Asymmetries of spin-flip electronic Raman scattering in a III-V semiconductor quantum well

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We have calculated the angular and the polarization dependence of spin-flip electronic Raman scattering from a III-V semiconductor quantum well with its conduction band spin split due to broken inversion symmetry. We found that the interference of light scattered from the longitudinal and transverse spin-density fluctuations leads to a dependence of the Raman spectrum on the direction of circular polarization of photons. This phenomenon at zero magnetic field is entirely due to the intrinsic electron spin dynamics in the spin-split band. The predicted asymmetric polarization dependence is preserved when the sample becomes dirty, such that the elastic-electron scattering rate exceeds the characteristic frequency of electron spin precession in the spin-split band. [S0163-1829(97)50204-9]

In semiconductors of zinc-blende structure, the absence of inversion symmetry lifts the spin degeneracy and splits the conduction band.[1] The electron energy split $h(\vec{k})=|\vec{h}(\vec{k})|$ depends on the direction of the wave vector $\vec{k}$, and grows as k^3 with increasing electron energy.[2] In bulk semiconductors, the amount of split has been deduced from optical orientation measurements,[1] and was found rather small. However, in narrow quantum wells, due to the confinement of electron motion along the growth direction, k can be sufficiently large to yield[3] a larger $h(\vec{k})$, which has been derived from the magnetoresistance in weak magnetic fields,[4] from the Shubnikov–de Haas oscillations,[5] and from the Raman spectra.[6]

The resonant Raman scattering is an effective tool for studying the electronic excitations in bulk semiconductors and in semiconductor microstructures.[7] In the depolarized geometry where the polarization of the incident light is perpendicular to that of the scattered light, the spin-flip Raman scattering detects the electron spin-density fluctuations, and the spin splitting of the conduction band shows up in the Raman spectrum as low-energy peaks, which correspond to the transitions between pairs of spin-split electron states. Measuring the positions of these peaks at different sample orientations and at different wave-vector transfers, one can determine the magnitude of the spin splitting and the structure of spin-split bands. Jusserand *et al.*[6] were the first to observe the spin splitting in the low-frequency spin-flip electronic Raman spectrum of GaAs/Al_xGa_{1-x}As quantum wells.

However, in this paper we will show that Raman scattering can provide additional information about the dynamics of electrons in the spin-split bands. This is due to the phases of electron spin excitations, the effect of which manifests itself in the interference of light inelastically scattered from different spatial components of the spin-density fluctuations. The contribution of this interference to the Raman cross section can be observed only if the incident and the scattered photon are circularly polarized. Furthermore, for a nongyrotropic material in the absence of a magnetic field, this observable interference term changes sign when the directions of circular polarizations are reversed. To demonstrate our theoretical finding, we will calculate the intensity of the electronic intrasubband spin-flip Raman scattering from a degenerate two-dimensional (2D) electron gas in a narrow quantum well in which only the lowest subband is occupied. Two limiting cases will be studied in detail: the low and the high mean elastic scattering rate of electrons as compared to $h(\vec{k})/\hbar$. In both cases the Raman scattering is asymmetric with respect to the right and the left circular polarization of photons.

For the convenience of mathematical presentation, we set $\hbar=1$. The electronic states in the lowest subband are specified as $|\vec{k},\alpha\rangle$, where $\vec{k}$ is the 2D wave vector and α is the spin projection onto the z axis which is along the growth direction. Let $\vec{e}_i$ (or $\vec{e}_s$) be the polarization vector of the incident (or scattered) photon, and $\vec{P}\equiv\vec{e}_i\times\vec{e}_s^{\,*}$ According to Hamilton and McWorter,[8] the spin-flip quantum amplitude of scattering a photon from the initial photon state $(\omega_i,\vec{k}_i)$ to the final photon state $(\omega_s,\vec{k}_s)$ can be expressed as

$$A_{\alpha\beta}(\vec{k},\vec{q})=\gamma\vec{P}\cdot\langle\vec{k}+\vec{q},\alpha|\vec{S}_q|\vec{k},\beta\rangle, \tag{1}$$

where $\vec{q}=(\vec{k}_i-\vec{k}_s)_\parallel$ is the component of $\vec{k}_i$-$\vec{k}_s$ parallel to the xy plane, and $\vec{S}_q$ is the corresponding Fourier component of the electron spin-density operator.

The matrix elements of $\vec{S}_q$ are characteristic to the spin dynamics of electrons in the spin-split conduction band of a III-V semiconductor described by the Hamiltonian[2]

$$H=E_k+\vec{h}(\vec{k})\cdot\vec{s}, \tag{2}$$

where $E_k=k^2/2m^*$. The second term in Eq. (2) has the form of an electronic spin $\vec{s}$ interacting with an effective *magnetic* field $\vec{h}(\vec{k})$ whose direction and magnitude vary with the electron wave vector. The explicit expression of $\vec{h}(\vec{k})$ depends on the crystallographic orientation of the quantum well system. If the growth direction z axis is along the [001] axis, then we have[3]

$$h_x(\vec{k})=\alpha k_x(k_y^2-\kappa^2), \quad h_y(\vec{k})=\alpha k_y(-k_x^2+\kappa^2), \tag{3}$$

where κ^2 is the expectation value of the operator $(i\partial/\partial z)^2$ with respect to the confined wave function along the [001] axis. If we choose this axis as the spin quantization axis, from Eq. (2) we easily derive the two spin-split subbands (SSSB's) as

$$E_{\pm,\vec{k}}=E_k\pm|\vec{h}(\vec{k})|/2. \tag{4}$$

The corresponding spin-dependent part of the eigenfunctions are

$$\psi_{\pm,\vec{k},\uparrow}=\frac{1}{\sqrt{2}}e^{-i\phi_k/2}, \quad \psi_{\pm,\vec{k},\downarrow}=\pm\frac{1}{\sqrt{2}}e^{i\phi_k/2}, \tag{5}$$

where ϕ_k is the angle between the vector $\vec{h}(\vec{k})$ and the x axis. The subscripts $\uparrow$ (or $\downarrow$) indicate that the projection of the electronic spin onto the z axis, s_z, is 1/2 (or $-1/2$).

Let us first consider very clean materials in which the elastic scattering time is sufficiently long that the corresponding broadening of electron energy is small compared to $h(\vec{k})$ and to $v_f q$, where v_f is the Fermi velocity. In this case the scattering cross section at the Stokes shift $\omega=\omega_i-\omega_s$ is proportional to

$$W(\omega,\vec{q})=\sum_{\vec{k}}\sum_{i,j=+,-}M_{\vec{k},i,j}(\omega,\vec{q})[1-f(E_{i,\vec{k}+\vec{q}})] \times f(E_{j,\vec{k}})\,\delta(E_{i,\vec{k}+\vec{q}}-E_{j,\vec{k}}-\omega), \tag{6}$$

where $f(E)$ is the Fermi distribution function, and

$$M_{\vec{k},i,j}(\omega,\vec{q})=\left|\sum_{\alpha,\beta=\uparrow,\downarrow}\psi'_{i,\vec{k}+\vec{q},\alpha}A_{\alpha\beta}(\vec{k},\vec{q})\,\psi_{j,\vec{k},\beta}\right|^2 \tag{7}$$

is the transition probability from the state $\vec{k}$ in the jth SSSB to the state $\vec{k}+\vec{q}$ in the ith SSSB. Let us define $\varphi_{\vec{k},\vec{q}}=\frac{1}{2}(\phi_{\vec{k}}+\phi_{\vec{k}+\vec{q}})$ and $\varphi'_{\vec{k},\vec{q}}=\frac{1}{2}(\phi_{\vec{k}}-\phi_{\vec{k}+\vec{q}})$. Then from Eqs. (1) and (5) we find the intra-SSSB ($i=j$) transition probabilities

$$M_{\vec{k},\pm,\pm}(\omega,\vec{q})=\gamma^2|P_x\cos(\varphi_{\vec{k},\vec{q}})+P_y\sin(\varphi_{\vec{k},\vec{q}}) \pm iP_z\sin(\varphi'_{\vec{k},\vec{q}})|^2 \tag{8}$$

and the inter-SSSB ($i\neq j$) transition probabilities

$$M_{\vec{k},\pm,\mp}(\omega,\vec{q})=\gamma^2|P_x\sin(\varphi_{\vec{k},\vec{q}})-P_y\cos(\varphi_{\vec{k},\vec{q}}) \pm iP_z\cos(\varphi'_{\vec{k},\vec{q}})|^2. \tag{9}$$

Since $q\ll k\simeq k_f$, we can set $\phi_{\vec{k}}\simeq\phi_{\vec{k}+\vec{q}}$ to simplify Eqs. (8) and (9) as

$$M_{\vec{k},+,+}(\omega,\vec{q})=M_{\vec{k},-,-}(\omega,\vec{q})=\gamma^2|\vec{P}\cdot\vec{n}_k|^2, \tag{10}$$

$$M_{\vec{k},\pm,\mp}(\omega,\vec{q})=\gamma^2|\vec{P}_\parallel\times\vec{n}_k\pm iP_z|^2, \tag{11}$$

where $\vec{n}_k=\vec{h}(\vec{k})/|\vec{h}(\vec{k})|$.

For intra-SSSB transitions, if we neglect the very small difference between the two 2D Fermi momenta in the two SSSB's, the delta function in Eq. (6) is simply $\delta(\vec{v}_f\cdot\vec{q}-\omega)$. The intra-SSSB excitations then give rise to a peak at $\omega=v_f q$ in the Raman spectrum, similar to the usual case of single particle intrasubband excitations in an electron gas.[7] However, the additional feature of the SSSB system is that, due to the spin splitting of electron energies, the intensity of this peak is angular dependent. This angular dependence is given by $M_{\vec{k}_q,+,+}(\omega,\vec{q})$ in (10) with $\vec{k}_q=k_f\vec{q}/q$.

The inter-SSSB transitions generate two more Raman peaks. We define $W_{i,j}(\omega,\vec{q})$ as the partial scattering cross-section in Eq. (6) contributed by the excitations with a hole in the jth SSSB and an electron in the ith SSSB. In order to demonstrate the essential physics with well-approximated analytical expressions, we assume zero temperature and obtain from Eqs. (4) and (6)

$$W_{\pm,\mp}(\omega,\vec{q})=N(E_f)\omega\int_0^{2\pi}\frac{d\phi}{2\pi} \times M_{\vec{k}_f,\pm,\mp}(\omega,\vec{q})\,\delta[\vec{v}_f\cdot\vec{q}\pm|\vec{h}(\vec{k}_f)|-\omega], \tag{12}$$

where $N(E_f)$ is the 2D density of states at the Fermi energy, and ϕ is the angle between the 2D $\vec{k}_f$ and the x axis. $W_{\pm,\mp}(\omega,\vec{q})$ have peaks (more precisely, inverse square root singularities) at the extremal points on the Fermi line where $\frac{d}{d\phi}(\vec{v}_f\cdot\vec{q}\pm|\vec{h}(\vec{k}_f)|)=0$. As was pointed out by Jusserand *et al.*,[9] the angular dependence of $|\vec{h}(\vec{k})|$ can be important in determining the precise positions of the peaks if $|\vec{h}(\vec{k})|>v_f q$. This allows[9] us to subtract a contribution of the Rashba term[10] which in asymmetric quantum wells adds to the intrinsic spin-orbit interaction given by Eq. (3).

The polarization dependence of the intensities of the inter-SSSB peaks is determined by $M_{\vec{k},\pm,\mp}(\omega,\vec{q})$ in Eq. (11) taken at the corresponding extremal points. This expression can be written as

$$M_{\vec{k},\pm,\mp}(\omega,\vec{q})=\gamma^2(|\vec{P}_\parallel\times\vec{n}_k|^2+|P_z|^2\pm i\vec{P}\times\vec{P}^*\cdot\vec{n}_k). \tag{13}$$

From this equation one can immediately see a drastic difference between two cases of linearly and circularly polarized incident and/or scattered light. In the former case the vector $\vec{P}$ is real and, hence, the third term in Eq. (13) is zero. However, in the latter case it is not zero. The term $\vec{P}\times\vec{P}^*\cdot\vec{n}_k$ is due to the interference of the light waves scattered by the $S_{\vec{q},z}$ component and the $S_{\vec{q},\parallel}$ component of the spin-density fluctuations. The contribution due to this term to the Raman cross section will disappear if the conduction band is not spin split. When the circular polarization directions of both the incident and the scattered light are reversed, then $\vec{e}_i$ becomes $\vec{e}_i^*$, $\vec{e}_s$ becomes $\vec{e}_s^*$, and so $\vec{P}$ becomes $\vec{P}^*$. As a result, the interference term in Eq. (13) exhibits their remark-

able property that they change sign under the reverse of the directions of circular polarizations.

To see the main qualitative features of the interference term we consider a simple case when the Rashba term in $\vec{h}(\vec{k})$ is absent and only linear terms with respect to $\vec{k}$ are taken into account in Eq. (3). This corresponds to a narrow symmetric quantum well where $\kappa \gg k_f$. In this case $|\vec{h}(\vec{k})|$ is angular independent and as can be seen from Eq. (12) the peaks in the Raman spectrum are at $\vec{k}=\vec{k}_q$ if $|\vec{h}(\vec{k})|<v_f q$ and $\vec{k}=\pm\vec{k}_q$ when $|\vec{h}(\vec{k})|>v_f q$. In the former case the two inter-SSSB Raman bands at $\omega=v_f q\pm|\vec{h}(\vec{k}_q)|$ are due to the both $W_{+,-}(\omega,\vec{q})$ and $W_{-,+}(\omega,\vec{q})$ contributions to the cross section. The corresponding interference terms have opposite signs, as can be seen from Eq. (13). On the other hand, if $|\vec{h}(\vec{k})|>v_f q$ we have $W_{-,+}(\omega,\vec{q})=0$, but $W_{+,-}(\omega,\vec{q})$ gives rise to two peaks at $\omega=|\vec{h}(\vec{k}_q)|\pm v_f q$. The higher-energy peak is due to excitations at $\vec{k}=\vec{k}_q$ while the lower-energy one is formed by the excitations at $\vec{k}=-\vec{k}_q$. Since $\vec{n}_{k_q}=-\vec{n}_{-k_q}$, the third term in Eq. (13) also has opposite signs for these two Raman bands. Therefore the interference is constructive for one of the inter-SSSB Raman bands, and it is destructive for the other band. If $\vec{h}(\vec{k})\to 0$, the two Raman bands merge and the corresponding interference terms cancel each other in the cross section. Hence, the interference contribution to the Raman spectrum is entirely due to the electron spin dynamics in the spin-split conduction band of quantum wells.

The signs of the interference terms can be reversed by changing the circular polarizations of incident and scattered light waves. Hence, taking difference of the two spectra with opposite circular polarizations, one can remove the peak corresponding to intra-SSSB excitations as well as the two first terms in Eq. (13), subtracting thus the interference contribution from the Raman spectrum. When the Rashba term as well as cubic terms give noticeable contribution to $\vec{h}(\vec{k})$ the relative intensities and positions of the Raman bands in the difference spectrum have more complicated angular dependence, as compared with the simple example considered above. Analysis of this dependence can give new additional information on the relative value of various contributions to $\vec{h}(\vec{k})$.

It should be noted that, besides the spin-flip term (1), a circularly polarized light can also be scattered from the electron charge-density fluctuations. This scattering is usually observed at parallel linear polarizations of incident and scattered light. However, charge-density fluctuations are spin independent and, hence, cannot give any contributions to the interference term and will not be observed in the difference spectrum.

After the study of clean samples, let us consider the opposite limit that the system contains a strong random elastic scattering potential such that both $v_f q$ and $h(\vec{k})$ are small as compared to the electron elastic scattering rate Γ. Under this condition, the Raman peaks due to various electron transitions merge and become difficult to be resolved. Nevertheless, we will show that the interference terms have the same dependence on the polarization of light as in the clean samples. This property allows us to derive the effects of spin splitting from Raman measurements.

The intensity of the spin flip Raman scattering can be calculated by making an average $\langle\cdots\rangle_{\rm ran}$ of Eq. (6) over a random potential due to impurities and imperfections. Using the fluctuation-dissipation theorem,[11] the scattering probability is expressed in terms of the spin-density correlation function as

$$W(\omega,\vec{q})=\frac{\gamma^2}{\pi}\mathrm{Im}\Bigg\{-i\sum_{i,j=x,y,z}P_iP_j^*\int_0^{2\pi}\frac{d\nu}{2\pi}\times\langle\mathrm{Tr}[S_{\vec{q},j}G(\nu,\vec{k},\vec{k}')\times S_{\vec{q},i}G(\nu+\omega,\vec{k}+\vec{q},\vec{k}'+\vec{q})]\rangle_{\rm ran}\Bigg\},\quad(14)$$

where the electron Green functions $G(\nu,\vec{k},\vec{k'})$ are matrices in the spin space. The off-diagonal components ($i=x,y$ and $j=z$) in the integrand represent the interference of the transverse (along the z axis) and the longitudinal (in xy plane) spin-density fluctuations. Because the conduction band is spin split, these components are finite at $q\neq 0$. When Γ is sufficiently large, the electron transport is dominately diffusive with the diffusion constant $D=v_f^2/4\Gamma$. Due to the D'yakonov-Perel mechanism,[12] the longitudinal and the transverse spin fluctuations relax with the respective rates $\Gamma_l=\langle h^2(\vec{k}_f)\rangle_{\rm dir}/2\Gamma$ and $\Gamma_t=\langle h^2(\vec{k}_f)\rangle_{\rm dir}/4\Gamma$, where $\langle\cdots\rangle_{\rm dir}$ is an average over the direction of $\vec{k}_f$. The electron diffusion and the spin relaxation can be described in the framework of quasiclassical approach when one ignores the quantum effects associated to the interference of quantum amplitudes of multiple scattering of electrons from defects. Within this approach the configuration average $\langle\cdots\rangle_{\rm ran}$ in Eq. (14) can be calculated with the so-called diffusion approximation,[13] which allows the off-diagonal components of the trace in Eq. (14) to be represented by the corresponding components of the diffusion propagator. Using the diffusion propagator which we have derived earlier,[14] the interference terms in the the scattering probability are obtained from Eq. (14) as

$$W_{if}(\omega,\vec{q})=i\gamma^2N(E_f)\vec{P}\times\vec{P}^*\cdot\vec{n}_{k_q}(\omega/4\Gamma)\times\mathrm{Re}\{m(q)/[D_l^{-1}D_t^{-1}-m^2(q)]\},\quad(15)$$

where $m(q)=|h(\vec{k}_q)|v_f q/(8\Gamma^2)$ and $D_{l,t}=4i\Gamma/(\omega+iDq^2+i\Gamma_{l,t})$. Because of the $\vec{P}\times\vec{P}^*$ term in Eq. (15), $W_{if}(\omega,\vec{q})=0$ for linearly polarized light which has $\vec{P}=\vec{P}^*$. On the other hand, with circular polarizations of both the incident and the scattered light, the interference $W_{if}(\omega,\vec{q})$ changes sign when the directions of polarizations are reversed. While this behavior is similar to that found in clean samples, an important distinction is that now the Raman band is structureless and its width depends on the values of Γ_l, Γ_t, and q.

We should mention that under the extreme resonance conditions of the spin-flip Raman scattering, the longitudinal and the transverse component contribute to the amplitude of scattering with different weights, and therefore the expression (1) with a scalar product $\vec{P}\cdot\vec{S}_q$ is no longer valid. However,

as long as these weights have equal phases, our theoretical analyses on the asymmetry with respect to the directions of circular polarizations remain unaffected qualitatively. A quantitative problem in connection to the extreme resonance conditions is perhaps the appearance of resonance denominators in γ. This question requires future study. Additional study is also required on the electronic exchange effect which gives rise to collective spin excitations in quantum wells.[15] We expect that this can result in new spectral features of the interference term.

To close this paper, we would like to emphasize our theoretical prediction that in both clean and dirty III-V semiconductor quantum wells, due to the coupling between the longitudinal and the transverse electron spin excitations in the spin-split subband, interference terms appear in the spin-flip inter-SSSB elecronic Raman scattering. This interference effect makes the Raman scattering asymmetric for the right and the left circularly polarized light, but not for linearly polarized photons. This asymmetry appears in nongyrotropic materials at zero magnetic field, and is entirely due to the intrinsic electron spin dynamics in the spin-split conduction band.

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