Nuclear Spin-Lattice Relaxation: A Microscopic Local Probe for Systems Exhibiting the Quantum Hall Effect

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A theory of magnetic quantum oscillations in the nuclear spin-lattice relaxation rate, T_1^{-1}, in quasi-two-dimensional conductors at low temperatures and under strong magnetic fields is presented. We show a close similarity between the magnetic field dependence of T_1^{-1} and that of ρ_{xx} in the quantum Hall effect. The shape and the amplitude of the oscillations in T_1^{-1} depend strongly on band anisotropy, electron mean free path, spin Larmor frequency, and temperature, and may provide rich information on the sample parameters.

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Magnetic quantum oscillations, originating from the discrete part of the electron energy spectrum in the magnetic field (Landau levels) are common to many transport properties in a three-dimensional electron gas. The most widely studied are the oscillations of the magnetoresistivity ρ_{xx} (Shubnikov–de Haas effect) and the oscillations of the magnetization (de Haas–van Alphen effect.[1] The remarkable features in two-dimensional conductors are the special size and the form of these oscillations: The Shubnikov–de Haas effect turns into the celebrated quantum-Hall-effect structures;[2-4] the magnetization oscillations have a saw-tooth form in the high-field region[1,5-10] and may drive a diamagnetic phase transition into a nondissipative state (ideally conducting phases[9]) in a dense two-dimensional electron gas.[11]

Apart from the drastic change of the form of the magnetic quantum oscillations in a 2D electron gas, one expects their appearance also in physical properties, which usually do not show any appreciable quantum oscillations in isotropic 3D systems. Indeed, we have shown previously[12] that the nuclear spin-lattice relaxation rate T_1^{-1} should exhibit strong magnetic oscillations in a quasi-two-dimensional metal. In isotropic metals, T_1^{-1} is due to the hyperfine interaction between the nuclear spin and the conduction-electron spin. It is known to obey the Korringa law,[13] that is, $T_1^{-1} \propto N^2(\epsilon_F) k_B T$, where $N(\epsilon_F)$ is the single electron density of states at the Fermi energy and T is the temperature. Deviations from the Korringa law are rare and may originate from strong electron correlations (e.g., superconducting correlations yield a logarithmic divergence of T_1^{-1} at T_c, the transition temperature[14,15]). To a high degree of accuracy, T_1^{-1} is, usually, field independent.[13] Ehrenfreund, Ron, and Weger[16] have observed, however, a strong magnetic field dependence of T_1^{-1} in $Hg_{3-x}AsF_6$, which was attributed by them to the quasi-two-dimensional character of the electron motion in this material.

Our purpose in the present paper is to study the magnetic field dependence of T_1^{-1} in quasi-two-dimensional conductors at sufficiently low temperatures and high magnetic fields where the Landau levels are well separated. We find a remarkable similarity between the magnetic field dependence of T_1^{-1} and that of ρ_{xx} in the quantum Hall effect, namely, exponentially vanishing T_1^{-1} when the chemical potential is in the mobility gap and maxima when a Landau level crosses the Fermi energy. The width of the maximum in T_1^{-1} is determined either by the band width in the k_z direction, Δ_z, or by the Zeeman splitting energy $\hbar\omega_e$ rather than by the temperature and scattering smearing of the Landau levels[17,18] as for ρ_{xx} in quantum Hall effect. This is due to the conservation laws governing the spin-flip process associated with the hyperfine interaction between the nuclear and conduction-electron spins.

The nuclear spin-lattice relaxation rate T_1^{-1}, caused by the hyperfine interaction between the nuclear spins and the conduction-electron spins, is related to the imaginary part of the local spin-spin correlation function through the equation

$$T_1^{-1} = \left(\frac{32\pi^2}{9}\right) \gamma_n^2 g^2 \mu_B^2 \, \mathrm{Im} \int_{-\infty}^{\infty} e^{-i\omega_n t} \{\langle S^+(\mathbf{R},t) S^-(\mathbf{R},0)\rangle\} dt, \tag{1}$$

where $S^+(\mathbf{R})$, $S^-(\mathbf{R})$ are the transverse components of the electron-spin-density operator at the nuclear position $\mathbf{R}$.

The time dependence is given in the Heisenberg representation with respect to the free-electron Hamiltonian in the presence of the static magnetic field. Here ω_n is the nuclear resonance frequency, γ_n is the nuclear gyromagnetic ratio, and μ_B is the Bohr magneton.

We consider an anisotropic electron gas having two equivalent easy axes (the x and y directions) and a perpendicular hard axis (in z direction). A static magnetic field **H** is applied along the z axis so that the single electron energies are given by

$$\epsilon(n,k_z)=\hbar\omega_c(n+\tfrac{1}{2})+\tfrac{1}{2}\Delta_z[1-\cos(k_z)d]+\tfrac{1}{2}\hbar\omega_e\sigma, \tag{2}$$

where ω_c is the cyclotron frequency, $n=0, 1, 2, \ldots$, d is the lattice constant in the z direction, ω_e is the electron-spin Larmor frequency, and $\sigma=\pm 1$. For a highly anisotropic motion, the maximal value of the longitudinal kinetic energy, Δ_z, is much smaller than the Fermi energy E_F.

For the sake of simplicity we shall use the parabolic band approximation in Eq. (2) so that the longitudinal kinetic-energy term there is of the form $\epsilon_z=\hbar^2k_z^2/2m_z^*$ with the longitudinal effective mass m_z^* defined by $m_z^*=2\hbar^2/\Delta_z d^2$.

Using the standard procedure outlined in Ref. 12, but now with the modification introduced by the presence of scattering mechanisms such as, for example, impurity scattering, we get

$$(T_1^{-1})_H=\frac{Am_z^*}{(2\pi)^4 2\hbar^2}a_H^{-4}\sum_{n,n'=0}^{\infty}\int_0^{\Delta_z}\frac{d\epsilon_z}{\sqrt{\epsilon_z}}\int_0^{\Delta_z}\frac{d\epsilon_z'}{\sqrt{\epsilon_z'}}\left(-k_BT\frac{\partial f^0}{\partial\epsilon}\bigg|_{\epsilon=\hbar\omega_c(n+1/2)}\right)\times\frac{\tau/\hbar}{1+[\epsilon_z-\epsilon_z'+\hbar\omega_c(n-n')-\hbar\omega_e]^2\tau^2/\hbar^2}. \tag{3}$$

In this equation $A\equiv(16\pi^3/9)(g\gamma_n\mu_B)^2\hbar$, $a_H\equiv(c\hbar/eH)^{1/2}$ is the magnetic length, $f^0(\epsilon)$ is the Fermi-Dirac distribution function in the absence of the scattering effect, T is the temperature, and τ is a phenomenological single electron relaxation time. In this formula the effect of scattering appears only in a Lorentzian smearing of the delta function characterizing the ideal energy-conserving situation considered in Ref. 12. The effect of scattering on the electron distribution function[19] is neglected. This is a reasonable assumption if the inverse relaxation time $\hbar/\tau$ is smaller than k_BT. A similar formula can be obtained by use of a fully microscopic approach.[20]

The magnetic field dependence of T_1^{-1}, based on the Eq. (3) for the case where $\Delta_z>\hbar\omega_e$, is presented in Fig. 1(a). The physics behind this figure can be understood from a schematic construction exhibited in Figs. 1(b) and 1(c): For a sufficiently high magnetic field, where $\hbar\omega_c\gg\Delta_z$, the spin-flip process takes place within a single Landau level [Fig. 1(c)]. This corresponds to neglecting all the terms in Eq. (3) for which $n'\neq n$. Under this condition the Lorentzian in Eq. (3) contributes significantly only within a strip of width $\Gamma=\hbar/\tau$ along the segment $\epsilon_z^{\uparrow}=\epsilon_z^{\downarrow}$, $\hbar\omega_e<\epsilon_z^{\uparrow}<\Delta_z$ in the $(\epsilon_z^{\uparrow},\epsilon_z^{\downarrow})$ plane [Fig. 1(b)]. The Pauli exclusion principle further restricts the possible spin-flip processes within a strip of width k_BT around $\epsilon_z=\epsilon-\hbar\omega_c(n_F+\frac{1}{2})$, where n_F is the number of fully occupied Landau levels [see Figs. 1(c) and 1(b)]. Thus the region in the (ϵ_z,ϵ) plane representing significant contributions to these processes is the parallelogram formed by the intersection of the above two strips [Fig. 1(c)]. "Vertical" processes within this region are exclusively due to the presence of the external energy reservoir (i.e., phonons, plasmons, etc.). "Horizontal" processes are associated with energy transfer between the electron-spin degrees of freedom and the orbital electron motion along the z direction. Now, if both the temperature and the Landau-level broadening are sufficiently small such that $k_BT,\hbar/\tau<\Delta_z$, while the Fermi energy is assumed to be fixed, the variation of the magnetic field is reflected in Fig. 1(c) by a horizontal motion of the small parallelogram between $\hbar\omega_e$ and Δ_z. Therefore the magnetic field dependence of the relaxation rate, shown in Fig. 1(a), reflects the one-dimensional density of states at the Fermi energy associated with the electron motion in the z direction. This is, of course, restricted to a $\Delta_z/\hbar\omega_c$ portion of the de Haas-van Alphen (dHvA) period, outside which T_1^{-1} is exponentially small,

$$\exp\{-\hbar\omega_c/k_BT\}.$$

The situation changes dramatically once the electron Zeeman splitting, $\hbar\omega_e$, becomes larger than Δ_z. Under this circumstance, and in the absence of any scattering process, the energy conservation requirement reduces the spin-lattice relaxation rate to zero. In the presence of scattering the contribution to the relaxation process is associated with the tail of the Lorentzian in Eq. (3) and therefore depends strongly on τ. Our numerical result for such a situation is shown in Fig. 2.

In the case where the chemical potential, μ, is pinned to a partially occupied Landau level ($N_0=$const) the expression for T_1^{-1}, Eq. (3), should be supplemented by

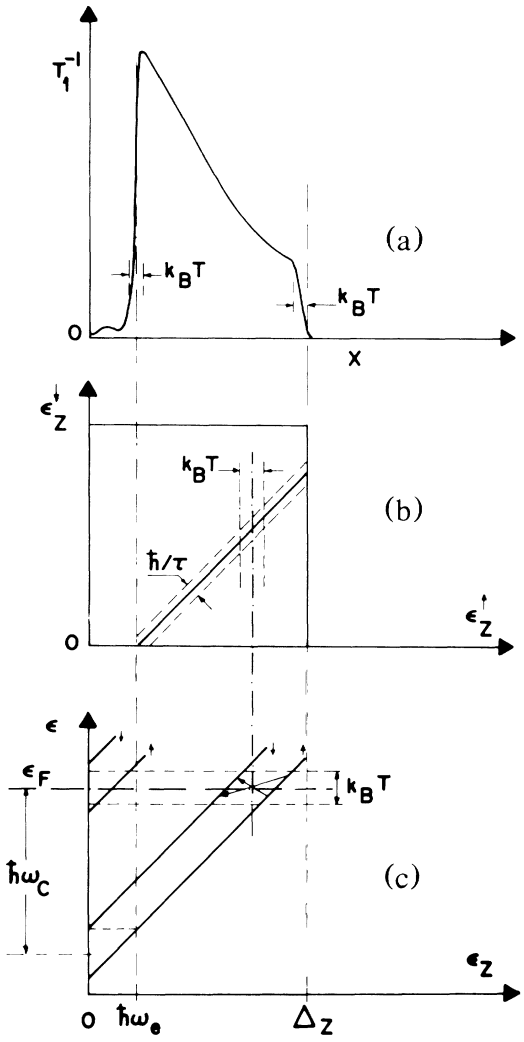

FIG. 1. A schematic construction explaining the physics behind the magnetic field dependence of the nuclear spin-lattice relaxation rate T_1^{-1} in a quasi-two-dimensional conductor: $(\hbar\omega_c > \Delta_z > \hbar\omega_e)$ within a $\Delta_z/\hbar\omega_c$ portion of a single dHvA period. In the rest of the period T_1^{-1} is exponentially small, $\exp\{-\hbar\omega_c/k_BT\}$. Low temperatures $(k_BT < \hbar\omega_e, \Delta_z)$ and narrow Landau levels $(\Gamma < \hbar\omega_e, \Delta_z)$ are assumed. (a) The magnetic field dependence of the relaxation rate T_1^{-1} reflects the one-dimensional density of states, at the Fermi level, associated with the electron motion in the z direction. Here $X = 1 - B^0/B$, and B^0 is the field, corresponding to the intersection of the spin-up Zeeman level with the Fermi level. (b) The Lorentzian in Eq. (3) contributes mainly along a strip of width Γ in the $(\epsilon_z^{\uparrow}, \epsilon_z^{\downarrow})$ plane. (c) Electron spin-flip processes in the Zeeman split Landau level are illustrated in the (ϵ, ϵ_z) plane.

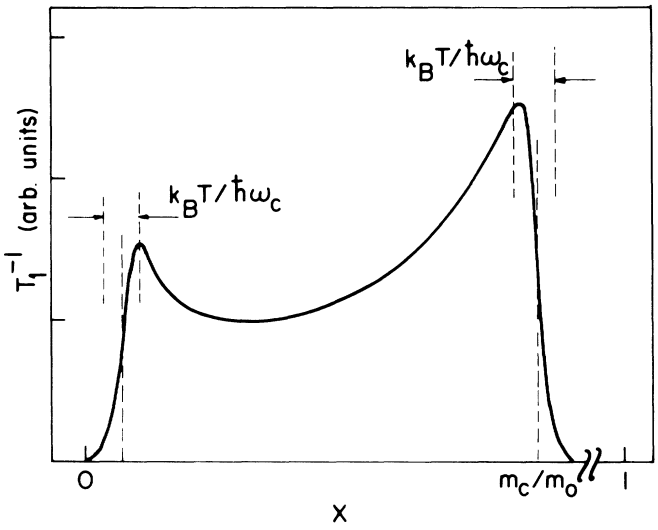

FIG. 2. Magnetic field dependence of the nuclear spin-lattice relaxation rate T_1^{-1} in a two-dimensional $(\hbar\omega_c > \hbar\omega_e \gg \Delta_z)$ conductor within a ω_e/ω_c portion of a single period.

the explicit dependence of μ upon B. This dependence can be calculated along the lines, presented in Ref. 9, where an analytic expression for the chemical potential in a 2D electron gas at finite temperatures is given [see Eq. (5) in Ref. 9]. Figure 3 exhibits $\mu(B)$ and $T_1^{-1}(B)$ in the case when the chemical potential is confined within the Zeeman split Landau level for the entire dHvA period.

To summarize, we find a remarkable similarity between the magnetic field dependence of the nuclear spin-lattice relaxation rate $T_1^{-1}(B)$ and $\rho_{xx}(B)$ in a two-dimensional electron gas at low temperatures and under strong magnetic fields, namely, exponentially small values of T_1^{-1} when the chemical potential is trapped between the Landau levels by localized states, as in conventional quantum Hall effect,[2-4] or by the diamagnetic-phase-transition-driven nondissipative state.[9]

Since the shape and the amplitude of the quantum oscillations in T_1^{-1} depend strongly on band anisotropy, electron mean free path, spin Larmor frequency, and the temperature, we conclude that the study of the magnetic field dependence of T_1^{-1} may provide rich microscopic information on the density of states in quasi-two-dimensional electronic systems, like quasi-2D synthetic metals, e.g., graphite intercalation compounds,[21] and layered transition-metal dichalcogenides, e.g., TaS_2 and $NbSe_2$.

Because the number of nuclei in the thin region, which gives rise to the quantum Hall effect, is small, measurement of the nuclear spin-lattice relaxation in a heterojunction presents a challenging experimental task. One may think about superlattices or multiple quantum wells with quite a number of layers to get enough nuclei, interacting with the electrons. The alternative possibility is to use nonconventional methods, like the observation of the electron-spin resonance[22] and of the Overhauser shift[23] in a heterojunction via measurements of the diag-

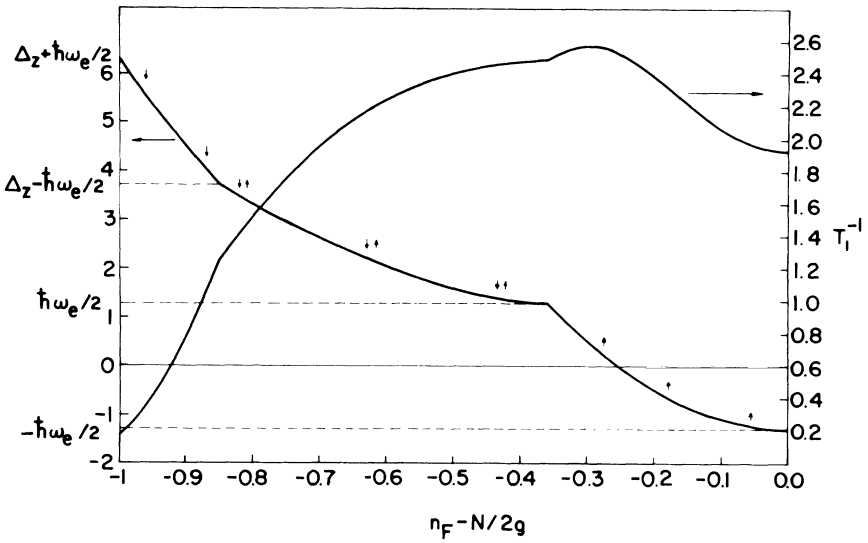

FIG. 3. Magnetic field dependence of the chemical potential, $\mu(B)$, and of $T_1^{-1}(B)$ when $\mu(B)$ is pinned to a Landau level. In this case the relaxation rate T_1^{-1} is finite over the entire dHvA period. Here $\hbar\omega_c/\hbar\omega_e=10$, $\Delta_z/\Gamma_0=15$, and $\hbar\omega_e/\Gamma_0=7.8$. On the horizontal axes n_F is the number of the Landau levels under the Fermi energy, N is the areal electron density, and g stands for the degeneracy of a Landau level.

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