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Surface Sum Frequency Generation of III-V Semiconductors[1]
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— Optical sum frequency generation (SFG) is a well-established technique for surface and interface studies but its use has been limited mainly to centrosymmetric materials so far. Here, we demonstrate that femtosecond broadband SFG spectroscopy has the ability to identify surface molecular vibrations on the archetypical non-centrosymmetric semiconductor GaAs (001), in which the bulk SFG signal typically dominates over surface SFG contributions. Azimuthal angle dependence of the second order SFG nonlinear response from GaAs (001) surface in the reflection geometry in vacuum for all eight polarization combinations are detected and analyzed. The results agree with and extend upon previous second harmonic generation (SHG) studies and phenomenological analysis. In addition, carbon monoxide and methanol are employed as molecular-markers on the GaAs (001) surfaces. The C-O stretching mode of carbon monoxide and the methyl group stretching modes of methanol are clearly observed even though the bulk contribution dominates the SFG signal. Coherent heterodyne interference is proposed as the mechanism for the surface signal enhancement. Two other zinc blende type III-V semiconductors, GaP and GaSb, are also studied and compared.

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