Magnetization in the Ultraquantum Limit

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(Received 10 December 2001; published 19 June 2002)

The magnetization below and far above the quantum limit for small Fermi surface orbits has been measured in the metallic compound $LaRhIn_5$. The magnetization due to a pocket of Fermi surface that comprises less than 1 part in 10^4 of the total Brillouin zone volume, and for which the quantum limit is approximately 7 T, leads to the appearance of an overall sample magnetic moment at fields between 7 and 32 T. This moment arises from diamagnetic currents produced by electrons in the ultraquantum limit. A model calculation of the origin and magnitude of the effect is in excellent agreement with the measured field dependence of the induced magnetization.

DOI: 10.1103/PhysRevLett.89.026401 PACS numbers: 71.18.+y, 71.27.+a

The prediction of magnetic field induced diamagnetism in metals, with a quantized oscillatory behavior, was first made by Landau in 1930 [1], and within the same year the discovery of magnetic quantum oscillations in bismuth by de Haas and van Alphen [2] was made. Since that time there have been numerous searches for the magnetic behavior of materials above the $n = 1$ Landau level (quantum limit) [3]. Many of these searches have been made on semimetallic Bi because it is known to have one of the smallest volume Fermi surfaces (FS), and therefore small quantum limit values of magnetic field, of any known pure material. However, these searches and their interpretations have been hindered because the low carrier concentration is magnetic field dependent making the analysis of the experimental results difficult. In this Letter we report measurements on $LaRhIn_5$ [4], a high electrical conductivity normal metal, in which we are able to observe quantum oscillations below the quantum limit from a piece of FS that occupies an extremely small part of the overall FS volume. The contribution to the overall sample magnetization due to the electrons in this FS pocket at fields far above the quantum limit (ultraquantum limit) has been measured and the results compared to a theoretical model that describes the measured data extremely well.

In the presence of a magnetic field, Onsager [5] showed that the orbital motion of electrons in metals is restricted to lie on tubes in k (wave vector) space, Landau tubes, determined by the condition of area quantization: $A(E, k_B) = (n + \frac{1}{2})\frac{2\pi eB}{\hbar c}$, where A is the area of the tube with its axis along B, n is an integer, and B is the magnetic field. As the applied field increases, the free energy of the metal oscillates as each Landau level is filled upon crossing the Fermi energy, giving rise to an oscillatory effect periodic in the reciprocal of the applied field, in all of the magnetic and thermodynamic properties of the metal. The resulting quantum oscillations in the magnetic moment of the sample are called the de Haas–van Alphen effect (dHvA). Only in a few systems has it been possible to measure dHvA oscillations up to the quantum limit where all of the electrons on a part of the FS reside in a single Landau level [3]. In $LaRhIn_5$ we have observed strong oscillating signals observable from near zero applied fields up to the quantum limit of 7 T for one small part of the FS. An induced net diamagnetic moment in the sample occurs at fields between 7 and 32 T above the quantum limit for these carriers in addition to quantum oscillations from the much higher frequency dHvA oscillations due to electrons occupying states in the overall large volume FS. Thus we have been able to examine the magnetic properties of a metal in which some of the electronic states are in the ultraquantum limit while the majority of the carriers are not. This unique situation means that the large majority of the electronic states holds the Fermi energy constant while the magnetization due to the 7 T quantum limit carriers is measured.

The magnetization measurements were made with a capacitance cantilever, a method that is now a textbook technique [6]. In the presence of a uniform applied dc magnetic field, H, the interaction of the magnetic moment of the sample results in a torque on the cantilever given by $\vec{\tau} = V(\vec{M} \times \vec{H})$, where V is the sample volume, so that the measured magnetization is always perpendicular to H, $M_\perp$. When there is a magnetic field gradient present an additional signal due to the force appears: $F = \mu_0 M_\parallel \frac{dH}{dz}$, where z is in the direction of the applied field. In the magnets used for these measurements the magnitude of field gradient is directly proportional to the applied field, so that the measured force is $F = cM_\parallel H$, where c is a constant, and the measured signal is proportional to the component of the magnetization parallel to H, $M_\parallel$.

For the results reported here measurements were made on a single crystal sample with the field applied nearly parallel to the [001] direction of the tetragonal structure. Measurements were carried out at temperatures from 1.5 K to greater than 20 K and in magnetic fields from zero to 32 T with the result at 1.5 K shown in Fig. 1. The large oscillations below 7 T are due to a small piece of FS showing

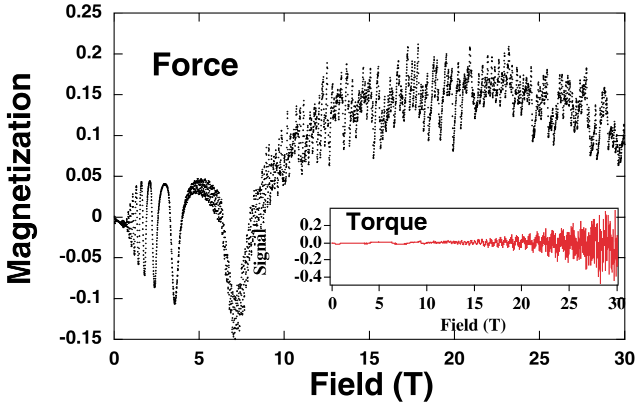

FIG. 1 (color online). The dHvA effect in $LaRhIn_5$ is shown as a function of field from zero to 30 T for measurements in the force position and for the torque position in the inset. The low frequency, 7 T FS orbit along with the high frequency oscillations seen clearly at low fields.

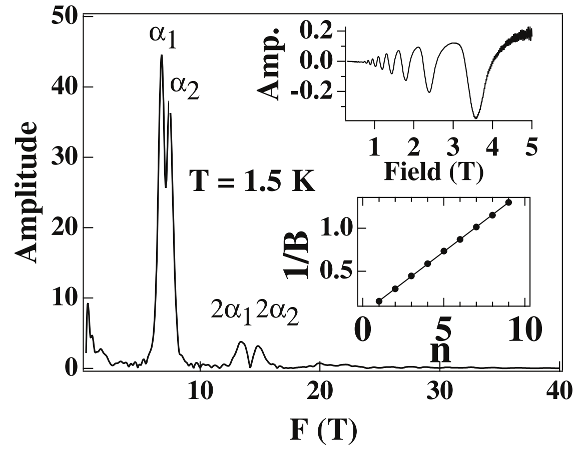

FIG. 2. The FT of the dHvA effect in $LaRhIn_5$ with the data at low fields is shown as a function of field in the top inset. The two peaks arise from two orbits on the FS. In the bottom inset the reciprocal fields of the zero crossings for the last nine ($n = 1-9$) oscillations in Fig. 1 are plotted as a function of integer Landau level n.

two dHvA frequencies of approximately 7 T and effective mass $m/m_0 = 0.067 \pm 0.001$. As can be seen in Fig. 1, the overall nonoscillatory magnetization parallel to the applied field of $LaRhIn_5$ at fields greater than 7 T is quite different from that observed below 7 T. In the inset to Fig. 1 it is seen that when the sample is in a uniform field so that only the torque proportional to $M_\perp$ is measured, the high frequency oscillations are of far greater amplitude (because there is no field inhomogeneity) and the background magnetization does not change above 7 T. The reason the field gradient decreases the amplitude of the high frequency oscillations is similar to the Dingle reduction factor due to impurity scattering in that it introduces an extra phase smearing in the detected signal. The ΔB across the sample causes the signals of a given frequency from different parts of the sample to be of different phases in an oscillation period, and adding signals of the same frequency, but varying phases reduces the amplitude. For low frequencies with large field periods this same ΔB corresponds to only a small phase shift and has less effect on the amplitude. This result was checked at several different values of the field gradient with the same result. The low frequency oscillations and a Fourier transform (FT) of them between 0.1 and 6 T are shown in Fig. 2. The FT shows two frequencies that we interpret as arising from a pocket of FS with two extremal area orbits. The second inset in Fig. 2 shows the reciprocal fields of the zero crossings plotted as a function of Landau level integer. Unlike Bi and other materials where oscillations near the $n = 1$ Landau level have been studied, deviations from periodicity in $\frac{1}{B}$ due to the field dependent Fermi energy do not occur. In the present case one can see that the oscillations remain perfectly periodic in $\frac{1}{B}$ up to $n = 1$.

Many high frequency dHvA oscillations are clearly observed at all fields, but, in addition, the sample acquires an overall field dependent magnetization parallel to H. The majority of the FS of $LaRhIn_5$ is quite complex with dHvA frequencies ranging from this 7 T frequency to near 12 000 T [7]. Here, we focus on a single sheet of FS: the 7 T pocket. Because the remainder of the FS comprises over 99.9% of the total FS volume, the electron concentration remains essentially constant in fields above the quantum limit for this single sheet of FS. We find that the shape of the overall curve above 7 T is due to a magnetization originating in the diamagnetic currents generated by the electrons on the 7 T orbits being in the ultraquantum limit.

The data in Fig. 1 cannot be explained by simple classical arguments. Above the quantum limit, the radius of an electron orbit is inversely proportional to the applied field, and the magnetic length is defined to be $\ell_B = (\frac{\hbar c}{eB})^{1/2}$. As the electrons orbit they produce a diamagnetic current proportional to the electric charge times the cyclotron frequency $\omega_c = \frac{eB}{mc}$. The resulting magnetic moment, $\frac{e\ell_B^2\omega_c}{2c} = \frac{e\hbar}{2mc}$, would be independent of field. Hence, we would expect the magnetic moment per carrier in the ultraquantum limit to be universal and independent of applied field.

A more realistic calculation of the field dependence of the magnetization above the quantum limit explicitly including quantization effects can be made to better differentiate the semiclassical answer from the "semi-quantum-mechanical" one. First we calculate the energy per unit volume of a metal in a magnetic field. The energy is divided into two components: $E_\perp$ and $E_\parallel$, where $\perp$ and $\parallel$ refer to the transverse and longitudinal directions with respect to the direction of the applied magnetic field. The transverse component is given by

$$E_\perp = \frac{N\hbar\omega_c}{2},$$

where ω_c is the cyclotron frequency and N is the number of electrons per unit volume. The longitudinal component is given by

$$E_\parallel = \frac{g_s}{2\pi \ell_B^2} \int_{-k_F}^{k_F} \frac{dk_z}{2\pi} \frac{k_z^2}{2m_\parallel} = \frac{1}{3} N \frac{\hbar^2 k_F^2}{2m_\parallel},$$

where g_s is the spin degeneracy factor, $m_\parallel$ is the longitudinal effective mass of the electrons contributing to the signal, and k_F is the Fermi momentum along the field. The total energy, therefore, can be expressed by

$$E = E_\perp + E_\parallel = \frac{N\hbar\omega_c}{2} + \frac{1}{3} N \frac{\hbar^2 k_F^2}{2m_\parallel}.$$

We now define the quantum limit field, B_q, to be the field that depopulates all but one Landau level. That is, it is a field at which the Fermi energy just touches the bottom of the $n = 1$ level where the last minimum in the magnetization oscillations occurs. Thus,

$$\hbar\omega_c(B_q) = \frac{\hbar^2 k_F^2(B_q)}{2m_\parallel}.$$

The Pauli principle requires that the number of electrons is equal to the number of occupied quantum states. In the ultraquantum limit fields, the latter is given by the product of the Landau degeneracy factor ($Ag_s/2\pi\ell_B^2$, where A is the sample area) and the number of quantum states for the motion along the field ($L2k_F/2\pi$). As a result

$$N = g_s \frac{1}{2\pi\ell_B^2} \frac{2k_F}{2\pi},$$

and $k_F = 2\pi^2\ell_B^2 N/g_s \propto B^{-1}$ for fixed N. Upon substitution of k_F and $\omega_c = eB/mc$ into the equation for the total energy, the total energy in terms of B_q becomes

$$E = \frac{n}{2}\hbar\omega_c(B_q)\left(\frac{B}{B_q} + \frac{2}{3}\left[\frac{B}{B_q}\right]^2\right).$$

Assuming an ellipsoidal FS and since the Fermi energy is not field dependent for this piece of FS over all of the measurement fields, the prefactor can be written in terms of the zero field E_F for an ellipsoid to be

$$\frac{\hbar\omega_c(B_q)}{2} = \left(\frac{2}{9}\right)^{1/3} E_F,$$

and the final expression for the total energy in this situation is

$$E = n\left(\frac{2}{9}\right)^{1/3} E_F\left(\frac{B}{B_q} + \frac{2}{3}\left[\frac{B}{B_q}\right]^2\right).$$

The magnetization in the quantum limit is given by $M = \frac{\partial E}{\partial B}$, with the final form for the magnetization being

$$M = \left(\frac{2}{9}\right)^{1/3} \frac{nE_F}{B_q}\left(1 - \frac{4}{3}\left[\frac{B_q}{B}\right]^3\right).$$

Note that the field-independent term in this last equation coincides with the estimate obtained from the qualitative picture of diamagnetic currents due to quantized electrons.

The overall magnetization data above 7 T were first fit to an expression that includes a term from the linear magnetization due to the cantilever material (BeCu), and this contribution was subtracted from the data. We find that this linear term exists with the same slope in fits to the data at fields far below 7 T, so it does not arise from the diamagnetic currents above the 7 T quantum limit. In Fig. 3 we show the resulting corrected data along with the fit to the magnetization given by $M = \alpha + \beta B^{-3}$, where α is a constant. From the fit we determine $\beta = 0.2363$ and $\alpha = -122.9$.

From the final equation for M the reduced magnetization is

$$m = \frac{M}{\left(\frac{2}{9}\right)^{1/3}\frac{nE_F}{B_q}} = -\left(1 - \frac{4}{3}\left[\frac{B_q}{B}\right]^3\right),$$

and the coefficient in front of the B^{-3} term is equal to $\frac{4}{3}B_q^3$. Taking the value of B_q to be the field of the last minimum of the oscillations, 7 T, this gives a value of the coefficient of the B^{-3} term to be -457. Dividing α by β from the fit to the experimental data to obtain the reduced magnetization, the experimental value is -520, giving agreement between experiment and theory to 10%.

The results of these measurements and calculations raise a rather important theoretical question [8]. According to current theories of Landau quantization at fields above the quantum limit the electrons in this small piece of FS should be transferred to other FS pieces below the Fermi energy. That is, if the $n = 1$ Landau level is raised above the Fermi energy by the application of fields greater than the quantum limit, electrons in this energy level should be depopulated into other Landau levels at the Fermi energy. As can be seen from the fit to the data, this clearly is not the case here. This is, in fact, a major point of these measurements and needs to be addressed by further theoretical arguments. We again point out that if depopulation of the last Landau level were the case, the field dependence of the magnetization above the quantum limit would be quite different from what is observed. The resolution of this apparent contradiction is the subject of current investigations.

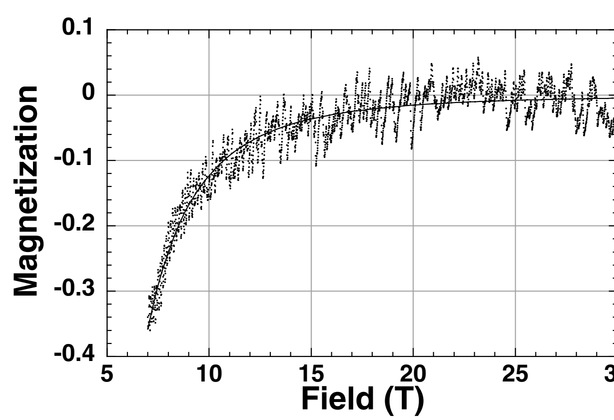

FIG. 3. The magnetization as a function of field in $LaRhIn_5$ above the quantum limit for the 7 T orbit. In this figure a linear in the B term has been subtracted from all of the data. The high frequency FS orbits in this material are seen clearly. The fit demonstrates good agreement with the interpretation that this moment arises from diamagnetic currents.

In summary, we have measured for the first time the induced magnetization due to diamagnetic currents of electrons in the ultraquantum limit, first predicted by Landau in 1930. Furthermore, a simple theory has been presented that predicts the field dependence of this contribution to within 10%. The reason this simple argument works in this case is that only a small portion of the FS is in the quantum limit while the vast majority of the electrons determining the overall Fermi energy and chemical potential remains constant. Therefore, no corrections need to be made for a field dependent Fermi energy or number of electrons contributing to the chemical potential. Thus, in looking for further examples of this effect, it would appear that what is needed is not a small Fermi surface due to a low carrier concentration material like bismuth but rather a material like $LaRhIn_5$ that presents a minority FS that can be studied in the limit in which the majority of the FS pins the chemical potential to its zero-field value. We point out that this new effect of a magnetic field on a 3D material in the quantum limit is similar to that of an electric field on a 2D gas, for example, a Si metal-oxide-semiconductor field-effect-transistor (MOSFET). In both cases one has a Fermi liquid or a non-Fermi liquid, whose parameters can be changed continuously with magnetic field or gate voltage. What the magnetic field does can be called "magnetic-field gating," and this Letter presents the simplest example of how a free-electron property is tuned by the field giving a way to tune correlation effects in one and the same sample analogous to the FET changing the number of carriers.

This work was supported in part by the National Science Foundation under Grants No. DMR-9971348 (Z. F.) and No. DMR-9705224 (A. F. H.). A portion of this work was performed at the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement No. DMR-9527035 and by the State of Florida. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy.

[1] L. Landau, Z. Phys. **64**, 629 (1930).
[2] W. J. de Hass and P. M. van Alphen, Proc. Netherlands R. Acad. Sci. **33**, 1106 (1930).
[3] For example, see N. L. Brignall and D. Schoenberg, J. Phys. C **7**, 1499 (1974); S. G. Bompadre, C. Biagini, D. Maslov, and A. F. Hebard, Phys. Rev. B **64**, 073103 (2001), and references therein.
[4] H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).
[5] L. Onsager, Philos. Mag. **43**, 1006 (1952).
[6] For references, see D. Schoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, London, 1984).
[7] Donavan Hall, E. Palm, T. Murphy, S. W. Tozer, C. Petrovic, Eliza Miller-Ricci, Lydia Peabody, Charis Quay Huei Li, U. Alver, R. G. Goodrich, J. L. Sarrao, P. G. Pagliuso, J. Wills, and Z. Fisk, Phys. Rev. B **64**, 064506 (2001).
[8] L. Gor'kov (private communication).