

SURFACE SCIENCE LETTERS

COMMENT ON ENHANCED RAMAN SCATTERING FROM ADSORBATES ON SEMICONDUCTOR SURFACES

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A recently published model of surface enhanced Raman scattering from adsorbed molecules on semiconductor surfaces is compared to our experimental results for amorphous carbon on PbTe. The model fails to account for the major features of the observed scattering.

In a recent letter [1] Ueba calculated the Raman polarizability of a molecule adsorbed on a semiconductor surface considering coupled molecule–semiconductor electronic excitations. As this work was motivated by our observation of enhanced Raman scattering from adsorbed amorphous carbon on PbTe [2] we feel compelled to comment on the applicability of his theory to our measurements.

An essential point in our paper was the fact that the enhanced scattering from the carbon overlayer appeared only with PbTe surfaces exhibiting an irregular array of triangular pits. For PbTe displaying no such surface roughness, the enhancement (if any) was smaller by a factor of ~ 100 . We have suggested that the enhanced scattering stems from roughness leading to a non-vanishing Raman matrix for the process involving the Coulomb interaction between the molecule dipole-moment and the electrons and holes confined in the substrate. Irrespective of the validity of this assumption, it is clear that the enhancement observed is much larger for rough surfaces. Ueba's mechanism, as he points out, does not involve roughness. Consequently, it cannot play a major role in the effects observed.

We also note that the Raman excitation profiles predicted by Ueba fail to account for the frequency dependence of the enhancement which we reported qualitatively. In all cases which he examines, the Raman enhancement factor peaks at energies below 2 eV, giving a greater enhancement in the red (~ 1.9 eV) than in the blue-green region of the spectrum (~ 2.5 eV). In contrast, our

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results show the intensity of the carbon-related band to be approximately 200 times larger in the blue-green region. In Ueba's work, the occurrence of a peak below the 2 eV-onset of interband transitions in PbTe results from considering coupling to a molecular transition at a higher energy, i.e., 3 eV. Instead, it appears that a transition of the molecule *below* 2eV would be needed to obtain better agreement with our data.

In summary, we feel that the model of enhanced Raman scattering proposed by Ueba fails to account for the major features of our experiments. There are, undoubtedly, many mechanisms which can lead to an enhancement; it is unlikely that any single process will explain all of the experimental observations. We believe that Ueba's mechanism can, at best, provide only a small contribution to the reported scattering.

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References

- [1] H. Ueba, *Surface Sci.* 133 (1983) L432.
- [2] J.E. Potts, R. Merlin and D.L. Partin, *Phys. Rev.* B27 (1983) 3905.