Raman mechanism for spin-current generation in a two-dimensional electron gas

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We propose the use of stimulated Raman scattering for the injection of a pure spin current in quantum wells, where due to the Dresselhaus and Rashba spin-orbit couplings a spin-flip Raman process is possible. We show that the stimulated Raman process is electron momentum resolved and, therefore, the electron spins can be directed to the given regions. The pure spin current can be engineered by changing the Rashba spin-orbit coupling with an external bias across the quantum well.

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Techniques used to optically manipulate electron spins in semiconductors are interesting from the perspective of fundamental physics. In addition, they are potentially useful for device applications in spintronics, particularly when a spin flow can be established that could provide a controllable delivery of spins. Spin currents can exist in the form of a spin-polarized electrical current, with both a net electrical current and a net spin polarization, or a pure spin current, where the net electrical current is zero due to electrons of opposite spins moving in opposite directions. The diverse physics of spin currents allows many possibilities for spin-current injection and measurements. Optical processes rely on the spin-orbit (SO) interaction, which allow the electric field of incident light to couple to the spin degrees of freedom. The orientation of electron spins in semiconductors by interband optical transitions, due to the absorption of circularly polarized light, leads to the generation of a net spin density in these crystals, which can then be accelerated using an applied bias. Spin currents can also be optically injected directly by taking advantage of quantum interference between one- and two-photon optical transitions across the band gap, and with one-photon absorption in crystals without inversion symmetry. This coherently controlled spin current has been observed experimentally by different techniques in the bulk and two-dimensional (2D) structures. Additionally, pure spin currents can be injected in quantum wells (QWs) by intersubband absorption of infrared radiation, or by direct spin-flip transitions resulting from far infrared absorption. All these are based on spin-current generation through absorption of visible or infrared radiation. The interband and intersubband transitions typically deposit about 1 eV and 100 meV in the crystal, respectively. Most of this energy is not associated with the velocity of the carriers, and hence is not accessible to aid in spin transport. It is essentially lost.

However, in such 2D structures electron spin slips can be achieved via a spontaneous Raman process, where an incident photon of frequency \( \omega_1 \) is absorbed and one with frequency \( \omega_2 \) is emitted. The energy difference \( \hbar \Omega = \hbar (\omega_1 - \omega_2) \) is then deposited to (Stokes process, \( \Omega > 0 \)) or extracted from (anti-Stokes process, \( \Omega < 0 \)) the system. The spectroscopy of spontaneous spin-flip Raman scattering has provided extremely valuable information about the SO interaction, but the process has a very small cross section. With a focus on spin-current generation, we here consider the stimulated Raman process in a 2D electron gas confined in a QW. Both the Rashba and Dresselhaus-type SO couplings are considered. We show that a pure spin current can be injected via the Raman process, since electrons of opposite momenta have opposite spin orientation. The characteristics of the spin current injection can be engineered by changing the Rashba SO coupling strength through an external bias, which determines the momentum-dependent spin states. We calculate the spin-flip and spin-current density injection rates, and argue that the spin current could be observed experimentally.

This effect should be very generally observable in quantum structures with spin splitting. As an example we consider an n-doped (110) GaAs QW, which is either asymmetrically doped or subject to an applied bias across the well. The Hamiltonian of the system is given by

\[
H = \hbar^2 k^2 / 2m_e + H_{SO},
\]

where \( m_e = 0.067 m_0 \) is the electron effective mass, for a bare mass \( m \). The SO Hamiltonian \( H_{SO} = H_D + H_R \) describes the system. Here the Dresselhaus term \( H_D \) and the Rashba term \( H_R \) originate, respectively, from the unit cell inversion asymmetry and structural asymmetry

\[
H_D^{[n]} = \alpha_D^{[n]} k_z \sigma_z F_{[n]}(k), \quad H_R^{[n]} = \alpha_R^{[n]} (\sigma_x k_y - \sigma_y k_x), \quad (1)
\]

where \( n \) is the subband index, \( k = k(\cos \phi, \sin \phi) \) is the plane wave vector of the electron envelope function, \( F_{[n]}(k) = 1 - (k_z^2 - 2k_x^2) \lambda_{[n]}^2 \), where \( \lambda_{[n]} \) depends on the QW width \( L \), and the \( \sigma \) are the Pauli matrices. The \( z \) axis is perpendicular to the QW plane and the in-plane axes are \( x = [001] \) and \( y = [1\overline{1}0] \). The Dresselhaus and Rashba parameters \( \alpha_D^{[n]} \) and \( \alpha_R^{[n]} \) depend on \( n \); in the rigid wall QW model one has \( \alpha_D^{[n]} = -\alpha_0 \pi^2 (\pi/L)^2 / 2 \), where \( \alpha_0 \) is the Dresselhaus constant for the bulk, and \( \lambda_{[n]} = L/n \pi \). The electron spins have a considerable out-of-plane component, and in particular when \( \alpha_R^{[n]} = 0 \), the spins are fully aligned along the [110] or [1\overline{1}0] directions. The \( k \)- and spin-dependent energy eigenvalues are given by

\[
e_{[n]}(k) = h^2 R^{[n]}(k) + \pm \varepsilon_{SO}^{[n]}(k),
\]

where

\[
\varepsilon_{SO}^{[n]}(k) = k \sqrt{ (\alpha_D^{[n]} F_{[n]}(k) \sin \phi)^2 + (\alpha_R^{[n]} \varepsilon_{SO}^{[n]}(k))^2 },
\]

with \( u \) and \( d \) states of energies

\[
e_{[n]} = \hbar^2 k^2 / 2m_e + \pm \varepsilon_{SO}^{[n]}(k),
\]

and

\[
e_{[n]} = \hbar \omega_0
\]
spin as the Fermi circle is split into two curves for electrons with different temperatures in the partially occupied states with the wave vectors $k_F$ and $k_F$, respectively. The minimum spin-splitting energy $\varepsilon_1^{[1]}(k_F)$ is achieved at $\phi = 0$, while the spin-splitting dependence on $\phi$ is determined by the product $k_F^2/2\alpha_0^2$. At low temperatures in the partially occupied $n=1$ subband the Fermi circle is split into two curves for electrons with different spins as $k_F^2(\phi) = k_F^2[1 + \varepsilon_1^{[1]}(k_F, \phi)/2E_F]$ with $k_F = \sqrt{2\pi N}$, where $N$ is the concentration (areal density) of electrons. The states with the wave vectors $k_F^2(\phi) < k < k_F^2(\phi)$ are singly occupied by spin-polarized electrons and, therefore, contribute to the spin-flip Raman scattering. At realistic spin-orbit couplings [$\alpha = 1$ meV, $\max(\alpha_0^{[1]}(n=1), \alpha_0^{[1]}(n=2))$] and electron concentrations of $N = 10^{12}$ cm$^{-2}$, the fraction of these states $\sim \alpha_F/E_F$ is of the order of 1%. A wave function in the subband $n$ has the form

$$\psi_n^{[1]}(k) = \frac{1}{\sqrt{S}} e^{i\mathbf{kr}} \phi_n^{[1]}(z) \xi_n^{[1]}(k_F^2)$$

where $S$ is the normalization area, $r$ is the 2D coordinate, and $\phi_n^{[1]}(z)$ is the envelope function. The spinor $\xi_n^{[1]}(k_F^2)$ for the spin-split transitions $\psi_n^{[1]}(k) \rightarrow \psi_n^{[1]}(k)$ demonstrates van Hove singularities $\nu(\Omega) \sim 1/\sqrt{\Omega-\Omega_M}$ when the energy approaches the maximum and minimum points ($\Omega_M$, $\Omega_M$) shown in Fig. 1. The shape of the spectrum and behavior of $\nu(E)$ is determined by the Rashba and the Dresselhaus parameters and $\lambda_2^{[1]}$. The conventional definition of spin current per electron is

$$j_{sd}(k_F^2) = \frac{1}{2} \langle \xi_n^{[1]}(k_F^2) | \mathbf{v}^s | \xi_n^{[1]}(k_F^2) \rangle$$

where $a$ and $b$ are the Cartesian indices. Velocity components $\nu^s = \hbar^{-1} (\partial H_{SO}/\partial k_a)$ are the sums of normal $\hbar k_a/m_c$ and anomalous $\hbar^{-1} \partial H_{SO}/\partial k_a$ spin-dependent terms. Both normal and anomalous contributions are important for understanding the physics of spin current and its injection. However, in the case of the Raman-induced spin current, which is our interest here, the anomalous velocity-related terms from the initial and final states cancel and hence do not contribute to the injection rates.

We consider the injection of pure spin current through a spin-flip Raman process involving virtual intersubband transitions. This is allowed since the subband spinors are not mutually orthogonal. The spinor $\xi_n^{[1]}(k_F^2)$ for the spin-split transitions $\psi_n^{[1]}(k) \rightarrow \psi_n^{[1]}(k)$ demonstrates van Hove singularities $\nu(\Omega) \sim 1/\sqrt{\Omega-\Omega_M}$ when the energy approaches the maximum and minimum points ($\Omega_M$, $\Omega_M$) shown in Fig. 1. The shape of the spectrum and behavior of $\nu(E)$ is determined by the Rashba and the Dresselhaus parameters and $\lambda_2^{[1]}$. The conventional definition of spin current per electron is

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electron energy and momentum selective. For \( \Omega \) in the interval \( 2\alpha_R^{[1]} k_F < \hbar \Omega < \max[2\epsilon_{\text{so}}^{[1]}(k_F)] \), there are four or eight \( k \) regions where spin-flip transitions are allowed by energy conservation (depicted in Fig. 1). One spin-flip process injects a pure spin current \( \delta J_{yz}(k) = \hbar^2 \sqrt{d_D^2} k^2 \sin^2 \theta F_{[1]}(k)/m \epsilon_{\text{so}}^{[1]}(k) \), which is of the order of \( \hbar v_F \), where \( v_F \) is the Fermi velocity. We use Fermi’s golden rule to calculate the spin-flip and spin-current density injection rates. The interaction of electrons with the light is taken in the dipole approximation \( H_{\text{int}} = -(e/mc) \mathbf{p} \cdot \mathbf{A}(t) \), where \( p = -\hbar \mathbf{V} \), is the momentum operator, \( \mathbf{A}(t) \) is a vector potential of the field, \( e \) is the electron charge and \( c \) is the speed of light. The intersubband velocity matrix element \( \langle \phi_{m}^{[1]} | \mathbf{v} | \phi_{n}^{[1]} \rangle = \epsilon_{m}^{[1]} \epsilon_{n}^{[1]} \) depends on the spin states in both subbands.

The external perturbation leads to injection rates of electron and hole (areal) density in the conduction subband, and the spin-current (areal) density given in the resonant approximation by

\[
\begin{align*}
\dot{n}(\Omega) &= \frac{1}{S} \sum_{k} \left[ \Gamma(k) \delta(\Omega - 2 \epsilon_{\text{so}}^{[1]} k/h) \right] \mathcal{E}_2^2, \\
J_{yz}(\Omega) &= \frac{\hbar}{2} \chi_I(\Omega) I_1 I_2.
\end{align*}
\]

(4)

The summation over \( k \) is performed in the singly occupied regions with \( k_F(\phi) < k < k_F(\phi) \) [shadowed area in the inset of Fig. 2(b)]. The injection rates are proportional to \( I_1 I_2 \), where \( I_{1,2} \) are the intensities of the field components in air. To characterize the injection rates in the considered above experimental geometry we write

\[
\dot{n}(\Omega) = \chi_{\alpha}^{[1]}(\Omega) I_1 I_2, \quad J_{yz}(\Omega) = \frac{\hbar}{2} \chi_{\gamma}(\Omega) I_1 I_2.
\]

(5)

With the aim of estimating the magnitude of the effect and demonstrating the possibility of spin-current engineering by changes in the Rashba parameter we present in Fig. 3 the results of calculations of \( \chi_{\alpha}^{[1]}(\Omega) \) and \( \chi_{\gamma}(\Omega) \) for three different sets of \( \alpha_R^{[\beta]} \) parameters. For small \( \alpha_R^{[\beta]} \) the transitions have a low probability that increases with the increase of \( \alpha_R^{[1]}/\alpha_D^{[1]} \), approximately as \( \max(\alpha_R^{[1]}/\alpha_D^{[1]}, \alpha_R^{[2]}/\alpha_D^{[2]})^2 \). The spectral range also changes as a function of \( \alpha_R^{[1]} \).

At small frequencies \( \Omega \), corresponding to \( k = 0 \), no spin-flip Raman transitions are allowed since the \( u \) and \( d \) spin states are orthogonal due to the vanishing \( H_D^{[3]} \) in the SO Hamiltonian. We only present results in Fig. 3 for the \( \hbar \Omega \) range where a significant number of spin flips occur. It is instructive to compare the spin-flip rates in the spontaneous \( \dot{n}_{sp} \) and stimulated Raman scattering. The spin-flip density injection rate via spontaneous Raman scattering can be estimated as \( \dot{n}_{sp} = (I_1/\epsilon^2 e^2 \hbar \omega_0 (r_0 \omega_0 / \gamma)^3 (N \epsilon_k F_{[1]} E_{[3]}) \), where \( r_0 \) is the classical radius of electron and the cross section per one spin-flip process at \( \max(\alpha_R^{[1]}/\alpha_D^{[1]}, \alpha_R^{[2]}/\alpha_D^{[2]})^2 = 1 \) is \( r_0 \). At intensity \( I_1 = 100 \text{ MW/cm}^2 \) and \( \omega_0 / \gamma = 20 \) we obtain \( \dot{n}_{sp} = 10^{13} \text{ cm}^{-2} \text{s}^{-1} \), whereas stimulated Raman scattering induces a corresponding number (for \( I_1 = I_2 \)) of \( \dot{n} = 10^{13} \text{ cm}^{-2} \text{s}^{-1} \). The spontaneous spin-flip Raman process has a much weaker injection rate, despite the fact that scattering at all frequencies in the interval \( 2\epsilon_{\text{so}}^{[1]} k_F < \hbar \Omega < \max[2\epsilon_{\text{so}}^{[1]}(k_F)] \) are allowed.

Now we discuss the experimental accessibility of the proposed Raman injected pure spin current. The spin-flip Raman scattering discussed here will produce two spots of electrons, with opposite spin orientations, after momentum relaxation. Provided that the spin-relaxation time \( \tau_s \) is longer than the momentum relaxation time \( \tau_m \), the distance of separation between these two spots is determined by momentum relaxation time \( \tau_s = \mu m / e \), where \( \mu \) is the density- and temperature-dependent mobility. We estimate the spot separation by \( d = (2 \pi / \hbar) \left| J_{yz} / \dot{n} \right| = \pi \chi_I / \chi_{\gamma} \). Using the result presented in Fig. 3, and for a low temperature mobility \( \mu = 5 \times 10^4 \text{ cm}^2 / \text{V s} \) the spot separation is about 100 nm. Distances of this order have been detected in experimental studies of pure spin currents injected using other optical processes, despite the fact that the beam spot size is much larger than this.4,5 We note that the concentration of the injected electron-hole pairs is limited by \( \Delta n = \dot{n}(\Omega) \min(\tau_s, \tau_r) \), where \( \tau_r \) is the pulse width. Our calculations assume zero temperature, but are expected to hold, at least, for temperatures corresponding to energies \( \ll \hbar \Omega \). Such temperatures would also guarantee high mobilities and large spot separations.

To conclude, we have shown that the stimulated Raman scattering is an efficient mechanism for injection of pure spin currents in quantum wells, with a small energy transfer per spin flip to the system. The calculated magnitude shows that
it should be experimentally accessible using spatially resolved pump-probe schemes. The spin current can be engineered by applying a bias and hence changing the Rashba SO coupling parameter by shifting the van Hove singularity in the joint density of states for the spin-flip transitions.

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