm. AFM allowed us to identify dots with pointed and truncated pyramid morphology [6]. A clear corre-
Correspondence between these morphological differences and the strain state existed, with pointed (truncated) pyramids being associated with coherently strained (relaxed) dots. We have thus concluded that the samples studied exhibit a relative volume of truncated pyramids ranging from 10 to 90%. Ge dots on Si(111) were deposited by MBE on a 7×7 reconstructed surface using a Knudsen cell with the substrate kept at 500°C. The samples were characterized in situ by AFM-STM with a commercial Omicron VT-STM. Two samples were deposited, with equivalent thicknesses of 1.0 and 1.7 nm. The surface reconstruction was 5×5 in the case of the 1.0-nm sample (essentially constituted by the wetting layer only), and a mixture of 5×5 and 7×7 in the case of the 1.7-nm sample.

3. X-Ray absorption experiments

XAFS spectra at the Ge K-edge were recorded at the ‘GILDA’ beamline at ESRF, Grenoble. The Ge absorption was monitored in the fluorescence mode by using a seven-element hyperpure Ge detector. XAFS data were analyzed using Fourier filtering methods, using known compounds as experimental standards [7]. Selected background-subtracted raw data are shown in Fig. 1; also shown are the spectra of the two model samples: bulk Ge and a Ge impurity in a crystalline Si matrix. In the low k region there are significant differences between the spectra of the samples and that of Ge. In particular at $k = 4 \ \text{Å}^{-1}$, Ge has a negative oscillation, while all the samples exhibit a positive oscillation, in good coincidence with the spectrum for a Ge impurity in Si. This feature is highlighted with an ellipse. This observation clearly suggests that a significant number of Si atoms bound to the average Ge atom exist. In Fig. 2, we show selected Fourier transforms of the data, the inspection of which is compatible with this conclusion.

The fitting of the first shell was performed in k-space using a linear combination of Ge–Ge and Ge–Si contributions. For Ge/Si(001), we have found that the average Ge atom is surrounded by $\sim 1.2 \pm 0.3$ Si atoms, independently of the morphology of the dots, which ranged from that typical of a strained dot to that associated with relaxed islands. For both samples deposited on Si(111), we found a Ge–Si coordination number of $2 \pm 0.3$. The XAFS analysis thus demonstrates considerable Si–Ge intermixing.

4. AFM and STM results

In Fig. 3, we show AFM plan views of Ge/Si(001)
dots deposited at 650°C (right) and 750°C (left) with the same growth rate and equal deposition times. It is clear that at higher temperatures, the dimensions of the islands are greater. In fact, in Fig. 4, we report a plot of the mean base width \( b_m \) for strained islands vs. the deposition temperature. Since \( b_m \) is inversely proportional to the square of the misfit, its increase is consistent with a reduced effective misfit between epilayer and substrate [8]. This effect can be explained with a temperature-enhanced diffusion of Si in the Ge deposited layer [9].

In Fig. 5a,b we show STM and AFM images of Ge islands on Si(111). \( T_d \) and equivalent thickness are 500 and 530°C, and 3 and 2 nm for (a) and (b), respectively. Both images were performed in-situ immediately after deposition and clearly show the erosion of the substrate around the island. The creation of this ‘trench’ (the depth of which is a few nanometers) indicates that Si atoms from the substrate diffuse into the Ge dots. This effect has been previously observed experimentally [10]; Kamins et al. could not, however, attribute it to a specific effect due to the oxidation of the substrate.

5. Conclusions

The XAFS results here presented clearly demonstrate and quantify the presence of Si–Ge intermixing for Ge quantum dots deposited on Si substrates. The results are complemented and further supported by AFM and STM studies which show an increase in the mean base width with deposition temperature and the presence of a ‘trench’ around the islands. Similar XAFS results have been recently obtained by some of the authors on the related system of InAs dots on GaAs(001) [11]. The conclusion of these studies is that interdiffusion is a general phenomenon in the growth of semiconductor quantum dots. The basic physics underlying this observation is that atomic interdiffusion can decrease the strain energy. The result is a modification of the Stranski–Krastanov growth mechanism in which the decrease of the strain energy due to interdiffusion must be considered.

References