Optical response of clean and hydrogen-covered vicinal Si(001) 2 × 1 surfaces

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Abstract
The optical response of the clean and di-hydrogenated Si(001) vicinal surface is experimentally investigated by two complementary optical techniques, reflectance anisotropy spectroscopy in normal incidence and surface differential reflectance spectroscopy in oblique incidence with s or p polarizations. The results are in good agreement with previous *ab initio* theoretical calculations. The components of the surface optical response tensor for polarization of light parallel and perpendicular to the dimers of silicon are determined from the experiments performed in s polarization. Moreover, it is shown that the component perpendicular to the surface is negligible and yields no specific effect in the p-polarization measurements.

1. Introduction

It was demonstrated a long time ago that linear optical methods can be very efficient probes for studying surfaces or interfaces of crystals, in various environments such as ultra-high vacuum (UHV), gas or liquids [1–8]. The two main surface-sensitive techniques that are intensively used are reflectance anisotropy spectroscopy (RAS), also called by some authors reflectance difference spectroscopy (RDS), and surface differential reflectance spectroscopy (SDRS). RAS, which measures the anisotropy of the optical response of a surface, can be used only with samples which display a crystalline or morphological anisotropy on a dimension range equal to the size of the optical beam, typically on several mm^2, except in some specific developments where microscopic spots can be used. SDRS consists in comparing the reflectance of a clean surface with a surface which has been modified, for instance by gas adsorption. It results in the determination of the relative change of reflectance induced by the adsorption, and therefore gives indirect information on the optical response of the clean surface.

The linear optical response of a surface is completely described by use of a complex diagonal response tensor $\bar{\bar{\Lambda}}(\omega)$ [9, 10], following the notation of Bagchi and Barrera, which has dimension of length, with components $\Lambda_x(\omega)$, $\Lambda_y(\omega)$ and $\Lambda_z(\omega)$. The theoretical determination of this tensor is a formidable task, which requires heavy calculation. Important progress has been obtained by different groups in the understanding of the optical response of semiconductor surfaces, either by use of the semi-empirical tight-binding approach (TB) [6, 11], or more recently by use of first-principles approaches such as the density-functional theory in the local-density approximation (DFT-LDA) [12]. Recent improvements have been made by introducing corrections due to excitonic and local-field effects [13].

The aim of the present study is to tentatively determine the three components of the response tensor of a well characterized surface, the vicinal Si(001) surface with a disorientation of $4°$. We combined both RAS and SDRS to investigate the clean reconstructed surface and the unreconstructed one obtained after adsorption of hydrogen at room temperature. The adsorption of hydrogen removes electronic surface states and permits us to get a bulk-like optical response for the surface, which can be compared to the clean one.

2. Experimental details

2.1. Preparation and characterization of the samples

The Si(001) 2×1 surface, when prepared under UHV, is formed by rows of Si dimers, which are all parallel [14]. However, the resulting crystalline anisotropy extends only on small regions with respect to the light beam, and the surface is actually composed of 2×1 and 1×2 domains rotated by $90°$ with respect to each other, yielding an overall isotropic surface. In order to obtain a single-domain surface, we have been using a well known and controlled method, namely the use of vicinal Si(001) surfaces with a disorientation of $4°$ along the [110] direction [15]. The samples were prepared by direct-current heating for several seconds at $1050\,°C$. The base pressure was 5×10^{-11} Torr and the pressure was maintained below 3×10^{-10} Torr during heating. The steps at the surface, which are aligned along the [$\bar{1}$10] direction, gather two by two to form double steps, separating terraces of average width of 3.9 nm, where the dimers are all parallel to the steps, as can be seen on the scanning tunnelling microscopy image of figure 1(a). The dimer rows can be distinguished and are perpendicular to the steps, therefore parallel to the [110] direction. They display a zigzag appearance, which is due to the alternative buckled dimers [16]. This procedure yields a mainly 2×1 single-domain surface, with minority 1×2 domains covering about 10%–20% of the surface. The LEED pattern drawn in figure 1(b) actually shows the 1×1 spots with 2×1 spots and faint 1×2 ones. Some of the principal 1×1 spots are double spots, which is due to the periodicity of the steps along the [110] direction. The adsorption of atomic hydrogen on the surface, maintained at room temperature, is obtained by dissociating molecules of hydrogen (with a pressure of 10^{-6} Torr of H_2) by use of a hot tungsten wire located in front of the sample. In order to avoid any contamination due to the tungsten wire, it was previously outgassed so that the increase of pressure was of the order of 10^{-11} Torr. This results in a unreconstructed and poorly ordered Si(001) 1×1:H surface where the Si dimers are broken and the Si atoms are bound to two H atoms [17], as confirmed by the 1×1 LEED pattern (figure 1(c)).

2.2. Optical techniques: RAS and SDRS

Both the RAS and the SDRS apparatuses are home-made spectrometers. The RAS is based on a photoelastic modulator, using the same principle as the one developed by Aspnes [18], and
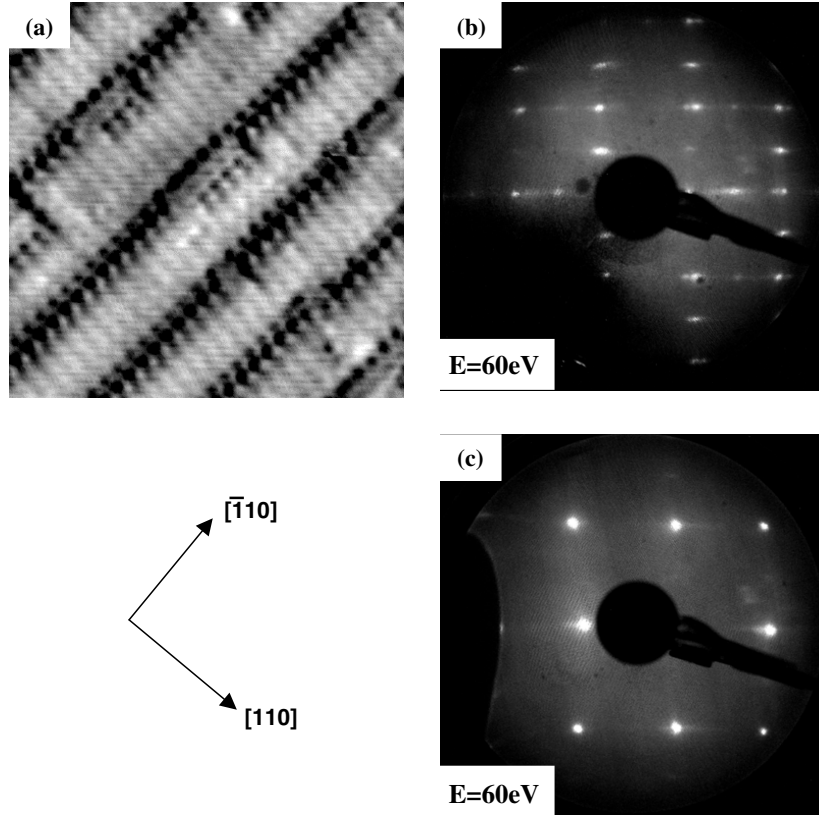
Figure 1. (a) STM image of the clean Si(001) 2 × 1 vicinal surface; (b) LEED pattern of the clean Si(001) 2 × 1 vicinal surface; (c) LEED pattern of the di-hydrogenated Si(001) 1 × 1:H vicinal surface.

permits us to deliver both the real and the imaginary parts of the reflectance anisotropy:

$$\frac{\Delta r}{r} = 2\frac{r_{[\bar{1}10]} - r_{[110]}}{r_{[\bar{1}10]} + r_{[110]}} \tag{1}$$

where the orientations $[\bar{1}10]$ and $[110]$ (respectively parallel and perpendicular to the dimers) are specified in figure 2. The reflectances $r_{[\bar{1}10]}$ and $r_{[110]}$ are the complex coefficients of amplitude reflection when the incident electric field is polarized along the two directions.

The SDRS was performed by use of a spectrometer, based on an optical multichannel analyser consisting in a Si photodiode array, described in detail previously [19]. It delivers the relative change of reflectivity of the substrate upon adsorption of hydrogen:

$$\left(\frac{\Delta R}{R}\right)_{s,p} = \frac{R_{Si} - R_{Si:H}}{R_{Si}} \tag{2}$$

where R_{Si} and $R_{Si:H}$ are the reflectivities (in intensity, i.e. the square of the reflectance: $R \equiv |r|^2$ in the case of normal incidence) of the clean Si surface and of the H-covered Si surface respectively. The measurements were performed in oblique incidence of 45°, in s or p polarization with the electric field along $[\bar{1}10]$ or $[110]$ (see figure 2).
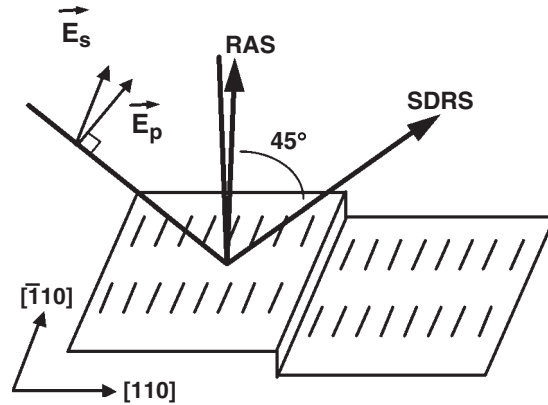
Figure 2. Scheme of the optical orientations.

3. Results and discussion

3.1. RAS and SDRS measurements

Figures 3(b) and (c) give the real and imaginary parts of the RA spectra for the clean 2×1 surface, whereas figure 3(a) displays the real part of the RA spectrum measured on the 1×1 di-hydrogenated surface. The real part of the 2×1 RA spectrum is similar to previous measurements performed by other groups [4, 20, 21]. Actually, the spectrum is indicative of the quality of the surface, and the intensity of the signal gives a good indication of the number of majority 2×1 domains and minority 1×2 ones. A slight contamination or a poorly reconstructed surface results in RA spectra which can differ strongly from the clean one. Although a complete understanding of RAS for Si(100) has not been achieved so far [22], it is known from recent *ab initio* calculations that the different features originates both from the dimers on the terraces and from the steps [23]. It also appears that there is no minimum at 1.6 eV in our spectra, although it was observed for anisotropic nominal surfaces displaying very large terraces [20, 24], and attributed to optical transitions between occupied and unoccupied surface bands corresponding to a hybridization along the dimer rows of the π and π^* states of the Si dimers [12, 23]. However, as can be seen here, the RA signal below 2 eV does not reduce to zero and a very slight shoulder is visible around 1.6 eV. It suggests that this transition is not suppressed for the vicinal surface but is rather broadened because of the presence of steps, which could induce some scattering. It can also be seen that the signal is much larger for the clean surface: the di-hydrogenated surface displays only a very small anisotropy, dominated by two structures at the bulk critical points of Si. They could originate from a small strain in the bulk induced by the surface [22].

The measurement of both the real and imaginary parts of RA permits us to determine the anisotropy of the surface dielectric response, namely the difference of the response tensor components $\Lambda_{[110]}(\omega) - \Lambda_{[\bar{1}10]}(\omega)$, by use of the following equation:

$$\frac{\Delta r}{r} = 2\frac{r_{[\bar{1}10]} - r_{[110]}}{r_{[\bar{1}10]} + r_{[110]}} = -2\mathrm{i}\frac{\omega}{c}\frac{\Lambda_{[\bar{1}10]}(\omega) - \Lambda_{[110]}(\omega)}{\varepsilon_{\mathrm{b}}(\omega) - 1} \tag{3}$$

where ε_{b} is the bulk dielectric function of silicon. Figure 4 shows the imaginary part of $\Lambda_{[110]}(\omega) - \Lambda_{[\bar{1}10]}(\omega)$, which is dominated by two main features. There is equilibrium between the positive and the negative parts, which is due to the fact that the weight of the different transitions, integrated over the whole range, does not depend on the orientation of the electric
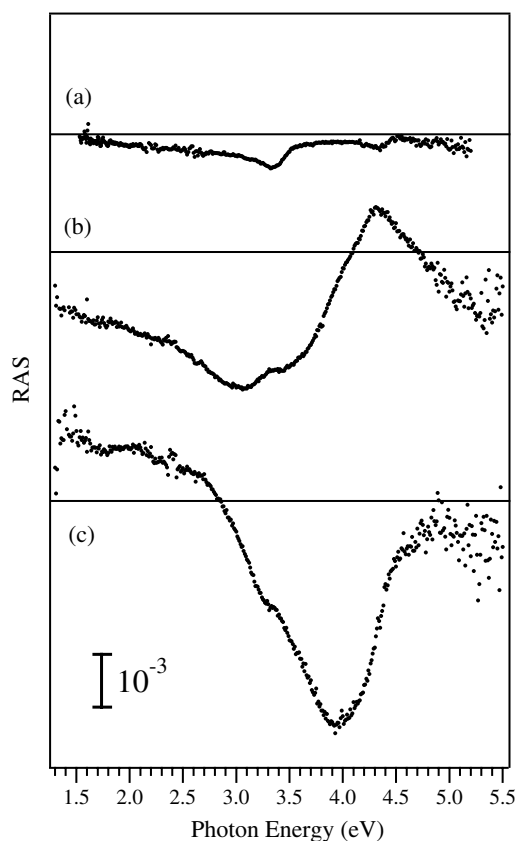
Figure 3. (a) Real part of the reflectance anisotropy spectrum measured on the di-hydrogenated Si(001) 1 × 1:H surface; (b) and (c) real and imaginary parts of the RAS measured on the clean Si(001) 2 × 1 surface.

field. The shape of this curve is different from the one of $\text{Re}\{\Delta r/r\}$, because the denominator in (3): $\varepsilon_b(\omega) - 1$ has real and imaginary parts. Finally, it can be seen that this calculated quantity is zero above 4.5 eV, which can be explained by the fact that surface effects, hence surface anisotropy, become negligible for large energies, where transitions are mainly bulk transitions.

$\Lambda_{[110]}(\omega) - \Lambda_{[\bar{1}10]}(\omega)$ is the largest piece of information concerning the optical response of a crystalline surface which can be obtained from RAS. The advantage of the method is that it gives the absolute anisotropy of the clean surface. But it gives only the anisotropy, which means that we do not get complete information on the optical response of the surface. The second technique we have been using here, SDRS, has the advantage of giving separate pieces of information on the optical response along the different directions of the surface [$\bar{1}10$], [110], and the normal direction [001]. However, this technique cannot deliver the absolute optical response, only the difference between the clean surface and the perturbed one. The validity of this approach is based on the fact that the optical response of the perturbed surface is very close to the one that would be expected for a surface having the same electronic structure as the bulk. This approximation appears to be reasonable in the case of a di-hydrogenated surface: it displays no specific atomic feature such as dimers, and all the dangling bonds which could introduce surface states are saturated by hydrogen. The small intensity of the experimental RA
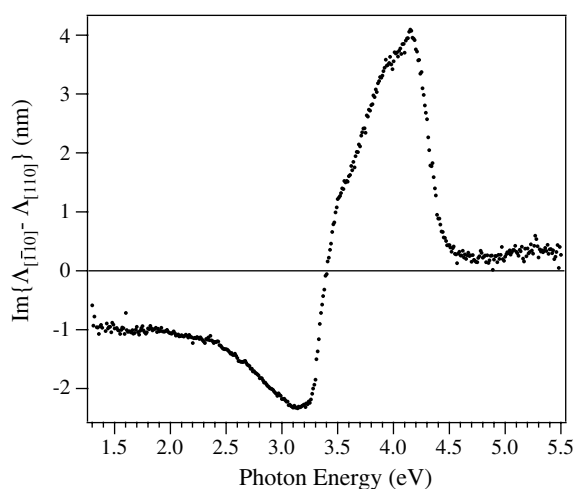
Figure 4. Imaginary part of the anisotropy of the surface optical tensor of the Si(001) 2 × 1 surface.
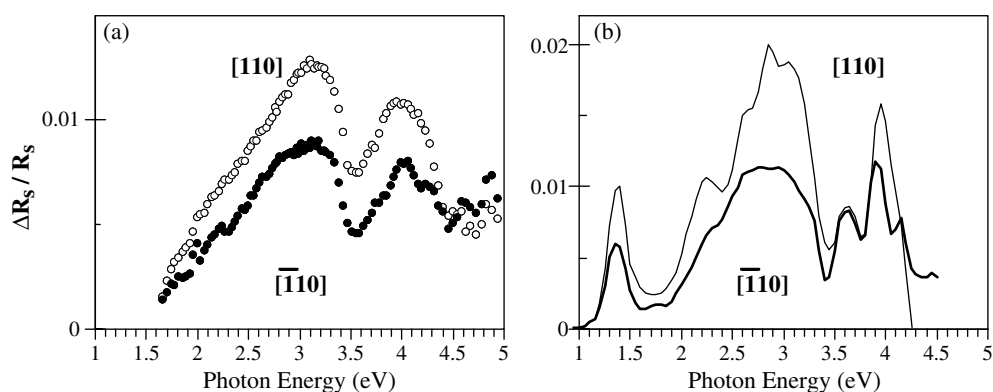

Figure 5. (a) Experimental surface differential reflectance spectra measured in s polarization, for light along the [$\bar{1}$10] (filled circles) and [110] (empty circles) directions. (b) DFT-LDA calculation for electric field along [$\bar{1}$10] (thick curve) and [110] (thin curve), after [26]. The signal has been divided by $\cos(\theta) = \cos 45°$ for accounting for the oblique incidence in the experiments.

spectrum for the 1 × 1:H surface with respect to the clean one (figure 3) is a good indication that the optical response of the hydrogenated surface is negligible with respect to the clean one. In consequence, we have measured the SDR spectra in s and in p polarizations, with two different orientations of the sample, either as shown in figure 2, or after rotating the sample by 90°. The spectra therefore reflect mainly the optical response of the clean surface when neglecting that from the hydrogenated one. Figure 5(a) presents the SDR spectra in s polarization for the electric field along the [$\bar{1}$10] direction, i.e. along the dimers, and for the electric field along the [110] direction, i.e. perpendicular to the dimers. Both spectra are very similar and are dominated by two main peaks around 3 and 4 eV. It can be shown that the 3 eV feature is mainly related to the dangling bonds of the dimers, while the 4 eV one is related to transitions of more internal electronic states of the dimers [25]. These experimental spectra can be compared with *ab initio* calculations performed in Del Sole's group a few years ago, where the same quantities have been obtained, namely the differential reflectivity between a clean

surface and the di-hydrogenated one [26]. The theoretical curves are drawn in figure 5(b). In order to compare the theory with the experiments, we have applied a shift of 0.5 eV towards larger energies in the theoretical curves, which compensates the well known underestimation of the bandgap of semiconductors in the *ab initio* approach. The same authors, in a more recent paper [12], have used an upward rigid shift of 0.5 eV to the conduction band, in agreement with previous theoretical results on the Si(001) surface. It can be seen that the agreement between the experiment curves and the theoretical ones is excellent, for the general feature as well as for the intensity which is only slightly smaller in the experiment than in the calculation. We do not get in the experiment the sharp peak at 1.4 eV seen in the calculated curves, due to the $\pi-\pi^*$ transition. This is due of course to the limited range we use here and to the fact that, as discussed for RAS, this optical transition is strongly damped or even suppressed for a vicinal surface, whereas the calculations have been done for a nominal single-domain surface without steps. Our results also confirm that the optical response of Si(001) is weakly anisotropic, and that the reflectance is larger for polarization along the dimer rows, in agreement with theory. In spite of the missing transition at 1.4 eV, the good agreement between experiment and theory for the 1.7–5 eV range reinforces our choice of working with the vicinal Si surface, which is much easier to prepare with a better reproducibility than the single-domain nominal one.

3.2. Determination of the optical response tensor of Si(001) 2 × 1 and discussion

The surface differential reflectance spectra in s and p polarizations are given by the formulae

$$\frac{\Delta R_s}{R_s} = 4\frac{\omega}{c}\cos\theta \, \text{Im}\left\{\frac{\delta\Lambda_{[\bar{1}10]}(\omega)}{\varepsilon_b(\omega)-1}\right\} \tag{4}$$

$$\frac{\Delta R_p}{R_p} = 4\frac{\omega}{c}\cos\theta \, \text{Im}\left\{\frac{(\varepsilon_b(\omega)-\sin^2\theta)\delta\Lambda_{[110]}(\omega)+\varepsilon_b(\omega)^2\sin^2\theta\,\delta\Lambda_{[001]}(\omega)}{(\varepsilon_b(\omega)-1)(\varepsilon_b(\omega)\cos^2\theta-\sin^2\theta)}\right\} \tag{5}$$

where θ is the angle of incidence (45°), $\varepsilon_b(\omega)$ is the bulk dielectric function of silicon, and the $\delta\Lambda(\omega)$ are the differences between the components of the response tensors of the clean Si(001) surface and the hydrogenated one, and are assumed to be very close to (and will be identified with in the following) the tensor components of the clean surface, as discussed above. When rotating the sample by an angle of 90°, the equations are similar, after exchanging the directions [$\bar{1}$10] and [110]. This response tensor, which has the dimension of length, has complex components, which, due to the principle of causality, fulfil the Kramers–Kronig relation [9]. Consequently, they can be expressed, for example, as sums of Lorentzian functions:

$$\Lambda(\omega) = \sum_j \frac{f_j}{\omega_j^2-\omega^2-i\Gamma_j\omega}. \tag{6}$$

The fitting of the Lorentzian function parameters, in order to reproduce the SDR spectra in s polarizations, allowed us to determine $\Lambda_{[\bar{1}10]}(\omega)$ and $\Lambda_{[110]}(\omega)$, i.e. the optical response parallel and perpendicular to the dimers. Their imaginary parts are drawn in figure 7 in comparison with the bulk dielectric function[1]. Both surface dielectric responses display a main peak lying below 3.5 eV, and a secondary peak centred around 4 eV. These peaks correspond clearly to the SDR peaks observed in figure 5. It is worthwhile to notice that, although the SDR spectrum for light polarization along the dimers ([$\bar{1}$10] direction) has a smaller overall intensity than the spectrum for light perpendicular to the dimers ([110] direction), this is not the case for Im$\{\Lambda_{[\bar{1}10]}(\omega)\}$ with respect to Im$\{\Lambda_{[110]}(\omega)\}$, the 4 eV peak being more intense for

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[1] Actually, there is some uncertainty on the intensity of $\Lambda(\omega)$: it can be seen from the equation that, if $\Lambda(\omega)$ is a solution of the fitting, any expression $\Lambda(\omega)+\alpha(\varepsilon_b-1)$ is also a solution. We have chosen the constant α so that Im$(\Lambda(\omega))$ does not display negative values.
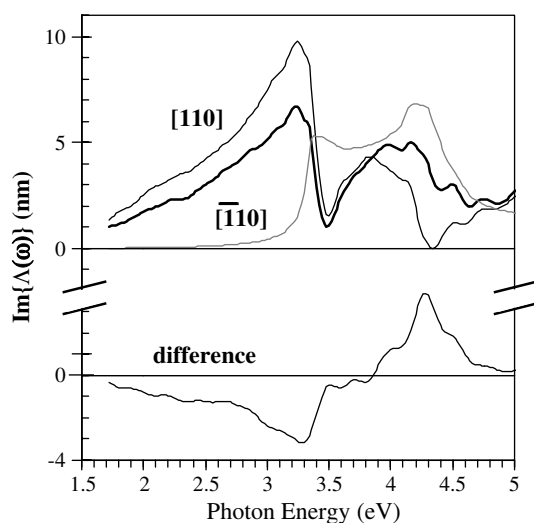
Figure 6. Imaginary part of the components parallel to the surface of the response tensor. Thin curve, $\Lambda_{[110]}(\omega)$; thick curve, $\Lambda_{[\bar{1}10]}(\omega)$; grey curve, $\text{Im}\{\varepsilon_b(\omega)\}$ (divided by 5). $\text{Im}\{\Lambda_{[\bar{1}10]}(\omega) - \Lambda_{[110]}(\omega)\}$ is also drawn.
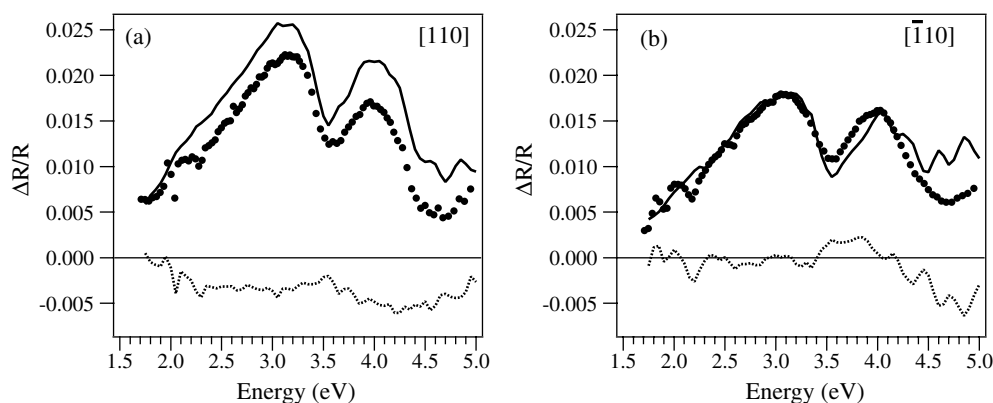

Figure 7. (a) Filled circles: experimental surface differential spectra in p polarization $\left(\frac{\Delta R_p}{R_p}\right)^{[110]}$, for the incidence plane along the [110] direction, as indicated in figure 2. Continuous curve: contribution $\left(\frac{\Delta R_p}{R_p}\right)_\parallel^{[110]}$ calculated from $\Lambda_{[110]}(\omega)$. Dashed curve: contribution $\left(\frac{\Delta R_p}{R_p}\right)_\perp$ obtained from the difference between the two previous spectra. (b) Same as (a), after having rotated the sample by 90°, with incidence plane in the [$\bar{1}$10] direction.

the parallel case, while the 3 eV one is more intense in the perpendicular case. Figure 6 also gives the difference $\text{Im}\{\Lambda_{[\bar{1}10]}(\omega) - \Lambda_{[110]}(\omega)\}$, which is to be compared to the same quantity directly drawn from RAS on clean surfaces (figure 4). The two curves are very similar, as they should be, with the same intensity and the same shape. The remaining differences are probably due to the fact that

(i) the SDRS measurements compare the hydrogenated surface and the clean one and
(ii) the result in figure 6 comes from two independent measurements for two orientations of light, on a sample which has been refreshed from one measurement to the next one.

After having determined these quantities, it could be possible in principle to get the component perpendicular to the surface from the SDR spectra measured in p polarization and using formula (5). The p-SDR spectra for electric field parallel to $[\bar{1}10]$ and $[110]$ are drawn in figure 7. As for s polarization the intensity is larger when the electric field is normal to the dimers ($[110]$). Equation (5) can be rewritten, for electric field along $[110]$ (respectively $[\bar{1}10]$):

$$\left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)^{[110]} = 4\frac{\omega}{c}\cos\theta\left[\mathrm{Im}\left\{\frac{(\varepsilon_{\mathrm{b}}(\omega)-\sin^2\theta)\delta\Lambda_{[110]}(\omega)}{(\varepsilon_{\mathrm{b}}(\omega)-1)(\varepsilon_{\mathrm{b}}(\omega)\cos^2\theta-\sin^2\theta)}\right\}\right.$$

$$\left.+ \mathrm{Im}\left\{\frac{\varepsilon_{\mathrm{b}}(\omega)^2\sin^2\theta\delta\Lambda_{[001]}(\omega)}{(\varepsilon_{\mathrm{b}}(\omega)-1)(\varepsilon_{\mathrm{b}}(\omega)\cos^2\theta-\sin^2\theta)}\right\}\right] \equiv \left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)^{[110]}_{\parallel} + \left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)_{\perp} \quad (7)$$

where the first part is the contribution of the parallel-to-the-surface electric field component in the $[110]$ (respectively $[\bar{1}10]$) direction, while the second one is the contribution of the perpendicular-to-the-surface one. Using the values of $\Lambda_{[\bar{1}10]}(\omega)$ and $\Lambda_{[110]}(\omega)$ determined above, $\left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)^{[110]}_{\parallel}$ and $\left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)^{[\bar{1}10]}_{\parallel}$ have been calculated and are drawn in figures 7(a) and (b). It is noteworthy that they are almost identical to the SDR spectra measured in p polarization. Figure 7 also gives the difference between the previous spectra, i.e. the contribution of the component normal to the surface $\left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)_{\perp}$, obtained from the two sets of treatments. This contribution is found in one case to be equal to zero, in the other case to be close to zero and featureless. The small difference obtained from the two treatments is due to the experimental imprecisions: these spectra are indeed obtained by comparing a freshly prepared Si sample to the same surface obtained after saturation of hydrogen. It is known that the quality of the clean surface is not perfectly reproducible and, hence, the optical response of the clean surface varies slightly from one sample to another. The good agreement between the so-determined contributions from both sets of measurements show that the reproducibility in preparing our samples is rather satisfactory. These results show that, for the Si(001) surface, measurements in p polarization do not provide any additional information with respect to s-polarization measurements. Because of the respective coefficients of $\Lambda_{[\bar{1}10]}(\omega)$ and $\Lambda_{[110]}(\omega)$ in equations (5) and (7), we would actually expect a larger contribution due to the perpendicular component that due to the parallel one. This can be easily checked by calculating the p-SDRS with the same given function, either for the normal component $\Lambda_{[001]}(\omega)$ only or for the parallel one $\Lambda_{[110]}(\omega)$ only. This is shown in figure 8 where the same Lorentzian function centred at 2.5 eV with a width of 0.5 eV has been chosen for both contributions. The normal contribution is actually ten times larger than the parallel one. The question now is why the normal contribution to the p-SDRS in our experiments is almost zero, with respect to the parallel contribution. Is this a general result, which could be valid for other crystalline surfaces, or is this a specific result for the Si(100) surface? This question cannot be answered here and would require theoretical calculations. The only remark which can be made at this point is that the perpendicular optical response for a surface appears to behave as the inverse of a dielectric function [9, 10]. It can indeed be written, for a crystal occupying the half-space $z > 0$, as

$$\Lambda_z(\omega) = \int_{-\infty}^{\infty}\mathrm{d}z\int_{-\infty}^{\infty}\mathrm{d}z'\,[\varepsilon_{zz}^{-1}(z,z';\omega) - \delta(z-z')\varepsilon_0^{-1}(z;\omega)] \quad (8)$$

where $\varepsilon_{zz}(z,z';\omega)$ is the z-diagonal component of the non-local dielectric tensor of the system and $\varepsilon_0(z;\omega)$ is the local dielectric function of the system in the absence of surface effects ($\varepsilon_0(z;\omega) = 1$ for $z < 0$ and $\varepsilon_0(z;\omega) = \varepsilon_{\mathrm{b}}(\omega)$ for $z > 0$). The dielectric function of crystals being usually much larger than unity, this would suggest the small intensity of $\left(\frac{\Delta R_{\mathrm{p}}}{R_{\mathrm{p}}}\right)_{\perp}$ to be a general result.
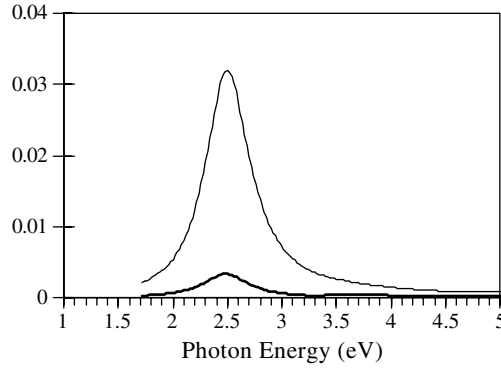
Figure 8. Normal contribution $\left(\frac{\Delta R_p}{R_p}\right)_\perp$ and parallel contribution $\left(\frac{\Delta R_p}{R_p}\right)_\parallel^{[110]}$ to the SDRS in p polarization, calculated from the same Lorentzian functions centred at 2.5 eV for $\Lambda_{[001]}(\omega)$ and $\Lambda_{[110]}(\omega)$.
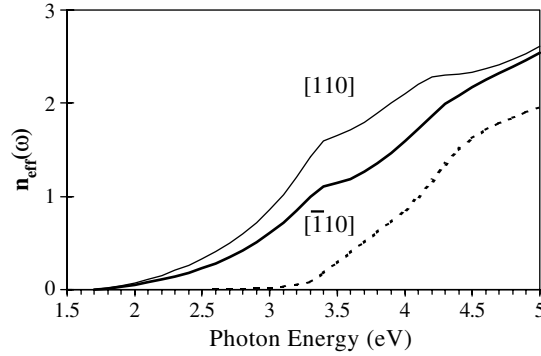

Figure 9. Effective number of electrons $n_{\mathrm{eff}}(\omega)$ calculated from $\Lambda_{[110]}(\omega)$ (thin curve) and $\Lambda_{[\bar{1}10]}(\omega)$ (thick curve), compared to $n_{\mathrm{eff}}(\omega)$ for bulk silicon (dashed curve).

Finally, the effective number of electrons per atom $n_{\mathrm{eff}}(\omega)$ involved in the optical transitions occurring below frequency ω [27, 28] can be obtained by applying the following sum rule to the previously determined quantities $\Lambda_{[\bar{1}10]}(\omega)$ and $\Lambda_{[110]}(\omega)$:

$$n_{\mathrm{eff}}(\omega) = \frac{m}{2\pi^2 e^2 N_s} \int_0^\omega \omega'[\mathrm{Im}\{\Lambda(\omega')\}]\,d\omega' \tag{9}$$

where N_s is the number of surface atoms. The result for the two orientations is drawn in figure 9. The integrals have been calculated using the spectral range available from the SDRS experiment, i.e. from 1.7 eV to ω, so the contribution of states below 1.7 eV is not taken into account. It is compared to the equivalent function determined for bulk silicon. In contrast to the case of other surfaces, such as the cleavage surfaces Si(111) 2×1 or Ge(111) 2×1 [1, 5], which are highly anisotropic, there is no important difference between the two orientations. In both cases, $n_{\mathrm{eff}}(\omega)$ increases to reach a plateau at a value of 1.1 or 1.6 around the energy of 3.5 eV. These plateaus correspond to the first maximum observed below 3.5 eV in $\Lambda_{[\bar{1}10]}(\omega)$ and $\Lambda_{[110]}(\omega)$, related mainly to transitions involving states of the dangling bonds of the Si dimers [25]. The values obtained for $n_{\mathrm{eff}}(3.5)$, slightly larger than unity, are in good agreement with the fact that one electron is involved in the dangling bond of each dimer atom, and suggest that valence states corresponding to the covalent bonds internal to the dimers also participate

partly in the optical response of the surface below 3.5 eV. Finally, the $n_{\text{eff}}(\omega)$ functions reach the same value around 5 eV: as could be expected, the same number of electrons per atom is involved in the interband transitions, whatever the orientation of the electric field is.

4. Summary and conclusion

We have investigated the optical response of the Si(001) 2 × 1 surface by two complementary optical techniques, RAS and SDRS. We could determine the optical response tensor of the surface. The components parallel to the surface are dominated by two peaks, due to optical transitions related to dimer states. They are slightly different when the electric field is along the dimers or normal to them. The component normal to the surface is found to be much smaller than the parallel ones, which results in the fact that p measurements give the same information as s measurements. The question of whether this result is specific to the Si(001) surface or can be generalized to other surfaces is open and would require a complete theoretical investigation.

References

[1] Chiaradia P and Chiarotti G 1995 *Photonic Probes of Surfaces* ed P Halevi (Amsterdam: Elsevier Science) chapter 3
[2] Borensztein Y 2000 *Surf. Rev. Lett.* **7** 399
[3] Aspnes D E and Studna A A 1985 *Phys. Rev. Lett.* **54** 1956
[4] Yasuda T, Mantese L, Rossow U and Aspnes D E 1995 *Phys. Rev. Lett.* **74** 3431
[5] Chiaradia P, Cricenti A, Selci S and Chiarotti G 1984 *Phys. Rev. Lett.* **52** 1145
[6] Noguez C, Beitia C, Preyss W, Shkrebtii A I, Roy M, Borensztein Y and Del Sole R 1996 *Phys. Rev. Lett.* **76** 49
      Beitia C, Preyss W, Del Sole R and Borensztein Y 1997 *Phys. Rev.* B **56** R4371
[7] Shkrebtii A I, Esser N, Richter W, Schmidt W G, Bechstedt F, Fimland B O, Kley A and Del Sole R 1998 *Phys. Rev. Lett.* **81** 721
[8] Mazine V and Borensztein Y 2002 *Phys. Rev. Lett.* **88** 147403
[9] Bagchi A, Barrera R G and Rajagopal A K 1979 *Phys. Rev.* B **20** 4824
[10] Del Sole R 1981 *Solid State Commun.* **37** 537
      Del Sole R 1995 *Photonic Probes of Surfaces* ed P Halevi (Amsterdam: Elsevier Science) chapter 4
[11] Shkrebtii A I and Del Sole R 1993 *Phys. Rev. Lett.* **70** 2645
[12] Palummo M, Onida G, Del Sole R and Mendoza B S 1999 *Phys. Rev.* B **60** 2522
[13] Schmidt W G, Glutsch S, Hahn P H and Bechstedt F 2003 *Phys. Rev.* B **67** 085307
[14] Hamers R J, Tromp R M and Demuth J E 1986 *Phys. Rev.* B **34** 5343
[15] Van Dijken S, Zandvliet H J W and Poelsema B 1998 *Surf. Rev. Lett.* **5** 15
[16] Wolkow R A 1992 *Phys. Rev. Lett.* **68** 2636
[17] Boland J J 1992 *Surf. Sci.* **261** 17
[18] Aspnes D E, Harbison J P, Studna A A and Florez L T 1988 *J. Vac. Sci. Technol.* A **6** 1327
[19] Borensztein Y, Lopez Rios T and Vuye G 1989 *Appl. Surf. Sci.* **41/42** 439
[20] Shioda R and van der Weide J 1998 *Phys. Rev.* B **57** R6823
[21] Jaloviar S G, Lin J-L, Liu F, Zielastek V, McCaughan L and Lagally M G 1999 *Phys. Rev. Lett.* **82** 791
[22] Hingerl K, Balderas-Navarro R E, Bonanni A, Tichopadek P and Schmidt W G 2001 *Appl. Surf. Sci.* **175/176** 769
[23] Schmidt W G, Bechstedt F and Bernholc J 2001 *Phys. Rev.* B **63** 045322
[24] Witkowski N, Pluchery O and Borensztein Y 2003 unpublished
[25] Borensztein Y, Witkowski N and Pluchery O 2004 unpublished
[26] Kress C, Shkrebtii A and Del Sole R 1997 *Surf. Sci.* **377** 398
[27] Wooten F 1972 *Optical Properties of Solids* (New York: Academic)
[28] Chiarotti G, Chiaradia P, Faiella E and Goletti C 2000 *Surf. Sci.* **453** 112