Linear Positive Magnetoresistance and Quantum Interference in Ferromagnetic Metals

A. Gerber, I. Kishon, I. Ya. Korenblit, O. Riss, A. Segal, and M. Karpovski

Raymond and Beverly Sackler Faculty of Exact Sciences, School of Physics and Astronomy, Tel Aviv University, Ramat Aviv 69978, Tel Aviv, Israel

B. Raquet

Laboratoire National des Champs Magnétique Pulsés, 143 Avenue de Rangueil, 31400 Toulouse, France

(Received 4 January 2007; revised manuscript received 28 March 2007; published 9 July 2007)

Positive, linear in field, and isotropic magnetoresistance in fields up to 60 T is found in geometrically constrained ferromagnets, such as thin films of iron, nickel, and cobalt and their granular mixtures with nonmagnetic materials. The resistivity measured as a function of temperature shows a minimum at temperatures reaching a remarkably high 92 K, followed by logarithmic dependence at low temperatures. We propose to explain both phenomena by a modified version of the quantum electron-electron interaction theory. The agreement is only qualitative while the observed magnitude of the magnetoresistance slope is much larger than the calculated one.

DOI: 10.1103/PhysRevLett.99.027201 PACS numbers: 75.47.−m, 75.50.−y, 75.70.Ak

The so-called ordinary magnetoresistance is caused by a Lorentz force that affects the carrier trajectories in conducting materials. The effect is positive (resistance increases with increasing field) and proportional to $B_\perp^2$, where $B_\perp$ is the magnetic induction component perpendicular to the current flow. Quasilinear magnetoresistance can be found in polycrystalline materials [1], in inhomogeneous materials with mixed components of the resistivity tensor [2], and in the extreme quantum limit where one Landau level dominates (e.g., bismuth) [3,4]. Since the Lorentz force depends on a relative orientation of field and charge velocity, the ordinary magnetoresistance is anisotropic, i.e., depends on a relative orientation of field and current. In all of the above-mentioned cases, magnetoresistance diminishes when the field is applied parallel to the current. Similar anisotropy was also observed in silver chalcogenides [5], which show nonsaturating magnetoresistance up to a magnetic field of 60 T [6]. Several more types of magnetoresistive behavior were found in magnetic materials. Anisotropic magnetoresistance, positive for current flowing parallel to magnetization, and giant magnetoresistance, typical for heterogeneous magnetic systems, are related to magnetic alignment and vanish at fields above magnetic saturation. At higher fields, magnetoresistance of ferromagnets can be isotropic, i.e., independent of a relative orientation of field and current, but is usually negative. The magnetic field supports the ferromagnetic order and suppresses the electron scattering by spin-wave excitations and local magnetic anisotropy, resulting in a decrease of the resistivity [7,8]. A shift of minority and majority spin bands under an applied magnetic field also produces a negative magnetoresistance in the framework of a naive Mott's *s-d* band scattering model [9] in which the mobility of majority spins is higher than that of minority ones.

Isotropic positive magnetoresistance was recently observed in disordered, low-carrier-density magnets FeCoSi [10], microsized FeC and NiC composites [11], and LaPbMnO perovskite-type manganites [12]. Quantum interference was proposed by Manyala *et al.* [10] as a possible source of the effect in FeCoSi. If this interpretation is correct, the quantum interference effects, at least in this material, reveal themselves at temperatures of the order of 100 K, about 2 orders of magnitude higher than in previously studied disordered semiconductors [13]. The quantum interference scenario was challenged by Onose *et al.* [14], who pointed out that most of the magnetoresistance data in FeCoSi scale with the magnetization alone. A model, in which the mobility of majority spins is smaller than the mobility of minority spins, was proposed [14] as an alternative explanation of the positive magnetoresistance in this material.

In this Letter, we will show that isotropic, positive, and linear in field magnetoresistance is an inherent property of geometrically constrained ferromagnets. We shall focus on Fe and Ni films 3–300 nm thick manufactured by electron beam deposition on room temperature glass or GaAs substrates. Films are polycrystalline with a few nanometer grain size and become electrically conductive at a thickness of about 3 nm. Samples were deposited either directly on precut substrates 5 mm long and 2 mm wide or through the 5-contact mask. The sample width in the latter geometry is 0.1 mm with 0.5 mm separation between voltage probes. Indium contacts were attached, and resistance was measured in the four-probe configuration in both dc and ac modes. The resistivity of films thicker than 20 nm is about 15 $\mu\Omega$ cm. At lower thicknesses, the resistivity increases up to 65 and 150 $\mu\Omega$ cm in 4 nm thick Fe and Ni films, respectively. A 3 nm thick Ni film on the edge of the percolation threshold reaches a resistivity of about 800 $\mu\Omega$ cm. Most of the magnetoresistance measurements were performed in a superconducting magnet at fields up to 16 T in both positive and negative field polarities. Signal

even in field was defined as magnetoresistance. The odd field component was negligible in the transverse field and below the measurement accuracy in the longitudinal field (see the upper inset in Fig. 1). Several samples were measured at fields up to 60 T at Toulouse Pulsed Magnetic Field Laboratory.

Figure 1 presents the magnetoresistance of 10 nm thick Ni film measured at 4.2 K with a magnetic field applied perpendicular to the film plane, i.e., perpendicular to the electrical current (transverse magnetoresistance) and with the in-plane magnetic field parallel to the current (longitudinal magnetoresistance). Anisotropic magnetoresistance effects are visible at low fields; however, beyond 1 T, both curves are identical within the measurement accuracy (the curves are shifted to match their values at 15 T). The longitudinal magnetoresistance of 9 nm thick Fe film is shown in the lower inset in Fig. 1 at fields up to 50 T. Beyond the magnetization alignment, the magnetoresistance is positive and strictly linear with field. Transverse magnetoresistance (not shown) is very close to longitudinal but additionally contains a minor quadratic in field component, probably due to the classic Lorentz force contribution. Isotropy of the positive linear magnetoresistance is observed in all cases discussed in the following; therefore, we shall present the longitudinal magnetoresistance only, if not specified otherwise.

The temperature dependence of the effect is illustrated in Fig. 2, where the high field magnetoresistance of 9 nm thick Fe film is shown at 4.2, 100, and 221 K as measured in the pulsed magnet. The inset presents the slow sweep measurement of the 4 nm thick Fe film performed in the superconducting magnet. The magnetoresistance slope $d\rho/dB$ measured at 77 K is practically identical to the one at 4.2 K; therefore, the magnetoresistance can be considered independent of temperature in this temperature range. Additional temperature-dependent resistivity sources, such as spin-wave scattering, become effective at higher temperatures [8]. As a result, the slope $d\rho/dB$ decreases with increasing temperature and eventually becomes negative.

In chemically homogeneous films, the linear positive magnetoresistance (LPMR) is observed in the entire field range in thin films only. Figure 3 presents the longitudinal magnetoresistance measured in 4, 15, 50, and 300 nm thick Fe films. While the 4 and 15 nm thick samples demonstrate a dominant LPMR starting from magnetic alignment at low fields, the behavior of thicker films is more complicated. Negative magnetoresistance is found in 50 and 300 nm thick films up to about 6 T, followed by a positive magnetoresistance at higher field. These samples were measured until 16 T only, which is not sufficient to estimate their high field behavior.

LPMR is not restricted to thin chemically homogeneous films of Ni and Fe only. Although not presented here, we find similar behavior in thin films of Co and in granular systems with ferromagnetic grains mixed with nonmagnetic metallic (Pb), insulating (SiO_2), and semiconducting (Ge) components. The magnetoresistance slope is of the order of $10^{-1}\%/\mathrm{T}$ in highly resistive $NiSiO_2$ and $10^{-3}\%/\mathrm{T}$ in NiPb.

LPMR was not found in test experiments with thin nonmagnetic Au and Al metallic films produced, contacted, and measured in the same way as ferromagnetic films. The magnetoresistance of these samples is similar to that reported earlier [15].

Quantum corrections to conductivity were mentioned recently [10] as relevant for magnetotransport in certain magnetic systems. Notably, the evidence of electron-electron interactions is found in all of our samples that

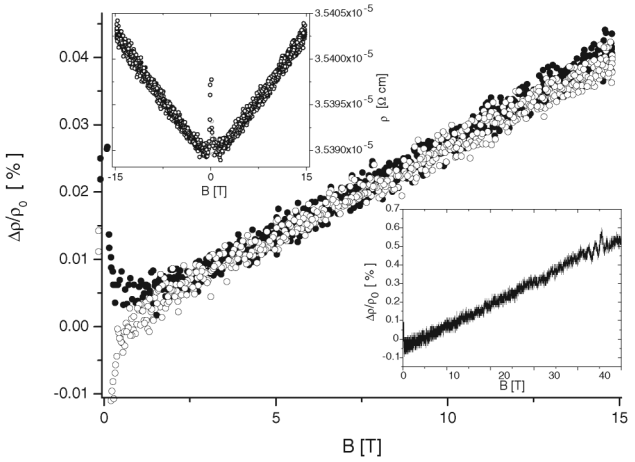

FIG. 1. Transverse (○) and longitudinal (●) magnetoresistance of 10 nm thick Ni film. Upper inset: Transverse magnetoresistance of 10 nm thick Ni film at both field polarities. $\rho_0 \approx 35\ \mu\Omega\,\mathrm{cm}$. Lower inset: High field longitudinal magnetoresistance of 9 nm thick Fe film. $\rho_0 \approx 27\ \mu\Omega\,\mathrm{cm}$. $T = 4.2$ K.

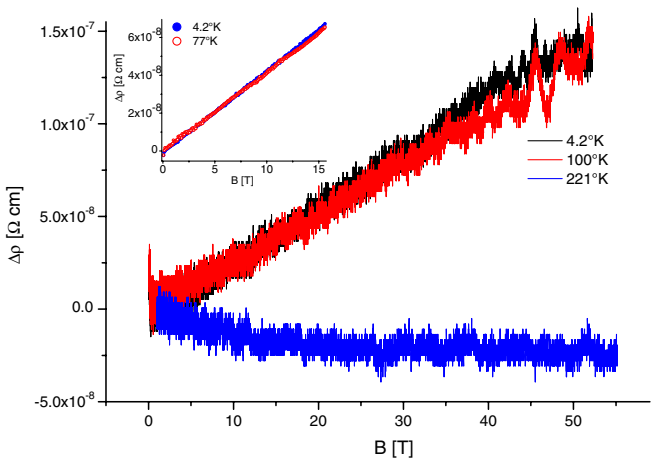

FIG. 2 (color online). High field longitudinal magnetoresistance of 9 nm thick Fe film at 4.2, 100, and 221 K. Inset: Longitudinal magnetoresistance of 4 nm thick Fe film at 4.2 and 77 K measured at a slow sweep in the superconducting magnet.

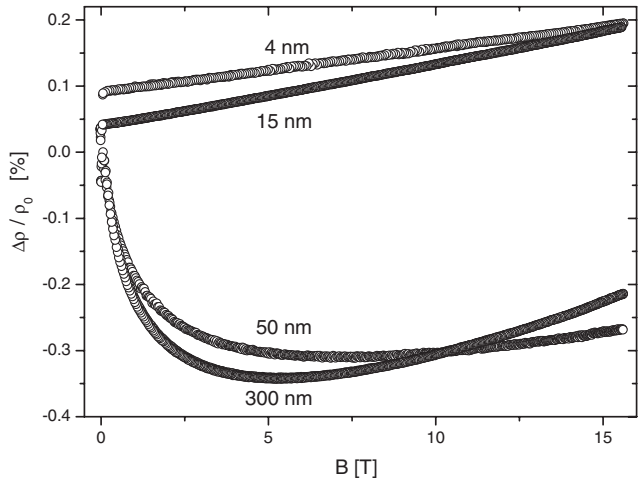

FIG. 3. Longitudinal magnetoresistance of 4, 15, 50, and 300 nm thick Fe films at 4.2 K.

show a dominant LPMR. Figure 4 presents the temperature dependence of resistivity measured at zero field and under 16 T field in several Fe and Ni films. The resistivity minimum is found in all of these samples, whereas the minimum temperature $T_{\min}$ is independent of the applied field. At temperatures below $T_{\min}$, the conductivity varies logarithmically with temperature (Fig. 4, inset) as $\Delta\sigma = \alpha \ln T$, with α close to $e^2/2\pi^2\hbar = 1.23 \times 10^{-5}\ \Omega^{-1}$, the slope predicted for both weak localization and electron-electron (e-e) interference corrections in two-dimensional systems [16,17]. The Kondo effect also predicts the logarithmic temperature dependence; however, it involves a spin-flip scattering of electrons by magnetic impurities and would not be expected in ferromagnetic materials. The independence of $T_{\min}$ on the applied magnetic field was argued [18] to indicate the e-e interactions being the origin of the phenomenon rather than the weak localization. Remarkably, the resistivity minimum in 3 nm thick Ni film is at 92 K, which is by 1–2 orders of magnitude higher than previously observed in nonmagnetic conductors. The increase of $T_{\min}$ in thin films can be explained by an enhancement of surface scattering in the thin film limit [19,20]. Superposition of the quantum corrections resistivity term $-\Delta\rho_{ee} = -\rho^2\Delta\sigma \approx -\rho_0^2\alpha \ln T$ with the thermally dependent scattering contribution to resistivity aT^ν results in the resistivity minimum at $T_{\min} = (\alpha/a\nu)^{1/\nu}\rho_0^{2/\nu}$, where ρ_0 is the remnant zero temperature resistivity, a is the coefficient, and ν is the power index corresponding to the major thermally dependent scattering mechanism. Thinner films have higher resistivity ρ_0 and therefore higher $T_{\min}$.

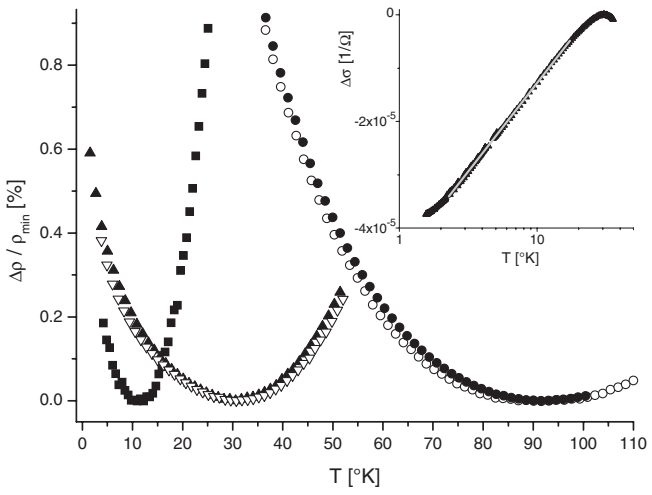

FIG. 4. Resistivity as a function of temperature of 10 nm thick Ni film (multiplied by 10) (■), 4 nm thick Fe film at zero field (▽) and 16 T field (▲), and 3 nm thick Ni film at zero field (○) and 16 T field (●). $\rho_{\min}$ is resistivity at its minimum and $\Delta\rho = \rho - \rho_{\min}$. $\rho_{\min}$ in 3 nm thick Ni is $7.85 \times 10^{-4}\ \Omega\,\mathrm{cm}$. Inset: Change of conductivity defined as $\Delta\sigma = \sigma - \sigma_{\max}$, where $\sigma_{\max} = 1/\rho_{\min}$, as a function of $\ln T$ for 4 nm thick Fe (▲). The slope is $1.43 \times 10^{-5}\ \Omega^{-1}$.

Prior to focusing on the quantum e-e interference scenario, we wish to clarify that no other mechanism known to us seems to provide an adequate description of the LPMR. The effect is isotropic; therefore, we exclude anisotropic properties related to electronic orbital motion or magnetostriction [21]. According to Mott's model [9], the conductivity σ of a ferromagnetic metal can be written as

$$\sigma = e^2\left(\frac{n_\uparrow\tau_\uparrow}{m_\uparrow} + \frac{n_\downarrow\tau_\downarrow}{m_\downarrow}\right), \tag{1}$$

where $n_\uparrow(n_\downarrow)$, $\tau_\uparrow(\tau_\downarrow)$, and $m_\uparrow(m_\downarrow)$ are the concentration, relaxation time, and effective mass of the s-electrons with spin-up (along the magnetization) and down, respectively. The change of the conductivity in magnetic field is

$$\frac{d\sigma(H)}{dH} = \frac{e^2 n}{m}\left[\tau_\uparrow\left(\frac{d\ln n_\uparrow}{dH} + \frac{d\ln\tau_\uparrow}{dH}\right) + \tau_\downarrow\left(\frac{d\ln n_\downarrow}{dH} + \frac{d\ln\tau_\downarrow}{dH}\right)\right]. \tag{2}$$

At this stage, we ignore relatively small differences between $n_\uparrow$ and $n_\downarrow$, $m_\uparrow$ and $m_\downarrow$, and define $n = n_\uparrow \approx n_\downarrow$ and $m = m_\uparrow \approx m_\downarrow$. In Ni, the derivative $d\ln n_\uparrow/dH$ is positive, while $d\ln n_\downarrow/dH$ is negative, and $\tau_\uparrow \gg \tau_\downarrow$, since its d band is fully polarized [22]. For the same reason, the density of states does not change with field and $d\ln\tau_\uparrow/dH = d\ln\tau_\downarrow/dH = 0$. We conclude, therefore, that $d\sigma/dH$ is positive and the expected magnetoresistance is negative. The situation can be different in some materials, as argued by Onose *et al.* [14] for FeCoSi.

Let us consider now the magnetic field effect on the quantum corrections to conductivity. The weak localization contribution is negative and is usually suppressed by rather low fields [16,17]. Antilocalization due to spin-orbit interactions can generate a positive magnetoresistance in normal metals but does not occur in ferromagnetic systems

[23]. The effect of a magnetic field on the electron-electron interaction was derived two decades ago for nonmagnetic conductors. In this case, the magnetic field induces the spin gap equal to $g\mu_B H$, where g is the g factor and μ_B is the Bohr magneton. The gap suppresses the contribution to conductivity originating from the electron-hole interaction in the diffusion channel and leads to a positive magnetoresistance. At high fields or low temperatures $g\mu_B H \gg kT$, the magnetoresistance is proportional to $\ln(g\mu_B H/kT)$ in two-dimensional and to $\sqrt{g\mu_B H}$ in three-dimensional space [24].

The situation is different in ferromagnets. A large exchange gap $\Delta(0)$ between the subbands of s electrons with spins up and down already exists in the absence of any applied field [$\Delta(0) > 0.1$ eV for Fe, Ni, and Co] [25]. The applied field H modifies the exchange gap to

$$\Delta(H) = \Delta(0) + \gamma g \mu_B H, \tag{3}$$

where $\gamma = 1$ if the moment of the s electrons is parallel to the moment of the d electrons (the case of Fe, Co, and Ni) and $\gamma = -1$ if the moments are antiparallel. In the fields and temperatures of our experiments, $g\mu_B H \ll \Delta(0)$ and $kT \ll \Delta(H)$. The field-induced correction to the resistivity in two dimensions is

$$\delta\rho = \frac{A_{2d}e^2}{2\pi^2\hbar} \ln\frac{\Delta(H)}{\Delta(0)} \rho^2 \approx \gamma \frac{A_{2d}e^2}{2\pi^2\hbar} \frac{g\mu_B H}{\Delta(0)} \rho_0^2 \tag{4}$$

and in three dimensions is

$$\delta\rho = \frac{A_{3d}e^2}{2\pi^2\hbar}\left(\frac{kT}{2\hbar D}\right)^{1/2}\left[\left(\frac{\Delta(H)}{kT}\right)^{1/2} - \left(\frac{\Delta(0)}{kT}\right)^{1/2}\right]\rho^2 \approx \gamma \frac{A_{3d}e^2}{4\pi^2\hbar} \frac{g\mu_B H}{[2\hbar D\Delta(0)]^{1/2}} \rho_0^2, \tag{5}$$

where D is the diffusion constant and A_{2d} and A_{3d} are constants of the order of unity that characterize the electron-electron interaction strength in two- and three-dimensional space, respectively. Thus, in ferromagnetic metals with parallel s and d moments ($\gamma > 0$), the quantum interference results in a positive magnetoresistance, which increases linearly with an increase of the field in both two and three dimensions. The linear dependence is independent of temperature and is expected at any laboratory magnetic field. All of these properties agree qualitatively with our observations. It should be noted that the same mechanism will generate a negative linear magnetoresistance if s and d moments are antiparallel ($\gamma < 0$).

To make a quantitative comparison, we compare the sample size with the length of the interference region $L_\Delta = \sqrt{\hbar D/\Delta(0)}$. We estimate D from the relation $D = 2\varepsilon_F/3ne^2\rho$, where ε_F is the Fermi energy and n is the electron concentration. With the values $\varepsilon_F = 10$ eV and $n = 10^{29}$ m^{-3} typical for Fe and Ni, we calculate $D \approx 10^{-3}$ m^2/sec. Using $\Delta(0) \approx 0.2$ eV, we obtain $L_\Delta \approx 1.5$ nm; i.e., the magnetoresistance of our samples should be given by Eq. (5). The coefficient $\delta\rho/H$ is then estimated as about 10^{-11}–10^{-12} Ω cm/T, which agrees well with the measurement in the 3 nm thick Ni sample. In thicker films, the measured slopes appear to be 1–2 orders of magnitude steeper than the calculated. This strong quantitative discrepancy as well as the unusually high temperature of the resistivity minimum are challenging.

To summarize, high field magnetoresistance of thin films of Fe, Ni, and Co and their granular mixtures with nonmagnetic materials is positive, isotropic, temperature independent below 100 K, and linear in applied magnetic field. We tentatively explain these findings by the quantum electron-electron interference which reproduces qualitatively both the resistance minimum and the linear magnetoresistance.

This work was supported by the Israel Science Foundation Grants No. 220/02 and No. 633/06 and by the EuroMagNET program.

[1] P. Kapitza, Proc. R. Soc. A **123**, 292 (1929).
[2] C. Herring, J. Appl. Phys. **31**, 1939 (1960).
[3] P. Kapitza, Proc. R. Soc. A **119**, 358 (1928).
[4] F. Y. Yang *et al.*, Science **284**, 1335 (1999).
[5] R. Xu *et al.*, Nature (London) **390**, 57 (1997).
[6] A. Husmann *et al.*, Nature (London) **417**, 421 (2002).
[7] G. Bergman, Phys. Rev. B **15**, 1514 (1977).
[8] B. Raquet *et al.*, Phys. Rev. B **66**, 024433 (2002).
[9] N. F. Mott, Adv. Phys. **13**, 325 (1964).
[10] M. Manyala *et al.*, Nature (London) **404**, 581 (2000).
[11] Q. Z. Xue, X. Zhang, and D. D. Zhu, J. Magn. Magn. Mater. **270**, 397 (2004).
[12] P. Chen, D. Y. Xing, and Y. W. Du, Phys. Rev. B **64**, 104402 (2001).
[13] T. F. Rosenbaum, Nature (London) **404**, 556 (2000).
[14] Y. Onose *et al.*, Phys. Rev. B **72**, 224431 (2005).
[15] G. Bergman, Phys. Rep. **107**, 1 (1984).
[16] P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[17] B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interaction in Disordered Systems*, edited by A. L. Efros and M. Pollak (North-Holland, Amsterdam, 1985), p. 1.
[18] M. Brands, A. Carl, O. Posth, and G. Dumpich, Phys. Rev. B **72**, 085457 (2005).
[19] E. H. Sondheimer, Adv. Phys. **1**, 1 (1952).
[20] A. Gerber *et al.*, Phys. Rev. B **65**, 054426 (2002).
[21] A. Ludwig and E. Quandt, in *Modern Trends in Magnetostriction Study and Application*, edited by M. R. J. Gibbs (Kluwer Academic, Dordrecht, 2000), p. 57.
[22] R. C. O'Handley, *Modern Magnetic Materials* (Wiley, New York, 2000).
[23] V. K. Dugaev, P. Bruno, and J. Barnas, Phys. Rev. B **64**, 144423 (2001).
[24] P. A. Lee and T. V. Ramakrishnan, Phys. Rev. B **26**, 4009 (1982).
[25] J. C. Phillips, Phys. Rev. **133**, A1020 (1964).