Observation of Coulomb Staircases in Arsenic Precipitates in Low-Temperature Grown GaAs

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(Received August 17, 1998; accepted August 27, 1998)

Gallium arsenide grown by molecular beam epitaxy (MBE) at low temperatures (LT) has recently attracted much attention [1 to 5], due to its unique electronic [1] and optical [2] properties. Unlike normal GaAs layers, LT-GaAs grown at 200°C is very nonstoichiometric with 1 to 2% excess As [3]. The quench-in excess As results in a high concentration of As antisite defects and causes an expansion of lattice constant detectable by X-ray diffraction. The resistivity of the LT-GaAs is as low as tens of ohms for as-grown LT-GaAs materials, and increases up to 10^6 to 10^7 Ωcm after post-growth annealing above 600°C [4]. The outstanding characteristics of LT-GaAs have generated great technological and scientific interest. For example, as a buffer layer, LT-GaAs can remarkably improve some critical characteristics of the GaAs metal–semiconductor field-effect transistor. It is also a suitable insulator layer for the metal–insulator–semiconductor field-effect transistor, and an active layer for fast photoconductive switches [5].

The high resistivity of the annealed LT-GaAs material has been explained by a defect compensation model [3] and also by the formation of overlapping buried Schottky barrier depletion regions resulting from As precipitates [6]. However, the mechanism for the high resistivity is still uncertain. It seems clear that an improved understanding of the properties of As precipitates will play a crucial role in resolving this problem. Besides, As precipitates are similar to self-organized quantum dots, the related investigation can lead to the potential application of single-electron devices. In this work, we report a study of the effect of the As clusters on the tunneling spectroscopy of LT-GaAs material. Quite interestingly, we have observed Coulomb staircases in tunneling spectroscopy. The deduced cluster size is found to be in good agreement with that obtained from transmission electron microscopy. Our observation shows that in addition to the great potential applications, LT grown GaAs materials provide good candidates for the study of Coulomb-staircase behavior.

The LT-GaAs in this study was grown by using a Riber 32P MBE system. In order to perform the tunneling spectroscopy, the sample was designed as follows: A 0.25 μm heavily Si-doped (3 × 10^{18} cm^{-3}) GaAs layer was first grown on a (100) n' GaAs substrate, followed by a LT-GaAs layer of thickness 100 Å grown at substrate temperature of 200°C. The annealing of the sample was performed in the growth chamber at 600°C under As pressure of 2.7 × 10^{-6} Pa for 30 min. The excess As within the LT-sample was measured by the double crystal X-ray diffraction curve, which shows the sample containing excess As of about 1%. For samples prepared by the same condition, transmission electron microscopy shows a high concentration of As precipitates with an average diameter of about 4 ± 2 nm distributed inside the LT-grown region [7]. The details of the sample preparation and characterization can be found in our previous report [8]. The tunneling spectroscopy was investigated with a commercial low-temperature scanning tunneling microscope (STM) (Park Scientific Inc.) operating in vacuum at 77 K. Tungsten tips were used in our STM measurements.

Fig. 1 shows I vs. V and dI/dV vs. V characteristics of the tunneling spectroscopy measured on the annealed LT-GaAs sample at 77 K. The dI/dV − V curve shows periodic oscillations with a voltage period ΔV = 64 meV as marked by arrows. The periodic structure becomes more pronounced with decreasing temperature. Because the oscillation behavior does not appear in the tunneling spectroscopy of the sample without As precipitates, we can attribute the oscillations to the effect of the As precipitates. To search the mechanism responsible for the observed oscillations, we keep in mind that the As precipitates are metallic and they are immersed in the insulating GaAs matrix. The average precipitate size is approximately 4 nm with an average precipitate spacing of about 20 nm [7]. Thus, in the measurement of the tunneling spectroscopy using STM, we can consider that the experiment probes the tunneling behavior of a single metallic cluster surrounded by an insulating region. This experimental configuration is tempting to recall that of periodic conductance oscillations of electron tunneling through a quantum dot [9]. If we apply the concept of the electron tunneling through a quantum dot, we can estimate the effective capaci-
tance of the probed single As precipitate from the equation $C_{\text{eff}} = \frac{e}{\Delta V}$, where $\Delta V$ is the voltage period obtained from the tunneling spectroscopy [10]. We then have $C_{\text{eff}} = 2.5 \times 10^{-18}$ F. Because the separation between the As precipitates is in a relative large distance [7] when compared with the diameter of As precipitates, we can assume the fact that charging an As precipitate will not affect the electronic properties of its neighboring precipitates. This approximation provides us a very simple experimental condition which can largely reduce the difficulty of our interpretation. We thus can estimate the size of the probed As precipitate from the equation $C_{\text{eff}} = \frac{4\pi\varepsilon r}{\varepsilon r}$ [9], where $\varepsilon = 12.9$ is the dielectric constant of the surrounding GaAs matrix, and $r$ is the radius of the As precipitate. The estimated diameter has the value of 3.6 nm, which is in good agreement with the average value (4.0 nm) of As precipitates obtained from transmission electron microscopy [7]. This agreement provides a strong evidence to support the fact that the oscillations observed in our tunneling spectroscopy can be explained in terms of the Coulomb-staircase behavior of electron tunneling through a quantum dot.

The above periodic oscillation behavior can be repeated by fixing the tungsten tip at different positions. However, the observed voltage periods do not have a constant value, which probably reflects the fact that the size of the As precipitates in LT-GaAs is not uniform as shown by transmission electron microscopy [7]. Indeed, the diameters of As precipitates deduced from tunneling spectroscopy are always within the range of 4 ± 2 nm obtained from transmission electron microscopy. In addition, the oscillation behavior does not appear each time when we change the position of the tungsten tip. We believe that this condition can occur when the tungsten tip is at a position between the precipitates.

In conclusion, through a designed structure of LT-grown GaAs, we have observed Coulomb staircases in the tunneling spectroscopy due to the existence of As precipitates. Our result shows that LT-grown GaAs materials not only have great potential technological applications, but they also provide a very good candidate for the study of striking Coulomb-staircase phenomena.

This work was partly supported by the National Science Council of the Republic of China. We wish to acknowledge useful discussions with Dr. T.D. Hu.

References