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Intensity oscillations have been found in the specular beam of reflection high-energy electron diffraction patterns during growth of Si(001) and Ge(001) by molecular beam epitaxy. The reported results demonstrate the dependence of the amplitude and damping of the oscillations on different parameters such as substrate temperature, electron beam angle of incidence, and azimuth.

In the past few years, one of the most practical aspects of reflection high-energy electron diffraction (RHEED) experiments during crystal growth by molecular beam epitaxy (MBE) has proven to be the phenomenon of oscillations in the intensity of the specular beam in RHEED patterns.[1] This effect is well known for GaAs(001) where the period of the oscillations was shown to correspond exactly to the growth of one monolayer[2] (meaning a complete layer of Ga plus a complete layer of As). Intensity oscillations were also observed during MBE growth of Ge (Ref. 3) and, more recently, of Si (Ref. 4). In the latter case the behavior seems less straightforward as both monolayer and bilayer oscillation modes were seen on the same Si(001) surface, measured for different azimuthal angles of the incident beam. This last observation emphasizes the fact that the fundamental principles underlying the occurrence of oscillations are still poorly understood.

In this letter we want to show the effect of the variation of some important experimental parameters upon the oscillatory behavior of the specularly reflected RHEED beam during MBE growth of Si(001) and Ge(001). The parameters concerned are the angle of incidence ϑ_i and the azimuthal angle of the electron beam, the substrate temperature, and the growth rate. Some of the observations reported here are at variance with observations reported in Ref. 4.

The measurements were performed in a cryopumped vacuum chamber with a base pressure of 3×10^{-8} Pa, which during growth conditions increased to 1–2×10^{-7} Pa. The system was fitted with an electron gun operated at 12.5 kV. The substrate was mounted on a sample holder in the usual RHEED geometry, allowing changes of both the incident and azimuthal angle. The RHEED pattern was displayed on a phosphor screen and monitored by a television camera for data processing. The Si substrates were cut from very accurately (better than 0.05°) oriented Si(001) wafers. They were chemically cleaned and, once inside the vacuum chamber, heated to about 900 °C for about 30 min.[5] This procedure resulted in a RHEED pattern consisting of very sharp spots of high intensity, indicating domains of 2×1 and 1×2 surface reconstructions, which is usual for Si(001). The spots became less sharp and more streaklike during growth but the pattern could be fully restored by subsequently annealing the sample at 800 °C. For growth, a small electrostatic Si evaporation cell was used, which only allows low evaporation rates, of the order of a few layers per minute. In the case of

Ge growth the same Si substrate was used, on which a few thousand layers of Ge were grown, this time using a Knudsen cell; the RHEED pattern then showed a lattice parameter increase of 4% (the difference between Si and Ge). RHEED patterns of the Ge surface were rather more streaky than in the case of Si. This is possibly caused by the fact that the orientation of the Ge buffer layer differed 0.3° from the orientation of the underlying Si substrate, as was found by x-ray diffraction after conclusion of the experiments. Intensity oscillations during growth could be detected on both the Si and the Ge surface. In Fig. 1 such oscillations are shown for both Si and Ge at different substrate temperatures and at an electron beam incidence angle of 0.6°. A number of features deserve special attention. First, for Ge, oscillations with the highest amplitude and smallest damping were found at room temperature. After careful preparation of the surface, up to 150 periods could be observed. Going to higher temperatures resulted in a smaller initial drop in intensity and a smaller amplitude. Above 400 °C, no oscillations could be detected, neither for the [010] nor for the [110] azimuth. Also for Si a maximum temperature for the occurrence of oscillations was found, in this case of about 600 °C. For Ge it was checked whether the maximum temperature depended on the molecular beam flux by increasing the growth rate from 12 s to 1 s per layer. This resulted in a few fast damping oscillations of small amplitude at 360 °C, only a trace of oscillatory behavior at 400 °C, and thus an increase of the maximum temperature by at most 20 °C.

Another important point is that at low growth temperatures (for Si around 300 °C, for Ge around room temperature) no change of the signal is found after stopping growth. At higher temperatures a (partial) change towards the original value is seen, but then the amplitude of the oscillations and the initial intensity decrease are smaller. This behavior is in contrast to that of GaAs(001) where large oscillations and fast recovery can both be observed.[1] The lack of recovery means that the phase of the oscillations can be kept constant if growth is stopped. This is demonstrated in Fig. 2, where the growth of Ge was interrupted in the third oscillation minimum for a period of 1 h. Oscillations then occurred again without change in phase and with only small decrease in amplitude. Obviously, this effect indicates a very small diffusion along the surface. This opens the possibility of monitoring the growth even at very low growth rates. Experimentally, growth rates as low as one layer per half-hour
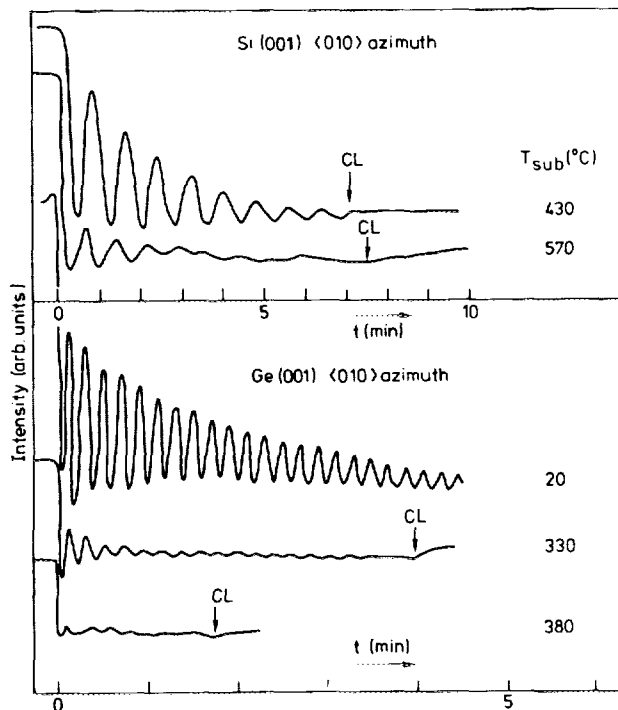
FIG. 1. Oscillations in the specular beam intensity during growth of Si(001) and Ge(001) for different substrate temperatures as indicated and for $\vartheta_i \sim 0.6°$. CL denotes interrupting growth by closing the shutter of the source.

were observed, which makes the system very well suited for investigations of the initial stages of crystalline growth. However, it is difficult to see how these results can be reconciled with models of layer-by-layer growth based on island growth formation, which should give rise to the intensity oscillations.[6]

The slow recovery also means that after each experiment the surface has to be prepared anew by heating to high temperatures. For Si a temperature of 800 °C was used and for Ge a temperature of 600 °C was sufficient, both for periods of 10 min. Either temperature is considerably higher than the highest temperature where oscillations were found.

The above experiments were all performed both in the [010] and in the [110] azimuth. In general, the behavior in the two directions was the same, although oscillations taken in the [010] azimuth were usually slightly less damped. In-
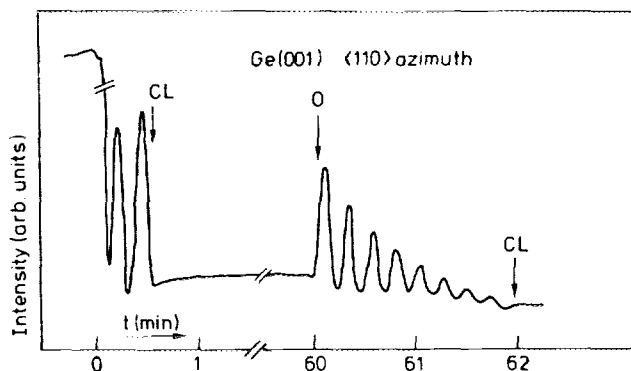
termediate values of the azimuth behaved similarly (for an incident angle of 0.6°) as shown in Fig. 3, in contrast to the results for Si reported in Ref. 4.

The final parameter to deserve attention is the angle of incidence ϑ_i of the electron beam. Both the intensity of the specularly diffracted beam and the phase and amplitude of the oscillations depend strongly on ϑ_i and their relationship is therefore of great interest. A plot of intensity versus ϑ_i ("rocking curve") for Ge(001) in the [010] azimuth is given in Fig. 4. Although the understanding of such curves is not complete, it is known that the main features are related to the surface topography.[7,8] For instance the maximum in intensity at 1.8° seems linked to the emergence of the (01) beam in the RHEED pattern, and not to the first or second allowed bulk Bragg reflection. Figure 5 shows the intensity changes during growth for different values of ϑ_i, as indicated in Fig. 4. At angles below 1°, amplitudes are large and slowly damping as can be seen in previous figures. Going to higher angle the amplitude decreases, while the direction of the initial response changes to an increase in intensity. This is already the case for 1.1° and more clearly so for 1.3°. Going to still higher angles, the initial response becomes smaller again. At the maximum in the rocking curve at 1.8° oscillations are fully absent. They return in a small region around 3°, but then disappear again for higher angles, among which the maximum at 4.2°. It appears, therefore, that increasing the angle of incidence mainly results in a decrease of the amplitude of the oscillations. The intermittent disappearance of the oscillations may be due to the fact that the oscillatory behavior does not scale with the increase in intensity.

In conclusion, we have made a number of observations concerning the occurrence of intensity oscillations during

FIG. 2. Intensity of the specular beam before and after interruption of growth on Ge(001) at room temperature for $\vartheta_i \sim 0.6°$. At CL the shutter is closed, at O it is opened again.
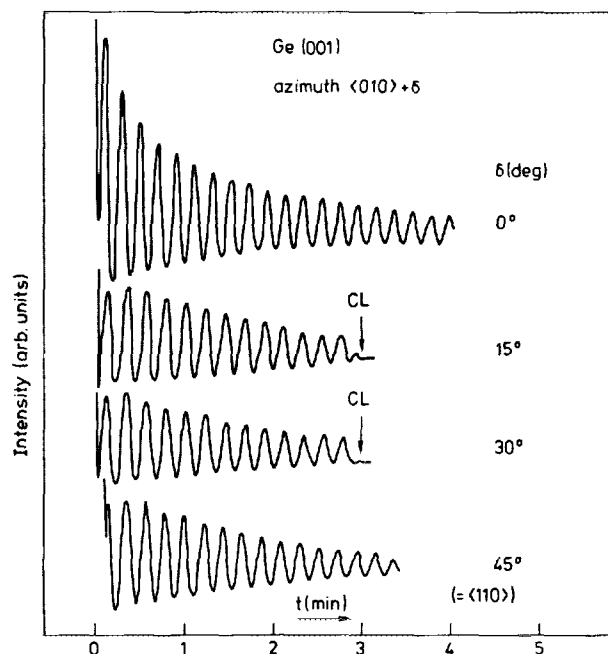

FIG. 3. Oscillations in the specular beam intensity during growth of Ge(001) at room temperature for different azimuthal angles as indicated, and for $\vartheta_i \sim 0.6°$. CL denotes closing the shutter.
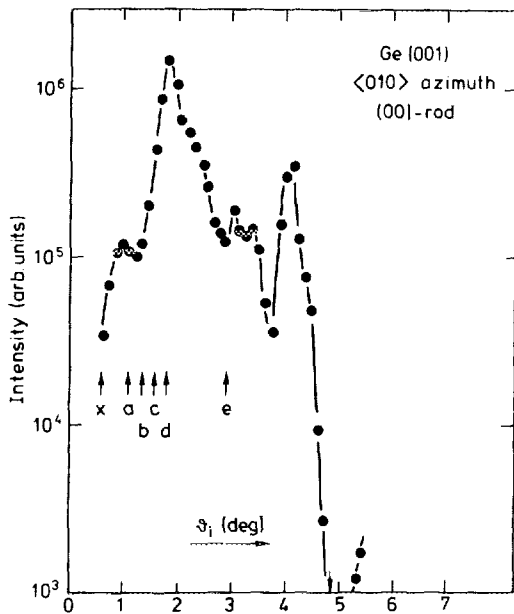
FIG. 4. Rocking curve for the (00) beam of Ge(001) at room temperature. Position x denotes the angle of incidence used for measurements shown in previous figures. Positions a–e correspond to the measurements shown in Fig. 5.
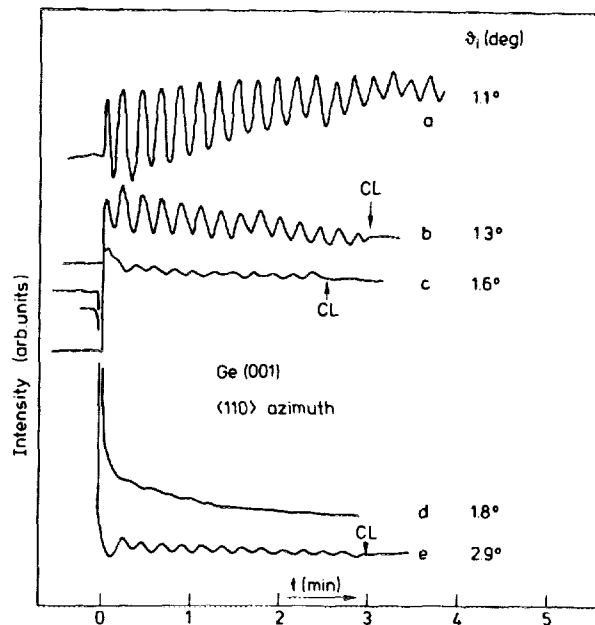

FIG. 5. Oscillations of the specular beam intensity during growth of Ge(001) at room temperature for different angles of incidence as indicated. The letters a–e correspond to the positions indicated in Fig. 4. CL denotes closing the shutter.

growth of Si and Ge. For Ge we found that the most prominent oscillations were obtained at room temperature for angles of incidence below 1°. At room temperature, recovery of the signal after growth is very small and the phase of the oscillation remains constant over long periods during interruption of growth. This must be of considerable importance in the formation of interfaces. For both Si and Ge a maximum temperature for the occurrence of oscillations was found, but no dependence of the azimuth upon the oscillatory behavior at small angles. These observations on Ge are in contrast to those of Ref. 4 on Si. Furthermore, the amplitude of the oscillations seems to decrease gradually with increasing electron beam angle of incidence, and do not follow the intensity changes of the rocking curve. Since this point is closely linked to the question of the cause of the oscillations, it will receive more attention in future investigations.

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