Spin-splitting surface states of strained GaAs(001) and spontaneous spin current from breaking of twofold symmetry

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1. Introduction

Ever since the intrinsic spin Hall effect was proposed [1,2], interest in spin properties has increased. The intrinsic spin Hall effect, proposed by Sinova et al. [1], originates from Rashba-type spin splitting. Materials that exhibit giant Rashba-type spin splitting are desirable for fabricating many semiconductor-based spintronic devices currently being studied [3]. Spin splitting occurs when inversion symmetry is broken, a behavior that can arise in five cases: first, when a crystal lacks a center of inversion in the bulk [4], in two-dimensional electronic states with structural inversion asymmetry [5], at surfaces that lack three-dimensional symmetry [6], at interfaces of semiconductor heterostructures at which chemical bonding is asymmetric [7,8], and at surface alloys on metals and an adlayer on semiconductor substrates with in-plane inversion asymmetry [9–12]. Though up to now in-plane inversion asymmetry is due to missing of one mirror plane by alloying or adsorbing, we introduce in-plane inversion asymmetry by applying stress at the topmost surface. We wish to search for novel spin properties at surfaces with in-plane inversion asymmetry.

In this paper we theoretically consider spin-splitting surface states from breaking of the inversion symmetry of the surface in GaAs, a zinc blende crystal. Our model is based on that of the quantum spin Hall effect, proposed by Bernevig and Zhang [13]. In their model, the quantum spin Hall effect is induced by the shear strain gradients in a quantum well that has the mesoscopic twofold symmetry of the interface. In contrast, in our model the mesoscopic twofold symmetry of the topmost surface of GaAs(001) is broken. Additionally, we discuss a novel phenomenon which can be utilized as a new mechanism to generate the spin current.

2. Theory

Assessing the conduction bands of strained zinc blende crystals, in the framework of perturbation theory, an electron Hamiltonian can be written as [14–18]

\[
H = H_0 + H_S + H_{SO},
\]

\[
H_0 = \frac{1}{2m}\mathbf{p}^2 + V_0(\mathbf{x}),
\]

\[
H_S = a(\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz}),
\]

\[
H_{SO} = \frac{\hbar}{2}\left\{\sigma\Omega_x + (\sigma\Omega_y + (\sigma\Omega_z)),
\right\}
\]

where \(m\) is the bare electron mass, \(V_0(\mathbf{x})\) is the potential in the unstrained crystals, \(a\) is the hydrostatic deformation potential, \(\varepsilon_{ij}\) is the strain tensor component, and \(\sigma\) are Pauli matrices. Additionally, in the perturbative Hamiltonian \(H_{SO}\), the first term describes the Dresselhaus spin–orbit coupling, while the second and third terms describe the strain-induced spin–orbit coupling. The Dresselhaus spin–orbit coupling originates from the periodic potential in the crystal, while the strain-induced spin–orbit coupling originates from the potential in the strained crystal. \(\Omega_x, \Omega_y, \Omega_z\) are vectors with components
\[ h\Omega_{J1} = 2E_{g}^{1/2} p_\gamma (p_{x}^{2} - p_{y}^{2}). \]
\[ h\Omega_{S1} = \nu (\epsilon_{xy} P_{y} - \epsilon_{xx} P_{x}), \]
\[ h\Omega_{2S} = \nu (\epsilon_{xy} P_{y} - \epsilon_{zz} P_{z}). \]  

(2)

The remaining components of these vectors are obtained by cyclic permutation of the indices \( x, y, \) and \( z \). In Eq. (2) \( E_{g} \) is the width of the band gap, and \( \nu, \nu' \) and \( \nu'' \) are constants that determine the magnitude of the spin–orbit coupling. It is reasonable that \( \Omega_{SO} \) can be written as Eqs. (1) and (2), because \( \Omega_{SO} \) is invariant, consisting of basis sets for the irreducible representation \( \Gamma_{3} \) of group \( T_d \) [16]. Experiments by Wu et al., investigating strain-induced spin–orbit coupling, indicated that for GaS the strain-induced spin–orbit coupling is more important than the Dresselhaus spin–orbit coupling [19]. The constants \( \nu, \nu' \) and \( \nu'' \) of the strain-induced spin–orbit coupling are \( 8 \times 10^{5} \text{m/s} \) and \( 6 \times 10^{5} \text{m/s} \) for bulk GaS [13,19], respectively.

We describe our model below. We choose one of the topmost lattice points of an ideal surface as the origin of the coordinate system and \( x, y, \) and \( z \) axes along the [100], [010], and [001] directions, respectively. Modifying Bernevig and Zhang’s model [13], we apply strain at the topmost surface by shifting the topmost surface atoms from its ideal surface position, described as

\[ u = (u_{x}, u_{y}, u_{z}) = (0, 0, T(y)2g_{xy}). \]  

(3)

where \( g \) is a constant that determines the magnitude of the displacement, and \( T(y) \) is an operator and defined as

\[ T(y)f(y) = \begin{cases} f(-y) & \text{if } y > 0 \\ f(y) & \text{otherwise} \end{cases}, \]

where \( f(y) \) is an arbitrary function. Our above model differs from that of Bernevig and Zhang by operating \( T(y) \) on \( u_{z} \) [13]. Modifying further, we use the strain tensor components of the small inhomogeneous confined strain, as described by

\[ \epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz} = 0, \]
\[ \epsilon_{xy} = \epsilon_{yx} = 0, \]
\[ \epsilon_{yz} = \epsilon_{zy} = S(y)
\]
\[ \epsilon_{xz} = \epsilon_{zx} = T(y)
\]
\[ S(y)f(y) = \left( \frac{\partial T(y)}{\partial y} \right)f(y) = \begin{cases} f(-y) & \text{if } y > 0 \\ f(y) & \text{otherwise} \end{cases}. \]

(4)

where \( x \) is a positive constant and \( n \) is a large enough integer. Here, GaAs is in the region \( z > 0 \), while the region \( z < 0 \) corresponds to the vacuum. Fig. 1 shows the displacement of the topmost surface atoms. Thus, in our model the mesoscopic twofold symmetry of the strained GaAs(001) surface is broken. We treat the effect of the displacement of the topmost surface atoms as a perturbation of the potential [20]. Here, the strain given by Eq. (4) can be realized by applying tensile stress within the first and fourth quadrants of the \( xy \)-plane at the topmost surface as well as compressive stress within the second and third quadrants of the \( xy \)-plane at the topmost surface, with fixed atoms on the \( x \) and \( y \) axes. By reconstruction of GaAs(001) surface, the distortion appears because of dimerization or dimer buckling. However, no strain is generated by representative reconstructions of the GaAs(001) surface, including the \( c(4 \times 4) \) reconstruction and the \( 2 \times 4 \) reconstruction. Thus, the strain tensor components (4) describe the strain of the strained GaAs(001) surface of our model if we consider reconstructions. Here, in the perturbative Hamiltonian \( H \) in Eq. (1), the first term, which describes the Dresselhaus spin–orbit coupling, can be omitted because the surface has a center of inversion in \( xy \)-plane. Additionally, \( H_{S} \) and the third term in \( H_{SO} \) does not contribute to \( H \), because \( \epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz} = 0 \). Therefore, the surface electron Hamiltonian of our model can be written as

\[ H = H_{0} + H_{SO}, \]
\[ H_{0} = \frac{\epsilon}{2} p^{2} + V_{0}(x), \]
\[ H_{SO} = \frac{1}{2} (\sigma_{x} \Omega_{S1x} + \sigma_{y} \Omega_{S1y} + \sigma_{z} \Omega_{S1z}), \]

where

\[ \Omega_{S1x} = -\nu g T(y) \exp[-x\sqrt{x^{2} + y^{2}}] \exp[-n^{2}z^{2}] y \exp[T^{\dagger}(y)T(y) - T(y)T^{\dagger}(y)], \]
\[ \Omega_{S1y} = -\nu g S(y) \exp[-x\sqrt{x^{2} + y^{2}}] \exp[-n^{2}z^{2}] y \exp[T^{\dagger}(y)T(y) - T(y)T^{\dagger}(y)], \]
\[ \Omega_{S1z} = -\nu g T(y) \exp[-x\sqrt{x^{2} + y^{2}}] \exp[-n^{2}z^{2}] y \exp[T^{\dagger}(y)T(y) - T(y)T^{\dagger}(y)] \sigma_{z}, \]

and \( V_{0}(x) \) is the potential in the ideal unstrained surface. The region \( z > 0 \) corresponds to the semi-infinite lattice. This perturbative Hamiltonian \( H_{0} \) only has a substantial effect near the topmost surface in \( z \) direction on the surface electron Hamiltonian. Because \( x(y) > 0 \) for \( x(y) > 0 \), we can simply consider the perturbative Hamiltonian \( H_{0} \) now.

\[ H'_{SO} = \frac{1}{2} \sigma_{z} \Omega_{S1z}, \]
\[ = \frac{1}{2} \nu g [\exp[-x\sqrt{x^{2} + y^{2}}] \exp[-n^{2}z^{2}] y \exp[T^{\dagger}(y)T(y) - T(y)T^{\dagger}(y)] \sigma_{z}, \]

Here, \( \Omega_{S1x} \) and \( \Omega_{S1y} \) are hermitian because

\[ \frac{\partial \exp[-n^{2}z^{2}]}{\partial x} = -2n^{2}z \exp[-n^{2}z^{2}] \]
\[ = 2 \left\{ \delta(z + (1/\sqrt{2}n)) - \delta(z - (1/\sqrt{2}n)) \right\}, \]

and because the wave function is continuous at \( z = 0 \) in \( z \) direction. We use this Hamiltonian \( H'_{0} + H'_{SO} \) to evaluate the electronic structure.

### 3. Results and discussion

We now qualitatively evaluate the electronic structure of surface-state conduction bands near the \( \Gamma \) point. Assuming a small perturbation, we consider that the surface electron is in Bloch states in the \( x \) and \( y \) directions as \( \Psi_{k}(x) = \exp[i(k_{x}x + k_{y}y)]u_{k}(x)(k_{x} = (k_{x}, k_{y})) \) in the region \( z > 0 \). The function \( u_{k}(x) \) satisfies the equation

\[ \left[ \frac{1}{2m} \partial^{2} + V_{0}(x) + \frac{\hbar^{2}}{2m} k_{x}^{2} \right] \Psi_{k}(x) = E(k)u_{k}(x), \]

where

\[ \Psi_{k}(x) = \psi_{k}(x) \exp[i(k_{x}x + k_{y}y)]. \]
Here, in the region $z > 0$ we use $u_i(x) = f_i(r)$, $u_i(x) = x f_i(r)$, and $u_i(x) = y f_i(r)$ satisfy the equation

\[
\begin{align*}
\left[ 1 - \frac{1}{2m} p^2 + V_0(x) \right] u_i(x) &= E_i(0) u_i(x), \\
\left[ 1 - \frac{1}{2m} p^2 + V_0(x) \right] u_i(x) &= E_p(0) u_i(x) \quad (i = x, y),
\end{align*}
\]

under the condition that in the region $z < 0$ $V_0(x) = V_0$ ($V_0$ is infinitely high), where $E_i(0)$ and $E_p(0)$ are the energy eigenvalue of the $s$ band of the surface-state conduction bands at the $\Gamma$ point and that of the $p$ band of the surface-state valence bands at the $\Gamma$ point, respectively. We expand the wave function of the $s$ band of surface-state conduction bands at $k = k_1$ near the $\Gamma$ point $u_{s1}(x)$ by $u_{i}(x)(i = x, y)$ by using $k \cdot p$ theory [21]. Then, we evaluate the energy eigenvalue of the $s$ band of the surface-state conduction bands at $k = k_1$ near the $\Gamma$ point $E_{s1}(k_1)$ by $k \cdot p$ theory. We do not consider dangling bonds of the topmost surface atoms, and we assume that bonding between adjacent (001) plane is weak. Additionally, we do not consider reconstruction of GaAs(001) surface. Up to the third order in energy, we obtain

\[
E_{s}(k_1) = E_{s}(0) + C_{k_1}^{(1)} k_x + C_{k_1}^{(2)} k_x^2 + C_{k_1}^{(3)} k_y^2 + C_{k_1}^{(4)} k_x k_y^2,
\]

where

\[
C_{k_1}^{(1)} = \frac{2 \sqrt{n} v_F \hbar}{n} \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r \, dr,
\]

\[
C_{k_1}^{(2)} = C_{k_1}^{(2)} = \frac{h^2}{2m} \int_{0}^{1} \left[ \frac{1}{E_i(0) - E_p(0)} \right]^2 \left[ \frac{1}{3} \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r^2 \, dr - \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r \, dr \right],
\]

\[
C_{k_1}^{(3)} = \frac{2 \sqrt{n} v_F \hbar}{n} \int_{0}^{1} \left[ \frac{1}{E_i(0) - E_p(0)} \right]^2 \left[ \frac{1}{3} \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r^2 \, dr - \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r \, dr \right],
\]

\[
C_{k_1}^{(4)} = \frac{2 \sqrt{n} v_F \hbar}{n} \int_{0}^{1} \left[ \frac{1}{E_i(0) - E_p(0)} \right]^2 \left[ \frac{1}{3} \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r^2 \, dr - \int_{0}^{1} (f_i(r))^2 \exp[-\alpha r^2] r \, dr \right].
\]

and the upper sign and lower sign denote the upspin and downspin, respectively. Here, we use $\exp[-n^2 z^2] = \exp[-n^2 (\cos \theta l)] = (\sqrt{n^2 - \delta_{s1}(\cos \theta)} + \delta_{s1}(\cos \theta))$ , where $\delta_{s1}(\cos \theta)$ is a delta sequence. Thus, the spin degeneracy of the bottom of the surface-state conduction bands is split. Figs. 2 and 3 show schematics of the electronic structure of the bottom of the surface-state conduction bands. Here, the spin-splitting is caused by the dependence of the spin direction in the $k \cdot p$ Hamiltonian. However dangling bonds rebond, and however GaAs(001) surface is reconstructed, the wave function is expanded in the (001) planes. Thus, the obtained electronic structure gives a qualitative insight into the real electronic structure of strained GaAs(001) surface with broken twofold symmetry.

Now, we discuss a model similar to Bernevig and Zhang’s model [13]. In this model, the perturbative Hamiltonian $H_{s0}^{(s)}$ is given as

\[
H_{s0}^{(s)} = \frac{1}{2} g \exp \left[ -\alpha \sqrt{x^2 + y^2} \right] \exp[-n^2 z^2] (y p_x - x p_y) \sigma_z.
\]
of zinc blende based semiconductor heterostructures grown along the [001] axis, atomic structures, whose threefold symmetry is broken, have $D_{3d}$ symmetry because of “common anion”. On the other hand, in “no common atom” heterostructures such as C1A1/C2A2 (C1 and C2 are cations, and A1 and A2 are anions.), there are two inequivalent interfaces to correspond to C1–A1–C2 and C2–A2–C1. Thus, atomic structures at interfaces have lower $C_{3v}$ symmetry, so that tight-binding calculations, which consider the full symmetry, explained optical anisotropy [22]. Additionally, in the framework of the envelope function approximation the “$H_{He}$ model” and the generalized boundary conditions demonstrated spin splitting of heavy hole or light hole subbands [7] and that of electron subbands [23], respectively. Here, in “$H_{He}$ model” two operators $B$ and $F$ corresponding to ‘backward’ and ‘forward’ bonds on each side of a C–A–C monolayer (C and A are cations and A is an anion.) are introduced and an asymmetric potential is added to the Hamiltonian as the perturbation [24]. In the generalized boundary conditions originally the term describing the heavy-light hole mixing due to the $C_{3v}$ symmetry is added to the Hamiltonian and the boundary conditions are changed [25]. On the other hand, we assume that the surface is not reconstructed and that atomic structures have $T_d$ symmetry, and we qualitatively evaluate the electronic structure by $k \cdot p$ theory.

Now, we discuss electron-doped n-type GaAs. In this case, the surface-state conduction bands are filled by electrons from lower to higher energy levels. Here, the expectation value of the velocity of an electron in a Bloch state of the $n$ band is given as $v_{n}^{\parallel}(k) = 1/h\partial E_{n}(k)/\partial k$ (k is the energy eigenvalue of the nband), using Feynman’s theorem. Because $v_{n}^{\parallel}(k) = 1/h\partial E_{n}(k)/\partial k$ in addition to Figs. 2 and 3, the spin current in x direction is to be expected. In our model, using the spin current operator $j_{x}^{(s)} = (1/2)\{v_{1} s_{1}, v_{2} s_{2}\} (v_{1}$ and $s_{1}$ are the velocity and spin operator, respectively.), we evaluate the spin current in the x direction of the s band electron $j_{x}^{(s)}$ and that in the y direction of the s band electron $j_{y}^{(s)}$ over Bloch states with the energy eigenvalue less than $E_{F}$ near $\Gamma$ point. Assuming that the Fermi surface is a circle with the radius of $k_{F}$, we obtain

$$j_{x}^{(s)} = \sum_{k} \left\{ \frac{\hbar}{2} v_{x}^{(s)}(k), 0(E_{F} - E_{1}(k)), -\frac{\hbar}{2} v_{y}^{(s)}(k), 0(E_{F} - E_{1}(k)) \right\}$$

$$= \frac{L}{2\pi} \int_{\Delta E} \left\{ \frac{\hbar}{2} \frac{\partial E_{1}(k)}{\partial k_{x}} 0(E_{F} - E_{1}(k)), \right\} dk_{x} dk_{y}$$

$$- \frac{1}{\hbar} \frac{\partial E_{1}(k)}{\partial k_{x}} 0(E_{F} - E_{1}(k))$$

$$\equiv \frac{L}{2\pi} \left( C_{1x} k_{x}^{2} - \frac{3}{4} C_{3x} k_{x}^{4} + \frac{1}{4} C_{4x} k_{x}^{4}\right).$$

$$j_{y}^{(s)} = \sum_{k} \left\{ \frac{\hbar}{2} v_{y}^{(s)}(k), 0(E_{F} - E_{1}(k)), -\frac{\hbar}{2} v_{x}^{(s)}(k), 0(E_{F} - E_{1}(k)) \right\}$$

$$= \frac{L}{2\pi} \int_{\Delta E} \left\{ \frac{\hbar}{2} \frac{\partial E_{1}(k)}{\partial k_{y}} 0(E_{F} - E_{1}(k)), \right\} dk_{x} dk_{y}$$

$$- \frac{1}{\hbar} \frac{\partial E_{1}(k)}{\partial k_{y}} 0(E_{F} - E_{1}(k))$$

$$= \frac{L}{2\pi} \left( C_{1y} k_{y}^{2} - \frac{3}{4} C_{3y} k_{y}^{4} + \frac{1}{4} C_{4y} k_{y}^{4}\right).$$

where $L$ is an appropriate length of the period for the evaluation. Thus, in our model the spin current flows spontaneously in the x direction. Being compared with previous study in in-plane inversion asymmetry, this is fundamentally new knowledge which our study brings. This phenomenon can be utilized as a new mechanism to generate the spin current by the use of nonmagnetic materials as the spin Hall effect, spin–rotation coupling [26], and helical spin polarization of topological insulators [27].

4. Conclusion

We proposed a model of spin-splitting surface states. In our model the spin degeneracy is split from breaking of the twofold symmetry of the surface. We derive these results by $k \cdot p$ theory. Additionally, for n-type GaAs the spin current flows in the parallel direction to a mirror plane spontaneously. This phenomenon can be utilized as a new mechanism to generate the spin current.

References