One-photon and two-photon spectroscopy and spin polarization

M. Idrish Miah

[1]*Department of Physics, University of Chittagong, Chittagong 4331, Bangladesh.*
[2]*Department of Mathematics, Chittagong University of Engineering and Technology, Chittagong 4349, Bangladesh.*

Abstract

On the basis of a higher-band Kane model for the energy-band structure and a multi-order perturbation theory of the spin-polarized electronic transport model a theoretical investigation has been carried out for the optically generation of spins via both one-photon and two-photon spectroscopy in materials with a zinc-blende structure (zinc-blende semiconductors). As the optically excited hole spin relaxation is extremely fast, the calculation for the polarization gives effectively the electronic spin polarization. It is found that the electron spin polarization depends on the mode of the spectroscopy and the excitation photon energy. On comparison between two spectroscopic modes, it is clear that the multiphoton spectroscopy enhances ~ 10% of the electron spin polarization. The reason for the possibility of enhancing the effect is discussed.

1. Introduction

Spin-polarized electron transport or spin transport is an essential part of spin physics, or spintronics, where the electronic spin is utilized in an equal footing as the electronic charge. Reliable spin transport or spin transport without spin relaxation, or the loss of the spin polarization, over distances that are comparable to the device dimensions is required for practical spintronic devices. However, once an electron spin imbalance is injected into (or generated in) a semiconductor, electrons experience spin-dependent interactions with the environment, i.e. with impurities, defects and excitations or phonons, which causes spin relaxation [1].

Spin relaxation generally refers to the process which brings a nonequilibrium electronic spin population to a spin equilibrium state. Since this nonequilibrium electronic spin in metals and semiconductors is used to carry the spin-encoded information, which is one of the important steps towards applied spintronics and possible future quantum computation, it is important to know how long the spin can travel without losing its initial spin orientation, both in distance as well as time. The determination of spin-flip rate is extremely important for electronic applications, because if the spins relax too fast, the distance travelled by the spin-polarized current will be too short to serve any practical purpose.

Proper understanding the spins of electrons and their transport in semiconductors is the cornerstone to developing the next generation of quantum computers, spin-based micro- and nano-technologies, and the spin transistor (the building block for future electronic devices). Considerable advances in understanding the transport of spins in solid state environment and semiconductors have been achieved in the recent years [2-9]. However, the efficient spin injection, or the generation of highly polarized spins, for a reliable transport still remains a major problem in semiconductor spintronics.

Because of the availability of high intense tunably lasers, multiphoton spectroscopy has been used to study the multiphoton absorption of the optical nonlinear processes, particularly in semiconductors [10]. A two-photon spin-excitation in lead chacogenides has been studied, where they predicted a high spin-polarization in these cubic materials [11]. For GaAs, the similar results have been observed recently in an experiment [5]. In the present investigation, we study theoretically the spin generation in zinc-blende structured semiconductor GaAs via the both one-photon and two-photon optical excitations/spectroscopic techniques. In the calculation, a higher-band Kane model for the energy-band structure [12] and a multi-order (first- and second-order) perturbation theory of the spin-polarized electron transport are used, where the electro-electron (e-e) and electron-phonon (e-ph) interactions are ignored.

2. Eight band model

Band-structure calculations for the direct gap (E_g) semiconductors with zinc-blende structures show that the electron wave functions at the valence band (VB*)* maximum and conduction band (CB) minimum have *p*- and *s*-type symmetry respectively [13]. There is a magnetic interaction between the magnetic moments associated with the spin and the orbital motions of the electrons – called the spin-orbit interaction – which removes the spin degeneracy, since it produces an energy difference between the states for which the orbital and spin magnetic moments are parallel and antiparallel. For each value of *j*, the total angular momentum, there are $2j+1$ possible values of m_j. In case of a single electron there are always two values of $j = l \pm 1/2$ (orbital angular momentum of electron plus/ minus its spin). For an *s*-electron (*l*=0) the only possible value of *j* is 1/2, which has the two projection $m_j = 1/2, -1/2$, and for a *p*-electron (*l*=1) *j* can have the values $j = l \pm 1/2 = 3/2$ or 1/2. Thus, the CB (*s*-symmetry) is twofold degenerate at the centre of the Brilloun zone (*k* =0), corresponding to spin-up and spin-down electrons ($m_j = \pm 1/2$) and the VB (*p*-symmetry) at *k*=0 is split by spin-orbit interaction into heavy-hole (HH) and light-hole (LH) bands and a split-off band (SO). The HH and LH bands with large and small effective masses are each twofold spin degenerate ($m_j = \pm 3/2, \pm 1/2$). Therefore, spin-orbit interaction splits the sixfold degenerate *p* band at *k* =0 into a fourfold degenerate level and twofold degenerate level which lies $\triangle$ (spin-orbit splitting). Figure 1 shows the energy bands (eight bands) for GaAs at *k* = 0. The basis sets for the eight band model are given in Table 1.
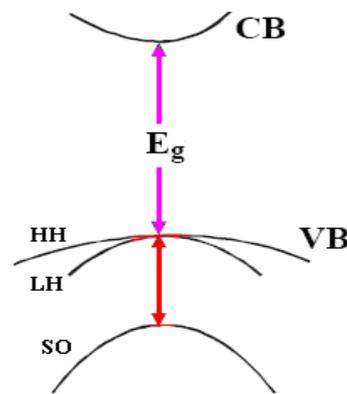
Figure 1. Eight band model: A scheme showing the heavy-hole (HH), light-hole (LH) and split-off (SO) bands of GaAs. The eight bands or states (counting one for each spin; up ↑ or down, ↓) comprise six *p*-like valence band (VB) states (the SO, HH and LH bands) and two *s*-like conduction band (CB) states.

Table 1. Bands, states $j,m_j>$ and their basis sets.

Band	$j,m_j>$	Basis set
CB	$1/2,1/2>$	$S{\uparrow}>$
CB	$1/2,-1/2>$	$S{\downarrow}>$
HH	$3/2,3/2>$	$(2)^{-1/2}(X+iY){\uparrow}>$
LH	$3/2,1/2>$	$-(6)^{-1/2}(X+iY){\downarrow} + (2/3)^{1/2} Z{\uparrow}>$
LH	$3/2,-1/2>$	$(6)^{-1/2}(X-iY){\uparrow} + (2/3)^{1/2} Z{\downarrow}>$
HH	$3/2,-3/2>$	$(2)^{-1/2}(X-iY){\downarrow}>$
SO	$1/2,1/2>$	$-(3)^{-1/2}(X+iY){\downarrow}- (3)^{-1/2} Z{\uparrow}>$
SO	$1/2,-1/2>$	$(3)^{-1/2}(X-iY){\uparrow}- (3)^{-1/2} Z{\downarrow}>$

3. Theory

The spin polarization due to two-photon spin generation has been calculated using the eight band Kane model in the limit of large spin-orbit splitting [13]. Here we estimate the electronic spin-polarization by calculating both the one-photon and two-photon photo-generation rates of electron spin densities using the perturbation theory of the spin transport model in the long wavelength limit, where the e-e and e-ph interactions are ignored. An illustration of the two-photon (one-photon) excitation scheme showing the pump pulse ω_2 (ω_1) coupling the initial and final states in the valance band (VB) and conduction band (CB) and the probe pulse

(ω_{Pr}) tuned to the band gap (E_g) excitation resonance ($\omega_{Pr} \approx E_g$) is shown in Fig. 1. For clarity, only the VB and CB are shown in Fig. 2
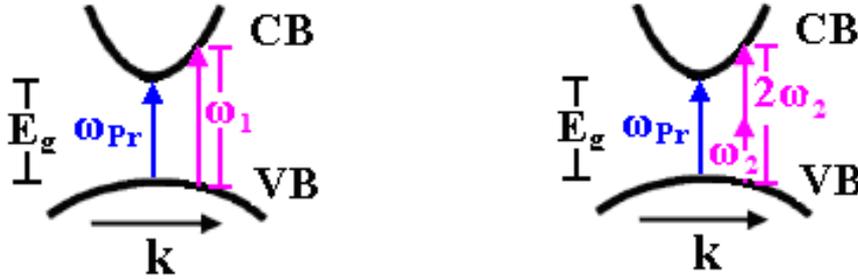
Figure 2. One-photon and two-photon spectroscopy: An illustration of the one-photon (left) and two-photon (right) excitation schemes showing the excitation pulse (ω_1 for one-photon and ω_2 for two-photon) coupling the initial and final states in the VB and CB bands and the probe pulse (ω_{Pr}) tuned to the band gap (E_g) excitation resonance. Here $E_1 = \hbar\omega_1$ is the one photon energy and $\omega_1 = 2\omega_2$. The two-photon energy is $E_2 = \hbar 2\omega_2 = E_1$ with a photon frequency of $\omega_2 = (1/2)\omega_1$ and an energy of $\hbar\omega_2$.

As optically excited hole spin relaxation is extremely fast, their polarization is effectively zero, and can be neglected. For an electric field :

$$\mathbf{E}(t) = \mathbf{E}_\omega (e^{-i\omega t} + e^{i\omega t}) \tag{1}$$

the two-photon spin generation rate can be written as:

$$dS^i / dt = \eta^{ijklm} E_\omega^j E_\omega^k E_\omega^{l*} E_\omega^{m*}, \tag{2}$$

where η^{ijklm} is a fifth rank pseudotensor symmetric on exchange of indices j and k, and on exchange indices l and m. The two-photon spin generation rate under the assumptions detailed above has been considered earlier for the doubly degenerate band case and is given as [14]:

$$d\mathbf{S} / dt = (2\pi / L^3) \sum_{c,c',v,\mathbf{k}} \left\langle c\mathbf{k} \left| \hat{\mathbf{S}} \right| c'\mathbf{k} \right\rangle C^{(2)*}_{c',v,\mathbf{k}} C^{(2)}_{c,v,\mathbf{k}} \delta\{2\omega - \omega_{cv}(\mathbf{k})\}, \tag{3}$$

where $\hat{\mathbf{S}}$ is the spin operator, L^3 is a normalized volume, $|n\mathbf{k}\rangle$ is a Bloch state with energy $\hbar\omega_n(\mathbf{k})$ and $C^{(2)}_{c,v,\mathbf{k}}$ is the two-photon amplitude given by :

$$C^{(2)}_{c,v,\mathbf{k}} = (e / \hbar\omega)^2 \sum_n \frac{\{\mathbf{E}_\omega . \mathbf{v}_{c,n}(\mathbf{k})\}\{\mathbf{E}_\omega . \mathbf{v}_{n,v}(\mathbf{k})\}}{\omega_{nv} - \omega(\mathbf{k})}. \tag{4}$$

Here $\mathbf{v}_{n,m}(\mathbf{k}) = \langle n\mathbf{k} | \hat{v} | m\mathbf{k} \rangle$ and $\hat{v}$ is the velocity operator. In the Fermi's golden rule, the photo-generation rate is time-independent and can be simplified if the spin-split bands are well-separated [15]. If the spin-split bands are well-separated, Fermi's golden rule gives $C^{(2)*}_{c',v,\mathbf{k}} C^{(2)}_{c,v,\mathbf{k}} = \left| C^{(2)}_{c,v,\mathbf{k}} \right|^2$ in Eq. (3). For GaAs, the spin-split pairs of bands should be treated as quasi-degenerate in Fermi's golden rule as the splitting is at most a few meV [20]. Thus:

$$d\mathbf{S} / dt = (2\pi / L^3) \sum_{c,c',v,\mathbf{k}} \left\langle c\mathbf{k} \left| \hat{\mathbf{S}} \right| c'\mathbf{k} \right\rangle C^{(2)*}_{c',v,\mathbf{k}} C^{(2)}_{c,v,\mathbf{k}} \times \frac{1}{2}[\delta\{2\omega - \omega_{cv}(\mathbf{k})\} + \delta\{2\omega - \omega_{c'v}(\mathbf{k})\}]. [5]$$

Similarly, the optical generation rate of electron-hole pairs can be obtained as :

$$dn / dt = (2\pi / L^3) \sum_{c,,v,\mathbf{k}} \left| C^{(2)}_{c,v,\mathbf{k}} \right|^2 \delta\{2\omega - \omega_{cv}(\mathbf{k})\}. \tag{6}$$

The two-photon energy is introduced by the frequency term "2ω" (=$2\omega_2$). For the generocity, the subscript 2 from ω is omitted). Defining $\eta_{2A} = -i\eta^{abccc}$ and $\eta_{2B} = \operatorname{Im}\eta^{aabac}$, the component of the spin generation rate along one of the cubic axes can be written as :

$$dS^i / dt = 2i \left(\mathbf{E}_\omega \times \mathbf{E}_\omega^* \right)^i \left[\eta_{2A} \left| \mathbf{E}_\omega \right|^2 + (2\eta_{2B} - \eta_{2A}) \left| \mathbf{E}_\omega^i \right|^2 \right]. \tag{7}$$

Here the indices *a*, *b*, and *c* denote components along the standard cubic axes [100], [010], and [001]. For a cubic isotropic material, the spin generation rate can be described by only one real parameter ($2\eta_{2B} = \eta_{2A}$) and thus :

$$dS^i / dt = 2i\left(\mathbf{E}_\omega \times \mathbf{E}_\omega^*\right)^i \left[\eta_{2A}\left|\mathbf{E}_\omega\right|^2\right]. \tag{8}$$

It should be noted that the cubic anisotropy means that the two-photon spin generation from circularly polarized light (CPL) depends on the angle of incidence of the light relative to the cubic axes [15]. For CPL incident along $\hat{\mathbf{z}}$ specified by polar angles θ and φ relative to the cubic axes,

$$d\mathbf{S} / dt \cdot \hat{\mathbf{z}} = \mp\eta_{2A}\left|\mathbf{E}_\omega\right|^4 \left(1 + \frac{2\eta_{2B} - \eta_{2A}}{4\eta_{2A}}\xi(\theta,\varphi)\right), \tag{9}$$

where the upper, − (lower, +) sign is the excitation for right, σ^+ (left, σ^-) circularly light:

$$\sigma^+ = (\hat{x} + i\hat{y}) / \sqrt{2}$$
$$\sigma^- = (\hat{x} - i\hat{y}) / \sqrt{2}$$

and

$$\xi(\theta,\varphi) = \sin^2(2\theta) + \sin^4(\theta)\sin^2(2\varphi). \tag{10}$$

The $\xi(\theta,\varphi)$ term is an important function which shows the cubic anisotropy in pumping, i.e. the angular dependence of pumping in spin-excitation in the cubic system. The term $\xi(\theta,\varphi)$ as a function of θ and φ is plotted in Fig. 3.
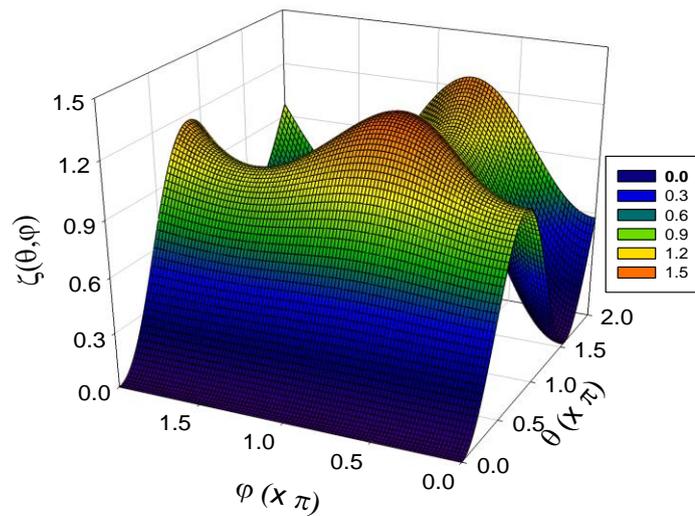
Figure 3. The term $\xi(\theta,\varphi)$ as a function of θ and φ.

This feature can be seen in the figure. Owing to the cubic anisotropy, the net generated spin is not always parallel to $\hat{\mathbf{z}}$. For example, for light along a <001> direction, $\left|d\mathbf{S}/dt\right| = 2\eta_{2A}\left|E_\omega\right|^4$, while for light incident along a <111> direction, $\left|d\mathbf{S}/dt\right| = (4/3)(\eta_{2A} + \eta_{2B})\left|E_\omega\right|^4$. Since hole spin polarization in bulk semiconductors is known to relax very fast, on a longer time scale one typically obtains only the electron spin polarization. The electronic spin polarization (P) can be estimated as:

$$P = -\left(\frac{2}{\hbar}\right)\frac{d\mathbf{S}/dt \cdot \hat{\mathbf{z}}}{dn/dt}, \tag{11}$$

where *P* is defined as $P = (n_\downarrow - n_\uparrow)/(n_\downarrow + n_\uparrow)$. Here the spin is opposite to the photon angular momentum (generated with a σ^+ light beam) and $n_\downarrow (n_\uparrow)$ is the density of spin-down (spin-up) electrons. The electronic spin-polarization due to one-photon excitation can also be obtained by replacing 2ω ($(=2\omega_2)$ by ω $(=\omega_1)$ and

the two-photon amplitude $C_{c,v,\mathbf{k}}^{(2)}$ by the one-photon amplitude $D_{c,v,\mathbf{k}}^{(1)}$. The approach can be applied for a wide class of semiconductors.

4. Results and discussion

Spin polarization as a function of one-photon and two-photon excitation energies was calculated in the long wavelength limit of the multi-order perturbation theory using values of the parameters for GaAs [16]. The assumption of a long wavelength limit is chosen to neglect both the e-e and e-ph interactions. Calculated results (solid lines) are shown in Figs. 4 and 5, where the spin polarizations are plotted as a function of the excess excitation photon energy for the one-photon (ΔE_ω) and two-photon ($\Delta E_{2\omega}$) excitations:

$$\Delta E_\omega = \hbar\omega_1 - \hbar\omega_{\mathrm{Pr}} \square \hbar\omega_1 - E_g = E_1 - E_g \tag{12}$$

$$\Delta E_{2\omega} = 2\hbar\omega_2 - \hbar\omega_{\mathrm{Pr}} \square 2\hbar\omega_2 - E_g = \hbar\omega_1 - E_g = E_1 - E_g . \tag{13}$$

Here $E_1 = \hbar\omega_1$ and $\omega_1 = 2\omega_2$. Symbols in the figures show the data taken from one-photon [17] and two-photon [5] spectroscopic experiments. On comparison, over the whole range of the excitation photon energy, a good agreement between experiment and theory is achieved.
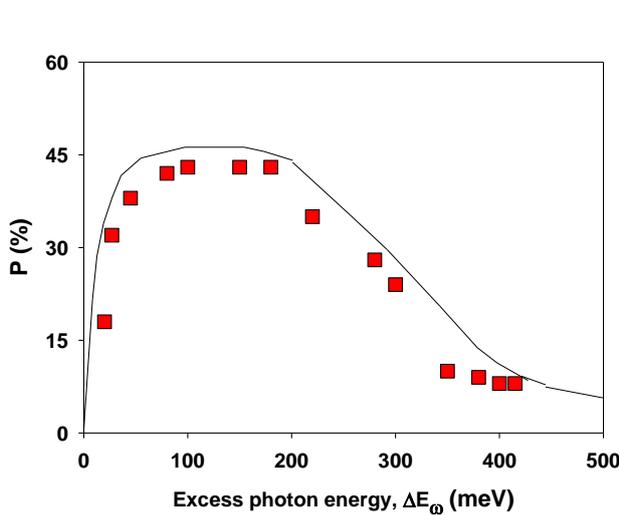
Figure 4. Spin-polarization as a function of one-photon energy (excess photon energy, $\Delta E_\omega \square \hbar\omega - E_g = E_1 - E_g$): experimental (symbol square) and theoretical values (black solid line).
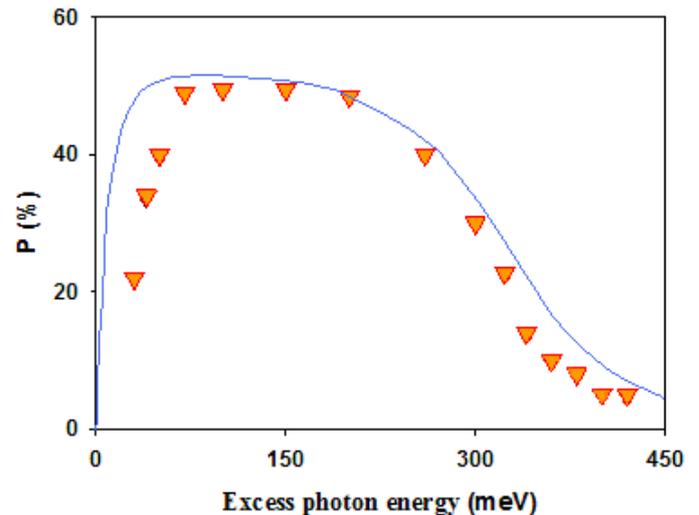
Figure 5. Spin-polarization as a function of two-photon energy (excess photon energy, $\Delta E_{2\omega} \square 2\hbar\omega_2 - E_g = E_1 - E_g$): experimental (down triangle) and theoretical values (blue solid line). Here $E_1 = \hbar\omega_1$ and $\omega_1 = 2\omega_2$.*

As expected, the spin polarization increases with excitation photon energy for low excess energy and remains almost constant up to ~200 meV, and then decreases. However, the maximum is obtained for the excess energy considerably less than the SO splitting energy of 0.340 eV for GaAs. The maximum value is ~50% for two-photon excitation. For the higher excess energy, the polarization decreases rapidly due to the mixture of LH and HH states with the SO valence band states which have an opposite sign. The LH and SO band transitions create the same electron spin orientation and the sum of their inter-band matrix elements is equal to the HH inter-band dipole-transition matrix element. There is still a sizeable degree of electron spin polarization even at excess energy higher than the spin-orbit splitting energy.

Although the maximum optical spin-polarization for an unstrained bulk sample is expected to be 50% in theory [15], the maximum has experimentally been observed to be less, as obtained spectroscopically with a single-photon excitation. In a bulk sample there might have some background unpolarized electrons, which might not be excited by single-photon excitation [18]. For an optically generated electron density $n(0)=n_\downarrow(0)+n_\uparrow(0)$, there is a background density of unpolarized electrons in bulk materials.

On a comparison between Figs. 4 and 5, it can be seen that a two-photon excitation enhances the spin polarization. This is because it takes the advantages over one-photon spin spectroscopy due to a much longer absorption depth, which allows spin excitation in the deep level, i.e. throughout the volume of a thin bulk sample.

It thus can be concluded that due to a much longer absorption depth highly spin-polarized electrons can be produced optically by the two-photon excitation of the bulk semiconductors. However, in a zinc-blende semiconductor, like GaAs, the spin polarization would decay with time due to the randomization of the initial spin polarization by the Dyakonov-Perel (DP) spin relaxation mechanism [1]. The DP spin relaxation occurs in semiconductors lacking inversion symmetry due to the spin precession about an intrinsic magnetic field induced by the presence of the spin-orbit interactions in a zinc-blende structure. However, the observed two-photon absorption is might be related to the excited e-ph interaction which is ignored in the present calculation.

Conclusion

An investigation for the optical spin polarization via both one-photon and two-photon spectroscopy in zinc-blende structured GaAs was carried out using a higher-band Kane model for the energy-band structure and a multi-order perturbation theory of the spin-polarized electronic transport. The electronic spin polarization for both spectroscopic modes was calculated as a function of circularly polarized excited photon energy. The polarization was found to be preserved within the excess photon energy of ~100 meV. However, it depolarized for the excitation photon energy equals to or larger than the energy gap of the SO band to the CB. On a comparison between two spectroscopic modes, it can be concluded that a two-photon spectroscopy enhances (~10%) the electron spin polarization over a single- or one-photon. The results in comparison between two spectroscopic modes are discussed. The results, however, suggest a possible way of enhancing the electronic spin polarization in semiconductors.

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