Optically Injected Spin Currents in Semiconductors

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We show that quantum interference of one and two photon absorption from a two color field allows one to optically inject ballistic spin currents in unbiased semiconductors. The spin currents can be generated with or without an accompanying electrical current and can be controlled using the relative phase of the two colors. We characterize the injected spin currents using symmetry arguments and an eight-band Kane model.

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The control of electronic spin in semiconductors is important for the study of spin dynamics in many-body systems, crucial for the development of new data storage and processing scenarios based on the spin degree of freedom, and essential as a first step towards a solid state implementation of a quantum computer [1]. There has been considerable work on achieving spin-polarized currents in semiconductors using transport in the presence of magnetic impurities [2–4] and injection from a ferromagnetic contact [5,6]. Optical manipulation of electron spin has largely been based on the fact that partially spin-polarized carriers can be injected in a semiconductor via one photon absorption of circularly polarized light [7]. This has been understood since the 1970s, but it is only recently that such optically oriented carriers have been dragged by a bias voltage to produce a spin-polarized current [8,9].

All these techniques rely on either an initial spin asymmetry or the use of transport effects to sort or accelerate the spins of interest. Moreover, the spin currents are always accompanied by charge currents. In this Letter we show that *direct* optical injection of spin currents in an unbiased, clean semiconductor should be possible—either with or without a concomitant charge current. Hence spin currents can be optically manipulated on the time scale of the ultrafast laser pulses now available. Admittedly, transport phenomena will modify the evolution of the spin current, but they are not essential for its generation.

The new effect we introduce here involves the quantum interference of one- and two-photon absorption processes in a two color light field. Such quantum interference can inject a k-space distribution of carriers with polar asymmetry—i.e., an electrical current—that can be controlled by adjusting the relative phase of the two beams [10,11]. In this Letter we show that by the appropriate choice of beam polarizations, the current is spin polarized. We also show that it is possible to generate spin currents with *no* associated electrical current. Here the quantum interference sorts the carriers as they are injected, sending those of one spin in one direction and those of the opposite spin in the opposite direction.

Unlike some coherent control schemes [12,13], the effect does not rely on any specific crystal symmetry; in this Letter we assume a bulk cubic crystal [14]. Since the asymmetry in spin populations that appears is a consequence of the spin-orbit coupling, spin injection due to one photon absorption decreases significantly when transitions from the split-off band become allowed [7]. A similar decrease occurs for the spin current injection; thus we here consider only light frequencies low enough so that there are no transitions from the split-off band.

In the independent particle approximation, the two color light field $\mathbf{E}(t) = \mathbf{E}_\omega e^{-i\omega t} + \mathbf{E}_{2\omega} e^{-i2\omega t}$ + c.c. causes transitions from the semiconductor ground state $|0\rangle$ to states $|csvp\mathbf{k}\rangle$ by exciting an electron from valence band v with spin index p to conduction band c with spin index s while leaving its crystal wave vector $\mathbf{k}$ unchanged. We write the state of the system as $|\psi\rangle = c_o(t)|0\rangle + \sum c_{csvp\mathbf{k}}(t)|csvp\mathbf{k}\rangle$ and calculate $c_{csvp\mathbf{k}}(t)$ to second order in perturbation theory, treating the light classically in the long wavelength limit. The injection rate of the expectation value of any observable $\hat{\theta}$ can then be written as

$$d\langle\hat{\theta}\rangle/dt = \dot{\theta}_1 + \dot{\theta}_I + \dot{\theta}_2\,,$$

where $\dot{\theta}_1$ is due to one photon absorption from the 2ω beam, $\dot{\theta}_2$ is due to two photon absorption from the ω beam, and $\dot{\theta}_I$ is an interference term. Each of these will have contributions from the injected electrons and holes, e.g., $\dot{\theta}_1 = \dot{\theta}_{1;e} + \dot{\theta}_{1;h}$. Through scattering and recombination processes, $\langle\hat{\theta}\rangle$ will relax to a steady state value. The injection terms discussed here can be used as a source in hydrodynamic equations [10,15], or a more exact quantum calculation can be made that treats both the generation and relaxation on an equal footing [16]; we defer these issues to a later communication. Electrical current injection experiments at room temperature are in good qualitative, and indeed semiquantitative, agreement with even the simpler transport description [15]; we would expect the same for spin current injection experiments. Since spin relaxation times for holes are much shorter than those for electrons [7], the electron contribution to the spin current will probably be the essential one in comparing with experiment, but we give both electron and hole contributions below.

Our measure of spin current is the pseudotensor $K^{ab} \equiv \langle\hat{v}^a\hat{S}^b\rangle$, where $\hat{\mathbf{v}}$ is the velocity operator and $\hat{\mathbf{S}}$ is the

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spin operator; superscript lowercase letters denote Cartesian components. For crystals of high enough symmetry, there is no spin current due to one or two photon absorption alone. The interference term survives, however, and is given by

$$\dot{K}^{ab}_{I;e} = \frac{2\pi}{L^3} \sum_{c,v,s,s',p,\mathbf{k}} \langle cs\mathbf{k}|\hat{v}^a \hat{S}^b|cs'\mathbf{k}\rangle \times \Omega^{(1)*}_{cs,vp,\mathbf{k}} \Omega^{(2)}_{cs',vp,\mathbf{k}} \delta[2\omega - \omega_{cv}(\mathbf{k})] + \text{c.c.},$$

$$\dot{K}^{ab}_{I;h} = -\frac{2\pi}{L^3} \sum_{c,v,s,p,p',\mathbf{k}} \langle vp'\mathbf{k}|\hat{v}^a \hat{S}^b|vp\mathbf{k}\rangle \times \Omega^{(1)*}_{cs,vp,\mathbf{k}} \Omega^{(2)}_{cs,vp',\mathbf{k}} \delta[2\omega - \omega_{cv}(\mathbf{k})] + \text{c.c.},$$

where $|ns\mathbf{k}\rangle$ is a Bloch state, the one photon amplitude $\Omega^{(1)}_{cs,vp,\mathbf{k}}$ and two photon amplitude $\Omega^{(2)}_{cs',vp,\mathbf{k}}$ are

$$\Omega^{(1)}_{cs,vp,\mathbf{k}} = i \frac{e}{2\hbar\omega} \mathbf{E}_{2\omega} \cdot \mathbf{v}_{cs,vp}(\mathbf{k}),$$

$$\Omega^{(2)}_{cs',vp,\mathbf{k}} = -\left(\frac{e}{\hbar\omega}\right)^2 \times \sum_{ns''} \frac{[\mathbf{E}_\omega \cdot \mathbf{v}_{cs',ns''}(\mathbf{k})][\mathbf{E}_\omega \cdot \mathbf{v}_{ns'',vp}(\mathbf{k})]}{\omega_{cv}(\mathbf{k})/2 + \omega_{vn}(\mathbf{k})},$$

where $\mathbf{v}_{ns,ms'}(\mathbf{k})$ is a velocity matrix element between Bloch states, $\hbar\omega_n(\mathbf{k})$ is the energy of band n, $\omega_{nm}(\mathbf{k}) = \omega_n(\mathbf{k}) - \omega_m(\mathbf{k})$, and L^3 is the normalization volume. We have assumed for simplicity that there is no degeneracy among bands except for spin degeneracy.

The form of $\dot{K}^{ab}$ is constrained by symmetry considerations. For cubic semiconductors with the point group T_d, O, or O_h, $\dot{K}^{ab}$ has the same symmetry it would have in an isotropic medium and can be written in terms of four independent parameters A_i, $i = 1$–4, as

$$\dot{K}^{ab}_I/D = A_1 E^a_\omega (\mathbf{E}^*_{2\omega} \times \mathbf{E}_\omega)^b + A_2 (\mathbf{E}^*_{2\omega} \times \mathbf{E}_\omega)^a E^b_\omega + A_3 \epsilon^{abc} E^{*c}_{2\omega} (\mathbf{E}_\omega \cdot \mathbf{E}_\omega) + A_4 \epsilon^{abc} E^c_\omega (\mathbf{E}^*_{2\omega} \cdot \mathbf{E}_\omega) + \text{c.c.},$$

where ϵ^{abc} is the completely antisymmetric Levi-Civita pseudotensor, and the repeated index c is to be summed over. A common factor D, defined below, has been separated so that the A_i are dimensionless.

We determine these parameters using an eight-band Kane model [7,17]. The model diagonalizes the Hamiltonian in a basis of eight zone center states. We neglect terms in the bands linear in k that can arise in the absence of inversion symmetry. We use the model to lowest order in k, obtaining the energy bands, states, and matrix elements in terms of three parameters: the fundamental band gap E_g, the split-off gap Δ, and the Kane energy E_P. The resulting four pairs of doubly degenerate bands, shown in Fig. 1, are parabolic. They are characterized by effective masses m_n, which we treat as additional independent parameters; note that the masses associated with the valence bands are negative. For the velocity operator we take $\hat{\mathbf{v}} = \hat{\mathbf{p}}/m$, neglecting a term due to the spin-orbit coupling.

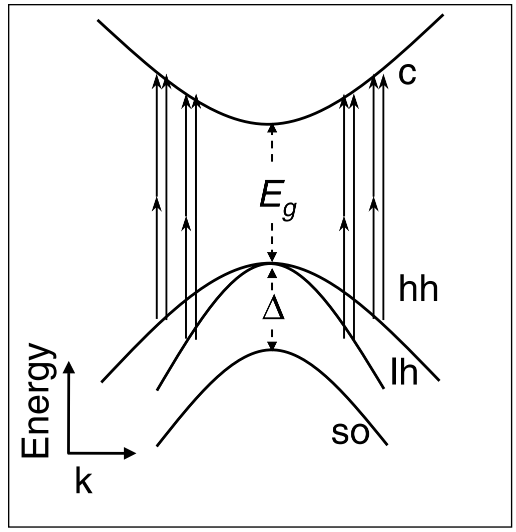

FIG. 1. Eight-band Kane model of a direct band gap semiconductor consisting of four pairs—conduction (c), heavy hole (hh), light hole (lh), and split-off (so)—of doubly degenerate bands. The fundamental gap E_g and split-off gap Δ are shown, and one and two photon transitions are indicated.

Because of the energy denominator in the two photon amplitude, intermediate states are less important the further in energy they are from the conduction and valence band of interest. We distinguish two types of terms: two-band terms, in which the intermediate band is the same as either the initial or final band, and three-band terms, in which it is different. Our calculation includes all the two-band terms, and only the three-band terms in which the intermediate band comes from the set {lh, hh, c}.

For the hh-c transition, the nonzero two-band electron terms are

$$A_{1;e:hh\text{-}c} = A_{2;e:hh\text{-}c} = \left(\frac{m_{c,hh}}{m}\right)^{3/2} \frac{m}{m_c} (1 + \zeta)$$

and the nonzero three-band electron terms are

$$A_{1;e:hh\text{-}lh\text{-}c} = \left(\frac{m_{c,hh}}{m}\right)^{5/2} \frac{m}{m_c} \frac{E_P}{3E_g} \frac{1 - \zeta}{1 + \varepsilon m_{c,hh}/m_{hh,lh}},$$

$$A_{2:e,hh\text{-}lh\text{-}c} = A_{4:e,hh\text{-}lh\text{-}c}/2 = -A_{1;e:hh\text{-}lh\text{-}c},$$

where $\varepsilon \equiv (2\hbar\omega - E_g)/(\hbar\omega)$, $m^{-1}_{n,m} = m^{-1}_n - m^{-1}_m$, and the quantity

$$\zeta \equiv \frac{1}{3} \frac{E_P \Delta}{E_g(\Delta + E_g)} \frac{m_c}{m}$$

is a measure of the extent to which the spin and velocity of an electron in the conduction band are entangled; setting ζ to zero gives the expressions that would have resulted if the approximation $\langle cs\mathbf{k}|\hat{v}^a \hat{S}^b|cs'\mathbf{k}\rangle \simeq \langle c\mathbf{k}|\hat{v}^a|c\mathbf{k}\rangle\langle cs\mathbf{k}|\hat{S}^b|cs'\mathbf{k}\rangle$ had been made. For the lh-c transition, the nonzero two-band electron terms are

$$A_{1;e:lh\text{-}c} = \left(\frac{m_{c,lh}}{m}\right)^{3/2} \frac{m}{m_c}\left(\frac{7}{3} - \zeta\right),$$

$$A_{2;e:lh\text{-}c} = \left(\frac{m_{c,lh}}{m}\right)^{3/2} \frac{m}{m_c}\left(\frac{7}{3}\zeta - 1\right),$$

and the nonzero three-band electron terms are

$$A_{1;e:lh\text{-}hh\text{-}c} = -\left(\frac{m_{c,lh}}{m}\right)^{5/2} \frac{m}{m_c}\frac{E_P}{3E_g}\frac{1}{1 - \varepsilon m_{c,lh}/m_{hh,lh}},$$

$$A_{2;e:lh\text{-}hh\text{-}c} = \zeta A_{1;e:lh\text{-}hh\text{-}c},$$

$$A_{3;e:lh\text{-}hh\text{-}c} = 2(\zeta - 1)A_{1;e:lh\text{-}hh\text{-}c}.$$

The factor

$$D \equiv \frac{\sqrt{2}}{60\pi}\frac{e^3 E_P}{\sqrt{m}}\frac{(2\hbar\omega - E_g)^{3/2}}{\hbar^4\omega^4}$$

contains the frequency dependence of the two-band terms.

For the holes, we give only the nonzero two-band terms

$$A_{1;h:hh\text{-}c} = A_{2:h,hh\text{-}c} = -\left(\frac{m_{c,hh}}{m}\right)^{3/2}\frac{m}{m_{hh}},$$

$$A_{1;h:lh\text{-}c} = -\frac{17}{9}\left(\frac{m_{c,lh}}{m}\right)^{3/2}\frac{m}{m_{lh}}\left(\frac{3E_P - 3E_g}{2E_P - 3E_g}\right),$$

$$A_{2;h:lh\text{-}c} = -\frac{1}{3}\left(\frac{m_{c,lh}}{m}\right)^{3/2}\frac{m}{m_{lh}}\left(\frac{7E_P + 3E_g}{2E_P - 3E_g}\right).$$

While the holes are injected with spin opposite that of the electrons [7], their velocity is also opposite, and thus the hole and electron spin currents have the same sign.

The same model used to calculate $\dot{K}^{ab}$ above gives an electrical current injection

$$\dot{J}_I^a = -iD\frac{e}{\hbar}[B_1 E_\omega^a(\mathbf{E}_\omega \cdot \mathbf{E}_{2\omega}^*) + B_2 E_{2\omega}^{*a}(\mathbf{E}_\omega \cdot \mathbf{E}_\omega)] + \text{c.c.},$$

where the parameters B_i (which include both electron and hole currents) are given by

$$B_{1:lh\text{-}c} = \frac{22}{3}\sqrt{\frac{m_{c,lh}}{m}}; \qquad B_{2:lh\text{-}c} = 2\sqrt{\frac{m_{c,lh}}{m}},$$

$$B_{1:hh\text{-}c} = 6\sqrt{\frac{m_{c,hh}}{m}}; \qquad B_{2:hh\text{-}c} = -2\sqrt{\frac{m_{c,hh}}{m}},$$

$$B_{1:lh\text{-}hh\text{-}c} = -\frac{4}{3}\left(\frac{m_{c,lh}}{m}\right)^{3/2}\frac{1}{1 - \varepsilon m_{c,lh}/m_{hh,lh}}\frac{E_P}{E_g},$$

$$B_{2:lh\text{-}hh\text{-}c} = -2B_{1:lh\text{-}hh\text{-}c},$$

$$B_{1:hh\text{-}lh\text{-}c} = 2\left(\frac{m_{c,hh}}{m}\right)^{3/2}\frac{1}{1 + \varepsilon m_{c,hh}/m_{hh,lh}}\frac{E_P}{E_g},$$

$$B_{2:hh\text{-}lh\text{-}c} = -\frac{1}{3}B_{1:hh\text{-}lh\text{-}c}.$$

Since we use an isotropic Kane model, there are two independent parameters in $\dot{J}_I^a$ rather than the three expected for crystals with point group T_d, O, or O_h. The electrical current injection $\dot{\mathbf{J}}_I$ has also been calculated using *ab initio* density-functional theory [10] and a simple three-band model [18]. A nonzero $\dot{\mathbf{J}}_1$ or $\dot{\mathbf{J}}_2$ requires lower crystal symmetry or surfaces [13].

We now examine the spin, current, and spin currents injected by various beam configurations. The beams are taken to be copropagating. Since our model is isotropic, there is no loss in generality in choosing the propagation direction as $\hat{\mathbf{z}}$.

Case 1: Same circular polarizations.—$\mathbf{E}_{\omega/2\omega} = E_{\omega/2\omega}e^{i\phi_{\omega/2\omega}}(\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})/\sqrt{2}$. The electron spin injection due to one photon absorption from the 2ω beam is $\dot{\mathbf{S}}_{1;e} = \mp(\hbar/4)\dot{n}_1\hat{\mathbf{z}}$ within the Kane model, where $\dot{n}_1$ is the one photon carrier injection rate [7]. The electron spin injected due to two photon absorption from the ω beam, also calculated within the Kane model, is

$$\dot{\mathbf{S}}_{2e} = \mp\frac{\hbar}{2}\frac{\sqrt{m_{c,hh}} + (7/3)\sqrt{m_{c,lh}}}{3\sqrt{m_{c,hh}} + (11/3)\sqrt{m_{c,lh}}}\dot{n}_2\hat{\mathbf{z}},$$

where only the two-band terms are included, and $\dot{n}_2$ is the two photon carrier injection rate. There is no interference term in either the spin or carrier injection when the crystal possesses inversion symmetry and thus the spin injection is simply $\dot{\mathbf{S}}_e = \dot{\mathbf{S}}_{1e} + \dot{\mathbf{S}}_{2e}$ [19]. The electrical current injection is $\dot{\mathbf{J}}_I = \sqrt{2}\,\hat{\mathbf{m}}DB_1E_\omega^2E_{2\omega}e/\hbar$, where the direction $\hat{\mathbf{m}} \equiv \hat{\mathbf{x}}\sin(2\phi_\omega - \phi_{2\omega}) \pm \hat{\mathbf{y}}\cos(2\phi_\omega - \phi_{2\omega})$. Its magnitude is comparable to the current from colinearly polarized beams; in fact, it is a factor of $\sqrt{2}$ smaller, since $B_1 \gg B_2$. The spin current injection is

$$\dot{K}_I^{ab} = \mp\sqrt{2}E_\omega^2E_{2\omega}D \times \{(A_1 - A_4)\hat{m}^a\hat{z}^b + (A_2 + A_4)\hat{z}^a\hat{m}^b\}.$$

Recalling that the first index of K^{ab} is associated with the carrier velocity, we see that the first term in $\dot{K}_I^{ab}$ shows that the electrical current is partially spin polarized. The extent of the spin polarization of the current f is defined by $\dot{K}^{az}/(\dot{J}^a/e) = \mp f\hbar/2$. In this case, $f = 2(A_1 - A_4)/B_1$. Using parameter values appropriate to GaAs [20], $f = 0.57$. The second term in $\dot{K}_I^{ab}$ represents spins pointing along $\hat{\mathbf{m}}$ that move along $\hat{\mathbf{z}}$. Since there is no net electrical current in the $\hat{\mathbf{z}}$ direction, this is a pure spin current. It arises because the electrons have a

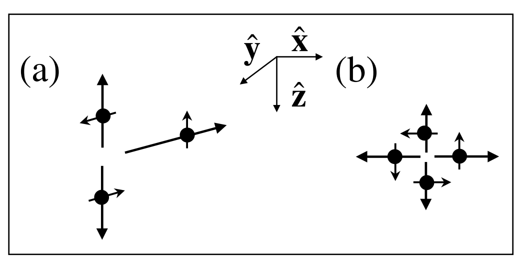

FIG. 2. Schematic illustrations of the net electron motion combining the information of $\dot{K}^{ab}$ and $\dot{J}^a$ for (a) case 1 with both beams right circularly polarized and (b) case 2, cross-polarized beams. The directions are specified in the text.

distribution of velocities such that those with positive $\hat{\mathbf{z}}$ components have opposite average spin to those with negative $\hat{\mathbf{z}}$ components. In GaAs, the second term is a factor of 7.7 smaller than the first. The situation is schematically indicated in Fig. 2(a).

Case 2: Cross linear polarizations.—$\mathbf{E}_\omega = E_\omega e^{i\phi_\omega}\hat{\mathbf{x}}$ and $\mathbf{E}_{2\omega} = E_{2\omega} e^{i\phi_{2\omega}}\hat{\mathbf{y}}$. In this case, since the beams are linearly polarized, there is no spin injection, $\dot{\mathbf{S}}_e = \dot{\mathbf{S}}_h = 0$, which can be verified by symmetry arguments. The electrical current depends sinusoidally on the relative phase of the two beams, $\dot{\mathbf{J}}_I = 2(e/\hbar)B_2 D E_\omega^2 E_{2\omega}\sin(2\phi_\omega - \phi_{2\omega})\hat{\mathbf{y}}$. According to the Kane model, its magnitude is much smaller than the current from cocircularly polarized beams, since the contributions from the heavy hole and light hole transitions largely cancel in B_2 rather than adding in B_1 [21]. In the notation of Atanasov *et al.* [10], the current in this case is proportional to η^{yxxy} while the current in case 1 is proportional to $\sqrt{2}\,\mathrm{Im}\eta^{xxyy}$, which is an order of magnitude greater than η^{yxxy} in their *ab initio* calculations. The spin current for cross linear polarizations is

$$\dot{K}_I^{ab} = -2E_\omega^2 E_{2\omega} D\cos(2\phi_\omega - \phi_{2\omega}) \times \{(A_1 + A_3)\hat{x}^a\hat{z}^b + (A_2 - A_3)\hat{z}^a\hat{x}^b\}.$$

Again there are two terms, the first arising from carrier motion along $\mathbf{E}_\omega$ with spins aligned along the beam propagation direction, and the second arising from carrier motion along the beam propagation direction with spins aligned along $\mathbf{E}_\omega$. Both of these are pure spin currents, since there is no electrical current in either direction. In GaAs, when the optical phase difference is zero, the first term is a factor of 1.3 larger and the second term is a factor of 3.3 smaller than the first term of the case 1 spin current injection. The electrical current injected along the $\mathbf{E}_{2\omega}$ polarization is always unpolarized. The situation is schematically indicated in Fig. 2(b).

There is no current or spin current injected when the beams have opposite circular polarizations, and there is only a small spin current injection, proportional to $A_3 + A_4$, when the beams are colinearly polarized.

In summary, we have shown that the quantum interference between one and two photon absorption can be used to inject and coherently control a spin current. This technique should prove an ideal way to study spin transport and relaxation, since spin current can be injected without a concomitant electrical current. And, as progress is made in extending relaxation times, it should lead to the all-optical manipulation of spin currents on an ultrafast time scale. For simplicity we have presented calculations only for bulk GaAs using an eight-band Kane model, but the effect will survive in nanostructure geometries, where there may be more applications to device physics.

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[1] D. D. Awschalom and J. M. Kikkawa, Phys. Today **52**, No. 6, 33 (1999).
[2] M. Oestreich *et al.,* Appl. Phys. Lett. **74**, 1251 (1999).
[3] R. Fiederling *et al.,* Nature (London) **402**, 787 (1999).
[4] Y. Ohno *et al.,* Nature (London) **402**, 790 (1999).
[5] P. R. Hammar *et al.,* Phys. Rev. Lett. **83**, 203 (1999).
[6] S. Gardelis *et al.,* Phys. Rev. B **60**, 7764 (1999).
[7] M. I. Dyakonov and V. I. Perel, in *Optical Orientation,* edited by F. Meier and B. P. Zakharchenya, Modern Problems in Condensed Matter Sciences Vol. 8 (North-Holland, Amsterdam, 1984), Chap. 2.
[8] D. Hägele *et al.,* Appl. Phys. Lett. **73**, 1580 (1998).
[9] J. M. Kikkawa and D. D. Awschalom, Nature (London) **397**, 139 (1999).
[10] R. Atanasov *et al.,* Phys. Rev. Lett. **76**, 1703 (1996).
[11] A. Haché *et al.,* Phys. Rev. Lett. **78**, 306 (1997).
[12] J. M. Fraser *et al.,* Phys. Rev. Lett. **83**, 4192 (1999).
[13] N. Laman *et al.,* Appl. Phys. Lett. **75**, 2581 (1999).
[14] In some cases, lowering the symmetry—for example, by using a strained sample—may increase the degree of spin polarization.
[15] A. Haché, J. E. Sipe, and H. M. van Driel, IEEE J. Quantum Electron. **34**, 1144 (1998).
[16] P. Král and J. E. Sipe, Phys. Rev. B **61**, 5381 (2000).
[17] E. O. Kane, J. Phys. Chem. Solids **1**, 249 (1957).
[18] M. Sheik-Bahae, Phys. Rev. B **60**, R11 257 (1999).
[19] Symmetry arguments restrict $\dot{\mathbf{S}}_{(I)}$ in T_d crystals to lie in the x-y plane.
[20] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Halsted, New York, 1988), Chap. 2.
[21] It is not zero, however, as reported for the parabolic band approximation in [10].