Quantum Spin Hall Effect and Enhanced Magnetic Response by Spin-Orbit Coupling

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We show that the spin-Hall conductivity in insulators is related to a magnetic susceptibility representing the strength of the spin-orbit coupling. We use this relationship as a guiding principle to search real materials showing quantum spin-Hall effect. As a result, we theoretically predict that two-dimensional bismuth will show the quantum spin-Hall effect, both by calculating the helical edge states, and by showing the nontriviality of the Z_2 topological number, and propose possible experiments.

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The spin-Hall effect (SHE) [1–3] has been attracting a lot of attention recently, partly due to its potential use for semiconductor spintronics. Its remarkable feature is to induce a spin current without breaking time-reversal symmetry. One of the interesting proposals is the quantum spin-Hall (QSH) phase [4–9], which is a 2D insulator with helical edge states. The edge states form Kramers pairs, with spin currents flowing oppositely for opposite directions of spins. The QSH phase can be regarded as a novel phase constrained by the Z_2 topological number I [6]. It is equal to a number of Kramers pairs of helical edge states modulo two. The QSH phase has $I =$ odd, while the spin-Hall-insulator (SHI) phase [10], topologically equivalent to a simple insulator, has $I =$ even. It is surprising that insulators without ordering, usually considered as featureless and uninteresting, can have a nontrivial topological QSH phase. Its nontriviality reveals itself, e.g., in a critical exponent [11]. For its interest akin to the quantum Hall systems, an experimental observation of the QSH phase is called for.

To search for candidates for the QSH phase among a vast number of nonmagnetic insulators, we need a guiding principle. In this Letter, we propose that the magnetic susceptibility would be a good measure, and we pick up bismuth as a candidate due to its strong diamagnetism. There are also other supporting clues: large spin splitting in the surface states of the 3D system, similar to edge states for the 2D systems, the crystal structure in the (111) plane similar the Kane-Mele model for the QSH phase [6]. Using a 2D tight-binding model, we show that this system has only one pair of edge modes. Its Z_2 topological number [6] is shown to be odd, i.e., nontrivial, which supports stability of the edge states. These aspects make bismuth a promising candidate for the QSH phase.

To relate magnetic susceptibility with spin-Hall conductivity (SHC), we derive here a spin-Hall analog of the Středa formula. The Středa formula [12,13] tells us that in insulators the Hall conductivity σ_{xy} is expressed as $\sigma_{xy} = \frac{e}{\Omega}\frac{dN}{dB}|_\mu$, where N is the number of states below the chemical potential μ, and Ω is the area of the system. A direct generalization to the SHC is to replace N by the spin s_z, i.e., $\sigma_s = \frac{1}{\Omega}\frac{ds_z}{dB}|_\mu$. This is physically reasonable from the following argument. Suppose we apply a magnetic field in some region, linear in time. Then a change of the total spin inside the region is proportional to $\frac{ds_z}{dB}|_\mu$. According to the Maxwell equation, the increasing magnetic field induces a circulating electric field. Therefore, by interpreting the spin change as due to a spin-Hall current by the electric field, the SHC is $\sigma_s = \frac{1}{\Omega}\frac{ds_z}{dB}|_\mu$. We can justify this argument by explicit calculations.

A key step is to use a "conserved" spin current [14]. In most papers on the SHC, the spin current is conventionally defined as $\vec{J}_s = \frac{1}{2}\{\vec{v}, s_z\}$. However, in the presence of the spin-orbit coupling, the spin is no longer conserved: $\mathcal{T} \equiv \dot{s}_z \neq 0$. Namely, the right-hand side of the equation of continuity, $\partial_t s_z + \vec{\nabla}\cdot\vec{J}_s = \mathcal{T}$, is nonzero, and the "conventional" spin current $\vec{J}_s$ is not directly related with spin accumulation. Instead, Shi *et al.* [14] defined a conserved spin current $\vec{\mathcal{J}}_s$ as follows. If the system satisfies $\int dV\mathcal{T} = 0$, as it does in a uniform electric field, one can write $\mathcal{T}$ as $\mathcal{T} = -\vec{\nabla}\cdot\vec{P}_\tau$. Thus a conserved spin current defined as $\vec{\mathcal{J}}_s \equiv \vec{J}_s + \vec{P}_\tau$ satisfies $\partial_t s_z + \vec{\nabla}\cdot\vec{\mathcal{J}}_s = 0$, and is calculated for several models [14,15].

To calculate the SHC for the conserved spin current, we consider an electric field with wave number q, and take the limit $q \to 0$ [14]. When we calculate a spin current flowing to the x direction in response to an electric field to the y direction, we take the vector potential $\vec{A} = A_y e^{iqx}\hat{y}$, and the response is calculated as

$$\sigma_s^{\mathcal{J}} = -\frac{e}{\Omega}\lim_{q\to 0} i\partial_q \sum_{n\neq m}\frac{f(\varepsilon_m) - f(\varepsilon_n)}{(\varepsilon_n - \varepsilon_m)(\varepsilon_n - \varepsilon_m + i\eta)} \times \langle n|[H, s_z e^{iqx}]|m\rangle\left\langle m\left|\frac{1}{2}\{v_y, e^{-iqx}\}\right|n\right\rangle. \quad (1)$$

By rewriting as $[H, s_z e^{iqx}] = \frac{1}{2}\{s_z, [H, e^{iqx}]\} + \frac{1}{2}\times\{[H, s_z], e^{iqx}\}$, the first and the second term correspond to the conventional and spin-torque terms [$\sigma_{\mu\nu}^{s0}$ and $\sigma_{\mu\nu}^{\tau}$ in Eq. (10) of Ref. [14]], respectively. By a calculation similar to [16], we get $\sigma_s^{\mathcal{J}} = \sigma_s^{\mathcal{J}(\mathrm{I})} + \sigma_s^{\mathcal{J}(\mathrm{II})}$ with

$$\sigma_s^{\mathcal{J}(\mathrm{I})} = \frac{ie}{8\pi\Omega}\int d\varepsilon \frac{df}{d\varepsilon}\mathrm{Tr}[([H,s_z]G_+\{v_y,x\} - \{v_y,x\}G_-[H,s_z] - 2[H,s_z x]G_+ v_y + 2v_y G_-[H,s_z x] + [x,(s_z,v_y)])(G_+ - G_-)],$$

$$\sigma_s^{\mathcal{J}(\mathrm{II})} = \frac{ie}{\Omega}\int \frac{d\varepsilon}{4\pi} f(\varepsilon)\,\mathrm{Tr}(s_z G_+ L_z G_+ - G_- L_z G_-), \quad (2)$$

where $G_\pm = (\varepsilon - H \pm i\eta)^{-1}$ and $L_z = xv_y - yv_x$ is an orbital-angular momentum. The term $\sigma_s^{\mathcal{J}(\mathrm{I})}$ is proportional to $\frac{df}{d\varepsilon}(G_+ - G_-)$. By noting $G_+ - G_- = -2\pi i\delta(\varepsilon - H)$, only the states at the Fermi energy contribute to $\sigma_s^{\mathcal{J}(\mathrm{I})}$. In insulators $\sigma_s^{\mathcal{J}(\mathrm{I})}$ vanishes identically. On the other hand, the second term $\sigma_s^{\mathcal{J}(\mathrm{II})}$ is expressed as

$$\sigma_s^{\mathcal{J}(\mathrm{II})} = \int \frac{d\varepsilon}{\Omega} f(\varepsilon)\,\mathrm{Tr}\left(s_z \frac{d\delta(\varepsilon - H)}{dB_{\mathrm{orb.}}}\right) = \frac{1}{\Omega}\frac{ds_z}{dB_{\mathrm{orb.}}}. \quad (3)$$

Equation (3) agrees with the above-mentioned physically expected form. This result (3) can be also written as

$$\sigma_s^{\mathcal{J}(\mathrm{II})} = \frac{-\hbar}{g\mu_B}\frac{1}{\Omega}\frac{dM_{\mathrm{orb.}}}{dB_{\mathrm{Zeeman}}} = \frac{1}{\Omega g}\frac{dL_z}{dB_{\mathrm{Zeeman}}}, \quad (4)$$

where g is the electron-spin g factor, $M_{\mathrm{orb.}}$ is an orbital magnetization, and μ_B is the Bohr magneton. These formulas are a spin analog of the Středa formula [13]. We note that a Středa formula for the SHC with the conventional spin current $\vec{J}_s$ [16] has extra terms involving $\dot{s}_z$, in addition to (3). These terms arise from spin nonconservation. Hence it is natural that they do not appear in (3), as we used the conserved spin current [17]. We remark that the definition of the spin current is still controversial. Since the spin is not conserved, there is no unique definition of the spin current. Because there is no established way of directly measuring the spin current, one way is to consider instead measurable quantities such as spin accumulation at edges. The spin accumulation depends crucially on boundary conditions, and the conserved spin current may correspond to smooth boundaries [14]. This point requires further investigation.

We calculate σ_s for the model on the honeycomb lattice proposed by Kane and Mele [6]. This model shows the SHI and the QSH phases, depending on parameters λ_R, λ_v, and λ_{SO}. We numerically evaluate the SHC by Eq. (4) using the formula of orbital magnetization [18,19],

$$M_{\mathrm{orb.}} = \frac{ei}{2\hbar}{\sum_n}' \int \frac{d^d k}{(2\pi)^d}\left\langle \frac{\partial u_{n\mathbf{k}}}{\partial \mathbf{k}}\right| (2\mu - \varepsilon_{n\mathbf{k}} - H) \left| \frac{\partial u_{n\mathbf{k}}}{\partial \mathbf{k}}\right\rangle, \quad (5)$$

where ${\sum}'_n$ is a sum over occupied bands. The calculated SHC (Fig. 1) is $\sigma_s \sim 0$ in the SHI phase and $\sigma_s \sim \frac{-e}{(2\pi)}$ in the QSH phase, except for the vicinity of the phase boundary. The SHC in the QSH phase, $\sigma_s \sim -e/(2\pi)$, is interpreted as a fundamental unit $e/(4\pi)$ times two, the number of the edge states. The quantization is exact when s_z is conserved, i.e., $\lambda_R = 0$, where the system decouples to two quantum Hall systems with $\sigma_{xy} = \pm e^2/h$. Remarkably, even when s_z is not conserved, the SHC remains almost quantized. Deviation from quantization is prominent near the phase boundaries, which is attributed to smallness of the band gap.

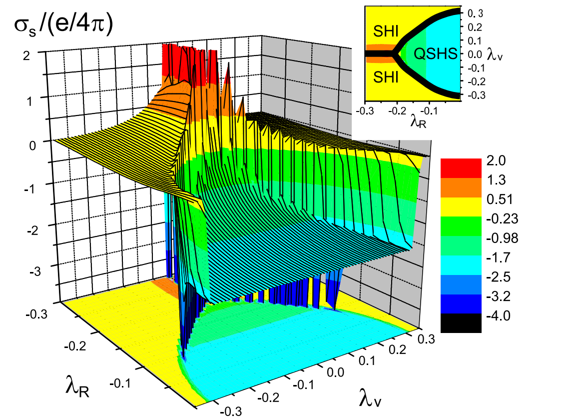

FIG. 1 (color online). Spin-Hall conductivity σ_s for the Kane-Mele model on the honeycomb lattice for various values of λ_R and λ_v in the unit of t. λ_{SO} is fixed as $0.06t$. As it is symmetric with respect to $\lambda_R \to -\lambda_R$, we show only the result for negative λ_R. The phase diagram is shown in the inset. Except for the vicinity of the phase boundary, $\sigma_s \sim 0$ in the SHI phase, and $\sigma_s \sim -e/(2\pi)$ in the QSH phase.

Therefore, materials with large susceptibility would be a good candidate for the QSH phase. Namely, if the susceptibility is large, σ_s should be large. Figure 1 then suggests that the system should be either in the QSH phase or near the phase boundary to the QSH phase. From this reason, we pick up some semimetals and related materials with large diamagnetic susceptibility, among which are bismuth and graphite.

Bi crystal has a rhombohedral structure, with trigonal symmetry around the (111) axis. Bi is a semimetal with a small hole pocket at the T point, and three electron pockets at the L points. Its strong diamagnetism has been studied experimentally and theoretically. It is theoretically attributed to massive Dirac fermions at the L and T points [20]. These Dirac fermions give a diamagnetic susceptibility which is enhanced as logarithm of the small energy gap [20]. Even when the Fermi energy is in the gap, this picture survives, and the susceptibility becomes even larger. Such Dirac fermions contribute to anomalous Hall effect and the SHE. Hence, it is no wonder such enhanced diamagnetic susceptibility implies an enhanced charge or spin-Hall conductivity.

Because the QSH phase is in 2D, we have to make Bi two dimensional, such as thin films and quantum wells. Such confinement discretizes the perpendicular momentum, and tends to open the gap. It was theoretically proposed that by making the Bi film thinner, it turns from semimetal to semiconductor [21,22]. Experiments show that the gap may open in thinner samples, whereas the gap is obscured by carrier unbalance between holes and electrons [23]. The Bi crystal can be viewed as a stacking of bilayers along the [111] direction. The interbilayer coupling is much smaller than the intrabilayer one, and the LEED analysis showed that the (111) surface of Bi is terminated with an intact bilayer [24]. A single bilayer [Fig. 2(a)] consists of two layers of triangular sublattices. This honeycomblike lattice structure [6] is a key for a nontrivial Z_2 topological number I; a crystal structure with high symmetry (e.g., square lattice) favors a trivial Z_2 topological number.

We demonstrate that 2D single-bilayer bismuth has a pair of helical edge states carrying spin currents with opposite spins. Furthermore, we show that the Z_2 topological number I is odd. For these purposes, we use the 3D tight-binding model [25] which well reproduces the band structure, and truncate the model by retaining only the hoppings inside the bilayer. The resulting 16-band model is regarded as a multiorbital version of the Kane-Mele model [6]. We first calculate the band structure for a single bilayer [Fig. 2(a)] for a strip geometry. The result [Fig. 2(b)] shows four edge states connecting between the bulk conduction and valence bands. They correspond to one Kramers pair of edge states, suggesting $I =$ odd. The spin Chern number [26] is calculated as $C_{\mathrm{sc}} = -2$, which is consistent with the existence of one pair of edge states. We also calculate the SHC from Eq. (4) and we get $\sigma_s \sim -0.74\frac{e}{4\pi}$. This value is reduced from $-2\frac{e}{4\pi}$ due to nonconservation of spin.

To confirm that the 2D bismuth is in the QSH phase, we also calculate the Pfaffian $\mathrm{Pf}(\boldsymbol{k})$ to calculate the Z_2 topological number I [6]. The bilayer system is inversion symmetric, allowing $\mathrm{Pf}(\boldsymbol{k})$ to be chosen real. The result is Fig. 2(c), where $\mathrm{Pf}(\boldsymbol{k})$ changes sign at the red curve, corresponding to Fig. 2a in [6]; it implies $I =$ odd. To further clarify the phase winding of $\mathrm{Pf}(\boldsymbol{k})$, we break the inversion symmetry by adding small on-site energies $\pm v$, for the atoms on the upper and the lower layers, respectively. This may correspond to a heterostructure or a single-bilayer thin film on a substrate. The result is shown in Fig. 2(d) for $v = 0.2$ eV. There is only one vortex for the phase of $\mathrm{Pf}(\boldsymbol{k})$ in the half Brillouin zone (BZ), which ensures $I =$ odd, like Fig. 2b in [6]. We note that the zeros of the Pfaffian do not follow the trigonal symmetry. It is because the Pfaffian is not covariant under unitary transformation of the Hamiltonian, and depends on a choice of the unit cell. Because this QSH phase is protected by topology, it cannot be broken unless the direct gap closes. This guarantees robustness of the nontrivial Z_2 index, despite the simplicity of the model. This odd Z_2 number guarantees stability of the helical edge states [7,8] against backscattering. We also found that bilayer antimony has even topological number, where edge states are fragile against opening a gap.

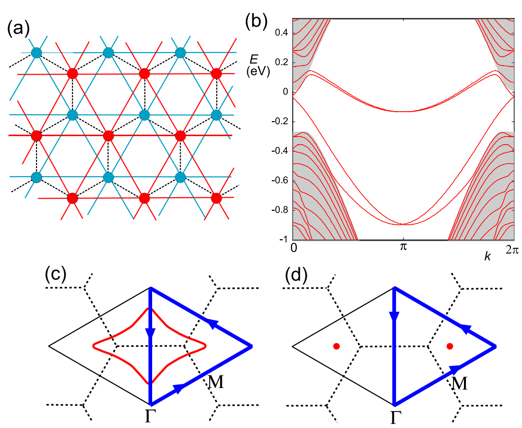

FIG. 2 (color online). Calculation on the bilayer tight-binding model of bismuth in the (111) plane. (a) Crystal structure of the (111)-bilayer Bi. The upper and lower layers are denoted by red and blue, respectively. The solid and broken lines represent intralayer and interlayer hoppings, respectively. (b) Band structure for the strip geometry with 20 sites wide. The gray region is the bulk bands, while the red curves are the states for the strip. All states are doubly degenerate. Zeros of the Pfaffian $\mathrm{Pf}(\boldsymbol{k})$ are shown in red for (c) the inversion-symmetric ($v = 0$) and for (d) the asymmetric ($v = 0.2$ eV) cases.

We now discuss a multilayer Bi thin film. Bismuth is suitable for pursuit of quantum size effects even at room temperature [27], thanks to a long mean-free path ($l \sim \mu$m mm) [28], large electron mobility up to 10^6 cm^2/V s at 5 K [29], and a small effective mass of electrons. Furthermore, Bi thin films can be synthesized with good quality. For example, a 10 μm-thick film shows magnetoresistance with a factor of few thousands at 5 K and a factor of 2–3 at room temperature [29,30].

By stacking N bilayers, the direct gap never closes, and the number of pairs of edge modes becomes N. When the film becomes thicker, the helical edge states are expected to evolve to 2D surface states on a 3D bulk Bi, which has been studied by the angle-resolved photoemission spectroscopy [31–33]. The surface states with large spin splitting are observed [33], which carry spin currents. If the film thickness D is less than the mean-free path l, e.g., $D \sim 10$ μm [29,30]; each of these edge modes has a quantized motion in the perpendicular direction. Hence, although the backscattering is relevant for even N, its effect on the edge states is almost negligible, and the system becomes gapless. By lowering the temperature or by increasing the disorder, the system will eventually become the SHI or the QSH phases depending on whether N is even or odd.

To observe the QSH phase experimentally, one way is to measure the spin current by an applied electric field. We note that the QSH phase does not show a quantization [6], unlike the quantum Hall effect (QHE). On the other hand, surprisingly, the critical exponent ν for the localization length is found to be different between the QSH and the SHI phases [11]. Thus, the QSH phase can be established via measurement of ν. In the QHE, ν is determined experimentally by changing the magnetic field across the plateau transition [34–36], because the change of the magnetic field controls the Fermi energy across the extended state. In the QSH phase, for example, by a change of a gate voltage, one may be able to control the Fermi energy to the extended or localized states. To determine ν, one has to see the range of the gate voltage ΔV_g showing nonzero σ_{xx} by varying the sample size [36] or the temperature [35].

Another way to establish the QSH phase is to observe the edge states by scanning tunneling microscopy or spectroscopy, as has been used for graphite [37,38]. There is one important difference between the edge states of the graphite and the QSH phase. In graphene the existence of edge states crucially depends on the edge shape; the zigzag edge has edge states while the armchair edge does not. For a rough edge with portions of zigzag and armchair edges, only at the zigzag edges can the signal of edge states be seen [38]. In contrast, the edge states in the QSH phase carry helical spin currents, and circulate along the whole edge around, irrespective of the details (e.g., the shape) of the edge.

Besides bismuth, graphite is another material with anomalously large diamagnetic susceptibility [39]. The spin-orbit coupling of graphite is small, and the diamagnetism is mostly carried by orbital motion. In the same token as the SHE, the orbital-angular-momentum (OAM) Hall effect can be studied [40]. The resulting OAM Hall conductivity is the susceptibility for the orbital: $\sigma_{\mathrm{OAM}}^{\mathcal{J}(\mathrm{II})} = \frac{1}{\Omega}\frac{dL_z}{dB_{\mathrm{orb.}}}$. Since this involves only the orbital, the spin-orbit coupling is not required for it to be nonzero. Because the orbital susceptibility is largely enhanced in the graphite due to massless Dirac fermions, graphite will show large OAM current.

In conclusion, we show that the SHC is directly related with a "spin-orbit" susceptibility, a response of the orbital magnetization by the Zeeman field. Hence the magnetic susceptibility can be a good measure for search of quantum spin-Hall systems. We theoretically predict that 2D bismuth will show the quantum spin-Hall effect, because the number of pairs of helical edge states is odd and the Z_2 topological number is nontrivial. The surface states with large spin splitting in bulk bismuth might be closely related with the edge modes.

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[1] M. I. D'yakonov and V. I. Perel', Phys. Lett. **35A**, 459 (1971).
[2] S. Murakami, N. Nagaosa, and S.-C. Zhang, Science **301**, 1348 (2003).
[3] J. Sinova *et al.*, Phys. Rev. Lett. **92**, 126603 (2004).
[4] B. A. Bernevig and S.-C. Zhang, Phys. Rev. Lett. **96**, 106802 (2006).
[5] X.-L. Qi, Y.-S. Wu, and S.-C. Zhang, Phys. Rev. B **74**, 045125 (2006).
[6] C. L. Kane and E. J. Mele, Phys. Rev. Lett. **95**, 146802 (2005); **95**, 226801 (2005).
[7] C. Wu, B. A. Bernevig, and S.-C. Zhang, Phys. Rev. Lett. **96**, 106401 (2006).
[8] C. Xu and J. E. Moore, Phys. Rev. B **73**, 045322 (2006).
[9] M. Onoda and N. Nagaosa, Phys. Rev. Lett. **95**, 106601 (2005).
[10] S. Murakami, N. Nagaosa, and S.-C. Zhang, Phys. Rev. Lett. **93**, 156804 (2004).
[11] M. Onoda, Y. Avishai, and N. Nagaosa, cond-mat/0605510.
[12] L. Smrčka and P. Středa, J. Phys. C **10**, 2153 (1977).
[13] P. Středa, J. Phys. C **15**, L717 (1982).
[14] J. Shi *et al.*, Phys. Rev. Lett. **96**, 076604 (2006).
[15] N. Sugimoto *et al.*, Phys. Rev. B **73**, 113305 (2006).
[16] M.-F. Yang and M.-C. Chang, Phys. Rev. B **73**, 073304 (2006).
[17] In [16] it is commented, without giving details, that even with the conserved spin current there appear extra terms (nonconserved part), which contradicts the present Letter. One can easily check the calculations in the present Letter, and the reason for this difference is not clear.
[18] D. Xiao, J. Shi, and Q. Niu, Phys. Rev. Lett. **95**, 137204 (2005).
[19] T. Thonhauser *et al.*, Phys. Rev. Lett. **95**, 137205 (2005).
[20] H. Fukuyama and R. Kubo, J. Phys. Soc. Jpn. **28**, 570 (1970); F. A. Buot and J. W. McClure, Phys. Rev. B **6**, 4525 (1972).
[21] V. N. Lutskii, Pis'ma Zh. Eksp. Teor. Fiz. **2**, 391 (1965) [JETP Lett. **2**, 245 (1965)].
[22] V. B. Sandomirskii, Zh. Eksp. Teor. Fiz. **52**, 158 (1967) [Sov. Phys. JETP **25**, 101 (1967)].
[23] C. A. Hoffman *et al.*, Phys. Rev. B **48**, 11 431 (1993); H. T. Chu, Phys. Rev. B **51**, 5532 (1995); C. A. Hoffman *et al.*, *ibid.* **51**, 5535 (1995); M. Lu *et al.*, Phys. Rev. B **53**, 1609 (1996).
[24] H. Mönig *et al.*, Phys. Rev. B **72**, 085410 (2005).
[25] Y. Liu and R. E. Allen, Phys. Rev. B **52**, 1566 (1995).
[26] D. N. Sheng *et al.*, Phys. Rev. Lett. **97**, 036808 (2006).
[27] E. I. Rogacheva *et al.*, Appl. Phys. Lett. **82**, 2628 (2003).
[28] D. H. Reneker, Phys. Rev. Lett. **1**, 440 (1958); W. S. Boyle and G. E. Smith, Prog. Semicond. **7**, 1 (1963).
[29] S. Cho *et al.*, Appl. Phys. Lett. **79**, 3651 (2001).
[30] F. Y. Yang *et al.*, Science **284**, 1335 (1999).
[31] C. R. Ast and H. Höchst, Phys. Rev. Lett. **90**, 016403 (2003); Phys. Rev. B **67**, 113102 (2003).
[32] T. K. Kim *et al.*, Phys. Rev. B **72**, 085440 (2005).
[33] Yu. M. Koroteev *et al.*, Phys. Rev. Lett. **93**, 046403 (2004).
[34] B. Huckestein, Rev. Mod. Phys. **67**, 357 (1995).
[35] H. P. Wei *et al.*, Phys. Rev. Lett. **61**, 1294 (1988).
[36] S. Koch *et al.*, Phys. Rev. Lett. **67**, 883 (1991).
[37] Y. Niimi *et al.*, Phys. Rev. B **73**, 085421 (2006).
[38] Y. Kobayashi *et al.*, Phys. Rev. B **71**, 193406 (2005).
[39] J. W. McClure, Phys. Rev. **104**, 666 (1956); **119**, 606 (1960).
[40] S. Zhang and Z. Yang, Phys. Rev. Lett. **94**, 066602 (2005).