An electron correlation originated negative magnetoresistance in a system having a partly flat band

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Inspired from an experimentally examined organic conductor, a novel mechanism for negative magnetoresistance is proposed for repulsively interacting electrons on a lattice whose band dispersion contains a flat portion (a flat bottom below a dispersive part here). When the Fermi level lies in the flat part, the electron correlation should cause ferromagnetic spin fluctuations to develop with an enhanced susceptibility. A relatively small magnetic field will then shift the majority-spin Fermi level to the dispersive part, resulting in a negative magnetoresistance. We have actually confirmed the idea by calculating the conductivity in magnetic fields, with the fluctuation exchange approximation, for the repulsive Hubbard model on a square lattice having a large second nearest-neighbor hopping.

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Negative magnetoresistance has provided fascination in diverse classes of systems, such as impurity band in semiconductors or Mn oxides. The problem is the interplay of the transport and the spin structure, and how the spins and carriers respond to magnetic fields can vary from system to system. In the physics of correlated electron systems, spin is a key ingredient. In fact, itinerant (metallic) ferromagnetism has been a central problem from the days of Kanamori, Hubbard and Gutzwiller. It is becoming increasingly clear that the criterion for the ferromagnetism becomes stringent as one incorporates correlation effects, but that a large density of states at the Fermi level does favor ferromagnetism.

In this paper, we propose a novel mechanism for negative magnetoresistance, which is realized as a combined effect of the electron correlation (proximity to ferromagnetism) and the band structure (coexistence of flat and dispersive parts). The idea is the following: We consider repulsively interacting electrons on a band, whose one-electron dispersion has a flat part in an otherwise dispersive band. For low electron densities with the Fermi level $E_F$ located near the flat part of the band, the system should have a strong tendency toward ferromagnetism. A large density of states $D(E_F)$ at the bottom of the band is indeed a situation originally considered by Kanamori and received a renewed interest recently for two and three dimensional systems as well as in one dimensional systems. When a magnetic field is applied, a weak field is then enough to drive the system into a significant magnetization (spin polarization), since the magnetic susceptibility is enhanced due to ferromagnetic fluctuations. The $E_F$ for the majority spins will then be shifted into the dispersive (i.e., lighter-mass) part of the band, and we expect that the system becomes more conductive for relatively weak magnetic fields. When the polarization is small enough there is a possibility that the effect of the increase in $v_F$ of the majority spin may be compensated by the decrease in $v_F$ of the minority spin, but such a compensation will not occur when the polarization is sufficiently large due to the enhanced susceptibility, since not only $v_F$ but also the number of the majority spin increase in the presence of the magnetic field. Further, there will be fewer electron-electron scatterings for a polarized state, at least for the Hubbard model where only up and down spin electrons interact. These will make the system more conductive. This idea has been inspired from a recent experimental result on a certain class organic conductors, called $\tau$-type conductors, for which Murata et al. have observed negative magnetoresistance, see below. The band structure calculation indicates that the dispersion is indeed flat at the bottom along $k_x$ and $k_y$ directions.

In the following, we confirm this idea for the single-band Hubbard model, a simplest model for repulsive electron correlation, on a simplest flat-bottomed tight-binding model, the square lattice having a large second nearest neighbor hopping, $t'$, along with $t$ between nearest neighbors. The one-electron dispersion becomes

$$\varepsilon_k = 2t (\cos k_x + \cos k_y) + 4t' \cos k_x \cos k_y, \quad (1)$$

and the dispersion is flat along $k_x$ and $k_y$ (right panel of Fig. 1) for $t' \approx 0.5 |t|$ with the van Hove singularity coming down to the bottom.

For $t' \approx 0.5$ (we take $|t| = 1$ hereafter), Hlubina has shown, with the T-matrix approximation for the Hubbard model, that the ground state is fully spin-polarized for $n \sim 0.4$. Since the T-matrix approximation is valid only in the low enough densities and for small $U$, here we start with obtaining the diagram for large $U$ with the exact diagonalization of a finite system ($4 \times 4$ sites with 8 electrons with the band quarter filled).

Detection of ferromagnetic states from total $S^2$ in finite systems requires some care as we have revealed in previous publications. Namely, ferromagnetic states in itinerant electron systems can accompany a spiral spin state in finite systems. The spiral state is a spin singlet state having the spin correlation wave length as large as the linear dimension of the system, which can lie in en-...
ergy below the ferromagnetic state. We found that the ground state of the 16-site system has always $S^z = 0$ in periodic boundary conditions in both $x$ and $y$, but if we look at the spin structure $S(q)$ (Fourier transform of $(S_x, S_y)$) the state turns out to be indeed spiral (i.e., $S(q)$ peaked at $(0, \pm \pi/L)$ and $(\pm \pi/L, 0)$) for large $U$ and $t' \approx 0.5$.

Although we can identify the spiral state as ferromagnetic in the thermodynamic limit, it has been shown that a faster approach to the thermodynamic limit is attained if we adopt an appropriate boundary condition (periodic in one direction and antiperiodic in another) to selectively push the spiral state above the ferromagnetic state in energy. If we look at the phase diagram thus obtained against $t'$ and $U$ in Fig. 4, the fully polarized ferromagnetic region is seen to exist for $t' \approx 0.5$ and $U > 6$.

Keeping this phase diagram in mind, we now discuss the conductivity in magnetic fields $B$. The conductivity of the interacting system is calculated here by the fluctuation exchange approximation (FLEX). The FLEX, introduced by Bickers et al., treats spin and charge fluctuations by starting from a set of skeleton diagrams for the Luttinger-Ward functional, based on the idea of Baym and Kadanoff. Then a $(k$-dependent) self energy is computed from RPA-type bubble and ladder diagrams self-consistently.

The dc conductivity is given in the Kubo formula as

$$\sigma_{\mu\nu} = \lim_{\omega \to 0} e^2 \sum_{\sigma} \int \frac{dk\,d^4q}{(2\pi)^6} \Im K_{kk'\sigma\sigma'}(\omega + i\delta) \frac{\Im K_{kk'\sigma\sigma'}(\omega + i\delta)}{\omega},$$

where $K_{kk'\sigma\sigma'}(\omega + i\delta)$ is the Fourier component of the retarded two-particle Green’s function and $v_{k\mu} = \partial c^0_{k\mu} / \partial k_{\mu}$ is the unperturbed velocity. We set $\hbar = 1$, $k_B = 1$ hereafter. If we follow Eliashberg for the analytic continuation of $K(\omega + i\delta)$ from $K(i\omega_n)$, where $\omega_n$ is the Matsubara frequency, the conductivity per spin read, for long enough life time of the quasi-particle,

$$\sigma_{xx} = e^2 \sum_{\sigma} \int \frac{dk}{(2\pi)^3} \left( \frac{\partial f}{\partial \varepsilon} \right) \Im K_{kk'\sigma\sigma}(\omega + i\delta) \frac{\Im K_{kk'\sigma\sigma}(\omega + i\delta)}{\omega},$$

where $v^*_{k\sigma}$ is the dressed velocity, $J^*_{k\sigma}$ the current, $\gamma_k$ the damping constant of the quasiparticle, and $f$ the Fermi distribution function.

Note that the conductivity of a clean system of interacting electrons can be finite even though the interaction is an internal force, since the momentum is dissipated through Umklapp processes. It has been shown by Yamada and Yosida that the conductivity diverges, as it should, when the Umklapp processes are turned off only if we consider the vertex correction for the current $J_k$ appropriately, so the correction should be included for a consistent treatment. Within the FLEX, which is a conserving approximation, there are three types of diagrams for irreducible vertices, namely two Aslamasov-Larkin (AL) type diagrams and one Maki-Thompson (MT) type.

One. Kontani et al. have shown that, when antiferromagnetic fluctuations are dominant, the AL contribution can be neglected. We can extend this argument to show that the MT term is also negligible when ferromagnetic fluctuations are dominant. Hence we consider only the MT term.

Let us now discuss the conductivity in magnetic fields. We have then to add the Zeeman term, $h \sum_{k\sigma} \text{sgn}(\sigma) c^0_{k\sigma} c_{k\sigma}$, to the Hamiltonian, where $h \equiv g\mu_B B$ is the Zeeman energy with $g \approx 2$. Green’s function and other quantities then become $\sigma$-dependent. To concentrate on the effect of the Zeeman splitting we assume here that the direction of the magnetic field is parallel to the current, so that we do not have to take account of the effect of the field on orbital motions.

The conductivity is obtained from the Bethe-Salpeter equation, which is a simple extension of the equations derived by Kontani et al. for the spin-independent case to the present spin-dependent case. We end up with the diagonal conductivity,

$$\sigma_{xx} = e^2 \sum_{k,\sigma} \int \frac{d\varepsilon}{\pi N} \left( -\frac{\partial f}{\partial \varepsilon} \right) \left\{ |G_{k\sigma}(\varepsilon)|^2 v_{k\sigma} J_{k\sigma}(\varepsilon) \right\} - \text{Re} \left[ G_{k\sigma}^2(\varepsilon) \right]|G_{k\sigma}(\varepsilon)|^2 J_{k\sigma}(\varepsilon),$$

where $N$ is the number of sites, $T$ the temperature, $G_{k\sigma}(\varepsilon)$ the dressed Green’s function, and the velocity $v_{k\sigma} = (\partial / \partial k_x) [\varepsilon^0_k + \text{Re} \Sigma_{k\sigma}(\varepsilon = 0)]$ with $\Sigma_{k\sigma}(\varepsilon)$ being the self energy.

The kernel, $V_{k\sigma\sigma'}(i\omega_n)$, which contains the effect of fluctuation exchanges, can be obtained by an analytic continuation of $V_{k\sigma\sigma'}(i\omega_n)$,

$$V_{k\sigma\sigma'}(i\omega_n) = \frac{U^2 \chi_{k\sigma\sigma'}^0(i\omega_n)}{1 - U^2 \chi_{k\sigma\sigma'}^0(i\omega_n)},$$

where $k \equiv (k, i\omega_n)$ in the last line.

Let us now present the results. We take the case of $t' = 0.5, U = 2$ with the band filling $n = 0.4$, which falls upon the paramagnetic region close to the ferromagnetic boundary in the phase diagram, Fig. 4. We first check that we do have strong ferromagnetic fluctuations. Figure 2 shows the wave number dependence of the spin susceptibility, $\chi_{k\sigma\sigma'}^{\text{RPA}} = 2\chi_{k\sigma\sigma'}^0(\omega = 0)/(1 - d_k^0(\omega = 0)\chi_{k\sigma\sigma'}^0(\omega = 0)) for
Let us next focus on the static, uniform magnetic susceptibility. Since we are sitting close to the ferromagnetic boundary, the susceptibility should be finite but enhanced. Here we compute the quantity in two ways: one is to calculate the derivative $\chi = \partial (n_d - n_{\alpha}) / \partial h$ with a finite difference $\delta h = 0.005$, and the other is $\chi^{\mathrm{RPA}}_0$. If we look at their temperature dependence in Fig. 3, for $t' = 0.5, n = 0.4$ with various $0.5 \leq U \leq 2.0$, they both sharply increase for $T \to 0$. There is a deviation between $\chi$ and $\chi^{\mathrm{RPA}}_0$ for larger $U$. The deviation itself indicates that the irreducible four-point vertex $\Gamma = \delta^2 \Phi / \delta G \delta G$ cannot be approximated by $U$. Since we are dealing with a case where the ferromagnetic phase appears for larger $U (> 6 \sim 7)$, $\chi$ underestimates the effect of external magnetic field for large $U$.

Now, we come to the key result for the magnetoresistance, given in Fig. 4 for $t' = 0.5, U = 2, n = 0.4$. The figure compares the resistivity against $T$ in the absence and in the presence of the magnetic field. We take the system size $N = 64^2$, with 512 Matsubara frequencies, which is checked to be sufficient for the temperature region studied here. We can see that we do have a negative magnetoresistance. The change in the resistance is of the order of 10% for the Zeeman energy $h = 0.05$. If we were not sitting close to the ferromagnetism, much larger fields would be required. The vertex correction does not alter the result significantly, which implies that Boltzmann’s transport picture (with no vertex correction) already has the negative magnetoresistance. The negative magnetoresistance should become more prominent at lower temperatures where the ferromagnetic fluctuation increases. This is not too noticeable in the result, which should be an artifact: as mentioned above, the spin polarization is underestimated in the FLEX at low temperatures for $U = 2$.

As touched upon at the beginning, Papavassiliou et al. has found experimentally that organic salts $D_2A_2A_\nu$, based on D (= P-S, S-DMEDT-TTF or EDO-S, S-DMEDT-TTF) in combination with linear anions $A$ (=AuBr$_2$, I$_3$, or IBr$_2$) with the fractional value of $y$ controlling the density of carriers are two-dimensional metals in the $\tau$ crystal form. The band structure of a single layer of the $\tau$-phase, calculated with the extended Hückel method, contains a flat-bottomed band.

In terms of the tight-binding model, we can regard that the in-plane molecular configuration is such that the next-nearest intermolecular hopping, $t_2 \simeq 0.02$ eV, appears in every other plaquetts in a checker-board manner on top of the nearest one $t_1 \simeq 0.2$ eV (Fig. 5). The checker-board makes the Brillouin zone folded, where the dispersion of the upper band in which $E_F$ resides is like the square root of Eqn. (1) with $t' = 0.5$ when the splitting due to $t_2$ is small. This way we can have an anisotropically flat dispersion along $k_x, k_y$ with a large $D(E_F)$ at the bottom of the band, so a ferromagnetic component in the spin fluctuation exists for this dispersion, as we have checked with FLEX. If we naively plug in $1/8$ ($\sim 0.05$ eV) of the width of the upper band calculated in ref. 20 for $t$, the $h = 0.05$ for $U = 2$ in Fig. 3 corresponds to $B \approx 30$ T. Since the divergence of $D(E_F)$ is weaker than that for Eqn. (1) because of the square root, large values of $U$ should be necessary to obtain appreciable negative magnetoresistance. In organic materials $U$ may be indeed large, but we have then to check whether the band mixing across the gap for large $U$ can smear out the ferromagnetism. Our preliminary result for the two-band model with the multi-band FLEX shows that the ferromagnetic component in the spin fluctuation does remain for $U \sim 4$, although it becomes weaker. Thus quantitative estimates of the magnetoresistance for the material should include these effects.

Murata et al. have in fact observed a negative magnetoresistance for $B \sim$ several tesla with some hysteretic behaviors in this material. Theoretically, however, we can argue that dominant ferromagnetic fluctuations are not a necessary condition for the negative magnetoresistance conceived here. In the present scheme, the the system becomes more conductive in moderate $B$ due to a combination of a correlation-enhanced spin susceptibility and the flat-bottomed dispersion. So all we have to have about the magnetism is a large enough susceptibility. Thus the negative magnetoresistance observed by Murata et al. is understandable provided the ferromagnetic component is present, if not dominant, in the spin fluctuation for the present mechanism to be relevant.

Although we have exemplified our idea so far in two dimensions, we believe that the present mechanism should be general, and can be found in other models with strong ferromagnetic spin fluctuations such as face centered cubic lattice, whose band structure has flat and dispersive parts as well.

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A preliminary result (K. Murata, private communications) indicates both an enhanced susceptibility from an $M - H$ curve and antiferromagnetic fluctuations from NMR. The present model for $\tau$-conductor should in fact be strongly antiferromagnetic if $t_2$ were absent, since the system reduces to a nearly half-filled square lattice. On the other hand ferromagnetic fluctuation is dominant if the lower band is neglected, so antiferromagnetic and ferromagnetic fluctuations should coexist for finite values of $t_2$, which will imply a coexistence of antiferromagnetism and the negative magnetoresistance.

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FIG. 1. The phase diagram of the $t$-$t'$ Hubbard model, determined by the exact diagonalization of 8 electrons in $4 \times 4$ system. The right panel is the one-electron dispersion for $t = -1, t' = 0.5$.

FIG. 2. The wave number dependence of $\chi^\text{RPA}_k$ for $t' = 0.5, U = 2, n = 0.4, T = 0.1$.

FIG. 3. The temperature dependence of the static magnetic susceptibility $\chi^\text{FLEX}_k$ (dashed line) and $\chi^\text{RPA}_k$ (full line) for $U = 0.5, 1.0, 2.0$ with $t' = 0.5, n = 0.4$.

FIG. 4. The temperature dependence of the resistivity of the $t$-$t'$ Hubbard model with (bottom) or without (top) vertex corrections for $t' = 0.5, U = 2, n = 0.4$ in zero magnetic field ($h = 0$, full line) and in a magnetic field ($h = 0.05$, dashed line).
FIG. 5. The in-plane molecular configuration in \( \tau\text{-D}_2\text{A}_1\text{A}_y \). The solid lines denote the nearest neighbor hopping while dashed lines the second nearest neighbor hopping between face-to-face molecules.