Optical anisotropy of Ge(001) 2×1

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We have measured the change in the optical reflection anisotropy of a clean Ge(001) surface upon exposure to molecular oxygen up to saturation coverage. Both phase and amplitude changes have been recorded with a normal-incidence ellipsometer. They have been found to be related by a Kramers–Kronig transformation. The change in the complex reflection ratio could be interpreted as an anisotropy of the clean Ge(001) 2×1 surface dielectric function, using a three-layer McIntyre–Aspnes approach and neglecting the oxygen overlayer. The surface dielectric function anisotropy can be described fairly well by optical selection rules, based on symmetry arguments. This model was applied to the possible optical transitions at this surface between filled dimers, dangling bonds and back-bonds and the empty dangling bonds and dimers.

1. Introduction

The contribution of the outermost surface to the total optical response of a material is only a few percent at maximum. Nevertheless, it is essential to understand the optical response of this top layer, if one tries to use optical techniques in surface science and technology. In these areas optical techniques have a high potential as they can monitor reaction processes at surfaces in situ. One of the many examples is the MBE growth of III–V semiconductors [1]. In surface-induced optical anisotropy (SIOA) the influence of the very surface layer on the optical response is studied.

In the past decade experiments have been performed in which the reflectance anisotropy, induced by a different geometric ordering of the atoms in two orthogonal directions parallel to the surface, has been measured. These experiments helped in clarifying the geometric structure of the optically anisotropic Si(111) 2×1 surface [2].

The theoretical understanding of SIOA phenomena, the interaction of electromagnetic waves with atoms in the surface area, has been developed along two lines: the continuous model in which an effective dielectric function is used for the description of a surface layer [3] and the discrete dipole model [4]. In the latter all atoms

Fig. 1. The asymmetric dimer reconstruction, (a) side view, (b) top view.

in the material studied are represented by dipoles with a characteristic polarizability. Recent calculations, applying the discrete dipole model to the Si(110) surface [5], have revealed that the internal electric field, resulting from the interaction between atoms, is sensitive to the geometric ordering of the atoms. To calculate surface optical features, the Maxwell equations therefore have to be treated on an atomic scale as carefully as the quantum-mechanical calculations of the polarizability of the atoms. It is expected that these local fields play an even larger role in the case of second harmonic generation, where almost the entire signal originates from the surface region. In order to investigate the influence of the electromagnetic field on an atomic scale at the surface, well defined model systems have to be used.

Undoubtedly one of the best known semiconductor surfaces is the (001) surface of silicon and germanium. This surface can lower its energy by the formation of a dimer bond between the two first-layer atoms [6–10]. There is still a discussion about whether the dimer is symmetric or not, in this work asymmetric dimers are assumed. This asymmetric dimer bond is depicted in fig. 1 and it is the building stone for the 2 × 1 reconstruction observed at room temperature. The electronic structure of the thus reconstructed surface has been found, theoretically and experimentally, to be different in the directions parallel and perpendicular to the dimer bond [7–10]. Hence the optical reflection in these two directions will also be different, while the bulk of the material is optically isotropic. Furthermore, the unit cell of this reconstruction is about the smallest possible one, which enhances the simplicity of theoretical evaluations and makes it highly suitable for the study of SIOA phenomena. However, the formation of dimers and their alignment in one direction induces anisotropic stress and strain, which is relieved by monatomic steps [11,12], because they induce a rotation of 90° of the dimer orientation, going from an upper terrace to a lower terrace.

In our SIOA study, one of the two terraces has to be eliminated, as the optical beam diameter cannot be focused below the dimension of the terraces, which is typically less than 100 nm. Such

a surface can be obtained by combining two single atomic steps into one double atomic step, which has the dimers on the upper as well as on the lower terrace parallel to the step edge [13,14]. These double steps are found on Ge(001) surfaces slightly misorientated towards the (110) direction.

The optical response was measured with a rotating analyzer ellipsometer (RAE) [15–17]. However, in contrast to standard RAE we measured at normal incidence. The common difference in reflection for s- and p-polarized light is now provided by the difference in reflection of the two geometric axes of the clean Ge(001)2 × 1 surface, the intrinsic $\bar{1}10$ and 110 axes. At normal incidence the complex anisotropic reflection ratio $\bar{\rho}^{\text{ni}}$ is defined as

$$\bar{\rho}^{\text{ni}} \equiv \frac{\bar{r}(\bar{1}10)}{\bar{r}(110)} \equiv \tan(\Psi) e^{i\Delta}. \quad (1)$$

The experimental configuration and the difference compared to classical ellipsometry will be discussed first. Special attention will be given to the measurement of the phase anisotropy, which has not been taken into account in most optical measurements so far. The changes in both amplitude and phase anisotropy, monitored upon exposure of the clean single domain Ge(001)2 × 1 surface to molecular oxygen up to saturation coverage, will be presented. We determined the optical anisotropy change because an absolute optical measurement in UHV environment is not easy to perform. In our interpretation we will make the usual assumption, provided by Bootsma and Meyer [18] and consider the germanium oxide layer as optically isotropic. The experimental data will be analyzed qualitatively within the continuous model. The position of the features recorded and their shape, will be shown to give good correspondence with a simple optical selection rules model, based on symmetry arguments, applied to data obtained from literature.

2. Experimental

In our experiments we used a Ge(001) wafer with a misorientation angle of 5° towards the

Fig. 2. Experimental configuration. (L) 75 W Xe lamp or 150 W halogen lamp, (M) Oriel 7240 monochromator, (F) 250 mm lens and 3 mm diaphragm, (P) Glan-Taylor polarizer, (Al₁, Al₂) aluminum mirrors, (S) sample, (A) Glan-Taylor rotating analyzer, (PM) EMI 9659 QB-S20 photomultiplier.

(110) direction. The surface was cleaned by ion bombardment (Ar⁺, 800 eV, dose $\approx 10^{13}$ ions/cm² · s, 10 min), followed by ion bombardment and simultaneous annealing [19] (≈ 700 K, 20 min) and finally annealing only (≈ 800 K, 1 h). The structure of the clean surface was determined by LEED measurements and a single domain 2 × 1 reconstruction was found. Several LEED spots were doubled, indicating regular terraces [20,21]. Fractional and doubled spots revealed that the dimers are parallel with the terrace steps, which agrees with DB steps [13,14]. The misorientation, estimated from the LEED spot splitting, was in reasonable agreement with the nominal angle of 5°.

The experimental configuration for optical anisotropy measurements is shown in fig. 2 and consists of a rotating analyzer ellipsometer at normal incidence. A parallel, monochromatic, linear polarized beam is reflected at an aluminum mirror (Al₁), passes a fused quartz, low-strain UHV window, which is tilted 5° off normal, and reflects on the germanium sample [29,30]. The sample is set in a position such that the $\bar{1}10$ axis corresponds to the s-polarization defined by the Al₁ mirror. The light incident on the Al₁ mirror is polarized at an angle of -45° with respect to the p axis. The reflected beam passes the UHV

window for the second time and reflects on the Al₂ mirror. The angle of the incident beam with the surface normal is less than 1° and therefore we consider the beam to be at normal incidence. The polarization state of the thus reflected beam is analyzed by a rotating analyzer at a frequency of $\omega_A = 67$ Hz. The change in polarization state $\tilde{\rho}_{\text{tot}}$ between polarizer and analyzer is described by [15]

$$\tilde{\rho}_{\text{tot}} = \tan(\Psi_{\text{tot}}) e^{i\Delta_{\text{tot}}} = \tilde{\rho}_{\text{Al}_2} \tilde{\rho}_{\text{W}_2} \tilde{\rho}_{\text{Ge}}^{\text{ni}} \tilde{\rho}_{\text{W}_1} \tilde{\rho}_{\text{Al}_1}. \quad (2)$$

Back-reflections and phase retardations between the components have been neglected. Al refers to the aluminum mirrors, W to the UHV window and Ge to the sample. The normalized intensity as a function of time at the detector is

$$I_n(t) = 1 + a(p) \cos(2\omega_A t) + b(p) \sin(2\omega_A t). \quad (3)$$

In this equation, p is the fixed polarization angle between the plane of incidence on the aluminum mirrors and the plane of transmission of the polarizer. The coefficients $a(p)$ and $b(p)$ are obtained from a Fourier analysis of the detector signal and from these $\tan(\Psi_{\text{tot}})$ and $\cos(\Delta_{\text{tot}})$ can be calculated according to

$$\tan(\Psi_{\text{tot}}) = \frac{1}{|\tan(p)|} \left(\frac{1 + a(p)}{1 - a(p)} \right)^{1/2} \quad (4a)$$

$$\cos(\Delta_{\text{tot}}) = \text{sign}(\tan(p)) \frac{b}{(1 - a^2(p))^{1/2}}. \quad (4b)$$

In a standard RAE configuration at normal incidence [22], the phase $\Delta_{\text{tot}} = 180^\circ$. Because we measure the cosine of this angle, we are extremely insensitive to small changes of Δ_{tot} in this region. This problem can be solved by using a phase retarder. In our case two aluminum mirrors were used as phase retarders and the resulting Δ_{tot} increases linearly from 201° at 2 eV to 232° at 5 eV. This not only increases the sensitivity to small phase changes, but it also allows us to use a standard RAE residue calibration procedure for the determination of the polarizer and analyzer offset angles and the influence of the detection electronics [16]. As a side effect the total signal strength increases due to the large

reflectance of the aluminum mirrors at the photon energies used (> 90%).

The influence of errors, introduced by imperfections of polarizer, analyzer and windows, has been discussed extensively by de Nijs and van Silfhout [17]. In the present work these errors are neglected. This is justified since we perform a relative measurement in which the change in $\tilde{\rho}_{\text{tot}}$ upon a large O₂ exposure of the clean Ge surface is recorded. Since only the Ge sample is affected by this oxidation process, the change in $\tilde{\rho}_{\text{Ge}}^{\text{ni}}$ is monitored (cl refers to the clean surface, ox refers to the O₂ exposed surface):

$$\frac{\tilde{\rho}_{\text{tot,cl}}}{\tilde{\rho}_{\text{tot,ox}}} = \frac{\tilde{\rho}_{\text{Ge,cl}}^{\text{ni}}}{\tilde{\rho}_{\text{Ge,ox}}^{\text{ni}}} = \frac{\tan(\Psi_{\text{cl}})}{\tan(\Psi_{\text{ox}})} e^{i(\Delta_{\text{cl}} - \Delta_{\text{ox}})} \equiv (1 + \delta \tan(\psi)) e^{i\delta \Delta}. \quad (5)$$

In our experiments the clean surface was exposed to molecular oxygen at room temperature until saturation coverage was reached. The change in $\delta \tan(\Psi)$ as a function of the photon energy is given in fig. 3a. Two broad peaks can be distinguished for $\delta \tan(\Psi)$ in the measured energy range, one around 1.8 eV and one around 3.2 eV. A minimum in $\delta \tan(\Psi)$ is observed at 2.3 eV.

Using the aluminum mirror it was possible to measure $\delta \Delta$ in the same photon energy region as well and the result is given in fig. 3b. The measured peak positions are the same as the ones observed by Zandvliet et al. [23]. They used a standard rotating analyzer ellipsometer and were able to relate their measurements to STM measurements and calculations.

3. Discussion

The amplitude and phase of a complex quantity should exhibit a Kramers–Kronig relation, based on a causality argument. This relation also holds for the change in the reflection ratio, yielding for the pair $\delta \tan(\Psi)$ and $\delta \Delta$ the following Kramers–Kronig transformation [24]:

$$\delta \Delta(\omega) = -\frac{2\pi}{\omega} \times \int_0^\infty \frac{\ln[\delta \tan(\Psi(\omega'))] - \ln[\delta \tan(\Psi(\omega))]}{(\omega')^2 - \omega^2} d\omega'. \quad (6)$$

This relation has been applied to the solid line in fig. 3a, the left- and right-hand tails have been extrapolated, as shown, in order to calculate the

Fig. 3. Change in ellipsometric angles as a function of photon energy $\hbar\omega$ upon exposure to molecular oxygen of a clean single domain Ge(001)2 × 1 surface up to saturation coverage. (a) $\delta \tan(\Psi)$: (●) measurements, the line is the approximation used in the Kramers–Kronig transformation, solid part: approximation of the data points, the hatched line is an extrapolation to zero. (b) $\delta \Delta$: (●) measurements, the solid line is the Kramers–Kronig transformation of the line in (a).

integral. The $\delta\Delta$ resulting from this transformation is depicted as the solid line in fig. 3b. From this figure it can be seen that a good agreement is obtained with the measured data points. This result implies that Kramers–Kronig relations are valid in this type of highly confined systems.

The analysis of the change in surface-induced optical anisotropy can be performed along two lines, the continuous model, using the dielectric constant (see, e.g., refs. [3,25]) and the discrete dipole model [4]. In the discrete dipole model, atoms are represented by dipoles with a certain polarizability derived from quantum mechanics. The electromagnetic interaction between the dipoles is governed by the local field as obtained from the Maxwell relations. In contrast, the continuous model uses solely a dielectric function to describe the problem. The influence of local fields on microscopic level has been very clearly shown by comparing the optical response of various reconstructed Si(110) surfaces [5]. Changes in the optical anisotropy were induced by a different geometric ordering of dipoles with the same polarizability. However, the local field variations only affect the intensity of anisotropic optical features. The energy positions and signs of these features are mainly governed by the quantum mechanics of the system, i.e., the polarizability of the dipoles. Therefore, if the intensities are wanted correctly, a full evaluation of the Maxwell relations on a microscopic scale is essential. For the Ge(001)2 × 1 case, treated in this article, such a detailed approach is not feasible, since the necessary detailed quantum-mechanical data are not available yet. So a crude form of the continuous model will be used to arrive at a simple but adequate picture to interpret our results. Hence we interpret the measured optical anisotropy in terms of the product of a surface layer d_{ss} and the anisotropy in the surface dielectric function in a McIntyre–Aspnes sense [1,26], as estimated from the quantum-mechanical selection rules of surface optical processes. However, one has to bear in mind that the continuous model can only give a qualitative interpretation of the measured optical features.

The clean germanium surface is described by three layers: the first layer is the semi-infinite

bulk Ge crystal with a dielectric function $\tilde{\epsilon}_b$, the second layer describes the surface states with a dielectric constant $\tilde{\epsilon}_{ss}$ of uniform thickness d_{ss} and the third layer is the vacuum region. Next, the clean Ge surface is exposed to molecular oxygen up to saturation coverage. This exposure changes the surface layer. Following Bootsma and Meyer [18] we will assume that this surface state layer is completely replaced by a thin optically transparent germanium oxide layer. In addition we also assume it to be isotropic. Then the oxidized surface can be effectively represented by two layers, the first layer being the semi-infinite bulk and the second layer the vacuum region.

Bulk germanium is optically isotropic and the recorded optical anisotropy change is related to the in-plane surface dielectric constant $\tilde{\epsilon}_{ss}$ [2,3,25]. The difference in dielectric function between the two orthogonal geometric axes parallel to the surface is given by [1]

$$\begin{aligned} \Delta\tilde{\epsilon}_{ss}d_{ss} &= (\tilde{\epsilon}_{ss}^{(\bar{1}10)} - \tilde{\epsilon}_{ss}^{(110)})d_{ss} \\ &= \frac{ic}{2\omega} (\tilde{\epsilon}_b - 1) \left(\frac{\tilde{\rho}_{Ge,cl}^{ni}}{\tilde{\rho}_{Ge,ox}^{ni}} - 1 \right). \end{aligned} \quad (7)$$

Eq. (7) is only valid when the Bootsma–Meyer assumption holds. The asterisks (*) in fig. 4 are the values of the imaginary part of $\Delta\tilde{\epsilon}_{ss}d_{ss}$ calcu-

Fig. 4. $\text{Im}(\Delta\tilde{\epsilon}_{ss}d_{ss})$, (*) calculated from fig. 3, the solid line is obtained by symmetry based optical selection rules.

lated from data of fig. 3, by applying eq. (7) and using values for $\bar{\epsilon}_b$ from ref. [27].

The imaginary part of the dielectric function reflects the absorption of electromagnetic waves in a medium and is related to the quantum-mechanical properties of the system through [28]

$$\text{Im}(\bar{\epsilon}_{ss}) \sim \frac{1}{\omega^2} \sum_{i,f} |\langle \Psi_f | \mathbf{e} \cdot \mathbf{p} | \Psi_i \rangle|^2 \mathcal{J}_{fi}(\hbar\omega), \quad (8)$$

where $|\Psi_{i(f)}\rangle$ is the initial (final) state, $\mathbf{e}$ is the polarization of the incident electric field (i.e., $(\bar{1}10)$ or (110)), and $\mathbf{p}$ is the electron momentum operator. $\mathcal{J}_{fi}(\hbar\omega)$ is the joint density of states of the initial and final states, separated by a photon energy $\hbar\omega$. We let $\mathcal{J}_{fi}(\hbar\omega)$ determine the energy position of an optical transition. As we want to give a qualitative picture using a continuous model, we will use the symmetry properties of the initial and final states to discriminate between optically active and non-active transitions. Therefore the matrix element equals zero unless the wave function of the initial and final states have unequal symmetries (i.e., even and odd) along the polarization direction $\mathbf{e}$ and equal symmetries perpendicular to $\mathbf{e}$ (both even or both odd) [28]. The difference in the two polarization directions of the electric field determines the sign of $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss})$ in eq. (7) and thus reflects the difference in electronic structure in the $(\bar{1}10)$ and (110) directions.

The electronic structure around the Fermi level of the clean Ge(001)2 × 1 surface is known from a number of theoretical and experimental investigations [7–10]. The surface states are attributed to filled (empty) dimer states $D_i(D_i^*)$, filled (empty) dangling bonds $D_{up}(D_{down})$ and the back-bonds B_1 and B_2 . Predominant symmetries have been derived for these six states from refs. [7–10] and are listed in table 1. In table 2 the transition energies estimated from these references for transitions between the four filled and two empty states are given. In table 2 we also listed the sign in $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss})$ of the eight possible transitions, as derived from symmetry-based optical selection rules. As an example the $D_{up} \rightarrow D_i^*$ transition will be described more precisely. From refs. [8,9] we derive an energy of 1.85 eV as an

Table 1

Predominant symmetries of the various surface states along the (110) and $(\bar{1}10)$ directions

	D_i^*	D_{down}	D_{up}	B_1	D_i	B_2
(110)	Even	Even	Even	Odd	Even	Odd
$(\bar{1}10)$	Odd	Odd	Even	Even	Even	Odd

estimate for the position of the $D_{up} \rightarrow D_i^*$ transition. The only change in the predominant symmetries of the two surface states involved is in the $(\bar{1}10)$ direction. Therefore, the optical absorption will mainly take place for $(\bar{1}10)$ polarized photons, which yields, according to eq. (8), that $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss}) > 0$. From the signs in table 2 derived in this way, we construct a qualitative spectrum for $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss})$ by setting all nonzero matrix elements in eq. (8) equal to +1 and taking for $\mathcal{J}_{fi}(\hbar\omega)$ a Gaussian line shape of fixed width. We take Gaussian peaks in order to reflect the Gaussian broadening of the energy levels. The result of this procedure has been scaled to the 1.85 eV value of the measured spectrum and is depicted as the solid line in fig. 4. We observe a fairly good agreement between our experimentally determined value for $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss})$ and the spectrum derived from optical selection rules. The peak signs and shapes of the observed transitions are correct as we would expect for a contin-

Table 2

Possible transitions at clean Ge(001)2 × 1, with their transition energies estimated from refs. [7–10]

Transition	Peak [eV]	Symmetry change	Peak sign in $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss})$
$D_{up} \rightarrow D_{down}$	1.1	$(\bar{1}10)$	+
$B_1 \rightarrow D_{down}$	1.6	$(110)\&(\bar{1}10)$	No peak
$D_{up} \rightarrow D_i^*$	1.85	$(\bar{1}10)$	+
$B_1 \rightarrow D_i^*$	2.2	$(110)\&(\bar{1}10)$	No peak
$D_i \rightarrow D_{down}$	2.6	$(\bar{1}10)$	+
$D_i \rightarrow D_i^*$	3.1	$(\bar{1}10)$	+
$B_2 \rightarrow D_{down}$	3.5	(110)	–
$B_2 \rightarrow D_i^*$	4.0	(110)	–

The change in symmetry of the two geometric directions on the surface determines whether an optical transition is possible by symmetry-based optical selection rules. The polarization dependence of such an optical transition is given by the sign in $\text{Im}(\Delta\bar{\epsilon}_{ss}d_{ss})$.

uous model. On the basis of our experience with the Si(110) surface [5] we expect that the other possible reconstructions of the Ge(001) surface will show an interesting change in optical anisotropy spectrum. The $D_{\text{up}} \rightarrow D_{\text{down}}$ transition at 1.1 eV is expected to have a positive value in $\text{Im}(\Delta \tilde{\epsilon}_{\text{ss}} d_{\text{ss}})$ however it falls, at the moment, outside the experimental photon energy range, due to limitations of the detector used.

4. Conclusions

With a minor adjustment it is possible to change a standard rotating analyzer ellipsometer into a powerful tool for the detection of surface-induced optical anisotropy. The change in the reflection anisotropy of a Ge(001)2 × 1 surface upon exposure to molecular oxygen can be interpreted straightforwardly as a difference in electronic structure in the two orthogonal directions at the surface. It was also possible to confirm the Kramers–Kronig relation in the highly confined system studied.

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