Calculation of linear and nonlinear optical susceptibilities in wide gap semiconductors

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CALCULATION OF LINEAR AND NONLINEAR OPTICAL SUSCEPTIBILITIES IN WIDE GAP SEMICONDUCTORS

DISSERTATION

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by

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ABSTRACT

An *ab initio* method is used to study the linear and the second-order nonlinear optical susceptibilities in wide band gap semiconductors. First, the density functional theory, implemented with the local-density approximation, is used to obtain the ground-state electronic structures of the materials. Next, time-dependent perturbation is applied to study the response of the electrons to an electromagnetic field. Over the past few years, Levine *et al.* developed a theory which reproduces the experimental values within 15% of the linear and second-harmonic susceptibilities for over ten semiconductors and insulators. The current study formally completes Levine's theory on the second-harmonic susceptibilities by including the corrections from a nonlinear local field which is proportional to the second power of the external electric field. The nonlinear-local-field corrections is shown to give a 50% increase to the linear-local-field corrections previously studied.

The calculations are extended to a series of wide gap semiconductors including four polytypes of SiC (2H, 4H, 6H, and 3C), BN, AlN, and GaN. A "scissors-operator" approach is used to shift the small band gaps given by the local density approximation to the experimental values. In the previous studies of small to medium gap semiconductors, using the scissors operators gives good agreement with experiments. The current study finds that, in the wide gap semiconductors, using the scissors operators result in about 10-20% underestimation of the linear

and second-harmonic susceptibilities.

The second-harmonic susceptibilities can vary by over 100% in different structures of a compound with identical chemical composition. Using this large structural dependence, one may be able to grow materials in a designed pattern to enhance their nonlinear optical properties.

Finally, the newly proposed method of using the "$2n+1$" theory to calculate the second-harmonic susceptibilities is demonstrated to be formally equivalent to our method. A scheme is suggested to incorporate the scissors operators into this new method even though the expressions do not have any explicit dependence on the conduction bands.

To My Parents

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