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OPTICAL SECOND-HARMONIC GENERATION FOR SURFACE STUDIES

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ABSTRACT

The development of optical second-harmonic generation as a tool for surface studies will be reviewed. Recent progress will be discussed.
OPTICAL SECOND-HARMONIC GENERATION FOR SURFACE STUDIES

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SUMMARY

Recently, it has been demonstrated that optical second-harmonic generation (SHG) is highly surface-specific, and can be used as a viable tool for surface studies. We shall review here the recent progress in the development of this surface probing technique. Specifically, we shall discuss the following problems.

First, we consider the application of SHG to the study of surface enhanced optical effects. The problem that Raman scattering from molecules adsorbed on rough surfaces of noble metals can be enhanced by many orders of magnitude has recently attracted a great deal of attention. It is believed that most of the enhancement comes from the local-field effect. The measurements, however, are generally limited to noble metals. We find that SHG should exhibit nearly the same local-field enhancement as Raman scattering, but it has much better sensitivity in the enhancement measurement and the enhancement factor of a rough surface relative to a smooth surface is independent of the adsorbed molecules. Therefore, we have used it to measure the local-field enhancement on roughened surfaces of 14 metals and semiconductors, with reference to Ag. It is found
that a few metals, other than the noble metals, show an enhancement comparable to Ag. They are clearly candidates for surface enhanced Raman studies. The experimental results are in fair agreement with a simple model calculation.

We consider the application of SHG to the better characterized surfaces. We find that SHG is often sensitive to submonolayers of molecular adsorbates at interfaces between any two media with inversion symmetry, and can be used to measure surface coverage, orientation, and arrangement of the adsorbates. On clear metal surfaces in ultrahigh vacuum, even time-resolved adsorption and desorption of atomic and diatomic species with less than 10% monolayer coverage can be monitored. The modified surface nonlinearity resulting from adsorption arises from different mechanisms in different cases. For large adsorbed molecules, it is usually dominated by the nonlinearity of the surface aligned molecules. For smaller molecules or atoms, it can be due to the newly established bonds between the adsorbates and the substrate, and/or due to modification of the surface properties of the substrate. Microscopic theories for calculating surface nonlinearities are desperately needed. In general, SHG from a surface has contributions from both the bulk and the surface. Using different polarization combinations and different crystalline surfaces, the two contributions can be separated.
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