Current-induced spin polarization at the surface of metallic films: A theorem and an ab initio calculation

I. V. Tokatly, E. E. Krasovskii, and Giovanni Vignale

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Spin-orbit coupling at the surface of metallic films: a theorem and an *ab initio* calculation

I. V. Tokatly,[1,2] E. E. Krasovskii,[1,3,2] and Giovanni Vignale[4,3,*]

[1] *Departamento de Física de Materiales, Universidad del Pais Vasco UPV/EHU, 20080 San Sebastián/Donostia, Spain*
[2] *IKERBASQUE, Basque Foundation for Science, E-48013 Bilbao, Spain*
[3] *Donostia International Physics Center (DIPC), E-20018 San Sebastián, Spain*
[4] *Department of Physics and Astronomy, University of Missouri, Columbia, Missouri 65211, USA*

The broken inversion symmetry at the surface of a metallic film (or, more generally, at the interface between a metallic film and a different metallic or insulating material) greatly amplifies the influence of the spin-orbit interaction on the surface properties. The best known manifestation of this effect is the momentum-dependent splitting of the surface state energies (Rashba effect). Here we show that the same interaction also generates a spin-polarization of the bulk states when an electric current is driven through the bulk of the film. For a semi-infinite jellium model, which is representative of metals with a closed Fermi surface, we prove as a theorem that, regardless of the shape of the confinement potential, the induced surface spin density at each surface is given by $\mathbf{S} = -\gamma\hbar\hat{\mathbf{z}} \times \mathbf{j}$, where $\mathbf{j}$ is the particle current density in the bulk, $\hat{\mathbf{z}}$ the unit vector normal to the surface, and $\gamma = \frac{\hbar}{4mc^2}$ contains only fundamental constants. For a general metallic solid γ becomes a material-specific parameter that controls the strength of the interfacial spin-orbit coupling. Our theorem, combined with an *ab initio* calculation of the spin polarization of the current-carrying film, enables a determination of γ, which should be useful in modeling the spin-dependent scattering of quasiparticles at the interface.

I. INTRODUCTION

Physical phenomena in which an electric current is converted into a spin polarization and/or a spin current, are receiving a great deal of attention in the context of orbital spintronics[1–17] – an appealing alternative to "classical" spintronics[18–22]. While in classical spintronics the spin dynamics is mainly controlled by exchange interactions, in orbital spintronics the central role is played by the spin-orbit (SO) interaction, which allows direct manipulation of the spins by electric fields.[23–34] In recent years both the exchange interaction-based approach and the spin-orbit interaction-based one have been shown to be viable for achieving current-induced switching of the magnetization of a ferromagnetic metal.[26,35–39] Although spin-orbit interactions are generally much weaker than exchange interactions, they are known to produce a characteristic linear in momentum spin splitting of surface states – the so-called Rashba effect[40], which is observed in semiconductor as well as metallic interfaces. The size of this splitting can be tuned by an external electric field, which creates the possibility of using the effect as the basis for a field-effect transistor.[41] In a different manifestation of the Rashba effect, a non-equilibrium spin accumulation in the surface states can produce a spin-galvanic current (or voltage) – a phenomenon that has been experimentally demonstrated in semiconductors[42] and, more recently, in metallic (Bi/Ag) interfaces.[43]

The Rashba splitting of surface states is by no means the only important manifestation of SO at a surface. Recently, it has been pointed out that the surface-induced SO coupling can have large effects also on the bulk states of a thin metallic film that is sandwiched between two insulating barriers[44–47]. **The bulk of such a metal is typically inversion-symmetric and its electronic states remain therefore doubly degenerate – at variance with the spin-split bands of the Rashba model. Nevertheless, it has been predicted that such films could exhibit large spin Hall angles, and that an electric polarization perpendicular to the surface should appear to second order in the electric field driving a current in the plane of the film.**[46]

In this paper we focus on the non-equilibrium spin polarization that appears in the immediate vicinity of the surface of a metal when a uniform current is driven throughout the bulk of the metal, parallel to the surface. The effect bears some similarity to the well known Edelstein effect,[2,3] which occurs in two-dimensional electron gases at the surface of semiconductors and metals. The crucial difference here is that the spin polarization of interest occurs in a continuum of bulk states scattering off the surface, with a relatively minor contribution from the surface state. The induced spin polarization is perpendicular to the current, parallel to the surface, and confined to a distance of order k_F^{-1} of the surface, where k_F is the three-dimensional Fermi wave vector of the bulk electrons. **The origin of the effect is spin-dependent electron scattering by the surface and, more specifically, the interference between the incident and the reflected wave.** For a semi-infinite jellium model, taken to be representative of metals with a simply connected Fermi surface, we prove as a theorem that the surface spin density (i.e., the spin density integrated over the coordinate perpendicular to the surface) is completely independent of the details of the confinement potential and is given by the elegant formula

$$\mathbf{S} = -\gamma\hbar\hat{\mathbf{z}} \times \mathbf{j}\,, \tag{1}$$

where $\mathbf{j}$ is the particle current density in the bulk, $\hat{\mathbf{z}}$ the

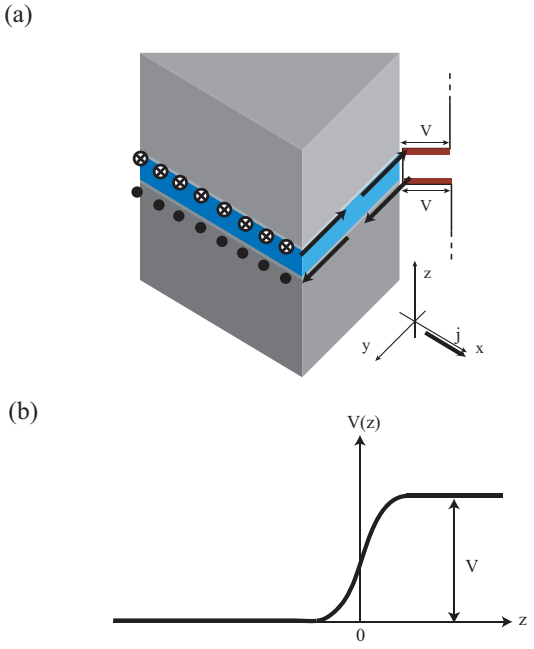

FIG. 1. (a) A metal film sandwiched between two insulators and separated from them by potential barriers of height V. The black arrows and dots indicate the direction of the spin polarization induced by the interfacial spin-orbit interaction when a current **j** flows along the x axis in the bulk of the film. (b) A more detailed view of the confining potential as a function of the coordinate z perpendicular to the interface. Only one interface is shown in this drawing, at $z = 0$, the other one being located far away on the negative z axis.

unit vector normal to the surface, and $\gamma = \frac{\hbar}{4mc^2}$ (with m the electron mass) contains only fundamental constants. The relation holds also for a general metallic solid, but γ then becomes a material-specific parameter that controls the strength of the interfacial spin-orbit coupling. In a thin metal film having two surfaces separated by a distance much larger than k_F^{-1} the two surfaces induce independent spin polarizations of opposite sign, such that the total integral of the spin density across the film vanishes. This situation can be described as a kind of bulk spin Hall effect, in which the current flowing in the bulk of the film induces spin accumulations of opposite signs on the two surfaces. But the driving force is not the spin-orbit interaction in the bulk of the metal, nor the spin-orbit interaction with impurities – rather, the spin-orbit interaction with the surface confinement potential, if one adopts the jellium model description, or, alternatively, in the real material, the combination of the atomic spin-orbit interaction with the loss of inversion symmetry induced by the termination of the bulk crystal. **This kind of spin Hall effect is different from the one investigated in Ref. 46 (where the accumulated spin was perpendicular to the plane of the film), but is quite similar (although conceptually distinct from it) to the standard spin-Hall effect, which arises from the spin-orbit interaction in the bulk of the metal.** It is remarkable that the final result has the simple and universal form of Eq. (1): such a structure is reminiscent of exact results about impurities in metals, where the summation of contribution from all occupied scattering states generates the Friedel sum rule or Fumi's theorem.[48]. **Similar results were previously obtained for the surface accumulation of spin in doped semiconductors[49] and for the edge-induced spin density in two-dimensional semiconductor quantum wells which break inversion symmetry[50–55], as well as for normal-metal/superconductor interfaces[56]. Ref. 53, in particular, presents strong numerical evidence for the robustness of the edge spin accumulation with respect to the form of the edge confinement potential. However, to the best of our knowledge, ours is the first work that demonstrates as a theorem the universality of the surface spin accumulation in the three-dimensional jellium model.**

There remains the fundamental problem of determining the value of the constant γ, which mimics in the jellium model the surface spin-orbit coupling of the real material. This constant is expressed in terms of the effective electron mass m and the effective Compton wavelength λ_c as follows:

$$\gamma = s\frac{m\lambda_c^2}{4\hbar}\,, \tag{2}$$

where $s = \pm 1$ is the overall sign of the expression (for a free electron in vacuum one has $s = +1$ and $\lambda_c = \hbar/mc \simeq 10^{-2}$Å, but these values can be dramatically different in a solid state environment: for electrons in GaAs, for instance, $s = -1$ and $\lambda_c \simeq 2$Å) To determine the quantities s and λ_c in any specific situation one must draw on detailed microscopic calculations, which take into account the effect of the atomic spin-orbit interaction on the electronic states. Here we propose a novel approach to the calculation of s and λ_c, based on the use of Eq. (1). The idea is to perform an *ab initio* calculation of the spin polarization of the bulk states of a thin metal film in the presence of a homogeneous current. Assuming the standard relaxation time approximation the current is introduced by shifting the Fermi surface in momentum space. The resulting coefficient of proportionality between the surface spin density and the particle current density yields an *ab initio* estimate of γ. In what follows, we apply this idea to the calculation of γ at the surface of a gold film. Although in Au(111) there is a Rashba-split surface state at the Fermi level, in our *ab initio* calculation we find its contribution to the surface spin polarization to be an order of magnitude smaller than the contribution from the bulk continuum. Moreover, a careful study of the three-dimensional spin density confirms that the induced spin is confined to a relatively small region ($\sim k_F^{-1}$) near the surfaces. We believe that Eq. (1), combined with *ab initio* theory, provides a remarkably simple approach to the determination of γ, a

crucial parameter for the spintronics of thin metal films.

II. PROOF OF THE THEOREM

We consider the setup of Fig. 1: a semi-infinite three-dimensional electron gas (jellium model) is confined to the half space with $z < 0$ by a potential that rises from $V(z) = 0$ for $z \to -\infty$ to $V(z) = V$ for $z \to \infty$. No assumption is made about the shape of this potential. It is further assumed that the chemical potential of the electrons μ is smaller than the barrier height V. Our objective is to calculate the integrated spin density induced by the interface in the infinite jellium. In the absence of spin-orbit interaction the electronic states are characterized by a conserved two-dimensional momentum $\mathbf{p}$ in the x-y plane (parallel to the interface) and by an asymptotic one-dimensional wave vector $k > 0$ in the z-direction (perpendicular to the interface):

$$\psi_{\mathbf{p},k}(\mathbf{r}, z) = e^{i\mathbf{p}\cdot\mathbf{r}}\varphi_k(z)\,, \tag{3}$$

where the wave functions $\varphi_k(z)$ are spinors of definite spin orientation ($\uparrow$ or $\downarrow$) and have the asymptotic form

$$\varphi_k(z) = \begin{cases} e^{ikz} + \hat{r}_k e^{-ikz}\,, & z \to -\infty \\ (1 + \hat{r}_k)e^{-\kappa z}\,, & z \to +\infty \end{cases} \tag{4}$$

where $\hat{r}_k$ is a phase factor and $\kappa = \sqrt{2mV - k^2}$.

This classification of states is essentially preserved by the spin-orbit interaction of form

$$H_{SO}(z) = \gamma V'(z)(\hat{\mathbf{z}} \times \mathbf{v}_\mathbf{p}) \cdot \boldsymbol{\sigma}\,, \tag{5}$$

where $\mathbf{v}_\mathbf{p}$ is the velocity operator and $V'(z)$ is the derivative of the potential with respect to z. The only difference is that $\hat{r}_k$ becomes a unimodular 2×2 matrix, mixing $\uparrow$ and $\downarrow$ spin states.

The spin polarization at position z is obtained from the trace of the spectral function

$$A_\mathbf{p}(z, \omega) = -2\Im m G^R_\mathbf{p}(z, z, \omega)\,, \tag{6}$$

where the retarded Green function $G^R_\mathbf{p}(z, z', \omega)$ is a 2×2 matrix in spin space, in the following manner

$$\mathbf{s}(z) = \frac{1}{2}\sum_\mathbf{p} \mathrm{Tr} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} f_\mathbf{p}(\omega) \left[\boldsymbol{\sigma} A_\mathbf{p}(z, \omega)\right]\,, \tag{7}$$

where $f_\mathbf{p}(\omega)$ is the average occupation of a state of parallel momentum $\mathbf{p}$ at energy ω. In equilibrium this would be the Fermi distribution at chemical potential μ and temperature T, $f(\omega) = [e^{\beta(\omega-\mu)} + 1]^{-1}$ independent of $\mathbf{p}$. In a current-carrying state, such as we are considering here, the occupation is given by a displaced Fermi distribution function $f_\mathbf{p}(\omega) = f(\omega - \mathbf{p} \cdot \mathbf{v}_d)$, where $\mathbf{v}_d$ is the average drift velocity of the electrons in the plane of the film. The surface spin density $\mathbf{S}$, defined as $\mathbf{s}(z)$ integrated over z is then given by

$$\mathbf{S} = -\sum_\mathbf{p} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} f_\mathbf{p}(\omega) \Im m \int dz \mathrm{Tr}\left[\boldsymbol{\sigma} G^R_\mathbf{p}(z, z, \omega)\right]\,. \tag{8}$$

Notice that we have set $\hbar = 1$ in these calculations.

This formula is exact and obviously yields zero spin polarization if spin-orbit coupling is absent. We will now proceed to evaluate $G^R_\mathbf{p}(z, z, \omega)$ to first order in the strength of spin-orbit coupling. The first-order expression for this is

$$G^R_\mathbf{p}(z, z, \omega) = \int dz' g^R_\mathbf{p}(z, z', \omega) H_{SO}(z') g^R_\mathbf{p}(z', z, \omega) \tag{9}$$

where $H_{SO}(z)$ is defined in Eq. (5) and $g^R_\mathbf{p}(z, z', \omega)$ is the retarded Green's function in the absence of spin-orbit coupling, i.e.,

$$g^R_\mathbf{p}(z, z', \omega) = \sum_k \frac{\varphi_k(z)\varphi^*_k(z')}{\omega - \epsilon_\mathbf{p}(k) + i\eta}\,, \tag{10}$$

where $\epsilon_\mathbf{p}(k) \equiv \frac{p^2}{2m_\parallel} + \frac{k^2}{2m}$. Making use of the explicit form of $H_{SO}(z)$ we find

$$\Im m \int dz \mathrm{Tr}\left[\boldsymbol{\sigma} G^R_\mathbf{p}(z, z, \omega)\right] = 2\gamma(\hat{\mathbf{z}} \times \mathbf{v}_\mathbf{p})\Im m \sum_k \frac{\langle\varphi_k|V'(z)|\varphi_k\rangle}{(\omega - \epsilon_\mathbf{p}(k) + i\eta)^2}\,, \tag{11}$$

where orthonormality of the states $\varphi_k(z)$ has been used. A crucial observation is that $-V'(z)$ is the operator of the force exerted by the interface on the electron. Its expectation value in the scattering state $|\varphi_k\rangle$ is therefore the negative of the pressure exerted by the electron of incoming perpendicular momentum k being reflected at the interface with outgoing perpendicular momentum $-k$. This pressure is simply the current of perpendicular momentum impinging on the surface, $\frac{2k^2}{m}$. **This can be easily understood by considering the flux of incident electrons $\propto k/m$, with each electron transferring momentum $2k$ to the surface.** Thus we have

$$\langle\varphi_k|V'(z)|\varphi_k\rangle = \frac{2k^2}{m}\,, \tag{12}$$

regardless of the detailed form of the potential. Armed with this result, we evaluate the momentum sum in Eq. (11). After an integration by parts we find

$$\Im m \sum_k \frac{\langle\varphi_k|V'(z)|\varphi_k\rangle}{(\omega - \epsilon_\mathbf{p}(k) + i\eta)^2} = 2\pi \sum_k \delta(\omega - \epsilon_\mathbf{p}(k))\,. \tag{13}$$

Plugging this into Eq. (11) and then Eq. (11) into Eq. (8), and reinstating physical units, we arrive at the promised "universal" result of Eq. (1), where the three three-dimensional particle current density $\mathbf{j}$ is given by $\mathbf{j} = n\mathbf{v}_d$, and n is the electron density.

III. AB INITIO CALCULATION FOR AU(111) SURFACE

To demonstrate the usefulness of our theorem we have performed an *ab initio* atomistic calculation of the spin-density profile induced by a current at the Au(111) surface. This is the classic system, in which the Rashba splitting of surface states was first observed[57], and for which a thorough theoretical analysis of both the surface states splitting[57] and the bulk continuum polarization[45] is available. The calculations were done for a finite-thickness slab of 19 atomic layers: the self-consistent (within the local density approximation) band structure was obtained with augmented plane waves method using the full-potential scheme of Ref.[59], and the spin-orbit coupling was included with the second variation technique of Koelling and Harmon[60]. The spin-resolved band structure was calculated in the $\bar{\Gamma}\bar{M}$ direction, and, to simplify the integration over the two-dimensional (2D) Brillouin zone, the hexagonal surface was assumed to be axially symmetric. In the finite-thickness slab formalism the eigenfunctions are two-component spinors labeled by a 2D Bloch wave vector $\mathbf{p}$ parallel to the surface and by a band index n, which subsumes the perpendicular-to-surface component of the Bloch wave vector. Each Bloch function contributes a spin density $\frac{\hbar}{2}\mathbf{s}_n(\mathbf{r},\mathbf{p})$, and we are interested in its in-plane y component $s_y(\mathbf{r})$, perpendicular to the current $\mathbf{j}=\hat{\mathbf{x}}j_x$. To calculate $s_y(\mathbf{r})$ we populate the electronic states with electrons according to a Fermi distribution shifted by an amount $\delta\mathbf{p}=\frac{4\pi e^2}{\hbar\omega_p^2}\mathbf{j}$, where ω_p is the plasma frequency entering the Drude conductivity $\sigma=\tau\omega_{\rm p}^2/4\pi$.[61] After the angular integration (assuming axial symmetry) we get

$$s_y(\mathbf{r}) = j_x \frac{e^2}{2\omega_{\rm p}^2} \sum_n s_n\left(\mathbf{r}, p_n^{\rm F}\right) p_n^{\rm F}, \tag{14}$$

where $p_n^{\rm F}$ is the Fermi wave vector in the 2D band of index n.

Figure 2(a) shows the depth profile $s_y(z)$, which is the average of $s_y(\mathbf{r})$ over the in-plane unit cell. The function $s_y(z)$ has a strong peak on the last atomic layer, and deep inside the slab it converges to a lattice-periodic function whose integral over the unit cell is zero. This spatially-dependent spin polarization, first observed in Refs.[44,45], arises from the asymmetric occupation of the bulk Bloch periodic states in the presence of a current j_x. The net spin polarization S_y (integrated over the unit cell) must be zero due to inversion symmetry (j_x is odd under inversion, S_y is even), but a spin-dipole density can and does appear in each unit cell, reflecting the intrinsic spin Hall effect of the material. This bulk effect is completely absent in the jellium model. The red curve in Fig. 2(a) is obtained from the total $s(z)$ by subtracting the lattice-periodic asymptotic function. This is the proper surface-induced spin polarization to be compared with the jellium-model calculations. It is thus seen that the effect of the surface extends over several atomic layers

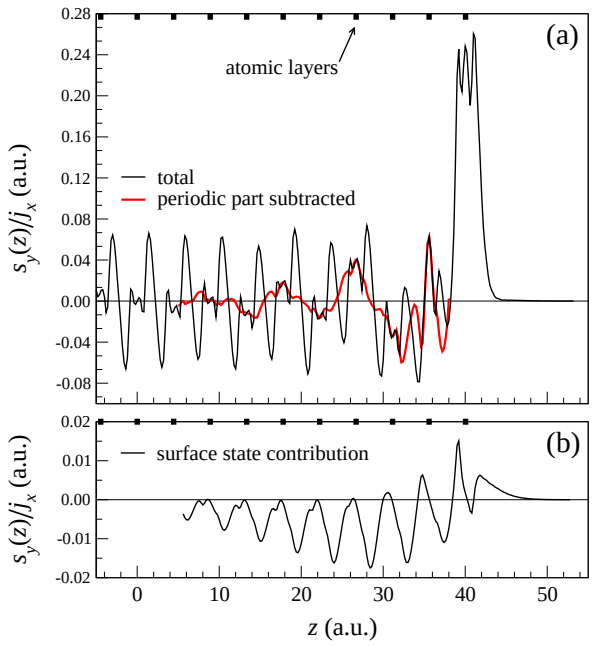

FIG. 2. (Color online) Spatially resolved induced spin density $s_y(z)$ per unit particle current density j_x at Au(111) calculated for a symmetric 19-layer slab: (a) total spin density and (b) surface state contribution. The center of the slab is at $z=0$. The three central layers are seen to be almost identical, which proves that the convergence with respect to the layer thickness has been achieved. The red curve shows $s_y(z)$ with the periodic asymptotic part subtracted.

into the interior of the crystal.[62] The calculations suggest that the main contribution to the surface-induced spin polarization comes from bulk-continuum states, which are not spin-split (they are Kramers degenerate). Somewhat unexpectedly, the contribution from the Rashba-split surface state is found to be an order of magnitude smaller, see Fig. 2(b), and opposite to the bulk spin polarization. **In particular, the strong peak of the spin density on the last atomic layer in Fig. 2a is seen to be unrelated to the surface state.**

The integral surface spin density S_y is related to the current j_x via Eq. (1) where the parameter γ is obtained by averaging Eq. (14) over the 2D unit cell and integrating over z. Expressed in Hartree atomic units (a.u.), for Au(111) the present calculation yields $\gamma = 0.7\,$a.u. (1 a.u. of time is $\hbar/1\mathrm{H} \simeq 2.42\times10^{-17}$ s). Equivalently, using the standard value $m=1.1\,m_e$ of the effective mass for Au in Eq. (2), we obtain $\lambda_c^2 \simeq 0.8$ Å^2. This value can now be used in the effective surface spin-orbit Hamiltonian Eq. (5) to reproduce, in the jellium model, the spin polarization obtained from the *ab initio* calculation. For a charge current density of 10^{10} A/m^2 (corresponding to a particle current density of 10^{29} m^{-2}s^{-1}) we find an induced spin density of the order of 10^{12} m^{-2} (in units of $\hbar$). It should be possible to observe this surface spin by Kerr rotation microscopy used e.g. to detect spin polar-

ization in 2D electron gases in semiconductors[28,29]

IV. CONCLUSION

Our analysis of the surface scattering problem in the jellium model has led to a direct connection, Eq. (1), between the spin-orbit coupling parameter γ and the spin density – a quantity that is not only measurable, but, more importantly from our point of view, computable in realistic systems. This connection is universal, in the sense of not depending on the form of the potential barrier in the jellium model. The value of this connection lies in the fact that it suggests a method to extract γ from a relatively simple *ab initio* calculation of the electronic structure. In turn, knowledge of γ allows many interesting and useful model calculations to be performed. Besides parameterizing the spin dependent surface scattering, the value of γ will, for example, be useful in modeling bulk effects of great interest in spintronics, such as spin diffusion and the spin Hall effect, particularly when a high degree of accuracy is not required. In fact, from our ab initio γ (or equivalently λ_c) the parameter $\frac{\lambda_c^2 k_F^2}{4}$ which controls the extrinsic spin Hall effect[63] is found to be $\simeq 0.2$, which is close the value fitted to experimental transport coefficients for Au (see Table I in Ref. 63).

V. ACKNOWLEDGMENTS

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* vignaleg@missouri.edu

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