

## Dynamics of coherent hole-population transfer between valence subbands in diamond- and zinc-blende-type semiconductors

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The time-dependent Schrödinger equation for the valence band that describes the hole dynamics in infrared (IR) laser fields is considered within Luttinger-Kohn Hamiltonian formalism. A unitary transformation matrix that substantially simplifies the initially overdetermined problem was found. For nonparabolic and spherical bands, the problem was reduced to three differential equations that describe the hole population transfer dynamics between heavy, light, and spin-orbit split-off subbands. The importance of a three-band approach in the analysis of ultrafast hole dynamics is discussed. The obtained results may be useful in analyzing the response of the hole population to a single ultrashort IR laser pulse of arbitrary shape or a combination of pulses. [S0163-1829(99)04807-9]

### I. INTRODUCTION

The response of holes in  $p$ -type semiconductors to external perturbations is usually treated within the degenerate valence-band formalism. The most popular valence-band Hamiltonian forms for semiconductors of diamond and zinc-blende structures are either  $4 \times 4$  or  $6 \times 6$  Luttinger-Kohn matrices.<sup>1-3</sup> As it is usual, spin splitting due to lack of inversion symmetry in the zinc-blende semiconductors is neglected. This results in a double degeneracy of all subbands. The interest in degenerate valence bands stems from two factors. First, because of the complex structure of the valence-band Hamiltonians, it is always desirable to find a unitary transformation that brings the Hamiltonian and the related Schrödinger system to a simpler form that is more suitable for either numerical calculations or further analytical simplification of the problem. With this in mind, a number of transformations were proposed.<sup>4-8</sup>

Second, with the advent of ultrashort-pulse optical spectroscopy, where pulse parameters such as amplitude, center time, center frequency, temporal width, type of chirping, etc. become accessible to the experimenter, there emerges a possibility of quantum control of the evolution of the wave function of the system under study (see, for example, Refs. 9 and 10 and literature cited therein). Population transfer using  $\pi$  pulses or adiabatic passage by pulse chirping, when after the optical pulse the final population resides almost entirely in a particular excited level, are well known in atomic physics.<sup>11-13</sup> The coherent phenomena in photoexcited semiconductors were considered, for example, in Refs. 14-18 and reviewed recently in Refs 19 and 20. The Coulomb correlations between electron-hole pairs determine a typical time scale on which the coherent phenomena can be observed in the case of photoexcitation of semiconductors. As discussed in Ref. 19, the coherent contribution to the considered phenomenon can be evaluated by means of a direct numerical integration while the incoherent one by a conventional Monte Carlo simulation. The approach where total dynamics is divided into coherent and incoherent parts was

used earlier in treating intravalence band infrared (IR) absorption<sup>21</sup> and hot hole noise.<sup>22</sup>

The purpose of this paper is to show how the three-band time-dependent and overdetermined  $6 \times 6$  Schrödinger system for the valence-band envelope wave function of diamond and zinc-blende semiconductors can be simplified. Earlier,<sup>4,23</sup> we described a simple parabolic two-band model in the infinite spin-orbit interaction limit and used it to explain the transparency of  $p$ -type germanium in intense  $\text{CO}_2$  and  $\text{NH}_3$  laser fields<sup>21</sup> and to predict tunnel noise in semiconductors.<sup>22</sup> As we shall see below, at least a three-band model is required to describe fast-hole transitions to the spin-orbit split-off valence subband or, what is not so evident, to include valence-band nonparabolicity correctly. In the next two sections, we present the transformed Schrödinger equation and the required transformation matrix. Finally, in Sec. IV we discuss the obtained results and give some illustrative examples.

### II. TRANSFORMED SCHRÖDINGER EQUATION

In analyzing the intervalence population dynamics, as will be seen, it is convenient to go over to a basis in which the Luttinger-Kohn Hamiltonian is a diagonal matrix and describes the dispersion laws of light ( $l$ ), heavy ( $h$ ), and spin-orbit split-off ( $s$ ) hole subbands. Such a basis appears natural if one starts with a hole, that initially with certainty was in a particular subband, and one wants to know the hole transition probability to other subbands after an external field or laser radiation was switched on. The other reason to go over to the energy representation is that in real situations, due to interaction of holes with an incoherent phonon bath, the holes populate light and heavy hole subbands before laser radiation is applied, not some intermediate energy states that are a mixture of the pure light hole, heavy hole, or split-off subband states. Of course, due to collisions of holes with phonons and a finite hole lifetime, the subbands are somewhat smeared: the smaller the hole coherence length and/or time is, the larger the subband smearing will be. In the following, the dynamical mixing of the valence subbands in

strong coherent laser fields will be treated in the wave-vector space, when the laser electric field  $F(t)$  is linearly polarized. For convenience we shall assume that the field is parallel to hole wave-vector component  $k_z$ . The electric field will be treated classically, i.e., its quantum nature will be neglected. Such an approximation is fully satisfactory, if the field amplitude is larger than a few volts per centimeter.<sup>23</sup> The hole transitions between the subbands will be described by the Schrödinger equation for the six-component envelope state vector  $\vec{\psi}$ :

$$i \frac{\partial \vec{\psi}}{\partial t} = \left( \hat{H}_0 + \frac{F(t)}{i} \frac{\partial}{\partial k_z} \right) \vec{\psi}. \quad (1)$$

Atomic units (a.u.) will be used:  $1_{\text{a.u.}}(\text{energy}) = 27.2$  eV,  $1_{\text{a.u.}}(\text{time}) = 2.42 \times 10^{-17}$  s,  $1_{\text{a.u.}}(\text{electric field}) = 5.14 \times 10^9$  V/cm,  $1_{\text{a.u.}}(\text{wave vector}) = 1.89 \times 10^8$  cm<sup>-1</sup>. In Eq. (1),  $\hat{H}_0$  is the Luttinger-Kohn Hamiltonian.<sup>1,2</sup> In the general case,  $\hat{H}_0$  is characterized by three parameters:  $\gamma_1$ ,  $\gamma_2$ , and  $\gamma_3$ . In the spherical approximation ( $\gamma_2 = \gamma_3$ ), the case considered in this paper,  $\hat{H}_0$  is

$$\hat{H}_0 = \begin{bmatrix} P+Q & L & M & 0 & \frac{i}{\sqrt{2}}L & -i\sqrt{2}M \\ L^* & P-Q & 0 & M & -i\sqrt{2}Q & i\sqrt{\frac{3}{2}}L \\ M^* & 0 & P-Q & -L & -i\sqrt{\frac{3}{2}}L^* & -i\sqrt{2}Q \\ 0 & M^* & -L^* & P+Q & -i\sqrt{2}M^* & -\frac{i}{\sqrt{2}}L^* \\ -\frac{i}{\sqrt{2}}L^* & i\sqrt{2}Q & i\sqrt{\frac{3}{2}}L & i\sqrt{2}M & P+\Delta & 0 \\ i\sqrt{2}M^* & -i\sqrt{\frac{3}{2}}L^* & i\sqrt{2}Q & \frac{i}{\sqrt{2}}Q & 0 & P+\Delta \end{bmatrix}, \quad (2)$$

where  $P = (\gamma_1/2)(k_x^2 + k_y^2 + k_z^2) \equiv (\gamma_1/2)k^2$ ,  $Q = (\gamma_2/2)(k_x^2 + k_y^2 - 2k_z^2)$ ,  $L = -i\sqrt{3}\gamma_2 k_z(k_x - ik_y)$ , and  $M = \gamma_2(\sqrt{3}/2)(k_x - ik_y)^2$ .

In Eq. (2),  $\Delta$  is the spin-orbit split-off energy. In contrast to the general case, in the spherical approximation the Hamiltonian (2) can be diagonalized to a relatively simple diagonal matrix. The wave-vector dependence of heavy  $E_h$ , light  $E_l$ , and split-off  $E_s$  subband energy in this case assumes the form

$$E_h = \frac{k^2}{2}(\gamma_1 - 2\gamma_2), \quad (3)$$

$$E_l = \frac{1}{2}(\Delta + \gamma_1 k^2 + \gamma_2 k^2 - r), \quad (4)$$

$$E_s = \frac{1}{2}(\Delta + \gamma_1 k^2 + \gamma_2 k^2 + r), \quad (5)$$

where

$$r = (\Delta^2 - 2\gamma_2 \Delta k^2 + 9\gamma_2^2 k^4)^{1/2}. \quad (6)$$

Figure 1 shows  $E_h$ ,  $E_l$ , and  $E_s$  versus  $k$  calculated from Eqs. (3)–(6), where a relatively strong nonparabolicity of  $l$  and  $s$  subbands can be seen. Here, and in the subsequent

calculations, the following parameters typical to InP were used:  $\Delta = 0.00397$  a.u. (=0.108 eV),  $\gamma_1 = 5$ ,  $\gamma_2 = 1.9$ . In the zero spin-orbit interaction limit, when  $\Delta = 0$ ,  $E_h$ , and  $E_l$  reduce to a fourfold-degenerate parabolic band and  $E_s$  to a doubly degenerate parabolic band. These new bands may be viewed, respectively, as new heavy- and light-mass subbands. In the opposite limit of strong interaction  $E_l$  and  $E_s$  reduce to  $E_l \approx \frac{1}{2}[(\gamma_1 + 2\gamma_2)k^2 - 9k^4\gamma_2^2/2\Delta]$  and  $E_s \approx \frac{1}{2}[2\Delta + \gamma_1 k^2 + 9k^4\gamma_2^2/2\Delta]$ . In the limit  $\Delta \rightarrow \infty$  the latter describes the parabolic bands as well.

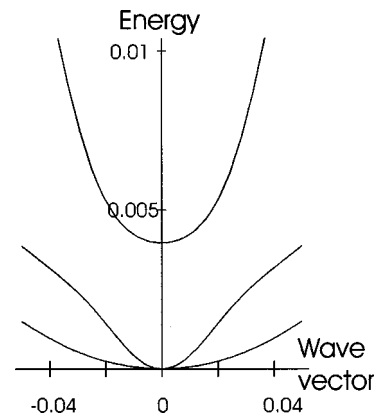


FIG. 1. Dispersion curves of heavy, light, and spin-orbit split-off subbands of InP in spherical approximation.

The unitary transformation matrix  $\hat{T}$  needed to diagonalize the Luttinger-Kohn Hamiltonian will be presented in the next section. Using the transformation matrix, the Schrödinger system (1) can be reduced to<sup>24</sup>

$$i\frac{\partial\vec{\varphi}}{\partial t} = \left[ \hat{H}_{0d} + \frac{F}{i} \left( \hat{T} \frac{\partial\hat{T}^+}{\partial k_z} + \frac{\partial}{\partial k_z} \right) \right] \vec{\varphi}, \quad (7)$$

where  $\hat{T}^+$  is Hermitian of  $\hat{T}$  and  $\vec{\varphi}$  is the new transformed state vector:  $\vec{\varphi} = \hat{T}\vec{\psi}$ . The new Hamiltonian  $\hat{H}_{0d}$  is the diagonal matrix

$$\hat{H}_{0d} = \text{diag}\{E_l, E_l, E_h, E_h, E_s, E_s\} \quad (8)$$

the elements of which are given by Eqs. (3)–(5). As it is known, in the absence of degeneracy a unitary transformation matrix can be found from normalized eigenvectors that correspond to their respective eigenvalues (dispersion laws) of the considered Hamiltonian.<sup>25</sup> However, in the general case, for example, when energy states are degenerate (in our case they are doubly degenerate) this is not true. In the following section, we shall show how the required transformation matrix can be constructed.

### III. THE TRANSFORMATION AND FIELD MATRICES

The Luttinger-Kohn Hamiltonian  $\hat{H}_0$  and the diagonalized Hamiltonian  $\hat{H}_{0d}$  are related by the transformation  $\hat{H}_{0d} = \hat{T}\hat{H}_0\hat{T}^{-1}$ . To find the matrix elements of  $\hat{T}$  we used an equivalent form  $\hat{H}_{0d}\hat{T} = \hat{T}\hat{H}_0$  along with the unitarity property of  $\hat{T}$ . In this way, the obtained 36 algebraic equations for the unknown elements  $T_{ij}$  are not independent. Therefore, the problem reduces to how to find a linearly independent set for  $T_{ij}$  elements from 36 coupled complex algebraic equations. The formulation as it stands is simple. Nonetheless, a straightforward attack by standard methods, e.g., by

elimination of variables or by methods used in algebraic computer packages, fails due to the linear dependence of the equations. This is largely connected with the presence of the square root (6) in dispersion laws. If an arbitrary algorithm is used, the dispersion laws (4) and (5) generate very complicated intermediate results, intractable for further calculations. Below, the algorithm used to find the transformation matrix  $\hat{T}$  is outlined briefly and only the final result is given.

Because of spherical symmetry of  $\hat{H}_0$  and  $\hat{H}_{0d}$ , it is convenient to reduce the matrix equation  $\hat{H}_{0d}\hat{T} = \hat{T}\hat{H}_0$  to spherical coordinates  $k$ ,  $\Theta$ , and  $\varphi$ :  $k_x = k \sin \Theta \cos \varphi$ ,  $k_y = k \sin \Theta \sin \varphi$ ,  $k_z = k \cos \Theta$ . Then representing the transformation matrix as a product of two matrices

$$\hat{T} = \hat{T}_\varphi \hat{t}, \quad (9)$$

where

$$\hat{T}_\varphi = \begin{bmatrix} 1 & e^{-i\varphi} & e^{-i2\varphi} & e^{-i3\varphi} & e^{-i\varphi} & e^{-i2\varphi} \\ e^{i\varphi} & 1 & e^{-i\varphi} & e^{-i2\varphi} & 1 & e^{-i\varphi} \\ e^{i2\varphi} & e^{i\varphi} & 1 & e^{-i\varphi} & e^{i\varphi} & 1 \\ e^{i3\varphi} & e^{i2\varphi} & e^{i\varphi} & 1 & e^{i2\varphi} & e^{i\varphi} \\ e^{i\varphi} & 1 & e^{-i\varphi} & e^{-i2\varphi} & 1 & e^{-i\varphi} \\ e^{i2\varphi} & e^{i\varphi} & 1 & e^{-i\varphi} & e^{i\varphi} & 1 \end{bmatrix}, \quad (10)$$

the azimuthal angle  $\varphi$  can be eliminated from the above matrix equations altogether. A close inspection of the resulting matrix equation shows that after equating the rows of the simplified matrix equation it is possible to construct six independent subsystems. In addition, it appears that the subsystems thus obtained, are similar in pairs and, therefore, it is enough to solve only three of them. A straightforward but lengthy calculation with MATHEMATICA gives the following final result:

$$\hat{T} = k^2 \gamma_2 \begin{bmatrix} \frac{\sqrt{3} \cos \Theta \sin \Theta}{s_1} & \frac{ix^{-1}(1+3 \cos 2\Theta)}{2s_1} & -\frac{3x^{-2} \sin 2\Theta}{2s_1} & -\frac{i\sqrt{3}x^{-3} \sin^2 \Theta}{s_1} & \frac{x^{-1}s_1}{\sqrt{2}r} & 0 \\ -\frac{i\sqrt{3}x \sin^2 \Theta}{s_1} & -\frac{3 \sin 2\Theta}{2s_1} & \frac{ix^{-1}(1+3 \cos 2\Theta)}{2s_1} & -\frac{\sqrt{3}x^{-2} \cos \Theta \sin \Theta}{s_1} & 0 & \frac{x^{-1}s_1}{\sqrt{2}r} \\ -\frac{x^2 \sin \Theta}{2k^2 \gamma_2} & 0 & \frac{\sqrt{3} \sin \Theta}{2k^2 \gamma_2} & -\frac{ix^{-1} \cos \Theta}{k^2 \gamma_2} & 0 & 0 \\ -\frac{ix^3 \cos \Theta}{k^2 \gamma_2} & \frac{\sqrt{3}x^2 \sin \Theta}{2k^2 \gamma_2} & 0 & -\frac{\sin \Theta}{2k^2 \gamma_2} & 0 & 0 \\ \frac{\sqrt{3}x \cos \Theta \sin \Theta}{s_2} & -\frac{i(1+3 \cos 2\Theta)}{2s_2} & \frac{3x^{-1} \sin 2\Theta}{2s_2} & \frac{i\sqrt{3}x^{-2} \sin^2 \Theta}{s_2} & \frac{s_2}{\sqrt{2}r} & 0 \\ \frac{i\sqrt{3}x^2 \sin^2 \Theta}{s_2} & \frac{3x \sin 2\Theta}{2s_2} & -\frac{i(1+3 \cos 2\Theta)}{2s_2} & \frac{\sqrt{3}x^{-1} \cos \Theta \sin \Theta}{s_2} & 0 & \frac{s_2}{\sqrt{2}r} \end{bmatrix}, \quad (11)$$

where  $x^n = \exp(in\varphi)$ ,  $s_1^2 = (k^2\gamma_2 - \Delta + r)r$ , and  $s_2^2 = (\Delta - k^2\gamma_2 + r)r$ . The matrix  $\hat{T}$  is unitary, i.e.,  $\hat{T}(\hat{T})^\dagger = \hat{1}$ . It should be noted that the concrete form of  $\hat{T}$  is not unique. The transformation (11) brings the Luttinger-Kohn Hamiltonian to the form (8). Other forms of  $\hat{T}$  will give different diagonalized Hamiltonians, e.g.,  $\text{diag}\{E_h, E_h, E_l, E_l, E_s, E_s\}$ ,  $\text{diag}\{E_s, E_l, E_h, E_h, E_l, E_s\}$ , etc.

Now it is possible to transform the field operator  $(1/i)(\hat{T}\partial\hat{T}^\dagger/\partial k_z + \partial/\partial k_z)$  to an energy representation using the standard operator equation  $\partial/\partial k_z = \cos\Theta\partial/\partial k - (\sin\Theta/k)\partial/\partial\Theta$ .

All the above described manipulations bring the system (1) to another system of six coupled differential equations for degenerate bands. Further simplifications are possible if one notes that one more transformation can be performed using the following diagonal matrix:

$$\hat{T}_\delta = \text{diag}\{e^{i3\varphi/2}, e^{i\varphi/2}, e^{-i\varphi/2}, e^{-i3\varphi/2}, e^{i\varphi/2}, e^{-i\varphi/2}\}. \quad (12)$$

$\hat{T}_\delta$  eliminates the azimuthal angle in the transformed field operator (this operator has the cylindrical symmetry) and at the same time preserves the diagonality of  $H_{0d}$  and, therefore, does not mix up the valence subbands. The transformation (12) brings the part  $\hat{d} = (1/i)\hat{T}\partial\hat{T}^\dagger/\partial k_z$  of the field operator to

$$\hat{d} = \begin{bmatrix} 0 & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{12} & 0 & d_{14} & d_{13} & d_{16} & d_{15} \\ d_{13}^* & d_{14} & 0 & d_{34} & d_{35} & d_{36} \\ d_{14} & d_{13}^* & d_{34} & 0 & d_{36} & d_{35} \\ d_{15}^* & d_{16} & d_{35}^* & d_{36} & 0 & d_{56} \\ d_{16} & d_{15}^* & d_{36} & d_{35}^* & d_{56} & 0 \end{bmatrix}, \quad (13)$$

where

$$d_{12} = \frac{6k^3 \sin\Theta \gamma_2^2}{s_1^2}, \quad (14)$$

$$d_{13} = \frac{\sqrt{3}k \sin^2\Theta \gamma_2}{is_1}, \quad d_{14} = -\frac{\sqrt{3}k \cos\Theta \sin\Theta \gamma_2}{s_1}, \quad (15)$$

$$d_{15} = \frac{i8k^3 \Delta \cos\Theta \gamma_2^2}{rs_1 s_2}, \quad d_{16} = -d_{12} s_1 / s_2, \quad (16)$$

$$d_{34} = -\sin\Theta / 2k, \quad (17)$$

$$d_{35} = d_{13} s_1 / s_2, \quad d_{36} = -d_{14} s_1 / s_2, \quad (18)$$

$$d_{56} = d_{12} s_1^2 / s_2^2. \quad (19)$$

Finally, with the help of the transformation

$$\frac{i}{\sqrt{2}} \begin{bmatrix} 1 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 1 \\ 1 & -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & -1 \end{bmatrix} \quad (20)$$

the coupling between degenerate valence subbands can be lifted off, i.e., the considered system of differential equations can be brought to a  $3 \times 3$  block-diagonal system. One of the two subsystems thus obtained is

$$i \frac{\partial}{\partial t} \begin{bmatrix} f_l \\ f_h \\ f_s \end{bmatrix} = \left\{ \begin{bmatrix} E_l & 0 & 0 \\ 0 & E_h & 0 \\ 0 & 0 & E_s \end{bmatrix} + F \begin{bmatrix} d_{12} & d_{13} + d_{14} & d_{15} + d_{16} \\ d_{13}^* + d_{14} & d_{34} & d_{35} + d_{36} \\ d_{15}^* + d_{16} & d_{35}^* + d_{36} & d_{56} \end{bmatrix} + \frac{F}{i} \frac{\partial}{\partial k_z} \right\} \begin{bmatrix} f_l \\ f_h \\ f_s \end{bmatrix}, \quad (21)$$

where  $f_l$ ,  $f_h$ , and  $f_s$  are state-vector components that describe dynamics of hole population in light, heavy, and split-off valence subbands. The other subsystem can be obtained from Eq. (21) after complex conjugation and change of variables:  $t \rightarrow -t$ ,  $F \rightarrow -F$ .

#### IV. DISCUSSION

It should be clear that now it is enough to solve only three differential equations instead of the initial six. Furthermore, in the obtained state vector  $\vec{f}$  formulation, the interpretation and the assumption about the initial hole population in various subbands is straightforward:  $|f_i|^2$  is the  $i$ th band population. The  $k_z$  component can be related to the time-dependent electric field through a characteristic equation of the subsystem (21).<sup>26</sup> In our case, the characteristic equation describes the Newton's law for  $k_z$  component:  $dk_z/dt = F(t)$ . The component perpendicular to  $k_z$  is conserved in the spherical approximation. With this in mind the partial derivatives in Eq. (21) may be replaced by the total derivative  $(i\partial/\partial t, i\partial/\partial k_z) \rightarrow id/dt$ , i.e., Eq. (21) can be reduced to a system of complex rate equations, in which the terms  $d_{ij}$  describe the coupling between population state vectors.

In the case of large spin-orbit interaction, the elements of the coupling matrix can be developed in powers of  $1/\Delta$ , then the field coupling matrix in Eq. (21) becomes

$$F \frac{\sin \Theta}{k} \begin{bmatrix} \frac{3}{2} & \frac{\sqrt{3}}{2} e^{-i\Theta} & \frac{3+i4 \cot \Theta}{\sqrt{2}} \frac{k^2 \gamma_2}{\Delta} \\ \frac{\sqrt{3}}{2} e^{i\Theta} & \frac{1}{2} & -\sqrt{3} e^{i\Theta} \frac{k^2 \gamma_2}{\Delta} \\ \frac{3-i4 \cot \Theta}{\sqrt{2}} \frac{k^2 \gamma_2}{\Delta} & -\sqrt{3} e^{-i\Theta} \frac{k^2 \gamma_2}{\Delta} & -3 \frac{k^4 \gamma_2^2}{\Delta^2} \end{bmatrix}. \quad (22)$$

In the limit of infinite spin-orbit interaction ( $\Delta \rightarrow \infty$ ) the matrix (22) reduces to that obtained earlier in Ref. 4 for two, heavy and light, parabolic subbands.

Selection rules and intersubband transition coupling strengths, determined respectively by  $(d_{13}+d_{14})$ ,  $(d_{35}+d_{36})$ , and  $(d_{15}+d_{16})$  terms in Eq. (21), depend on the angle  $\Theta$  between the electric field  $\vec{F}$  and the hole wave vector  $\vec{k}$ . For  $\Theta=0$ , i.e., when  $\vec{k}$  and  $\vec{F}$  are parallel, only  $l$ - $s$  transitions are allowed. For  $\Theta=\pi/2$ , all transitions are allowed. For large  $\Delta$  values the transitions between heavy- and light-mass subbands will predominate, as can be seen from Eq. (22).

Figure 2 shows the time dependence of the split-off subband population  $|f_s|^2$ , when initially the hole with certainty was in the heavy-hole subband<sup>27</sup> with  $\Theta=\pi/2$  and  $k=0.0259$ . Curve 1 corresponds to exact resonance, i.e., when the laser frequency is  $\omega=E_l(k)-E_h(k)=4.5 \times 10^{-3}$ . The steps correspond to half periods of the electric field. The curve labeled ‘‘Off-resonance’’ shows the evolution of  $|f_s|^2$ , when the laser frequency was detuned by 10% from the resonance. The electric field amplitude for both cases was  $F=10^{-5}$  and the laser was turned on at  $t=0$ .

Figure 3 shows nearly total transfer of the hole population from the heavy to the light-hole subband of  $p$ -type InP, when  $\Theta=\pi/2$  and  $k=0.0178$ , and when the hole was acted on by about three periods of Gaussian-shaped infrared-laser pulse of the angular frequency  $10^{-3}$ , as shown in the upper left corner in the figure. The pulse width at half maximum  $t_{1/2}=0.126$  ps and the maximum electric field amplitude  $F_0$  of the Gaussian envelope is 32 kV/cm. The transfer of 95% of

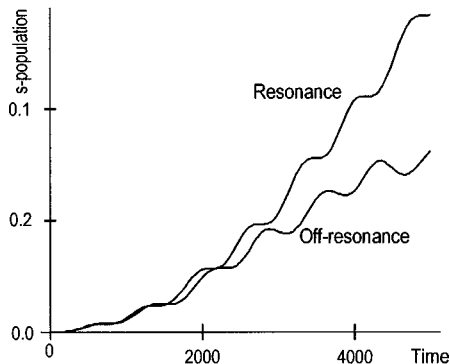


FIG. 2. The probability to find the hole in the spin-orbit split-off subband at resonance and after laser was detuned by 10%. The sinusoidal electric field in both cases was switched on at the moment  $t=0$ .

the population was achieved by tailoring  $F_0$  and  $t_{1/2}$  of the pulse. To obtain 100% transfer, what would correspond to  $\pi$  pulse, inclusion of some frequency chirping is probably required. Figure 3 clearly demonstrates that in semiconductors a nearly total hole transfer between subbands is possible if ultrashort laser pulses, consisting of only few periods, are used. Calculations show that in the considered case the maximum  $s$ -band population  $|f_s|^2$  during the IR pulse does not exceed 0.2% and drops down to 0.05% at the end of the pulse. Thus, if one is interested in  $h$ - $l$  transitions only, one may neglect in the transformed Schrödinger system (21) the spin-orbit split-off subband and the respective coupling terms associated with this subband even in the case of very short, femtosecond IR pulses. However, it should be noted that in such a two-band approximation, the splitting energy  $\Delta$  does not vanish as can be seen by inspection of the matrix elements  $d_{12}$ ,  $d_{34}$ ,  $(d_{13}+d_{14})$ . Negligible  $s$ -subband population in this case is in a large part associated with a strong coupling between  $h$  and  $l$  subbands. Here, it is also important to remark that in the discussed two-band approximation, the subband nonparabolicity and the subband coupling are taken fully into account. In the standard two-band approximation, where at first one takes the limit  $\Delta \rightarrow \infty$  and then only diagonalizes the remaining Hamiltonian, the nonparabolicity is lost. This can also be seen from Eq. (22), if the limit  $\Delta \rightarrow \infty$  is taken.

Figure 4 shows the light and split-off subband populations as a function of time when the hole initially was in the same state as in Fig. 3, i.e., in the  $h$  subband with the same  $k$  and  $\Theta$ , but the laser now was tuned to a  $h$ - $s$  transition:  $\omega=E_s(k)-E_h(k)=0.0048$ . In this case after optimization of  $t_{1/2}$  and  $F_0$  it appeared possible to transfer as much as 82%

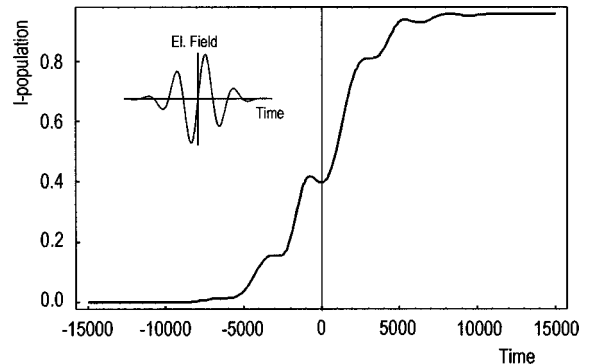


FIG. 3. Transfer of hole population from heavy- to light-hole subband induced by ultrashort,  $t_{1/2}=126$  fs, IR laser pulse shown in the upper-left corner. Electric field envelope amplitude  $F_0$  is 32 kV/cm.

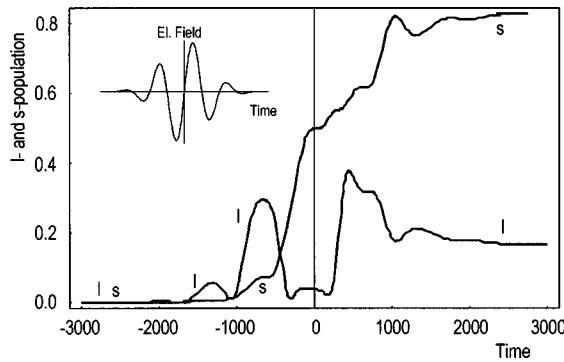


FIG. 4. Split-off ( $s$ ) and light ( $l$ ) subband population induced by ultrashort,  $t_{1/2} = 24.2$  fs, IR laser pulse shown in the upper-left corner.  $F_0 = 478$  kV/cm.

of the  $h$ -subband hole population to the split-off subband. It should be remarked here that, due to strong  $h$ - $l$  coupling, the light-hole population  $|f_l|^2$  during laser excitation may reach rather high (up to 37% in Fig. 4) intermediate values. To have the hole population in the  $s$  subband close to 100% at the end of the laser pulse, a more complicated shape of the electric field, which may be found with the help of the quantum optimal control theory,<sup>9,10</sup> is required. In general, one also would like to know whether in principle it is possible to

transfer a hole (electron) to a higher energy subband without exciting other valence (conduction) subbands during the action of the laser pulse. This property may be important if the phonon emission rate from other subbands is larger than from the final one.

In conclusion, the overdetermined time-dependent Schrödinger system described by  $6 \times 6$  Luttinger-Kohn Hamiltonian was reduced to the much simpler system (21), assuming that the valence band is spherical ( $\gamma_2 = \gamma_3$ ) and that the laser electric field is linearly polarized. The nonparabolicity was included fully. The components of the wave function of the reduced equation directly reflect the hole population in the respective valence subbands, what may be advantageous in analyzing the population dynamics under action of arbitrarily shaped ultrashort IR laser pulses, for example, linearly or quadratically chirped pulses. Furthermore, the obtained exact equations may be a starting point for further analytical simplifications. If needed, the warping of the subbands may be included as a perturbation, by applying the transformation matrix (11) to that part of the Hamiltonian, which describes the nonsphericity of the valence band.

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