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Optical second-harmonic generation from GaAs(100) surfaces: the influence of H_2

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Abstract. The interaction of H_2 with GaAs(100) surfaces has been monitored using optical second-harmonic generation (SHG) excited using an Nd:YAG laser operating on the sub-bandgap fundamental line at 1064 nm. The SHG response has been recorded both before and after treatment of the substrate in H_2 to remove contamination and under a high overpressure of H_2 (10 mbar). It is found that the polar SHG response from a GaAs(100) surface contaminated with carbon and oxygen is essentially structureless apart from the appearance of a peak in SHG intensity when the incident electric vector is aligned with a ⟨110⟩ azimuth. After cleaning in H_2, the polar SHG intensity response is both more intense and more structured, showing maxima corresponding to the situation in which the incident electric vector is aligned with both the ⟨110⟩ and ⟨−110⟩ azimuths. No further changes are observed in the polar SHG response when measurements are made with the GaAs(100) surface under 10 mbar H_2. While these data are preliminary and may not be directly interpreted in terms of a particular two-dimensional surface overlayer, they illustrate the high surface SHG response obtainable from GaAs(100), which is discussed in terms of the presence of a highly non-linear surface layer.

1. Introduction

In our laboratories we are concerned with the study of the mechanisms of compound semiconductor growth from vapour phase precursors, i.e. processes such as metallorganic phase epitaxy (MOVPE), which typically involve the use of pressures of up to 1000 mbar and temperatures of up to 1000 K. Clearly, the *in-situ* study of either surface or gas-phase MOVPE processes is not trivial, but recently there has been much work on the development of optical techniques which may bridge the pressure gap that exists between real semiconductor growth conditions and the environment required for the operation of conventional surface science techniques [1]. One such technique which may provide surface-specific information and be unhampered by the use of relatively high pressures is optical second harmonic generation (SHG) [2, 3]. While dynamic changes in SHG intensity can be used to monitor adsorption and desorption processes, McGilp has shown that studies of the polarization anisotropy in the SHG response from a surface can also reveal structural information [3, 4].

In this present paper we report the results of a study of the anisotropic response of the SHG intensity from a GaAs(100) in response to the structural changes that occur upon removal of surface contamination by reduction under H_2 and the subsequent

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influence of a high overpressure of H_2 on the surface structure. While these data are preliminary and require much further processing, they provide the first indications of the structure of a GaAs(100) surface recorded under high overpressure conditions, where techniques such as LEED and RHEED may not operate.

2. Experimental details

The experimental system employed in this work has been described in detail elsewhere [5]. Briefly, SHG was produced from a GaAs(100) substrate mounted in a UHV chamber using the fundamental line of an Nd:YAG laser (1064 nm), operating at 20 Hz repetition rate, with a pulse duration of 10 ns and average pulse surface energy density of approximately 5 mJ cm^{-2}. The laser impinged on the GaAs(100) surface at an angle of 45° to the surface normal. The laser pulse energy was normalized using a photodiode arrangement and after optical filtering, the second harmonic signal at 532 nm was measured using a photomultiplier tube connected to a gated boxcar integrator. Polarization anisotropy measurements were made by manually rotating a polarizer. No polarization analyser was used in the output stage of the experiments.

3. Results and discussion

Figure 1 depicts typical polarization anisotropy plots recorded from a GaAs(100) surface, where the zero on the angle of polarization axis corresponds to the configuration in which the polarization of the electric vector of the incident radiation was aligned with a $\langle 110 \rangle$ surface azimuth. The data have been corrected for the variation in amplitude of the electric vector of the incident radiation as a function of angle of rotation while most of the asymmetry in the plot along the $\langle 110 \rangle$ direction in fact arises from an experimental artifact. Despite these factors and the presence of possibly several layers of carbon and oxides of Ga and As, it is clear from this figure that prior to surface cleaning, the surface possesses some considerable rotational anisotropy in χ^{2s}, the second-order surface susceptibility, which is the parameter that determines the efficiency of SHG. Figure 1 depicts analogous data recorded from the substrate in UHV, after heating to approximately 800 K in 1.5 mbar H_2 for some 10 min. Auger data taken before and after this treatment revealed that as we have reported previously [6], a clean surface is generated which is severely depleted of arsenic. From figure 1 it can be seen that the overall SHG response has been dramatically increased (factor of 3) by this cleaning procedure, while additional structure has also appeared in the form of two new lobes have aligned with the orthogonal axis. Finally the figure reveals that no further changes occur to the surface under a constant hydrogen overpressure of 10 mbar.

The pattern of SHG intensity for the clean surfaces depicted in figure 1 intuitively describes the four-fold symmetry that would be expected for unreconstructed GaAs(100) surfaces, although great care must be taken in interpreting such data since while the total SHG intensity can be measured, the relative contributions from in-plane and out-of-plane components of the non-linear surface susceptibility cannot be easily separated. Thus it may be that the pattern depicted in figure 1 may be entirely fortuitous. Experiments are now under way in our laboratories which utilize a polarizer on the output side of the experiment which will enable us to separate out the various contributions and make a reliable measurement of the effective surface symmetry.

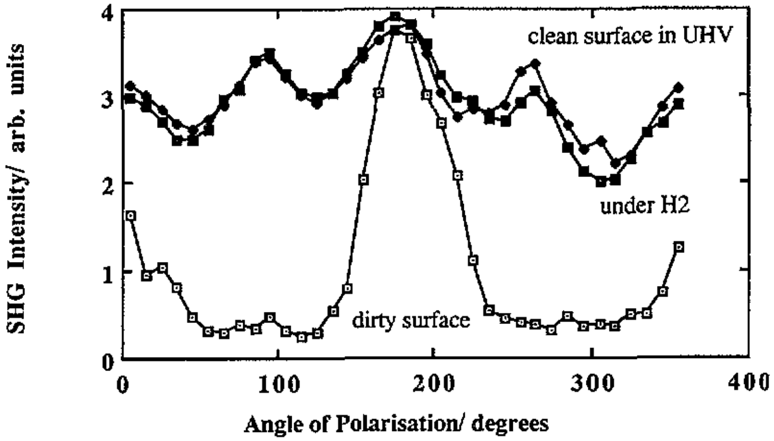

Figure 1. Variations in SHG intensity at 532 nm as a function of angle of polarization, recorded from a GaAs(100) surface under various conditions as described in the text. The excitation source was an Nd:YAG laser employed at near normal incidence. The laser was operated at 1064 nm, with a pulse duration of 10 ns, repetition rate of 20 Hz, and a surface power density of less than 5 mJ cm^{-2}.

It is interesting to consider the apparent ease with which a surface chemical change has been observed via the SHG response from a substrate which is expected to generate an intense bulk SHG response due to the lack of inversion symmetry in the bulk unit cell together with the observation that the SHG response increased dramatically as surface contamination was removed. Considering the first of these observations, we note that in a previous publication we reported the observation of an intense surface SHG response from a GaAs(100) surface, which allowed us to monitor a surface adsorption process [5]. Stehlin *et al* have demonstrated for the Sn/GaAs(100) system, that via the correct choice of input and output polarizations and angle of incidence it is possible to discriminate against the bulk SHG response and observe only the surface SHG response [7]. However, the conditions employed in these present experiments do not meet these requirements. Thus it is likely that some alternative explanation exists as to why the signals observed in our work are apparently surface-specific. We propose here that the most likely reason for the high degree of surface specificity observed in our experiments lies in the nature of the optical response of the polar GaAs surface, which results in the presence of a highly non-linear surface overlayer. As yet there is little evidence aside from relative SHG intensities to support this proposal but we are aware that theoretical calculations by Cini and coworkers also lend support to the proposal that polar surfaces such as GaAs(100) will give rise to an enhanced SHG response as compared to less polar surfaces such as Si(111) [8]. This same argument may also be applied to the observation regarding the increase in SHG intensity for the clean surface where removal of an essentially amorphous deposit with little in-plane contributions to the SHG response generates a surface with somewhat higher anisotropy in the in-plane components of the susceptibility tensor. We note that a similar enhancement of surface SHG response has been reported by Hollering in a comparison of data for clean and oxide covered Si(100) surfaces [9].

4. Conclusions

We have demonstated for the first time that the technique of SHG may be used to study structural changes occuring at a GaAs(100) surface following chemical treatment. We have also made measurements under relatively high overpressure conditions and while it is somewhat disappointing that no further changes in surface structure were observed, the data serve to illustrate the potential of the SHG technique for obtaining surface structural information under high-pressure conditions, where electron diffraction techniques may not operate.

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