4 Optical nonlinear effects in semiconductors

C. N. IRONSIDE

4.1 Introduction

It is hard to exaggerate the technological importance of semiconductors; as a group of materials they have found extensive application in electronics, where the key feature is the ability to alter radically their electronic properties by employing small amounts of dopants. Their optical properties have also attracted considerable interest, and semiconductors have been shown to be very useful in opto-electronics, where they are widely employed in the detection and generation of light. As a consequence their linear optical properties have been extensively studied, and since the invention of the laser in 1960 their nonlinear optical properties have also attracted a great deal of interest. In particular, the large magnitude of the nonlinear optical properties at photon energies close to the band-gap energy has generated considerable attention; with the demonstration of optical bistability, all-optical switching and optical computing are causing excitement.

The semiconductors which are extensively employed in electronic and opto-electronic applications are covalent inorganic solids. They are composed of elements in groups II to VI of the periodic table combined as II–VI, III–V and IV. In this chapter we will focus on the III–V inorganic semiconductors which are used in many opto-electronic applications, because they have a mature growth and process technology that makes them particularly attractive for nonlinear optical studies.

Third-order optical nonlinearities arise in semiconductors in some profusion. The third-order effects give rise to an intensity-dependent refractive index and absorption coefficient. These effects are divided into two broad classes called resonant and nonresonant optical nonlinearities (Bloembergen, 1965). Resonant effects deal with photon energies close to the fundamental absorption edge in semiconductors and are caused by photogenerated carriers, whereas nonresonant effects are at photon energies well below the fundamental absorption edge. With resonant optical nonlinearities, real photogenerated carriers are produced which give rise to the optical nonlinearities. However, the optical nonlinearity associated with the production of real carriers does not fit neatly into the theory outlined in chapter 2. In particular, the theory is not usually cast in a suitable way for dealing with free mobile charge and the resultant nonlocal spatial effects and time-dependent effects.
When considering resonant nonlinearities, in some ways it is more appropriate to take a broad definition of nonlinear optical effects and to regard them generally as intensity-dependent optical properties. Perhaps the most significant aspect of resonant optical nonlinearities is the insight that is provided into the dynamics of free carriers in semiconductors.

Third-order nonresonant effects are more easily described by the usual nonlinear theory outlined in chapter 2 and more directly relate to $\chi^{(3)}$. Recently, (Sheik-Bahae et al., 1990 a,b, 1991) there has been a breakthrough in the theory of nonresonant third-order nonlinear optical effects in semiconductors. It is now apparent that two-photon absorption in semiconductors is related to the imaginary part of $\chi^{(3)}$ (intensity-dependent absorption) and that from previous work (Pidgeon et al., 1979) on the dispersion of the two-photon absorption, the dispersion of the real part of $\chi^{(3)}$ (intensity-dependent refractive index) can be calculated via a Kramers–Kronig transformation. This work and its implication for all-optical switching are discussed in this chapter.

The zinc blende (or sphalerite) tetrahedrally coordinated structure of the III–V semiconductor is illustrated in Figure 4.1, where it is compared with the diamond-like structure of group V semiconductors. The crystal structure is cubic with a 43m point group symmetry. Group III–V and II–VI semiconductors lack a centre of symmetry and therefore, as explained in chapter 2, they have a second-order nonlinearity. From the symmetry of the point group it can be determined that they only have a $\chi_{14}$ element of the second-order nonlinearity tensor. The usefulness of second-order nonlinearities in semiconductors is restricted by their limited range of transparency and by lack of sufficient birefringence for phase matching. However, with semiconductors, an extensive range of material technologies is available that can provide some solutions, and second-order nonlinear optics of semiconductors is still an active area of research.

The development of semiconductors for electronic and opto-electronic applications has resulted in a very sophisticated growth technology, where

Figure 4.1 Tetrahedral crystal structure of group IV and III–V semiconductors: (a) crystal structure of diamond, typical of Si, Ge; (b) crystal structure of zinc blende, typical of GaAs, InSb.