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LETTER TO THE EDITOR

Interband Bloch–Siegert shift in p-type GaAs and InP at terahertz frequencies

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Abstract. The shift of resonance energy in direct optical interband transitions is evaluated. The case of hole transitions between heavy and light mass valence bands is considered, using the full form of the Luttinger–Kohn Hamiltonian. In addition to anisotropy, very large Bloch–Siegert shift, of the order of 100%, was found at high IR light intensities in the terahertz frequency range. The shift is related to a periodic hole motion in the Brillouin zone.

1. Introduction

In semiconductors and dielectrics the optical interband transitions usually occur in those points of the Brillouin zone where photon energy $\hbar\omega$ matches vertical separation between two resonant bands; i.e., when condition $\hbar\omega = \varepsilon_i(\mathbf{k}) - \varepsilon_j(\mathbf{k})$ is satisfied, where $\varepsilon_i(\mathbf{k})$ and $\varepsilon_j(\mathbf{k})$ are the energies of the i th and j th bands at electronic wave vector \mathbf{k} and \hbar is the Planck constant. At low light intensities such a resonance approximation is fully satisfactory. However, at high light intensities the resonance may occur at a different photon energy. This is a well known effect in atomic physics, where the shift in resonance energy is known as the Bloch–Siegert (BS) shift [1–3]. At not too high intensities the shift is proportional to light intensity and the square of the dipole moment between the considered atomic energy levels. Higher order perturbation theory is required in finding the shift analytically [2, 3].

In this paper the BS shift is evaluated for the case of semiconductors, where optical transitions are between energy bands rather than discrete energy levels. As an example, intervalence hole transitions between heavy and light mass valence bands in p-type semiconductors are considered. Because of warping and nonparabolicity of the valence band, even in the lowest order perturbation treatment the transition matrix elements are anisotropic and have rather complicated structure [4, 5]. For this reason a numerical method was used in the present paper to evaluate the BS shift. In the first part of this Letter the method of calculation is briefly described and in the second part the results for p-GaAs and p-InP are presented.

2. Method of calculation

The intervalence population dynamics were investigated with the following Schrödinger equation

$$i\hbar \frac{\partial |\psi\rangle}{\partial t} = \left[H_0(\mathbf{k}) + e \frac{\mathbf{F}(t)}{i} \frac{\partial}{\partial \mathbf{k}} \right] |\psi\rangle \quad (1)$$

which describes the evolution of the six-component state vector $|\psi\rangle$. In (1) H_0 is the Luttinger–Kohn Hamiltonian [6], which is a 6×6 complex matrix whose elements are expressed through hole wave vector components, $\mathbf{k} = \{k_x, k_y, k_z\}$, measured with respect to the crystallographic axis. $\mathbf{F}(t)$ is the time-dependent electric field of radiation, e is the elementary charge and $i = \sqrt{-1}$. The quantization of electromagnetic field was neglected. Semiclassical calculations are correct to order n^2 , where n is the average number of electromagnetic field quanta [7]. Even for moderate electromagnetic field densities n is larger than 100 in a volume of micrometer dimensions.

The partial derivatives in equation (1) may be reduced to the total derivative if one notes that the equation of motion for the wave vector

$$\hbar \frac{d\mathbf{k}}{dt} = e\mathbf{F}(t) \quad (2)$$

at the same time is a characteristic equation of (1) [8]. As a result, equation (1) can be transformed to the following form

$$i\hbar \frac{d|\psi\rangle}{dt} = H_0(\mathbf{k})|\psi\rangle. \quad (3)$$

Equations (2) and (3) were solved numerically, assuming that electric field is parallel to the $\langle 100 \rangle$ crystallographic axis and changes harmonically with time, $\mathbf{F}(t) = \mathbf{F}_1 \cos \omega t$, where ω is the cyclic frequency. The state vector $|\psi\rangle$, in general, is a linear combination of doubly degenerate heavy (h), light (l) and spin-orbit split-off (s) band wave functions. Normally one would like to start with a given hole wave vector \mathbf{k} and band index, and then observe the time dependence of probability for a hole to be in the h -, l - or s - band. To obtain the hole population in a particular band from $|\psi\rangle$, one must know the unitary transformation matrix T that connects H_0 with the band-diagonal valence band Hamiltonian H_d :

$$H_d = T H_0 T^\dagger \quad (4)$$

where T^\dagger is Hermitian and at the same time the inverse of T . In our case H_0 and H_d are time dependent; therefore, T must depend on time too. At this point it should be noticed that the transformation (4) also is the complex singular value decomposition, a procedure that is well-known in numerical mathematics for finding an orthonormal basis [9]. Both H_0 and H_d depend on the same wave vector; therefore, if one knows \mathbf{k} at some moment t , one can immediately find H_0 . With the help of the singular value decomposition the matrix T can then be found and, if needed, H_d . The population in the respective bands can be calculated from the state vector

$$|f(t)\rangle = T(t)|\psi(t)\rangle. \quad (5)$$

The j th component of $|f\rangle$ is the eigenvector of H_d

$$H_d(\mathbf{k})|f(\mathbf{k})\rangle_j = \varepsilon_j(\mathbf{k})|f(\mathbf{k})\rangle_j \quad (6)$$

where $\varepsilon_j(\mathbf{k})$ is the dispersion law of the j th band. If $|\psi\rangle$ is normalized to unity, then $|f\rangle$ is also normalized: $\langle f|f\rangle = \sum_j |f_j|^2 = 1$, where summation is over all valence bands, including the degeneracy factor.

Equations (2) and (3) were solved numerically as a single set of nine complex differential equations. The transformation (5) was used to calculate the initial state vector $|\psi(0)\rangle$ from $|f(0)\rangle$. Throughout the calculations we have assumed that $|f(0)\rangle = \{0, 0, 0, 0, 0, (1+i)/2\}$, which corresponds to a hole initially being in one of the degenerate h -bands. The evolution of the hole population remained unchanged if other degenerate bands or a linear combination of degenerate h -band wave functions were used as a starting point. The transformation (4) was also employed to find the evolution of the population in the l -band from state vector $|\psi(t)\rangle$ at some convenient intermediate communication moments.

3. Bloch–Siegert shift in p-GaAs and p-InP

The above described procedure was applied to find hole transition dynamics between valence bands. Because near the degeneracy point at $k = 0$ the photon energy required for the transition is small, one expects that near the degeneracy point the BS shift will be large. Valence band nonparabolicity and warping were taken into account through Luttinger constants γ_1 , γ_2 and γ_3 and spin-orbit splitting energy Δ . The following values were used in the present calculation [10]: $\gamma_1 = 6.85$, $\gamma_2 = 2.1$, $\gamma_3 = 2.9$, $\Delta = 0.341$ eV for GaAs, and $\gamma_1 = 5$, $\gamma_2 = 1.6$, $\gamma_3 = 2.1$, $\Delta = 0.108$ eV for InP. Although we were interested only in heavy–light band transition dynamics, nonetheless, in calculations we have included the split-off band as well. The effect of the s -band is especially important in p-InP, where due to small spin-orbit splitting energy the s -band gives a large contribution to h - and l -band nonparabolicity at free hole energies close to Δ . The initial hole wave vector was parallel to either the $\langle 111 \rangle$ or $\langle 110 \rangle$ direction. In simulations two kinds of hole population oscillations were observed. The first one was fast and synchronous with changes of electric field and had small amplitude. The second kind of oscillation was slow and had large amplitude. The latter can be identified with Rabi oscillations, which are very well known in atomic physics [3]. It was observed that, as in atoms, the intervalence Rabi oscillation period was inversely proportional to electric field strength. At resonance the maximum oscillation amplitude was equal to one. The dependence of slow variation of population with time in all investigated cases could be rather well approximated by the squared-sine law. Figure 1 shows the dependence of Rabi oscillation amplitude on the IR photon energy in GaAs, when $F_1 = 10$ kV cm $^{-1}$ (this value corresponds to light intensity 4.9×10^5 W cm $^{-2}$ in a bulk of GaAs) and when the hole wave vector modulus is 3.78×10^6 cm $^{-1}$. The points are the results of simulation and solid line is a fitting with the Lorentzian curve. The dashed vertical line shows the energy difference, $\varepsilon_{lh} = \varepsilon_l(\mathbf{k}) - \varepsilon_h(\mathbf{k}) = 56.79$ meV, between light and heavy mass valence bands at the same wave vector, $|\mathbf{k}| = 3.78 \times 10^6$ cm $^{-1}$. In this particular case the absolute BS shift, which is the difference between the position of Lorentzian curve maximum and vertical line, is $\delta\varepsilon = 2.7$ meV. The relative shift in this case is $\delta\varepsilon/\varepsilon_{lh} = 0.0475$. In atoms usually one observes much smaller BS shift [3].

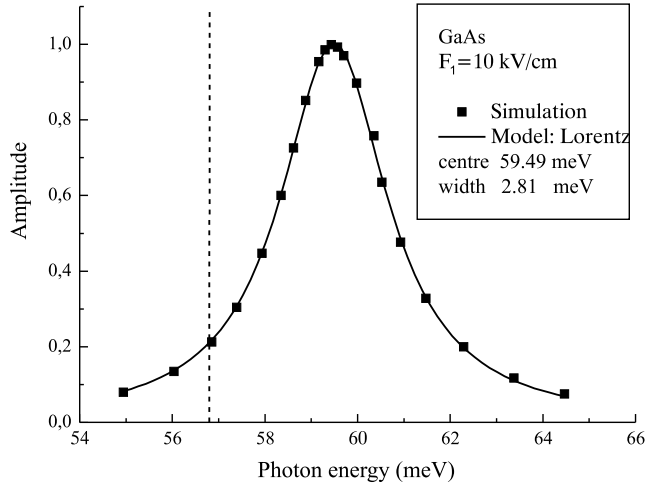


Figure 1. Dependence of Rabi oscillation amplitude on IR photon energy for light–heavy intervalence hole transitions in GaAs at hole wave vector $|\mathbf{k}| = 3.78 \times 10^6$ cm $^{-1}$. The vertical dashed line shows the value of energy between light and heavy mass bands at the same wave vector.

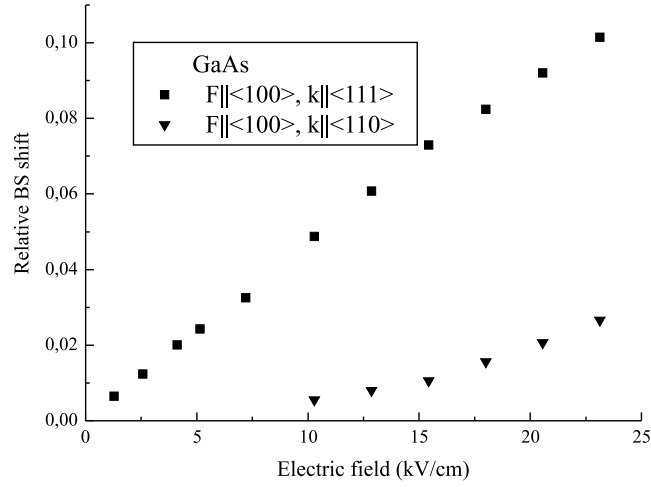


Figure 2. Relative Bloch–Siegert shift as a function of electric field amplitude in GaAs for two directions of hole wave vector, $|k| = 3.78 \times 10^6 \text{ cm}^{-1}$.

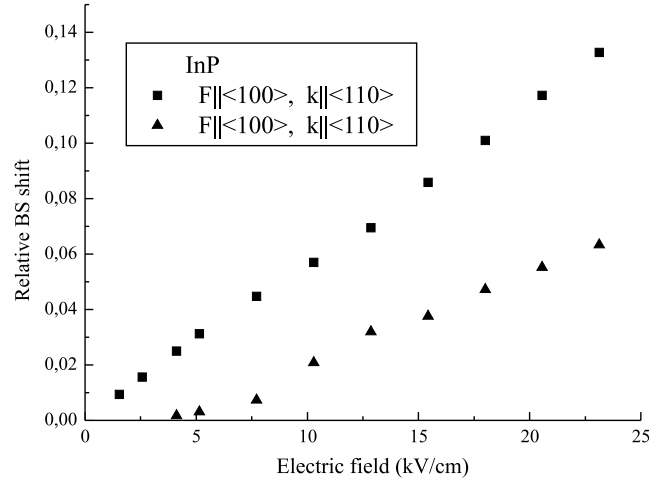


Figure 3. Relative Bloch–Siegert shift as a function of electric field amplitude in InP for two directions of hole wave vector, $|k| = 3.78 \times 10^6 \text{ cm}^{-1}$.

The dependence of relative BS shift on electric field amplitude is shown in figures 2 and 3 for p-GaAs and p-InP, respectively, for two directions of hole wave vector. The modulus of k was the same as in figure 1. First of all, in figures 2 and 3 one can see a large anisotropy in the BS shift. The magnitude of anisotropy correlates with the light and heavy mass constant-energy surface anisotropy defined by the expression

$$a = 2[\varepsilon_{lh}(111) - \varepsilon_{lh}(110)]/[\varepsilon_{lh}(111) + \varepsilon_{lh}(110)].$$

For GaAs at $|k| = 3.78 \times 10^6 \text{ cm}^{-1}$ one has $\varepsilon_{lh}(111) = 58.21 \text{ meV}$, $\varepsilon_{lh}(110) = 54.67 \text{ meV}$ and $a = 6.3\%$, while for InP one has $\varepsilon_{lh}(111) = 34.82 \text{ meV}$, $\varepsilon_{lh}(110) = 34.03 \text{ meV}$ and $a = 2.3\%$. Secondly, the growth of the BS shift with the field strength is closer to linear rather than to quadratic dependence. Since the Rabi oscillation period is inversely proportional to electric field strength, long simulation times are required at low fields. Due to accumulation

of errors (in figures 2 and 3 the scatter of points from averaged values suggests a typical error in the present simulation), it was impossible to check whether the BS shift changes over to quadratic dependence at low intensities in the case of interband transitions. Thirdly, the width of the Lorentz line in figure 1 was observed to be narrower at lower intensities, which is in agreement with the concept that Rabi oscillations result from a competition between the absorption and induced emission.

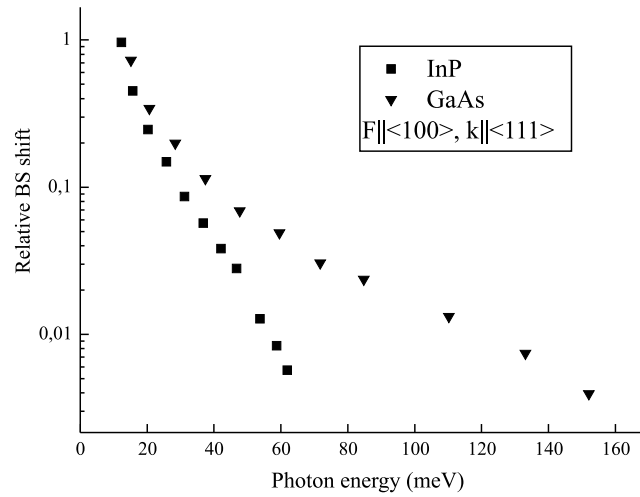


Figure 4. Relative BS shift in InP and GaAs as a function of photon energy at electric field amplitude $F_1 = 10 \text{ kV cm}^{-1}$.

Figure 4 shows that intervalence BS shift is a rather strong function of the photon energy. At $\hbar\omega > 100 \text{ meV}$ the shift is smaller than a percent; however, at the energies of the order of 10 meV the shift is as large as 100%.

As known from atomic physics, strong time-varying optical electric fields may give rise to the optical Stark effect. In the case of p-type semiconductors a similar effect, i.e. lifting of the valence band degeneracy, may be expected. The rearrangement of the band structure at the centre of the Brillouin zone, due to strong IR optical radiation field, was considered in [11]. It was shown that the action of the laser field on critical points is similar to some effective deformation, the magnitude of which is proportional to light intensity. In general the effect was found to be rather weak, about 1 meV at IR light intensity 10 MW cm^{-2} . Therefore, one expects that the valence band splitting, if any, will have negligible effect on the Bloch–Siegert shift values calculated within Luttinger–Kohn formalism. The influence of valence band degeneracy at $k = 0$ on hole velocity was considered in [12].

In conclusion, this investigation shows that the Bloch–Siegert shift is present in interband transitions as well. In the case of intervalence transitions it is a strong function of photon energy, and in the terahertz frequency range and at intensities about 1 MW cm^{-2} the shift may be larger than 100%. Probably, this conclusion also applies to two- or lower dimensional structures, where IR optical transition energies between subbands originating from spatial quantization are also small. Finally, it should be noted that the BS shift in atoms comes directly from the time dependent perturbation term, whereas in case of intervalence transitions the contribution is indirect, through oscillations of the wave vector in the Luttinger–Kohn Hamiltonian.

References

- [1] Bloch F and Siegert A J 1940 *Phys. Rev.* **57** 522
- [2] Cohen-Tannoudji C, Dupont-Roc J and Fabre C 1973 *J. Phys. B: At. Mol. Opt. Phys.* **6** L214
- [3] Allen L and Eberly J H 1975 *Optical Resonance and Two-level Atoms* (New York: Dover) ch 2
- [4] Normantas E 1982 *Fiz. Tekh. Poluprov.* **16** 630
- [5] Dargys A 1989 *J. Phys.: Condens. Matter* **1** 9653
- [6] Luttinger J M and Kohn W 1955 *Phys. Rev.* **97** 869
- [7] Stenholm S 1973 *J. Phys. B: At. Mol. Opt. Phys.* **6** 1650
- [8] Godunov S K 1979 *Mathematical Physics Equations* (Moscow: Nauka)
- [9] Press W H, Teukolsky S A, Vetterling W T and Flannery B P 1992 *Numerical Recipes in FORTRAN* (Cambridge: Cambridge University Press)
- [10] Dargys A and Kundrotas J 1994 *Handbook on Physical Properties of Ge, Si, GaAs and InP* (Vilnius Science and Encyclopedia Publishers)
- [11] Rebane Yu T 1985 *Fiz. Tverd. Tela* **27** 1364
- [12] Dargys A 1989 *Phys. Status Solidi b* **155** 615