Study of Structural Phase Transition of Pt-Induced Nanowire on Ge(001) Surface using Reflection High-Energy Positron Diffraction

The atomic configuration and the structural phase transition of Pt-induced nanowires on a Ge(001) surface are investigated using reflection high-energy positron diffraction (RHEPD). A previously proposed theoretical model, composed of Ge dimers on the top layer and buried Pt arrays in the second and fourth layers [1], is confirmed to be the fundamental structure of the nanowires. Analysis of the temperature-dependent RHEPD intensities suggests that the structural phase transition is very similar to a new type of phase transition [2]. That is, the precursory order-disorder behavior for the thermal-fluctuating low-temperature phases triggers the displacive transition to the high-temperature phase.

Self-assembled nanowires have attracted a great deal of interest not only for their potential applications in nano-devices, but also for providing a fundamental understanding of one-dimensional properties. Recently, Gürlu et al. [3] found that well-ordered and defectless nanowires are formed on Ge(001) surfaces by depositing Pt atoms. They first proposed a structural model of the Pt-induced nanowires based on their scanning tunneling microscopy (STM) observations, called the Pt-dimer (PD) model [Fig. 1(a)]. van Houselt *et al.* [4] reported that the nanowires undergo a Peierls transition at around 80 K. However, recent theoretical calculations suggested different structural models to explain those STM studies. The tetramer-dimer-chain (TDC) model [Fig. 1(b)] and the nanowire (NW) model [Fig. 1(c)] were suggested by Stekolnikov *et al.* [5] and Vanpoucke *et al.* [1], respectively. Despite extensive studies using STM and *ab initio* calculations, the atomic configuration and the phase transition mechanism of the nanowires are still controversial.

Reflection high-energy positron diffraction (RHEPD) is a surface-sensitive tool owing to the positive charge of the positron. Positrons are repelled by a positive crystal potential so that they cannot enter crystals at low-angle grazing incidences (total reflection). Therefore, RHEPD is very sensitive to the topmost layer of crystals. Recently, we developed a new RHEPD apparatus [6] on beamline SPF-B1 of a linac-based intense positron beam at the Slow Positron Facility (SPF). In this study, we investigated highly ordered Pt-induced nanowires on Ge(001) surfaces by using the RHEPD apparatus.

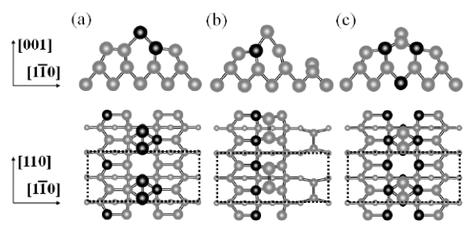

Figure 1
Schematic illustrations of Ge(001)-p(4×2)-Pt structures; (a) PD, (b) TDC and (c) NW models. The black and gray spheres represent Pt and Ge atoms, respectively. The dotted rectangles represent p(4×2) unit cells.

Pt was deposited on a clean Ge(001) surface kept at 720 K under ultra-high vacuum condition. The Pt coverage was 1.2 monolayer (ML), where 1 ML corresponds to ~6.3×10^{14} atoms/cm^2. RHEPD experiments were performed using a 10 keV positron beam generated from a ^{22}Na source at the JAEA and the intense positron beam at the SPF. The incident azimuth was 22.5° away from the [1$\bar{1}$0] direction (one-beam condition). The glancing angle (θ) for total reflection is approximately 2.2°.

Figure 2 shows the RHEPD rocking curve of the specular spot at 35 K [7]. The solid and broken lines are calculated rocking curves assuming the PD, the TDC, and the NW models, respectively. Under the assumptions of the PD and the TDC models, the calculated curves exhibit dips in the total reflection region due to the interference effects of the positron waves reflected by the first and second surface layers [8]. The features do not appear in the experimental curve. On the contrary, the calculated curve for the NW model (thick solid line) is in good agreement with the experimental data. We also examined other structural models with the Pt coverage ranging from 0.25 to 1 ML [7]. Among these examined models, the NW model exhibits the best reliability factor. Thus, the fundamental structure of the Pt-induced nanowires is explained by the NW model.

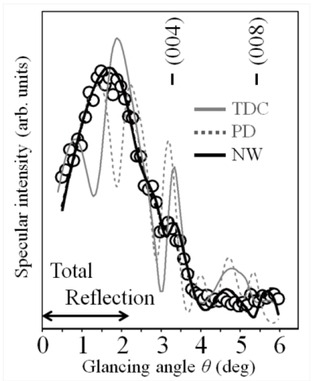

Figure 2
RHEPD rocking curves measured from Pt-induced nanowires under the one-beam condition at 35 K. The open circles denote experimental data. The solid gray, broken gray, and solid black line are the calculated curves for the TDC, PD and NW models, respectively.

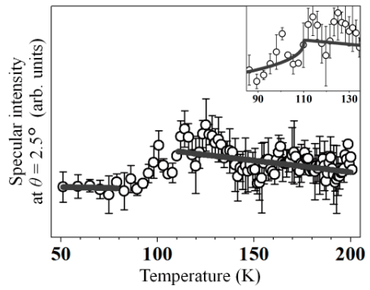

Figure 3
The temperature-dependent one-beam specular spot intensity at θ = 2.5° for the temperature range 50-200 K. The solid lines indicate the calculated temperature dependence assuming Debye temperatures of 210 and 130 K. The inset shows a close-up of the temperature dependence around T_c. The solid curve and solid line indicate the optimum temperature dependence below and above T_c, respectively

Figure 3 shows the temperature dependence of the specular spot intensity at θ = 2.5° in the temperature range from 50 to 200 K. The intensity gradually increases from 80 to 110 K, indicating the progress of the displacive transition for the topmost Ge dimers [7], and thereafter, a conventional Debye-Waller-like temperature dependence is observed within experimental error. From the slopes of the relation between specular intensity and temperature, the surface Debye temperatures are determined to be 210 ± 80 K for the low-temperature phase and 130 ± 40 K for the high-temperature phase. The amplitudes of the surface-normal vibrations of the topmost Ge dimers are 0.06 Å at 50 K and 0.15 Å at 120 K. The temperature dependence of the intensity between 80 and 110 K is somewhat continuous and would be reproduced by a power-law, as shown by the inset in Fig. 3. The order parameter is proportional to $|1-T/T_c|^{\beta}$ with β = 0.36 ± 0.15 and T_c = 111 ± 10 K. The value of β is smaller than that anticipated for the mean field approximation (β = 0.5). This is probably due to the coexistence and fluctuations of the low-temperature phases and the high-temperature phase as observed by using STM [7].

From the ARPES measurements, we observed the change in electronic band dispersions due to the atomic displacements of the topmost Ge dimers [7]. The results are also consistent with the scenario of the phase transition with the precursory order-disorder behaviors.

In the present results, the structural phase transition is very similar to a new type of phase transition [2]. That is, the order-disorder behavior for thermal-fluctuating low-temperature phases triggers the displacive transition from the low-temperature phases to the high-temperature phase.

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BEAMLINE

SPF-B1

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