Berry Phase and de Haas–van Alphen Effect in $LaRhIn_5$

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We explain the experimental data on the magnetization of $LaRhIn_5$ recently published by Goodrich *et al.* [Phys. Rev. Lett. **89**, 026401 (2002)]. We show that the magnetization of a small electron group associated with a band-contact line was detected in that Letter. These data provide the first observation of the Berry phase of electrons in metals via the de Haas–van Alphen effect.

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In recent years the concept of the so-called Berry phase [1] has attracted considerable attention thanks to its fundamental origin; see, e.g., Refs. [2,3], and references therein. According to Berry, if a Hamiltonian of a quantum system depends on parameters, and if the parameters undergo adiabatic changes so that they eventually return to their original values, the wave function of the system can acquire the so-called geometrical phase in addition to the familiar dynamical one. This additional phase (the Berry phase) differs from zero when the trajectory Γ of the system in the parameter space is located near a *point* at which the states of the system are degenerate [1]. In analyzing this situation, Berry assumed that the Hamiltonian of the system is a Hermitian matrix which is linear in deviations of the parameters from the point, and he presented his final result in the pictorial form. He found that such a point can be considered as a "monopole" in the parameter space when the geometrical phase is calculated. In other words, the point "generates" a field which coincides in the form with that of the monopole, and the flux of this Berry field through the contour Γ gives the geometrical phase of the system. Evidence for this phase was obtained in experiments with various physical systems [2,3]. However, an experimental observation of the Berry phase for electrons in crystals has proved a challenging problem (some progress in this direction was achieved only recently [4–6]).

It is well known (see, e.g., Ref. [7]) that the semiclassical motion of an electron in a crystal in the magnetic field H can be represented as the motion of the wave vector $\mathbf{k}$ in an orbit in the Brillouin zone. This orbit is the intersection of the constant-energy surface, $\varepsilon(\mathbf{k}) =$ const, with the plane, $k_z =$ const, where z is the direction of the magnetic field $\mathbf{H}$ and $\varepsilon(\mathbf{k})$ is the electron dispersion relation in the crystal. Berry's result is applicable to such an electron, with the Brillouin zone playing the role of the parameter space [8]. However, in crystals with the inversion symmetry and a weak spin-orbit interaction, the Berry phase of the electrons has specific features [9] which are due to the fact that the electron states are invariant under the simultaneous inversion of time and spatial coordinates. This invariance permits one to transform the Hermitian Hamiltonian of the electron into the real form for any point of the Brillouin zone. As a consequence, the character of the energy-band degeneracy differs from that considered by Berry. Now the electron energy bands $\varepsilon_l(\mathbf{k})$ contact along *lines* in the Brillouin zone, and the lines need not be symmetry axes [10]. In other words, the degeneracy is not lifted along these lines, and the monopole in the $\mathbf{k}$ space disappears. As it was shown in our Letter [9], in such a situation the band-contact lines play the role of infinitely thin "solenoids" which generate the Berry field with the flux $\pm\pi$. Although this field is zero outside the solenoids, if the electron orbit surrounds the line, the flux threads the orbit, and the electron acquires the Berry phase $\phi_B = \pm\pi$ when it moves around this line. It is clear that in this case the Berry phase does not depend on the shape and the size of the electron orbit but is specified only by its topological characteristics (there is either a linking of the orbit with the band-contact line or there is not).

The Berry phase of the electron modifies [9] the well-known semiclassical quantization rule [11] for the electron energy in the magnetic field, ε,

$$S(\varepsilon, k_z) = \frac{2\pi e H}{\hbar c}(n + \gamma), \tag{1}$$

where S is the cross-sectional area of the closed electron orbit in the $\mathbf{k}$ space, n is a large integer ($n > 0$), e is the absolute value of the electron charge, and the constant γ is given by

$$\gamma = \frac{1}{2} - \frac{\phi_B}{2\pi}. \tag{2}$$

The meaning of formula (2) is the following: When the electron makes a complete circuit in its orbit, the change of the phase of its wave function consists of the usual semiclassical part $\hbar cS/eH$, the shift $-\pi$ associated with the so-called turning points of the orbit where the semiclassical approximation fails, and the Berry phase. Equating this change to $2\pi n$, one arrives at Eqs. (1) and (2). Thus, when the electron orbit links to the band-contact line, one obtains $\gamma = 0$ (the values $\gamma = 0$ and $\gamma = 1$ are equivalent) instead of the usual value [11]

$\gamma = 1/2$. This change of γ has to manifest itself in the de Haas–van Alphen effect [9].

In a recent experimental investigation [12] of the de Haas–van Alphen effect in $LaRhIn_5$, the oscillations of magnetization associated with a small cross section of the Fermi surface of this metal were detected. The electron cyclotron mass m^* corresponding to this cross section was also small compared with electron mass m, $|m^*| \approx 0.067m$. Authors of that paper attributed these oscillations to a small electron pocket of the Fermi surface. Besides, in that paper, the magnetization of the electrons of the pocket was studied in the ultraquantum limit, and the following intriguing contradiction between the obtained experimental data and the existing theory was discovered: Since in the ultraquantum limit the electrons occupy only the lowest Landau level, they have to migrate into large sheets of the Fermi surface when this level is raised above the Fermi energy. Hence, the magnetization M of the electrons of the pocket has to vanish with increasing magnetic field H. However, the experimental data [12] reveal a finite contribution to the magnetization even in such magnetic fields.

In this Letter, we resolve this contradiction. We show that in fact a small electron group associated with a band-contact line was detected in Ref. [12], and the results of Ref. [12] provide the first observation of the Berry phase of the electrons via the de Haas–van Alphen effect.

A small cross section of a Fermi surface appears near that point $\mathbf{k}_0$ of the Brillouin zone for which the two conditions are fulfilled: Topology of this surface changes at the $\mathbf{k}_0$ if the Fermi energy ε_F is shifted past some critical energy ε_0, and this ε_0 is close to the initial Fermi level of the crystal. In the case of the degeneracy of two electron energy bands of the crystal [say, $\varepsilon_+(\mathbf{k})$ and $\varepsilon_-(\mathbf{k})$] along a line in the Brillouin zone, these bands near the $\mathbf{k}_0$ always can be represented in the form [13]

$$\varepsilon_\pm(\mathbf{k}) = \varepsilon_0 + \frac{\hbar^2 k_3^2}{2m_3} + \hbar(\mathbf{v}_\perp \cdot \mathbf{k}) \pm \sqrt{(\hbar V_1 k_1)^2 + (\hbar V_2 k_2)^2}, \quad (3)$$

where the wave vector $\mathbf{k}$ is measured from the $\mathbf{k}_0$; the constants m_3, V_1, V_2, and $\mathbf{v}_\perp = (v_1, v_2, 0)$ are some parameters of the spectrum; the k_3 axis coincides with the tangent to the band-contact line at the point $\mathbf{k}_0$; and the $\mathbf{k}_0$ is defined by the condition that the band energies in the line [$\varepsilon_+(\mathbf{k}) = \varepsilon_-(\mathbf{k})$ there] reach the extremal value ε_0 at this point. A small extremal cross section can exist only under the condition $a_\perp^2 \equiv (v_1/V_1)^2 + (v_2/V_2)^2 < 1$, which we imply to hold below.

If $\mathrm{sgn}(m_3)(\varepsilon_F - \varepsilon_0) < 0$, the Fermi surface has the shape of a neck, with the band-contact line being inside the neck [Fig. 1(b)]. Here $\mathrm{sgn}(z) = 1$ for $z > 0$ and $\mathrm{sgn}(z) = -1$ if $z < 0$. As the Fermi energy passes the critical energy, $\mathrm{sgn}(m_3)(\varepsilon_F - \varepsilon_0) > 0$, the neck is broken,

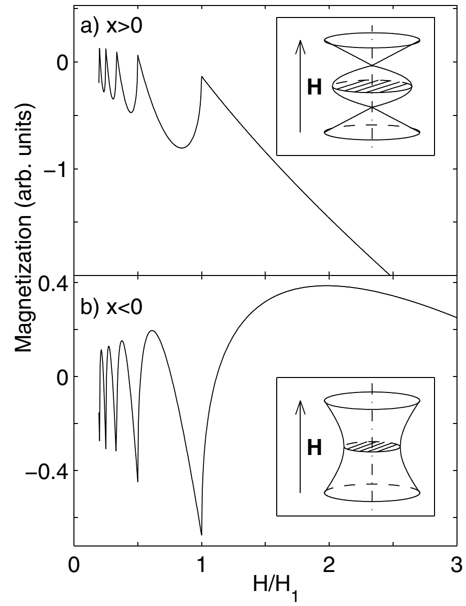

FIG. 1. The electron magnetization associated with the band-contact line, Eq. (4), for $x > 0$ (a) and for $x < 0$ (b). A sign of x coincides with the sign of $(\varepsilon_F - \varepsilon_0)m_3$. The electron spectrum is described by Eq. (3) with $a_\perp^2 \equiv (v_1/V_1)^2 + (v_2/V_2)^2 < 1$. The field H_1 is given by Eq. (6). The insets show the appropriate Fermi surfaces and the extremal cross sections; the dash-dotted lines depict the band-contact line.

and immediately a new pocket appears, i.e., the Fermi surface takes the self-intersecting shape, with the band-contact line still lying inside it [Fig. 1(a)]. Thus, in the case of the degeneracy of the bands, the $\mathbf{k}_0$ is the point where a self-intersecting Fermi surface appears (or disappears) at $\varepsilon_F = \varepsilon_0$.

Let the magnetic field H be in the k_3 direction. In this case, the exact Landau levels were found in a vicinity of the critical energy ε_0 [14]. Interestingly, these exact levels obtained from the appropriate Schrödinger equation coincide with the semiclassical levels, given by Eq. (1) and $\gamma = 0$, at *all* n (even at $n \sim 1$), and not just for $n \gg 1$. On the basis of this spectrum, the magnetization M of the electrons with spectrum (3) was calculated for an *arbitrary* strength of H [14]:

$$M = -\left(\frac{e}{c}\right)^2 \frac{2^{3/2}|m_3(\varepsilon_F - \varepsilon_0)|^{1/2}}{\pi^2 \hbar |m^*|} H_1^{1/4} H^{3/4} f(x), \quad (4)$$

where

$$x = \mathrm{sgn}[m_3(\varepsilon_F - \varepsilon_0)](H_1/H)^{1/2},$$

and the universal function $f(x)$ is completely independent of the spectrum,

$$f(x) = \frac{1}{4}\int_{-x}^{\infty} dt \left(\frac{1}{2} - \{t^2\}\right) \mathrm{sgn}(t) \frac{7t + 6x}{\sqrt{x + t}}. \quad (5)$$

Here $\{z\}$ means the fractional part of the number z. The

H_1 is one of the fields given by the formula

$$\frac{e}{c\hbar}H_n = \frac{S_{\rm ex}}{2\pi}\frac{1}{n} = \frac{m^*}{2\hbar^2}\frac{(\varepsilon_F - \varepsilon_0)}{n}, \quad (6)$$

where $n = 1, 2, \ldots$, $S_{\rm ex}$ is the area of the extremal cross section of the Fermi surface (see Fig. 1), and the cyclotron mass m^* is proportional to $\varepsilon_F - \varepsilon_0$,

$$m^* = \frac{\varepsilon_F - \varepsilon_0}{V_1 V_2 (1 - a_\perp^2)^{3/2}}, \quad (7)$$

and is small compared to the electron mass m at $|\varepsilon_F - \varepsilon_0| \ll m V_1 V_2 \sim 1$–10 eV. The meaning of the fields H_n will become clear from the subsequent analysis. Note that we consider $M(H)$ at a fixed ε_F since, in a metal, large sheets of its Fermi surface provide such large density of states that ε_F is practically independent of the magnetic field.

If $|x| \gg 1$, the magnetization (4) splits into the oscillation part, which completely agrees with the well-known Lifshits-Kosevich formula [11,15], and the smooth contribution χH, with the magnetic susceptibility χ coinciding with that of Ref. [13]. At low temperatures,

$$\delta\varepsilon_H \equiv \frac{e\hbar H}{|m^*|c} \gg 2\pi^2 T, \quad (8)$$

the oscillations of the magnetization M prevail over the smooth part, many harmonics in the Lifshits-Kosevich formula are relevant, and sharp peaks of M occur when the Landau levels cross the Fermi energy. It is the field H_n defined by Eq. (6) that gives the position of the peak at crossing ε_F by the nth Landau level. Note that at $x > 0$ the oscillations of M result from the *maximum* cross section of the pocket with $k_3 = 0$, and the peaks of the magnetization are directed *upward* [Fig. 1(a)], while at $x < 0$ the oscillations result from the *minimum* cross section of the neck with $k_3 = 0$, and the peaks are directed *downward* [Fig. 1(b)].

Formula (6) provides the possibility to find the band-contact lines in metals, using the Shoenberg procedure [11]. Plotting experimental values of $1/H_n$ versus n, one can state that a band-contact line has been detected if this dependence is extrapolated to the origin of the coordinate. If the γ were different from zero, the dependence would be extrapolated to $-\gamma$. We emphasize that since for the electrons near the point $\mathbf{k}_0$ the exact spectrum in the magnetic field coincides with the semiclassical spectrum, formula (6) defines the positions of the peaks in $M(H)$ not only for large n but also for $n \sim 1$. Thus, it is sufficient to use several last oscillations of $M(H)$ in this detection. This enables one to find γ with maximal accuracy. Note also that the observation of the sharp peaks for the last oscillations ($n \sim 1$) is the most favorable since $\delta\varepsilon_H \sim |\varepsilon_F - \varepsilon_0|/n$ in Eq. (8).

The ultraquantum limit occurs when $|x| \leq 1$, i.e., when $H \geq H_1$. In this limit the magnetization $M(H)$ cannot be decomposed into the oscillation parts and the term χH [e.g., $f(x) \approx 0.156$ at $|x| \ll 1$]. In this case the magnetization is determined by the Landau levels of the lower band $\varepsilon_-(\mathbf{k})$, which are *all* occupied by the electrons. It is important that at $H \sim H_1$ the magnetization far exceeds that of the usual small pockets and necks which contain no band-contact line. Indeed, for such the pockets and necks the appropriate magnitude of M is of the order of the prefactor before the function $f(x)$ in Eq. (4), but m^* in such situations is not proportional to $\varepsilon_F - \varepsilon_0$ and generally not small, $m^* \sim m$. Moreover, at $H \sim H_1$ the M in Eq. (4) generally *is not small* compared with the smooth part of the magnetization, $\chi_0 H_1$, caused by the *large electron groups* in metals. Estimating the smooth part of the magnetic susceptibility of the large electron groups, χ_0, by the Landau formula for the electron gas [11], $\chi_0 \sim (e/c)^2(\varepsilon_F)^{1/2}/\hbar\sqrt{m}$, we find that, at $H \sim H_1$,

$$\frac{M(H)}{\chi_0 H} \sim \frac{m V_1 V_2}{(\varepsilon_F|\varepsilon_F - \varepsilon_0|)^{1/2}} \gg 1.$$

In the last inequality, we have assumed that $m V_1 V_2$ and ε_F are of the order of the characteristic energies in metals, 1–10 eV.

Above we have neglected the spin-orbit interaction in the crystal. With this interaction, the semiclassical quantization rule (1) is modifies as follows [11]:

$$S(\varepsilon, k_z) = \frac{2\pi e H}{\hbar c}\left(n + \gamma \pm \frac{g m^*}{4m}\right), \quad (9)$$

where $\gamma = 1/2$, and g is the so-called g factor of the electron orbit. Besides, the spin-orbit interaction generally lifts the degeneracy of the bands $\varepsilon_+(\mathbf{k})$ and $\varepsilon_-(\mathbf{k})$. But if this interaction is not too strong, so that the gap between these bands is essentially smaller than the energy gaps between $\varepsilon_\pm(\mathbf{k})$ and other bands of the crystal, the concept of the band-contact line is still valid approximately. As it was shown in our paper [16], if the semiclassical electron orbit in the magnetic field surrounds such a band-contact line, one has $g \approx 2m/m^*$, and formula (9) is equivalent to Eq. (1) with $\gamma = 0$ for all n. In other words, Eq. (6) for the peak positions is robust to "switching on" the spin-orbit interaction. Note that the g factor is *large even for a very weak spin-orbit interaction*, and this result is the other manifestation of the nonzero Berry phase (instead of $\gamma = 0$). The spin-orbit interaction also modifies formula (4) [14]. However, if the splitting of the bands $\varepsilon_\pm(\mathbf{k})$ is small near the point $\mathbf{k}_0$, the modification is negligible, and it increases rather *slowly* with the strength of the spin-orbit interaction.

We now apply the above results to the experimental data of Ref. [12]. These data obtained at a low temperature (1.5 K) reveal the sharp peaks in the magnetization of $LaRhIn_5$ when the magnetic field H is parallel to the [001] direction of this tetragonal compound (Fig. 2). The analysis of the peak positions (see the inset of Fig. 2)

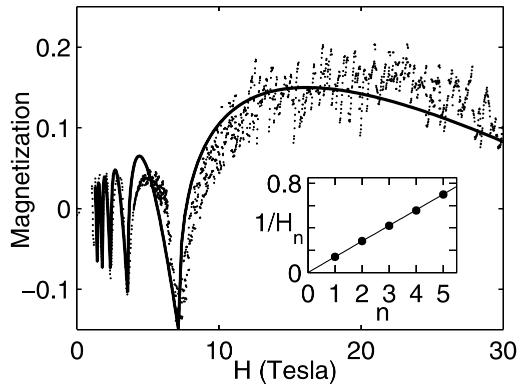

FIG. 2. The experimental data [12] on the magnetization of $LaRhIn_5$ (dots) and the magnetization calculated from Eqs. (4) and (10) (solid line) for $\mathrm{sgn}(m_3)(\varepsilon_F - \varepsilon_0) = -25$ meV, $|m^*| = 0.067m$ ($H_1 = 7$ T), and χ_0 given in the text. The inset shows the dependence of the experimental values of $1/H_n$ [12] on n. This dependence gives $\gamma = 0$.

gives $\gamma = 0$ [17]; i.e., we conclude that the oscillations in the magnetization result from some small electron group near the band-contact line [18]. Using the experimental value [12] of the cyclotron mass, $|m^*| = 0.067m$, and the position of the last peak $H_1 \approx 7$ T, we find from Eq. (6) that $|\varepsilon_F - \varepsilon_0| \approx 25$ meV. The downward peaks mean that we deal with the situation shown in Fig. 1(b).

To verify this conclusion, we also compare the theoretical $M(H)$ with the experimental data (Fig. 2). The experimental magnetization M_{exp} has been approximated by

$$M_{\mathrm{exp}}(H) = \chi_0 H + M(H), \tag{10}$$

where $M(H)$ is given by Eq. (4), while χ_0 is the smooth part of the magnetic susceptibility of the large electron groups in $LaRhIn_5$. Thus, we have only the two *constants* to fit the experimental data: the prefactor in Eq. (4) and χ_0. In Fig. 2 we show the theoretical curve calculated under the condition $\chi_0 H_1/M(H_1) = 0.14$. Note that $M(H_1)$ is noticeably larger than $\chi_0 H_1$. It is also evident that the curve reproduces the experimental data sufficiently well even without any corrections to M due to the spin-orbit interaction.

Although the band structure of $LaRhIn_5$ was calculated in Ref. [19], the data presented in that paper do not permit one to find the band-contact lines in this crystal. To locate these lines, it would be well to calculate the bands lying near ε_F over the Brillouin zone and to trace the evolution of these bands with the strength of the spin-orbit interaction. Such an analysis could also clarify one more point: It turns out that two small and almost equal cross sections determine the oscillations of M in $LaRhIn_5$ [12]. This can occur if the direction of the magnetic field slightly differs from the [001] axis, and if the directions of the band-contact lines at the equivalent points $\mathbf{k}_0$ do not coincide with this axis. Note that, when the magnetic field is tilted away from the k_3 axis, the component H_3 has to be inserted in the above formulas [14]. Thus, an experimental investigation of the angular dependences of the two cross sections could also assist in clarifying this result of Ref. [12].

In summary, we resolve the contradiction discovered in Ref. [12]. It turns out that a small neck of the Fermi surface with *the band-contact line inside the neck* [see the inset of Fig. 1(b)] was discovered in Ref. [12]. In this case the magnetization in the ultraquantum limit does not vanish, while the positions of the peaks in the oscillation part of $M(H)$ depend on the nonzero Berry phase for the electron orbits in the magnetic field. In other words, the results of Ref. [12] provide essentially the first observation of the Berry phase via the de Haas–van Alphen effect.

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[17] A close inspection of the inset of Fig. 2 of Ref. [12] leads to the same conclusion.
[18] It is possible that this line is almost straight and parallel to the [001] direction, with these properties of the line being due to the quasi-two-dimensional electron energy spectrum of the crystal.
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