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Time-resolved reflection high-energy electron diffraction study of the Ge(111)-c(2×8)−(1×1) phase transition

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The dynamics of the Ge(111)-c(2×8)−(1×1) phase transition is investigated by 100-ps time-resolved reflection high-energy electron diffraction. A laser pulse heats the surface while a synchronized electron pulse is used to obtain the surface diffraction pattern. Slow heating shows that the adatoms in Ge(111)-c(2×8) start to disorder at ~510 K and are converted to a disordered adatom arrangement at 573 K. For heating with 100-ps laser pulses, the Ge(111)-c(2×8) reconstructed adatom arrangement starts to disorder at 584±16 K, well above the onset temperature of ~510 K for the disordering of Ge(111)-c(2×8) observed for slow heating.

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FIG. 1. Normalized RHEED intensities, $I/I(T=304 \text{ K})$, of the $(0\bar{1})$ and $(01)$ diffraction streaks versus temperature are plotted on a semilogarithmic scale. The $(01)$ streak ($\triangle$) shows the Debye-Waller behavior over $300 \text{ K}$ to $650 \text{ K}$, with an effective surface Debye temperature $\Theta_s = 98 \text{ K}$. The $(0\bar{1})$ and $(01)$ streaks were examined at the same time. RHEED streak intensities normalized to that at $304 \text{ K}$ versus surface temperature are shown in Fig. 1. The exponential Debye-Waller behavior of the $(0\bar{1})$ streak is observed below $\approx 510 \text{ K}$. At higher temperatures, deviation from the exponential behavior occurs, indicating the onset of adatom disorder in the $c(2 \times 8)$ structure. The coexistence of disordered and ordered regions on the surface at temperatures well below the $573\text{ K}$ transition temperature was previously observed by STM.\textsuperscript{3} Our results agree with the STM observations. The Debye-Waller behavior of the $(01)$ streak persists above $573 \text{ K}$. The Debye-Waller factor is used to extract the transient temperature on the surface during laser pulse heating, since it is not affected by the Ge$(111)$-$c(2 \times 8)$–$(1 \times 1)$ phase transition. The MD simulation of Takeuchi, Selloni, and Tosatti showed that the mean square displacement of $110$, $109$, $111$, $27$ reflectivity equal to $1.8 \times 10^3 \text{ m}^{-1}$,\textsuperscript{29} and a $100$-ps FWHM Gaussian laser pulse with peak fluence equal to $1.8 \times 10^8 \text{ W/cm}^2$. The experimental results agree well with the heat diffusion model. Figure 2 also relates the maximum transient temperature rise on the Ge$(111)$ surface to the peak fluorescence of the heating laser pulse. This is used to determine the maximum surface temperature rise which is proportional to the laser peak fluorescence.

We next raised the sample temperature close to the onset temperature of the reconstruction phase transition and used a fixed laser fluence to further raise this surface temperature in a transient manner. The time-resolved RHEED intensity was monitored by fixing the delay time at the time at which the RHEED intensity is minimum, which is temporally close to the time of maximum surface temperature rise. RHEED streak intensity of the $(0\bar{1})$ streak normalized to that at the base temperature was obtained for various peak laser fluences. Results are shown in Fig. 3 for three pump-probe scans representing base temperatures successively closer to $510 \text{ K}$. In each of these scans, the exponential behavior with temperature remains for lower laser peak fluences with a corresponding surface Debye temperature $\Theta_s$ of $110$, $109$, and $87 \text{ K}$ for base temperatures of $442$, $473$, and $507 \text{ K}$, respectively. This is within $\pm 12\%$ of $\Theta_s = 98 \text{ K}$ observed for the slow heating. Deviation from exponential behavior occurs at higher peak laser fluences depending on the base temperature, indicating that the adatoms are no longer bonded to the $T_s$ sites by a harmonic oscillator and start to diffuse as the temperature is increased. From Fig. 1, we see that this starts at $\approx 510 \text{ K}$, which is well below the thermodynamic transition temperature of $573 \text{ K}$. This behavior was explained as premelting,\textsuperscript{7} or as a result of surface strain.\textsuperscript{12} A MD simulation showed that the energy barrier for the adatom diffusion is decreased when surface vacancies are present.\textsuperscript{30} It is reasonable to expect that adatom diffusion starts at a lower temperature than the thermodynamic transition temperature, because surface vacancies are always present on a
real Ge(111) surface. STM observations showed that the disorder starts from the domain boundaries where surface vacancies are present.\(^4\)

In Fig. 3, the deviations from exponential Debye-Waller behavior occur at laser peak fluences of \(13.4 \pm 1.6 \times 10^7\) W/cm\(^2\) for a base temperature of 507 K and 17.3 \(\pm 2.1\times10^7\) W/cm\(^2\) for a base temperature of 473 K. The indicated errors are due to nonuniformity of the laser beam across the probed sample area. These two fluences correspond to maximum transient temperature rises of 82 \(\pm 10\) and 106 \(\pm 13\) K, respectively. Therefore, for 100-ps laser heating, the Debye-Waller behavior of the \(c(2 \times 8)\) lattice remains up to 589 \(\pm 10\) and 579 \(\pm 13\) K for the two curves showing reconstruction in Fig. 3, giving an average of 584 \(\pm 16\) K. For slow heating, the \(c(2 \times 8)\) lattice starts to disorder at \(\sim 510\) K. For both slow heating and 100-ps laser heating, the Ge(111)-\(c(2 \times 8)-(1 \times 1)\) phase transition occurs over a temperature range of \(\Delta T\), which we define as the interval between the onset temperature of the phase transition and the temperature at which the RHEED intensity is 10\% of that at the onset temperature. The onset temperature of the phase transition is that when the RHEED intensity deviates from the Debye-Waller behavior. For 100-ps laser heating, Fig. 3, the Ge(111)-\(c(2 \times 8)-(1 \times 1)\) phase transition starts at 584 \(\pm 16\) K and spreads over a temperature range of \(\Delta T\) of 58 K, whereas for slow heating, Fig. 1, the phase transition starts at 510 K and spreads over a temperature range of 55 K. Therefore, we conclude that the \(c(2 \times 8)\) structure is overheated by 74 \(\pm 16\) K above the onset temperature of adatom disordering observed under thermodynamic equilibrium conditions of slow heating. MD simulations showed that the Ge(111)-\(c(2 \times 8)\) structure survives for 3 ps at 1200 K,\(^3\) whereas our experimental results show that the \(c(2 \times 8)\) lattice persists up to 584 \(\pm 16\) K for 100-ps laser pulse heating. However, the MD simulation assumes an ideal step heating which brings the surface to high temperature instantaneously and keeps it at that temperature thereafter; while in our experiment, the sample is heated to a high temperature in a time comparable to the laser pulse width and the temperature decreases after that by heat diffusion to the bulk.

Further experiments were performed to examine the temporal behavior of the Ge(111)-\(c(2 \times 8)-(1 \times 1)\) phase transition. The normalized RHEED \(0\overline{2}\) streak intensities were obtained at various delay times between the arrival of the heating laser pulse and the electron probe pulse at the surface of the sample. Results for different incident laser peak fluences are shown in Fig. 4. The base temperature of the sample is 507 K. The solid line is from a one-dimensional heat diffusion model, converting the obtained temperature rise to normalized RHEED intensity using the Debye-Waller factor of that order. The surface is kept at a base temperature of 507 K. (a) \(I_p=10.2 \pm 1.2 \times 10^7\) W/cm\(^2\), consistent with that expected from heat diffusion. (b) \(I_p=14.2 \pm 1.7 \times 10^7\) W/cm\(^2\) and (c) \(I_p=16.3 \pm 2.0 \times 10^7\) W/cm\(^2\) deviate from that expected from heat diffusion, indicating partial disordering of Ge(111)-\(c(2 \times 8)\). (d) \(I_p=18.3 \pm 2.2 \times 10^7\) W/cm\(^2\), near total conversion to Ge(111)-(1\( \times 1\)) structure.
extinction of the (0½) RHEED intensity is thought to be due to the combination of the effects of the growth of the disorder from domain boundaries, and perhaps microscopic laser fluence variation at the surface beyond that measured by scanning the laser beam profile. The decrease in the surface temperature by heat diffusion results in a surface temperature of 559±6 and 566±7 K, respectively, for (c) and (d) at ~4.5 ns from the time of the peak laser fluence. For these temperatures the surface disorder persists. The Ge(111)-c(2×8) structure is observed, however, to fully recover before the next laser pulse for 50-Hz repetition rate. In all of the experiments reported here, no surface damage was observed.

In summary, we have investigated the dynamic behavior of the Ge(111)-c(2×8)–(1×1) phase transition. For slow heating, our RHEED results show that the adatoms in the Ge(111)-c(2×8) reconstruction state start to disorder at ~510 K and are converted to a disordered adatom arrangement at ~573 K. This is consistent with previous LEED and STM observations. However, by 100-ps laser pulse heating, time-resolved RHEED measurements show that the disorder starts at 584±16 K, 74±16 K above the onset temperature for the disordering under thermodynamic equilibrium. This result is in qualitative agreement with the overheating of Ge(111)-c(2×8) which was previously predicted from MD simulations.

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