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The dynamic interaction of triethyl gallium with GaAs(100) surfaces monitored using optical second-harmonic generation

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Abstract. We report the results of a preliminary investigation into the use of optical second-harmonic generation (SHG) as a probe of surface processes involved in semiconductor growth. It is demonstrated that a predominantly surface SH signal may be generated from GaAs(100) substrates. The dynamic interaction of triethyl gallium, a typical reactant in MOVPE growth of GaAs, with an As-rich GaAs(100) surface at 300 K has been monitored using SHG. It is shown that this interaction gives rise to a permanent increase in the SH intensity.

1. Introduction

Within the dipole approximation, SHG is forbidden from the bulk of any material possessing a centrosymmetric structure [1–3]. Consequently in such materials any SH signal produced must originate solely from the surface region where the symmetry is broken and the SHG process becomes allowed. GaAs, on the other hand, has a bulk structure that is *centro-asymmetric* and therefore an SH signal would be expected from the bulk, which may be several orders of magnitude greater than the surface contribution [4]. It has recently been suggested that this problem can be overcome by polarisation discrimination of the bulk signal which may restore the surface sensitivity of the SHG experiment on materials with a centro-asymmetric structure [3, 5]. This approach has been exploited in [5] in order to monitor the adsorption of Sn on GaAs(100). Alternatively, in [6] it was shown that for Si(111) surfaces, predominantly surface SHG is obtained if sub-band-gap primary radiation is employed, implying that surface state resonances may dramatically influence SHG. Here we demonstrate that surface SHG may be obtained from GaAs(100) under conditions where either or both mechanisms may apply, reporting only the second observation of surface-specific SHG from GaAs (100).

2. Experimental details

The experiments were carried out in a UHV system described in detail elsewhere [7]. The samples used were GaAs(100), Si-doped 1×10^{17} cm^{-3} (MCP) and the triethyl gallium (TEG) was electronics grade (Epichem). The substrates were prepared so as to be clean and As-rich as described in [7, 8].

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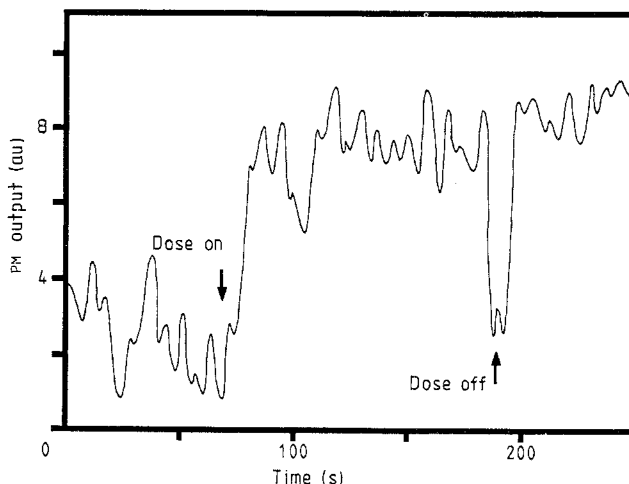

Figure 1. The change in the SHG intensity from a GaAs(100) surface during a dynamic exposure to TEG at 1×10^{-7} mbar pressure, at 300 K. There is a clear rise in the SHG intensity which persists after the TEG supply has been turned off, showing the surface to have undergone an irreversible change due to adsorption of TEG. The sudden fall and rise in SHG as the TEG dose is stopped is due to an experimental artefact only.

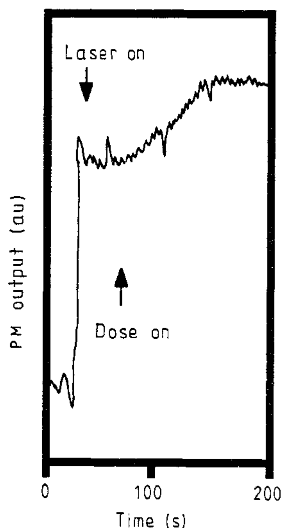

Figure 2. The change in the SHG intensity from a GaAs(100) surface during a dynamic exposure to TEG at 5×10^{-9} mbar pressure, at 300 K.

The 1064 nm line of an Nd:YAG laser (Quantel) was incident on the GaAs(100) substrate at an angle of 45° to the surface normal. The reflected IR light plus the frequency-doubled component were then separated using a dichroic mirror. The light was then passed through a suitable filter into the photomultiplier tube.

3. Results and discussion

The data obtained during the dynamic exposure of the As-rich GaAs(100) surface to TEG at 300 k at two different TEG pressures are shown in figures 1 and 2. In both cases,

an increase in SH intensity was observed with increasing TEG exposure. The net increase in the SHG intensity upon exposure to TEG at 300 K cannot be adequately accounted for at this stage; however, we note that similar behaviour was observed following adsorption of Sn on GaAs(100) [5]. By analogy with experiments performed on metal surfaces, an increase in SHG intensity would signify an increase in the second-order surface susceptability, i.e. a decrease in the work function [9]. Although we have previously shown that TEG is adsorbed on GaAs(100) at 300 K [10], we are uncertain at this time whether the adsorption is molecular or dissociative although, recently. McCaulley and co-workers have indicated, from desorption experiments, that adsorption on a Ga-rich surface is dissociative at room temperature [11], which would be consistent with the presence of an overlayer with highly metallic character.

Several aspects of this work need to be investigated further—in particular, the mechanism by which the SHG is localised to the substrate surface. However, it has been shown that SHG offers the possibility of monitoring, in real time, the adsorption of a typical MOVPE precursor at the surface of a compound semiconductor. It also suggests that SHG may become a useful *in situ* diagnostic technique for use for the harsh environment of the MOVPE-growth reactor.

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