Optical nonlinear processes in semiconductors in the presence of a DC field: an ab-initio description

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A deep understanding of the nonlinear optical properties of solids is crucial for the improvement of nonlinear devices and provides an opportunity to search for new materials. Among all the non-linear phenomena existing in nature, an important role is played by the electro-optic effect. The electro-optic effect produces a change of the refractive index in a medium using a DC electric field and has attracted particular interest for the development of optoelectronic devices. In the linear electro-optic effect (LEO) or Pockels effect, the change is proportional to the applied electric field. It may be seen as a second-order polarization and then described by a second-order susceptibility, which is known to be zero in the dipole approximation for centro-symmetric materials. Therefore a peculiarity of the LEO effect comes from the fact that it only occurs in materials without inversion symmetry or originates from symmetry-breaking regions.

From the theoretical point of view, most of the calculations for second-order susceptibilities have been done in the framework of Second Harmonic Generation (SHG), [1]. In that case, the frequency of the incoming field is considered as high with respect to vibrational frequencies and the lattice is kept static. Therefore, one has to evaluate only the electronic contribution, obtained directly from the optical susceptibility, coming from the interaction of the valence electrons and the electric fields. The knowledge of the electro-optic tensor implies in principle the evaluation of two additional contributions, the ionic and piezoelectric parts. The ionic contribution is linked to the ionic displacements and depends on the variation of the dielectric tensor induced by these displacements. The piezoelectric contribution comes from the possible modification of the shape of the unit cell due to the electric forces, [2]. Following the same idea, a similar phenomenon happens also for SHG, corresponding to a second harmonic response in the presence of a dc-field, called "Electric Field Induced Second Harmonic" (EFISH) and described in terms of a third-order polarization.

I will show our results for the second order susceptibility describing the LEO tensor, within the ab initio framework of time-dependent density-functional theory. I will present our analytic derivation of the macroscopic polarization up to second order in terms of the electric fields, including the effect of a scissors operator to account for the quasi-particle effect. Excitonic effects will be included on the basis of a simple approach, [3]. The ionic contribution will be evaluated within the same framework and included explicitly. These results will be compared with experimental data. Furthermore, while the ionic part is often evaluated through the Faust-Henry coefficient [4], the validity of this approximation will also be discussed. Finally, preliminary results will be presented for the EFISH process.

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