Berry phase and the phase of the Shubnikov–de Haas oscillations in three-dimensional topological insulators

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Within the semiclassical approach, we calculate contributions of the Berry phase and of the Zeeman coupling of an electron moment with the magnetic field to the phase of the Shubnikov–de Haas oscillations for the surface electrons in the Bi_2X_3 family of three-dimensional topological insulators (X stands for Te or Se). We also discuss a relation of the obtained results with published experimental data on the Shubnikov–de Haas oscillations for this family of topological insulators.

PACS number(s): 73.20.−r, 71.18.+y, 03.65.Vf

Three-dimensional topological insulators (TI) attract considerable interest as a new state of solids.[1] These materials are insulating in the bulk and are metallic on their surfaces. Recently, a new important class of these materials was discovered that consists of Bi_2X_3 compounds where X stands for Te or Se (Refs. [2] and [3]). The angle-resolved photoemission spectroscopy revealed that the energy bands of the surface electrons are practically linear in the wave vector **k** in these compounds, forming a massless Dirac cone in their surfaces.[2,4,5] It is necessary to emphasize that, although a similar Dirac cone exists in graphene, the surface electron states in the TI essentially differ from the electron states in graphene. The surface states of the TI have no inversion symmetry, and the spin-orbit coupling is not weak in these crystals. These features of the TI lead to a locking of electron spins with **k** for the surface states, giving rise to Dirac fermions without spin degeneracy.

Recently, the quantum oscillations of conductivity in the magnetic field (the Shubnikov–de Haas oscillations) were observed in $(Bi_{1-x}Sb_x)_2Se_3$ (Ref. [6]), Bi_2Te_3 (Refs. [7] and [8]), Bi_2Te_2Se (Refs. [9] and [10]), $Bi_{1.5}Sb_{0.5}Te_{1.7}Se_{1.3}$ (Ref. [11]), and Bi_2Se_3 (Ref. [12]). It was found that the observed oscillations result from the surface electrons of these TI. For such a two-dimensional metal the part of the conductivity describing these oscillations in the semiclassical limit $\delta\sigma_{xx}$ has the form[13–16]

$$\delta\sigma_{xx}(1/H) = \sum_{l=1}^{\infty} A_l \cos\left[l\left(2\pi\frac{F}{H} + \varphi\right)\right], \quad (1)$$

where A_l are positive amplitudes of the harmonics of the periodic in $1/H$ signal, F and φ are the frequency and the phase of the oscillations, the x-y plane coincides with the surface of TI, and the magnetic field $H = H_z$ is perpendicular to this surface. The frequency F is determined by the area $S(\varepsilon_F)$ of the closed orbit of an electron with the Fermi energy ε_F in the space of the wave vectors **k**, $2\pi F = \hbar c S(\varepsilon_F)/e$ (Refs. [13] and [14]), where e is the absolute value of the electron charge. The phase φ is given by the constant γ, $\varphi = -2\pi\gamma$ (Ref. [16]), that enters the semiclassical quantization condition for the electron energy in the magnetic field, Eq. (2). Both F and φ can be experimentally determined by Fourier analysis of $\delta\sigma_{xx}(1/H)$ (Ref. [17]). It was found in the experiments[8,10,12] that the phase of the oscillation φ practically coincides with the appropriate phase for the electrons in graphene (i.e., $\varphi \approx 0$).

On the other hand, this phase measured in other experimental investigations[6,7,9,11] generally differs both from the phase for the usual two-dimensional electron gas ($\varphi = -\pi$) and from the phase in graphene. As will become clear below, the constant γ (i.e., φ) depends on the so-called Berry phase[18] of electrons and on the Zeeman coupling of an electron moment with the magnetic field. In Refs. [6], [7], and [9] it is the Zeeman coupling that was discussed as a possible source of the nonzero φ. Taskin and Ando[19] suggested that a deviation of the dispersion relation $\varepsilon(\mathbf{k})$ for the surface charge carriers from an ideal cone can shift their Berry phase Φ_B from its usual value π characteristic of the Dirac electrons and hence can change the phase of the oscillations φ as well. In this Brief Report, using the semiclassical approach, we calculate both the Berry phase and a contribution of the Zeeman coupling to the constant γ for the surface electrons in the Bi_2X_3 family of TI. We also discuss a relation of these results with the published experimental data.

The semiclassical quantization condition for the energy levels ε_n of an electron in the magnetic field H reads[14,20]

$$S(\varepsilon_n) = \frac{2\pi eH}{\hbar c}(n + \gamma), \quad (2)$$

where n is a large integer. The quantization condition (2) is obtainable from the one-band Hamiltonian[21] of a semiclassical electron in the magnetic field. This Hamiltonian $\hat{H}$ can be schematically represented in the form

$$\hat{H} = \varepsilon_0(\mathbf{k}) - \frac{eH}{c}\left(\mu_0^{(1)} + \mu_0^{(2)}\right), \quad (3)$$

where the subscript 0 marks a two-dimensional electron-surface band under study, the term

$$\mu_0^{(1)}(\mathbf{k}) = [\mathbf{v}_0 \times \mathbf{\Omega}_0]_z \quad (4)$$

is proportional to the intraband part of the orbital electron moment, $\mathbf{v}_0 = (1/\hbar)(\partial\varepsilon_0/\partial\mathbf{k})$ is the electron velocity,

$$\mathbf{\Omega}_0(\mathbf{k}) = i\int u_{\mathbf{k},0}^*(\mathbf{r})\frac{\partial}{\partial\mathbf{k}}u_{\mathbf{k},0}(\mathbf{r})d\mathbf{r}, \quad (5)$$

and $\mu_0^{(2)}$ is the sum of the z components of the interband orbital moment of the electron and of its spin. In Eq. (5) the integration is carried out over a unit cell of the crystal lattice, and $u_{\mathbf{k},0}(\mathbf{r})$ is the periodic factor in the Bloch wave function of the zeroth band

$$\psi_{\mathbf{k},0} = \exp(i\mathbf{k}\mathbf{r})u_{\mathbf{k},0}.$$

Before proceeding to TI, let us briefly review the possible situations that can occur in crystals and that have an effect on the form of Eq. (2). If the spin-orbit interaction in a crystal is negligible, and if the electron states under study have the inversion symmetry, the constant γ is given by the formula[22]

$$\gamma = \frac{1}{2} - \frac{\Phi_B}{2\pi}, \qquad (6)$$

where Φ_B is the Berry phase of the electron orbit Γ

$$\Phi_B = \oint_\Gamma \mathbf{\Omega}_0 d\mathbf{k}, \qquad (7)$$

$d\mathbf{k} \equiv d\kappa[\mathbf{i}_z \times \mathbf{v_0}]/|\mathbf{v_0}|$; $d\kappa$ is the length of an infinitesimal element of the orbit Γ, and $\mathbf{i}_z$ is the unit vector parallel to $\mathbf{H}$. It is this situation that occurs in graphene. The part $\Phi_B/2\pi$ of Eq. (6) results from the term $\mu_0^{(1)}$ in Hamiltonian (3). The term $\mu_0^{(2)}$ is identically equal to zero in this case (if one neglects the electron spin). As was shown in our paper,[22] the Berry phase Φ_B of an electron is always equal to $\pm\pi$ when it moves around a Dirac point (i.e., if the electron orbit Γ surrounds this point). Otherwise, one has $\Phi_B = 0$. Importantly, the Berry phase does not depend on the shape and the size of the electron orbit since the inversion and time-reversal symmetries lead to rot $\mathbf{\Omega}_0 = 0$ everywhere except the Dirac point. In other words, the result $\Phi_B = \pm\pi$ remains true at *any dependence $\varepsilon_0(\mathbf{k})$ in the vicinity of the orbit*, and γ can take on only the universal values 0 or 1/2.

If the spin-orbit interaction is not weak, but still there is an inversion symmetry for the electron states, all these states are double degenerate in spin. In this case one has $\gamma = 1/2$, but the quantization rule (2) contains an additional term associated with the so-called electron g factor, g (i.e., in this case $\gamma \rightarrow 1/2 \pm gm_*/4m$ where m is the electron mass and m_* is its cyclotron mass). One part of this g factor is still determined by the Berry phase, whereas its second part depends on the term $\mu_0^{(2)}$ which is not vanishing now. The complete theory of the g factor for electrons in metals was presented in our papers.[23,24] It is important that if the strength of the spin-orbit interaction is small, the g factor approaches a limiting form that leads to the same electron spectrum described by Eq. (2) with $\gamma = 1/2$ or 0 as in the absence of this interaction.[25]

When the inversion symmetry is absent, the formulas for the constant γ can be derived by a simple modification of the expressions given in Refs. 23 and 24. In particular, when the spin-orbit interaction is not weak, and hence when the electron states are not degenerate in the electron spin, we obtain

$$\gamma - \frac{1}{2} = -\frac{1}{2\pi}\oint_\Gamma \mathbf{\Omega}_0 d\mathbf{k} - \frac{1}{2\pi}\oint_\Gamma \frac{\mu_0^{(2)}(\mathbf{k})}{v_0(\mathbf{k})}d\kappa. \qquad (8)$$

Here the first integral coincides with $-\Phi_B/2\pi$, while the second integral can be considered as a result of the Zeeman coupling of the electron moment $\mu_0^{(2)}$ with the magnetic field. For the *ideal* Dirac cone, Φ_B is still equal to $\pm\pi$ when the electron orbit surrounds the Dirac point.[18] But if the electron band $\varepsilon_0(\mathbf{k})$ is not described by a strictly linear dependence in the whole energy interval from the Dirac point to the Fermi energy ε_F, this result for Φ_B generally fails, and the Berry phase may differ from π. It is this possibility that was discussed in Ref. 19

since the spin-orbit interaction is strong, and the inversion symmetry is broken for the electron surface states of TI.

The functions $\mathbf{\Omega}_0(\mathbf{k})$ and $\mu_0^{(2)}(\mathbf{k})$ can be calculated[23,25] using a $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian of electrons at a point $\mathbf{k}_0$ located near the electron orbit. This calculation is also possible with an effective two-band Hamiltonian that contains nonlinear terms in $\mathbf{k} - \mathbf{k}_0$ (Refs. 24 and 26). Such terms are usually introduced into the Hamiltonian to take into account the other bands different from the two considered explicitly. In the magnetic field H the effective two-band Hamiltonian should also contain a term linear in H. This term is required for the accurate calculation of $\mu_0^{(2)}(\mathbf{k})$.

In Refs. 27 and 28 the effective two-band Hamiltonian was found for the surface electrons of the Bi_2X_3 family in the vicinity of the center of their two-dimensional Brillouin zone,

$$\hat{H} = bk^2\hat{\mathbf{1}} + v(k_y\hat{\sigma}_x - k_x\hat{\sigma}_y) + c(k_+^3 + k_-^3)\hat{\sigma}_z + \frac{\mu_B}{2}g_{sz}H_z\hat{\sigma}_z, \qquad (9)$$

where b, v, c, and g_{sz} are some material-dependent parameters; μ_B is the Bohr magneton; $k^2 = k_x^2 + k_y^2$; $k_\pm = k_x \pm ik_y$; $\hat{\mathbf{1}}$ is the unit matrix; and $\hat{\sigma}_i$ are the Pauli matrices. At $H = 0$ this Hamiltonian describes the conduction and valence surface bands

$$\varepsilon_{c,v}(\mathbf{k}) = bk^2 \pm E,$$
$$E^2 \equiv v^2k^2 + 4c^2k_x^2(k_x^2 - 3k_y^2)^2, \qquad (10)$$

which touch each other at the point $k = 0$, and which we denote by the indexes c and v. Note that these bands are not strictly linear functions of k. They also contain the quadratic and cubic corrections to the Dirac cone. The values of the parameters defining Eq. (9) for the crystals of the Bi_2X_3 family were estimated in Ref. 28.

Using Hamiltonian (9) and formulas (A2), (A3), (A7), (A8) of our paper,[23] we calculate $\mu^{(2)}(\mathbf{k})$ and $\mathbf{\Omega}(\mathbf{k})$, for example, for the conduction band c,

$$(\Omega_c)_{x,y} = \pm\frac{v^2k_{y,x}}{2E[E + 2ck_x(k_x^2 - 3k_y^2)]}, \qquad (11)$$

$$\mu_c^{(2)}(\mathbf{k}) = \frac{ck_x(k_x^2 - 3k_y^2)}{E}\left(\frac{2v^2}{E\hbar} - \frac{\hbar g_{zs}}{2m}\right), \qquad (12)$$

where m is the electron mass, and the signs $+$ and $-$ correspond to $(\Omega_c)_x$ and $(\Omega_c)_y$, respectively. Note that $\mu_c^{(2)}$ is proportional to the parameter c defining the hexagonal warping term in Hamiltonian (9). Without this term, the electron moment lies in the x-y plane,[27] and so it has no component along the magnetic field.

Using formulas (11) and (12) and an expression for the velocity $v_c = |\partial\varepsilon_c(\mathbf{k})/\partial\mathbf{k}|$ that is directly obtainable from Eq. (10), one can find γ, calculating the integrals over the orbit Γ in Eq. (8). This orbit is defined by the condition $\varepsilon_c(\mathbf{k}) = \varepsilon_F$ where the constant ε_F is the Fermi energy. It is significant that the orbit is symmetric relative to the transformation $k_x \rightarrow -k_x$, $k_y \rightarrow -k_y$, while the term $\mu_c^{(2)}$ is antisymmetric to this transformation. Thus, the second integral in Eq. (8) is always equal to zero. Representing $\mathbf{\Omega}$ as a sum of the symmetric $\mathbf{\Omega}^s \equiv [\mathbf{\Omega}(\mathbf{k}) + \mathbf{\Omega}(-\mathbf{k})]/2$ and antisymmetric $\mathbf{\Omega}^a \equiv [\mathbf{\Omega}(\mathbf{k}) - \mathbf{\Omega}(-\mathbf{k})]/2$ parts, we find that only the antisymmetric

part gives a nonzero contribution to γ. A direct calculation with Eq. (11) yields

$$\left(\Omega_c^a\right)_{x,y} = \pm\frac{k_{y,x}}{2k^2}. \tag{13}$$

This $\mathbf{\Omega}_c^a$ leads to $\Phi_B = -\pi$, and hence one obtains $\gamma = 0$ for any electron orbit in the conduction band (the values $\gamma = 1$ and $\gamma = 0$ are equivalent in the semiclassical limit). A similar calculation gives $\Phi_B = \pi$, and $\gamma = 0$ for the valence band. Interestingly, if the Dirac cone in the Brillouin zone were shifted from the point $\mathbf{k} = 0$ or were deformed asymmetrically, the Berry phase and the Zeeman term in Eq. (8) would not have the universal values and would depend on ε_F, see below.

In fact, the obtained result $\gamma = 0$ is exclusively caused by the time-reversal symmetry of the Hamiltonian. This symmetry alone dictates that any electron orbit is symmetric relative to the transformation $\mathbf{k} \to -\mathbf{k}$, and that the Hamiltonian should have the form

$$\hat{H} = h_0(\mathbf{k})\hat{\mathbf{1}} + \sum_{i=1}^{3} h_i(\mathbf{k})\hat{\sigma}_i$$
$$+ \frac{\mu_B}{2}H_z\left[g_0(\mathbf{k})\hat{\mathbf{1}} + \sum_{i=1}^{3}g_i(\mathbf{k})\hat{\sigma}_i\right], \tag{14}$$

where the arbitrary functions $h_0(\mathbf{k})$, $h_i(\mathbf{k})$ and $g_0(\mathbf{k})$, $g_i(\mathbf{k})$ satisfy only the requirements: $h_0(\mathbf{k}) = h_0(-\mathbf{k})$, $g_0(-\mathbf{k}) = -g_0(\mathbf{k})$, and $h_i(\mathbf{k}) = -h_i(-\mathbf{k})$, $g_i(-\mathbf{k}) = g_i(\mathbf{k})$. With Eq. (14) we find the following expressions for $\mathbf{\Omega}_c(\mathbf{k})$ and $\mu_c^{(2)}(\mathbf{k})$ generalizing formulas (11) through (13)

$$\mathbf{\Omega}_c^a = \frac{1}{2\left(h_1^2 + h_2^2\right)}\left(h_2\frac{\partial h_1}{\partial \mathbf{k}} - h_1\frac{\partial h_2}{\partial \mathbf{k}}\right), \tag{15}$$

$$\mathbf{\Omega}_c^s = -\frac{h_3}{\sqrt{h_1^2 + h_2^2 + h_3^2}}\,\mathbf{\Omega}_c^a, \tag{16}$$

$$\mu_c^{(2)} = -\frac{1}{2\hbar\left(h_1^2 + h_2^2 + h_3^2\right)}\sum_{i,j,l=1}^{3}\varepsilon_{ijl}h_i\frac{\partial h_j}{\partial k_x}\frac{\partial h_l}{\partial k_y}$$
$$- \frac{\hbar}{4m}\left(g_0 + \frac{1}{\sqrt{h_1^2 + h_2^2 + h_3^2}}\sum_{i=1}^{3}g_ih_i\right), \tag{17}$$

where ε_{ijl} is the completely antisymmetric unit tensor with $\varepsilon_{123} = 1$. A direct calculation shows that $\text{rot}\,\mathbf{\Omega}_c^a = 0$ everywhere except the Dirac point for any functions $h_1(\mathbf{k})$, $h_2(\mathbf{k})$. Hence, for the calculation of the Berry phase, it is sufficient to consider $\int_\Gamma \mathbf{\Omega}_c^a d\mathbf{k}$ for a small orbit Γ in the immediate vicinity of the Dirac point where these functions reduce to linear ones. For linear functions $h_1(\mathbf{k})$, $h_2(\mathbf{k})$ one indeed obtains $\Phi_B = \pm\pi$. As to the term $\mu_c^{(2)}(\mathbf{k})$, it is antisymmetric relative to the transformation $\mathbf{k} \to -\mathbf{k}$ and does not contribute to γ. Nevertheless, formulas (14)–(17) which are, in principle, applicable to a general case (i.e., not only to the Bi_2X_3 family of TI) can lead to nontrivial values of γ if the appropriate electron orbits in some TI become asymmetric with respect to the point $\mathbf{k} = 0$.

The asymmetry of the orbits can appear when the external magnetic field is not strictly perpendicular to the surface of

TI (i.e., when it has an in-plane component $\mathbf{H}_\parallel$). In this case Hamiltonian (9) contains an additional term[27]

$$\delta\hat{H} = g_\parallel\frac{\mu_B}{2}(H_x\sigma_x + H_y\sigma_y), \tag{18}$$

where $g_\parallel$ is the in-plane g factor, $g_\parallel \sim 1$ (Ref. [28]). This term shifts the Dirac cone of the surface electrons in their Brillouin zone from the point $\mathbf{k} = 0$ to the point $\mathbf{k}_* = g_\parallel(\mu_B/2v)[\mathbf{H} \times \mathbf{i}_z]$ (Ref. [27]), and the electron orbits become asymmetric. This asymmetry generally leads to a nonzero value of γ. However, in the first order in the small parameter $g_\parallel\mu_BH_\parallel/\varepsilon_F$ the correction to γ vanishes for the case of Hamiltonian (9) since the angular dependence $\cos 3\phi$ characteristic of the factor $h_3(\mathbf{k})$ in Eq. (16) differs from the angular dependence $\cos\phi$ (or $\sin\phi$) that describes the deformation of the electron orbits due to the field $H_\parallel$. Here ϕ is the angle between $\mathbf{k}$ and the x axis. Due to this difference in the angular dependences, the appropriate integrals over ϕ determining the correction vanish. Thus, for the family Bi_2X_3 of TI the effect of the in-plane magnetic field on γ is negligible. This conclusion is in an agreement with the experimental data[6,7,9] showing that the phase of the Shubnikov–de Haas oscillations does not alter after applying the field $H_\parallel$.

We now discuss the relation of the obtained results with the published experimental data. In Ref. [6] it was shown that in $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Se}_3$ the electron Landau levels ε_n at small n can be well described by the formula

$$\varepsilon_n = \pm\sqrt{\left(\frac{\mu_B}{2}g_{sz}H\right)^2 + \frac{2neHv^2}{c\hbar}}, \tag{19}$$

where $g_{sz} \approx 50$. This formula gives the exact Landau levels of Hamiltonian (9) at $b = c = 0$. Let us rewrite expression (19) in the form of quantization condition (2)

$$S(\varepsilon_n) = \frac{2\pi eH}{\hbar c}\left(n + \frac{Hg_{sz}^2\mu_B^2c\hbar}{8ev^2}\right), \tag{20}$$

where $S(\varepsilon) = \pi k^2 = \pi(\varepsilon/v)^2$ is the area of the electron orbit in the $\mathbf{k}$ space, and consider the case of large n (i.e., of relatively small H). This case just corresponds to the semiclassical approximation used in our calculations. Now the second term in the right-hand side of formula (20) is small as compared to the first one, and we may express H via n as follows: $H \approx \hbar cS/(2\pi en)$. Inserting this expression for H in the second term of Eq. (20), we arrive at

$$S(\varepsilon_n) \approx \frac{2\pi eH}{\hbar c}\left(n + \frac{Sg_{sz}^2\mu_B^2c^2\hbar^2}{16\pi e^2v^2}\frac{1}{n}\right). \tag{21}$$

Strictly speaking, the right-hand side of the quantization condition (2) is a series in $1/n$ [i.e., it has the form $n + \text{const} + (1/n) + \cdots$] in which only the first two terms $n + \gamma$ are usually kept. It is this cut of the series that yields Eq. (2). Formula (21) shows that spectrum (19) in the semiclassical limit ($n \gg 1$) leads to $\gamma = 0$ in agreement with our results obtained above and that in contrast with the case of the usual electron gas, a finite g_{sz} in Eqs. (9) and (19) gives a contribution only to the term of the order of $1/n$. In other words, the nonzero values of γ found in the experiments[6,7,9] are due to the difference of the exact spectrum from the semiclassical one at not too large values of n. This conclusion is supported by an

analysis of the Shubnikov–de Haas oscillations carried out by Taskin and Ando[19] for various TI. Note that in graphene one has a relatively small value of $g_{sz} \sim 2$, and the semiclassical spectrum defined by Eq. (2) practically coincides with the exact spectrum (19) even at $n = 0$ and 1. The large value of $g_{sz} \approx 50$ for $(Bi_{1-x}Sb_x)_2Se_3$ is probably due to a relatively small gap between the Dirac-point energy and the energies of some other surface bands at $\mathbf{k} = 0$ for this TI.

In summary, within the semiclassical approach, we derive formula (8) for the constant γ that enters the quantization condition (2) and defines the phase $\varphi = -2\pi\gamma$ of the Shubnikov–de Haas oscillations in TI. This γ is determined by the Berry phase of an electron orbit and by a part of the electron moment averaged over the orbit. Since the electron orbits are symmetric with respect to the Dirac point $\mathbf{k} = 0$, we find that the Berry phase is equal to π, while the averaged moment is zero, and hence one always has $\gamma = 0$ for the Bi_2X_3 family of TI. The nonzero values of γ found in some experiments appear to be due to the difference of the exact electron spectrum from the semiclassical one at not too large quantizing numbers n.

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