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THEORETICAL STUDIES OF OPTICALLY INDUCED HIGH SPIN POLARIZATION AND CUBIC ANISOTROPY IN CUBIC SEMICONDUCTORS

Mohammad Idrish Miah*
Department of Physics, University of Chittagong, Chittagong–4331, Bangladesh
*m.idrish.miah@cu.ac.bd

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ABSTRACT

Theoretical studies of optical spin polarization and cubic anisotropy in cubic semiconductors GaAs and GaSb are performed. We use Kane wave functions to calculate the eight-level transition matrix elements permitted by the optical selection rules. It is found that the spin polarization depends on semiconductor, the pumping direction as well as the photon energy. A high spin polarization of about 55% is obtained for GaSb. The anisotropic effect in these cubic crystals is clearly seen. The band edge polarization is found to be flatter and higher for GaAs than that for GaSb. The results are discussed in comparison with the experimental data available in the literature.

Keywords: Semiconductors; Multiphoton absorption; Spin generation.

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INTRODUCTION

Much interest in spin transport in semiconductors has been observed recently [1-5], after a long wait since its fist study (now recognized as the first prediction of the spin Hall effect, or the Dyakonov and Perel (DP) spin relaxation mechanism in semiconductors in 1971 [6]. This interest was accelerated by the observation of the long spin lifetime [3,7]. Considerable interest in spin transport in semiconductors emerges a new subfield in condensed matter physics called spintronics or spin electronics [5,8], or spin physics in broader sense [2], aiming at realizing high performance devices where carriers' spin may play an active role. However, one of the important requirements necessary in developing these devices is the transporting spin polarized carriers reliably over reasonable distances, i.e. without spin polarization or spin relaxation. This requires to inject, create/generate a high spin polarization is semiconductors. Therefore, spintronic researchers are looking for possible methods for enhancing the spin polarization. Of them, the electrical method by a ferromagnetic coupling shows low spin injection efficiency due to a conductivity mismatch between the ferromagnet and semiconductor [4]. On the other hand, the optical method by a circularly polarized is successful [9-10].

As an optical method, the single photon spectroscopy has widely been used [11-13]. Recently, multiphoton spin spectroscopy also used to study the multiphoton absorption in optical nonlinear processes of indirect bandgap semiconductors [14]. Particularly, a two-photon spin-excitation in lead chacogenides has been studied, where they predicted a high spin-

polarization in these cubic materials [15]. For GaAs, the similar results have been observed recently in an experiment [8]. In the recent investigation, we study two-photon spin generation GaAs both theoretically and experimentally [16]. Here, in the present investigation, we study the electronic spin generation, polarization, and cubic anisotropy in cubic semiconductors GaAa and GaSb based on the eight-band model. It is found that the electron spin polarization depends on the pumping direction as well as photon energy and that the band edge polarization is flatter and higher for GaAs than that for GaSb.

METHOD

Spin generation

Semiconductors like GaAs have the zinc-blende structures, in which the lattice consists of two interpenetrating fcc lattices, one occupied by Ga and the other by As. Ions. We know from both experiment and band-structure calculations for the direct gap (E_g) semiconductors that the electron wave functions at the valence band (VB*)* maximum and conduction band (CB) minimum have, respectively, *p*- and *s*-type symmetry respectively. At Γ, where k =0, the symmetry of the point group is not reduced. For the optical spin generating process the important requirement is that the spin-orbit interaction does split the six-fold degenerate *p* band at Γ into a four-fold degenerate $P_{3/2}$ level and two-fold degenerate $P_{1/2}$ level (Figure 1).

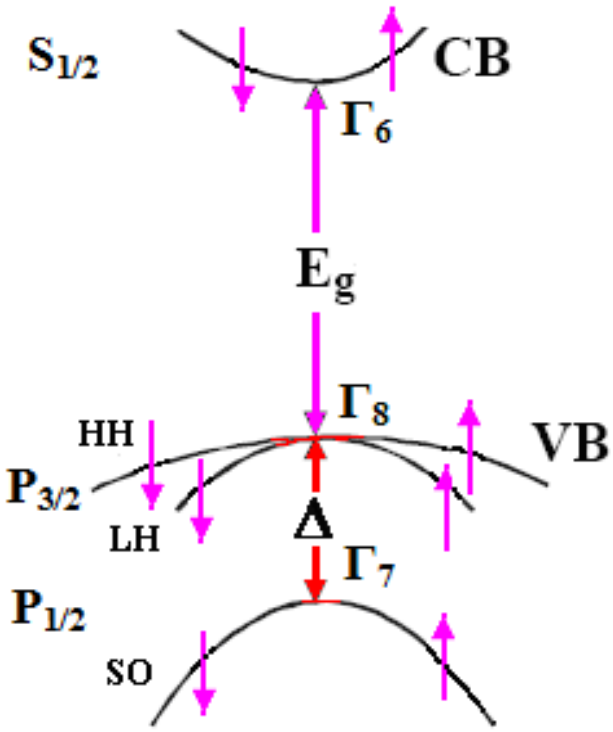

Figure 1. A scheme showing the heavy-hole (HH), light-hole (LH) and split-off (SO) bands of a semiconductor. One band, two bands and one band at symmetry points Γ_6, Γ_7 and Γ_8 respectively. Total eight band-states (counting each spin state: spin up ↑ or spin down, ↓) comprise six *p*-like valence band (VB) states (HH, LH and SO bands) and two *s*-like conduction band (CB) states. The $S_{1/2}$, $P_{3/2}$ and $P_{1/2}$ levels are shown on the right.

The CB (*s*-symmetry) is twofold degenerate $S_{1/2}$ at the centre of the Brilloun zone (k =0), corresponding to spin-up and spin-down electrons and the VB (*p*-symmetry) at k=0 is slit 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 mass are each twofold spin degenerate.

For a spin polarized electron system, if $n_\uparrow$ and $n_\downarrow$ are the numbers of spin up and spin down electrons, the degree of spin polarization is calculated from the relation

$$P(\vec{r},t)=\frac{n_\uparrow(\vec{r},t)-n_\downarrow(\vec{r},t)}{n_\uparrow(\vec{r},t)+n_\downarrow(\vec{r},t)} \qquad (1)$$

According to the optical selection rules, the transition rate from the HH states to CB is three times larger than from the LH states. As the LH transition generates opposite spins, the optical excitation produces a 3:1 ratio of to spin-up to spin-down CB electrons for a left/right circularly polarized light provided that the photon energy is low enough to avoid exciting carriers from the split-off band. The straightforward calculation based on this gives a 50% spin polarization in bulk semiconductors [11]. Optical excitation with a σ^+(σ^-) orients spins along the direction parallel (antiparallel) to the direction of the light propagation. A 50% spin polarization has not yet been obtained by an optical experiment in a bulk material. However, recent studies with multiphoton excitation and full-band calculation show that more than 50% spin polarization could be achieved in some materials [8,16].

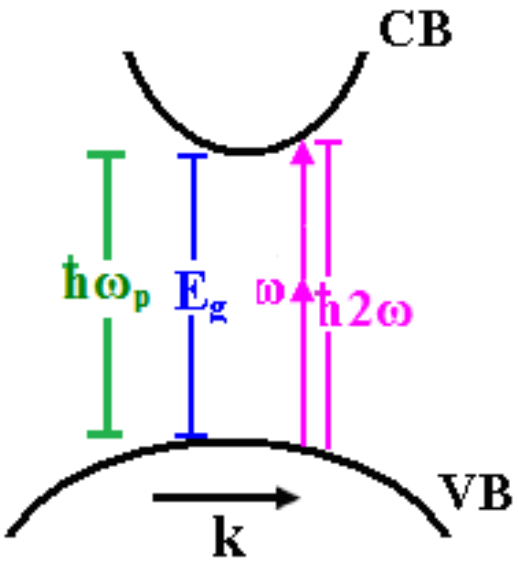

Figure 2. Two-photon spectroscopy: An illustration of the two-photon absorption schemes showing the excitation pulse for the two-photon (2ω) coupling the initial and final states in the VB and CB bands. The probe pulse (ω_p) tuned to the band gap (E_g) excitation resonance.

As a multiphoton process, an illustration of the two-photon excitation schemes showing the excitation pulse for two-photon (2ω) coupling the initial and final states in the VB and CB bands is given in Figure 2, where $E_{2\omega}=\hbar 2\omega$ and $E_p=\hbar\omega_p$ are the two-photon and pump

beam energies, respectively. The probe pulse (ω_p) tuned to the band gap (E_g) excitation resonance ($\omega_p \approx E_g$)

Calculations

We estimate the electronic spin-polarization by calculating the both one-photon and two-photon photo-generation rate of electron spin density ($d\mathbf{D}/dt$) using the perturbation theory in the long wavelength limit, where electron-electron and electron-phonon interactions are ignored. As optically excited hole spin relaxation is extremely fast, their polarization is effectively zero, and can be neglected. For an electric field ***E*** the two-photon spin generation rate can be written as $dD^i/dt = \eta^{ijklm} E_\omega^j E_\omega^k E_\omega^{l*} E_\omega^{m*}$, 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 [16,17]

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

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

$$\mathrm{C}^{(2)}_{\mathrm{c,v,k}} = \left(\frac{e}{E_\omega} \right)^2 \sum_n \frac{\{\mathbf{E}_\omega . \mathbf{v}_{c,n}(\mathbf{k})\}\{\mathbf{E}_\omega . \mathbf{v}_{n,v}(\mathbf{k})\}}{\omega_{nv} - \omega(\mathbf{k})} \tag{3}$$

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 [18]. If the spin-split bands are well-separated, Fermi's golden rule gives $\mathrm{C}^{(2)*}_{\mathrm{c',v,k}} \mathrm{C}^{(2)}_{\mathrm{c,v,k}} = \left| \mathrm{C}^{(2)}_{\mathrm{c,v,k}} \right|^2$ in equation 4. 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 [17]. Thus

$$\frac{d\mathbf{D}}{dt} = \frac{2\pi}{V} \sum_{c,c',v,\mathbf{k}} \left\langle c\mathbf{k} \left| \hat{\mathbf{D}} \right| c'\mathbf{k} \right\rangle \mathrm{C}^{(2)*}_{\mathrm{c',v,k}} \mathrm{C}^{(2)}_{\mathrm{c,v,k}} \times \frac{1}{2} [\delta\{2\omega - \omega_{cv}(\mathbf{k})\} + \delta\{2\omega - \omega_{c'v}(\mathbf{k})\}] \tag{4}$$

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

$$\frac{dn}{dt} = \frac{2\pi}{V} \sum_{c,,v,\mathbf{k}} \left| \mathrm{C}^{(2)}_{\mathrm{c,v,k}} \right|^2 \delta\{2\omega - \omega_{cv}(\mathbf{k})\} \tag{5}$$

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

$$\frac{dD^i}{dt} = 2i \left(\mathbf{E}_\omega \times \mathbf{E}_\omega^* \right)^i \left[\eta_{2A} \left| \mathbf{E}_\omega \right|^2 + (2\eta_{2B} - \eta_{2A}) \left| \mathbf{E}_\omega^i \right|^2 \right] \tag{6}$$

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

$$\frac{dD^i}{dt} = 2i\left(\mathbf{E}_\omega \times \mathbf{E}_\omega^*\right)^i \left[\eta_{2A}\left|\mathbf{E}_\omega\right|^2\right] \tag{7}$$

It should be noted that the cubic anisotropy means that the two-photon spin generation from circularly polarized light depends on the angle of incidence of the light relative to the cubic axes [19-21]. For the light incident along $\hat{\mathbf{z}}$ specified by polar angles θ and φ relative to the cubic axes,

$$\frac{d\mathbf{D}\cdot\hat{\mathbf{z}}}{dt} = \mp\eta_{2A}\left|\mathbf{E}_\omega\right|^4\left(1+\frac{2\eta_{2B}-\eta_{2A}}{4\eta_{2A}}a_p(\theta,\varphi)\right) \tag{8}$$

where the upper, – (lower, +) sign is the excitation for $\boldsymbol{\sigma}^+ = (\hat{x}+i\hat{y})/\sqrt{2}$ ($\boldsymbol{\sigma} = (\hat{x}-i\hat{y})/\sqrt{2}$) light and $a_p(\theta,\varphi) = \sin^2(2\theta)+\sin^4(\theta)\sin^2(2\varphi)$.The term $a_p(\theta,\varphi)$ 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.

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, $\left|d\mathbf{D}/dt\right| = 2\eta_{2A}\left|E_\omega\right|^4$, while for light incident along a [111] direction, $\left|d\mathbf{D}/dt\right| = (4/3)(\eta_{2A}+\eta_{2B})\left|E_\omega\right|^4$. 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*, as defined in equation 1, can be estimated as

$$P = -\left(\frac{2}{\hbar}\right)\frac{d\mathbf{D}\cdot\hat{\mathbf{z}}}{dt}\Big/\frac{dn}{dt} \tag{9}$$

For the calculations of the spin polarization, we calculate the interband transition matrix elements using the eight-band model, as shown in Figure 1. Kane wave functions [22] for CB and VB bands in the **k.p** approximation are

$$\varphi_{i\alpha} = a_i S_\downarrow + b_i\left\{\frac{(X-iY)_\uparrow}{\sqrt{2}}\right\} + c_i Z_\downarrow \tag{10}$$

$$\varphi_{i\beta} = a_i S_\uparrow + b_i\left\{-\frac{(X+iY)_\downarrow}{\sqrt{2}}\right\} + c_i Z_\uparrow \tag{11}$$

where *i* takes the values 1, 3 and 4 to give the wave functions for the CB, LH and SO bands respectively, and for the HH band (Γ_8 point) takes the value of 2 in the following equations

$$\varphi_{i\alpha} = \frac{(X+iY)_\uparrow}{\sqrt{2}} \tag{12}$$

$$\varphi_{i\beta} = \frac{(X-iY)_\downarrow}{\sqrt{2}} \tag{13}$$

The basic functions *X*, *Y*, *Z* and *S* refer to a coordinate system depending on **k**. The coefficients *a*, *b* and *c* also depends on **k**, and for k =0 they reduces to the following set of values (a = 1, b = 0, c = 0), (a = 0, $b=\sqrt{2/3}$, $c=-1/\sqrt{3}$) and (a = 0, $b=1/\sqrt{3}$, $c=\sqrt{2/3}$) for CB band (Γ_6 point), SO band (Γ_7 point) and LH band (Γ_8 point) respectively. The parameter-values used for this calculation are taken from[23] and are given in the Table 1.

Table 1: Parameter-values used for calculation.

		GaAs	GaSb
E_g (eV)	Energy between Γ_{6C} and Γ_{8V} bands	1.519	0.813
E_0 (eV)	Energy between Γ_{7C} and Γ_{8V} bands	4.488	3.300
Δ (eV)	Energy between Γ_{8V} and Γ_{7V} bands	0.341	0.750
Δ’ (eV)	Energy between Γ_{8C} and Γ_{7C} bands	0.171	0.330
Δ^- (eV)	Interband SO coupling	-0.061	-0.280
ζ_{1L}	Luttinger parameter	7.797	13.200
ζ_{2L}	Luttinger parameter	2.458	4.400
ζ_{3L}	Luttinger parameter	3.299	5.700
Q_k (meV Å)	*k*-linear term	-3.400	0.430

RESULTS AND DISCUSSION

Electron spin polarization as a function of excess photon energy for multiphoton excitations was calculated for semiconductors GaAs and GaSb. The calculated results are shown in Figure 3, where the spin polarization *P* is plotted as a function of the excess photon energy. As expected, the spin polarization increases with the excess photon energy for the lower excess energy and remains almost constant up to ~150 meV and then decreases. The maximum value of *P* is ~55% for GaSb. However, for GaAs, *P* is about 50%. 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. The results agree well with those for GaAs [16].

Although the maximum optical spin-polarization for an unstrained bulk sample is expected to be much more in the present theory, the maximum has experimentally been observed to be less because of the unexciting carriers in the bottom of a bulk. In a bulk sample there might have some background unpolarized electrons, which might not be excited by single-photon excitation [24]. For an optically generated electron density there is a background density of unpolarized electrons in bulks. However, *P* obtained by a single-photon pumping is much less [9].

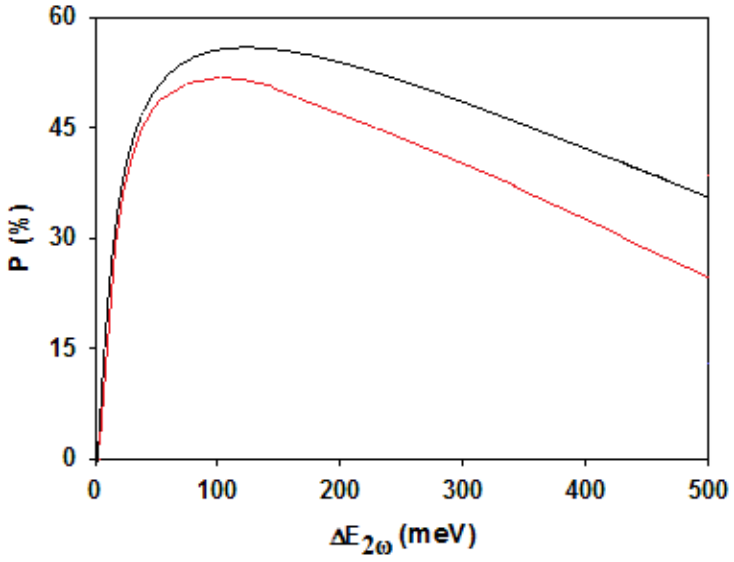

Figure 3. Dependence of spin polarization on the photon energy excess to the bandgap for GaSb (upper curve) and GaAs (lower curve) and for the direction [111].

However, the spin polarization would decay due to the randomization of the initial spin polarization by the DP spin relaxation mechanism [2,6]. The DP spin relaxation occurs in cubic and noncentrosymmetric semiconductors lacking inversion symmetry due to the spin precession about an intrinsic effective magnetic field induced by the presence of spin-orbit (SO) in a zinc-blende structure. This effective magnetic field, $\mathbf{h}_{\text{eff}} = \kappa\{k_x(k_y^2 - k_z^2)\hat{x} + cp\}$ (*cp* stands for cyclic permutation), originated from the SO interaction $\mathrm{H}_{SO} = (\hbar/2)\,\boldsymbol{\sigma}\cdot\mathbf{h}_{\text{eff}}$ [25], is a vector depending on the orientation of the electron momentum vector with respect to the crystal axes xyz and causes the electron spin to process with the equation of motion $d\boldsymbol{\sigma}/dt = \mathbf{h}_{\text{eff}} \times \boldsymbol{\sigma}$. Here κ is the SO coupling coefficient and is given by

$$\kappa = \frac{\xi\hbar^2}{\sqrt{2(m^*)^3 E_g}}\left\{1 - \frac{E(\mathbf{k})}{E_g}\frac{9 - 7\gamma + 2\gamma^2}{3 - \gamma}\right\}, \tag{14}$$

where m^* is the electron effective mass, ξ *is* a dimensionless material-specific parameter which gives the magnitude of the SO splitting and $\gamma = \Delta/(E_g + \Delta)$. The scattering changes the direction of the procession axis, which leads to spin dephasing or relaxation.

Figure 4 shows the dependence of spin polarization on the direction of the light excitation relative to the cubic axis of the crystals for GaAs and GaSb. As can be seen, spin polarization increases from the direction [001] to a direction [111] by an amount of about 5%, and the maximum spin polarizations are about 50% and 55% for GaAs and GaSb, respectively, for the direction [111]. As expected, a pumping in the random direction [---] gives an average.

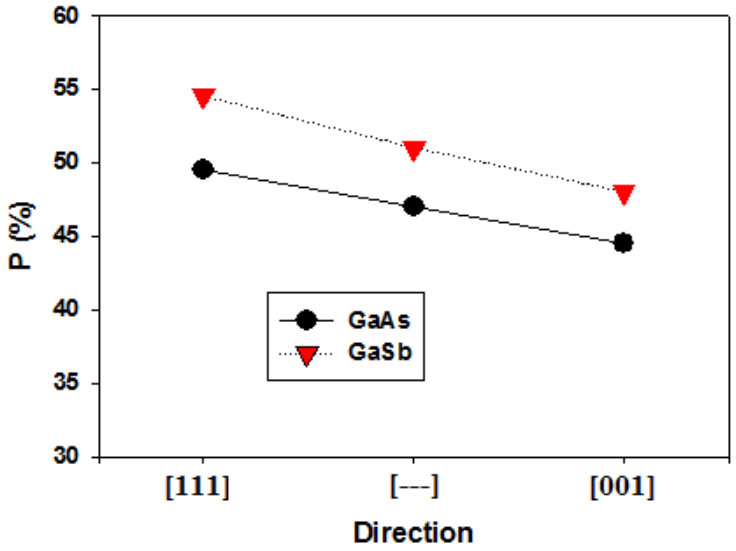

Figure 4: Dependence of spin polarization on the direction of the light excitation relative to the cubic axis of the crystals, where [---] denotes the random direction.

The dependence of the average spin polarization on the excess photon energy within 100–200 meV for the random direction is shown in Figure 5. As available for GaAs [16], an experimental trend for GaAs is also shown for the comparison with the theoretically obtained data. As can be seen, a good agreement between theory and experiment is obtained. The trend is almost the same, but theoretical polarization is about 3% higher than the experiment value. On a comparison with the single photon excitation [9], a two-photon excitation enhances the spin polarization significantly. This is because the two-photon excitation takes the advantages over one-photon spin spectroscopy due to a much longer absorption depth [26], which allows spin excitation in the deep level (i.e. throughout the volume of a thin bulk sample).

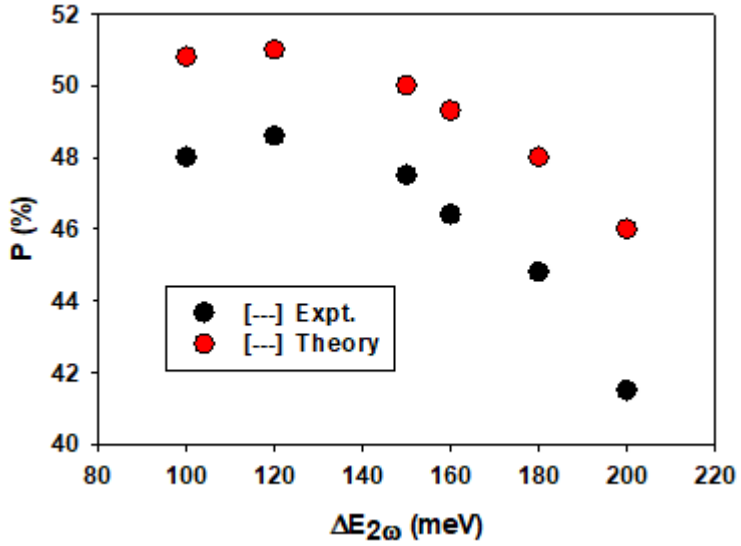

Figure 5 Dependence of the average spin polarization on the excess photon energy within 100-200 meV for the random direction [---]. An experimental trend for GaAs compares the theoretically obtained data.

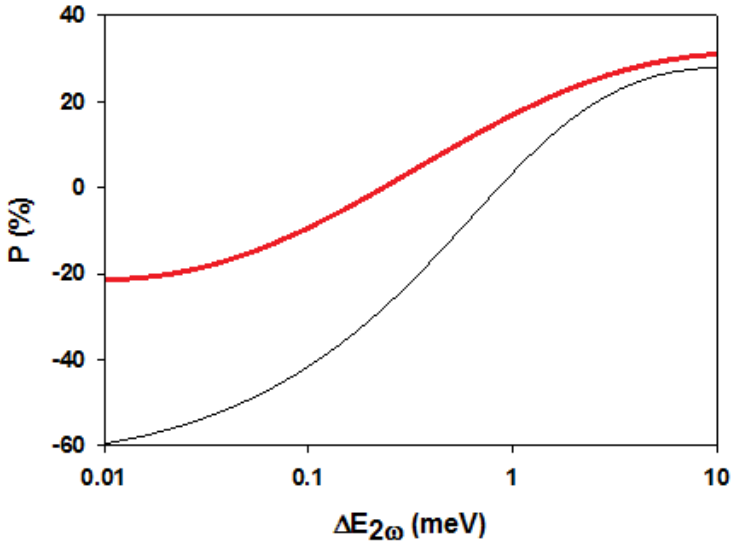

Figure 6. Dependence of the average spin polarization near the band edge for GaSb (black/lower curve) and GaAs (red bold/upper curve).

Figure 6 shows the dependence of the average spin polarization near the band edge for GaSb and GaAs. There is a feature of small photon energy dependence, which can not be seen in Figure 3. As can be seen here, the band edge polarization is flatter and higher for GaAs than that for GaSb. This is because of the different in their band structures and band energies. The band edge polarization was also found to depend on the anisotropy of pumping or crystal orientations. The results are consistence with those obtained in other investigations [27-28].

CONCLUSIONS

Studies of optical spin polarization and cubic anisotropy in cubic semiconductors GaAs and GaSb were theoretically performed. For the calculations, we used Kane wave functions for the eight-level energy bands. From the calculated results it was found that the spin polarization depends on the pumping direction and photon energy. A maximum 55% spin polarization was obtained for achieved. The anisotropic effect in these cubic crystals was also observed. The band edge polarization was found to be flatter and higher for GaAs than that for GaSb. The results were discussed in comparison with the experimental data available in the literature and based on the spin relaxation mechanism.

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