Almost-Localized Electrons in a Magnetic Field

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It is shown that, within a Gutzwiller type of approach to correlated electrons, the effective mass of quasiparticles composing an almost-localized Fermi liquid is spin dependent and varies strongly with magnetic field. The magnetization of such a system saturates in physically accessible fields. The results are used to explain the field dependence of both the effective mass and of the specific heat in the heavy-fermion systems at low temperature.

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A spectacular magnetic-field dependence of the effective mass has been observed in the heavy-fermion system CeB_6 by Joss *et al.*[1] In this paper we provide a theoretical explanation of this behavior which is assumed to take place in the low-temperature (coherent) regime of the Kondo lattice, i.e., when a narrow band is formed due to the hybridization of localized ($4f$) and itinerant ($2p$) states, with an almost integer valency of the $4f$ states. To achieve that goal we discuss first some unique novel features of almost-localized electrons in an applied magnetic field. Namely, we show that the magnetization of an almost-localized Fermi liquid (ALFL) saturates in a relatively low field, a behavior usually ascribed to the presence of localized moments in the system. This property, as well as the large values of the effective mass (which is also spin dependent) discussed below, demonstrates that ALFL's form a separate class of metals.

Our point of departure is the quasiparticle picture of narrow-band electrons obtained by a reformulation[2] of the Gutzwiller-Brinkman-Rice theory.[3] We assume that the band is almost half filled, i.e., the filling is $n=1-\delta$, with $\delta \ll 1$. The single-particle energy in an applied magnetic field H_a is then

$$E_{\mathbf{k}\sigma}=\Phi_\sigma \epsilon_{\mathbf{k}} - \tfrac{1}{2} g\mu_B H_a \sigma , \quad (1)$$

where $\epsilon_{\mathbf{k}}$ is the bare band energy, and Φ_σ is the band narrowing factor and is related to the $\mathbf{k}$-independent, ω-(energy-) dependent self-energy via $\Sigma_\sigma(\omega)=(1-\Phi_\sigma)\omega$, so that the effective-mass renormalization is $m_\sigma^* \equiv (1-\partial\Sigma/\partial\omega)^{-1}=\Phi_\sigma^{-1}$. The factor Φ_σ can be calculated explicitly by a straightforward generalization of our previous[4] approach to the case with $H_a \neq 0$. Explicitly,

$$\Phi_\sigma=(1-n_\sigma)^{-1}\{(1-n)+(2/n_\sigma)\eta-(1/n_\sigma^2 n_{-\sigma})\eta^2\} , \quad (2)$$

where $n_\sigma=\langle n_{i\sigma}\rangle$ is the average number of particles (per site) with spin $\sigma \pm 1$, and $\eta \equiv \langle n_{i\uparrow} n_{i\downarrow}\rangle$ is the probability of double occupancy at site i. This expression contains the correlation function $\eta \ll 1$ which is determined[2,3,5] by minimizing the system energy E which contains both the band and the short-range Coulomb-repulsion parts, i.e.,

$$E/N=(1/N)\sum_{\mathbf{k}\sigma} E_{\mathbf{k}\sigma}\bar{n}_{\mathbf{k}\sigma}+U\eta , \quad (3)$$

where $\bar{n}_{\mathbf{k}\sigma}$ is the Fermi-Dirac distribution function, and U is the magnitude of the intra-atomic Coulomb repulsion. One should note that Φ_σ is determined by calculating first variationally the quantity η from the balance between the band (the first term) and the Coulomb repulsion ($U\eta$) energies since close to the Mott-Hubbard localization they are of comparable magnitude.[2] For $n=1$ and $H_a=0$ it is found that $\Phi_\sigma \equiv \Phi = 8\eta(1-2\eta)$; thus, the present formulation reproduces the earlier result[3] for $n=1$ and provides leading terms in the limit of interest, namely, $\delta \gg \eta$.

For $H_a \neq 0$, Eq. (2) must be additionally minimized with respect to the magnetization $m \equiv \langle n_{i\uparrow}-n_{i\downarrow}\rangle$ per site; the latter procedure yields the equilibrium magnetic polarization $\bar{m}=m/n$. The optimal values of η and m are subsequently used to determine $\Phi_\sigma=\Phi_\sigma(m,\eta)$, and hence the many-body part of the mass enhancement Φ_σ^{-1}.

The principal point we make at this stage is that Φ_σ depends strongly on H_a. Physically, such a big change is caused by the circumstance that for ALFL the band and Coulomb energies are comparable and of opposite sign; hence much smaller (Zeeman, thermal) contributions to the total energy trigger a large change in the optimal values of m and η.[2] This triggering effect is illustrated explicitly in Fig. 1, where the density of quasiparticle states is drawn for both majority ($\sigma=\uparrow$) and minority ($\sigma=\downarrow$) spin directions. From Eq. (2) it follows that the mass $m_\uparrow^*$ will grow with H_a and approach the value $m_\uparrow^* \simeq 1/2\delta \gg 1$ near the saturation value of $m=1-\delta$, where $\eta=0$. On the contrary, the $\sigma=\uparrow$ electrons will acquire the bare band mass m_B^* in the magnetically saturated state. This is because for $\bar{m}=1$ the short-range Coulomb-interaction term vanishes. In the absence of the field, $m=0$, while $\eta=(n^2/4)(1-U/U_c)$ for $U \leq U_c$ and zero for $U \geq U_c$. Then, $m_\uparrow^*=m_\downarrow^* \sim \{2(1-n)$

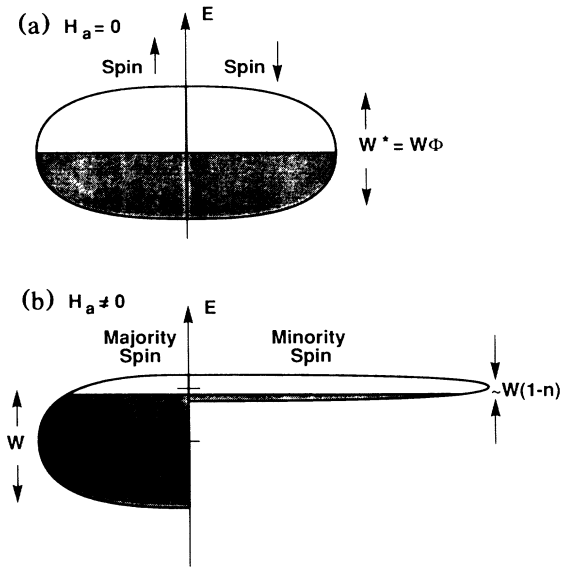

FIG. 1. Schematic representation of quasiparticle states in band narrowed by electron-electron interactions for both the magnetic field $H_a = 0$ (top) and $H_a \neq 0$ (bottom).

$+1-(U/U_c)^2\}^{-1}$ for $U \leq U_c$, and $m_\uparrow^* = m_\downarrow^* = \{2\delta/(1+2\delta)\}^{-1}$ for $U \geq U_c$.

The results of calculations outlined above for the mass enhancement Φ_σ^{-1} are presented in Fig. 2. The featureless (rectangular) form of bare density of states (DOS) with the width W has been chosen in the computations. The effect of Landau quantization on the DOS has been ignored when calculating the total energy since its oscillating component averages out. The results for $n=1$ (top panel) differ remarkably from those for $\delta > 0$. The full Gutzwiller form[3] of $\Phi_\sigma \equiv q_\sigma$ has been taken in the $n=1$ case. The absence of spin dependence of the mass for $n=1$ reflects a precursory localized-moment behavior at and above the threshold U_c ($=2W$ for featureless DOS) for the Mott-Hubbard localization since we do not expect any spin splitting in the localized phase. Furthermore, the results for $n < 1$ plotted in the bottom panel of Fig. 2 when compared with the corresponding mass splitting calculated for the interacting electron gas[6] demonstrate that the present m_σ^* changes exceed those in Ref. 6 by more than 2 orders of magnitude. These dramatic effects are due solely to correlation effects close to the Mott-Hubbard localization, i.e., by the circumstance that for $U \to U_c$ and $n \to 1$ the electric conduction is realized mainly through hole states in the lower Hubbard subband, not by the empty states at and above the Fermi level (note that $\delta \gg \eta$).

The results presented in Fig. 2 provide quantitative

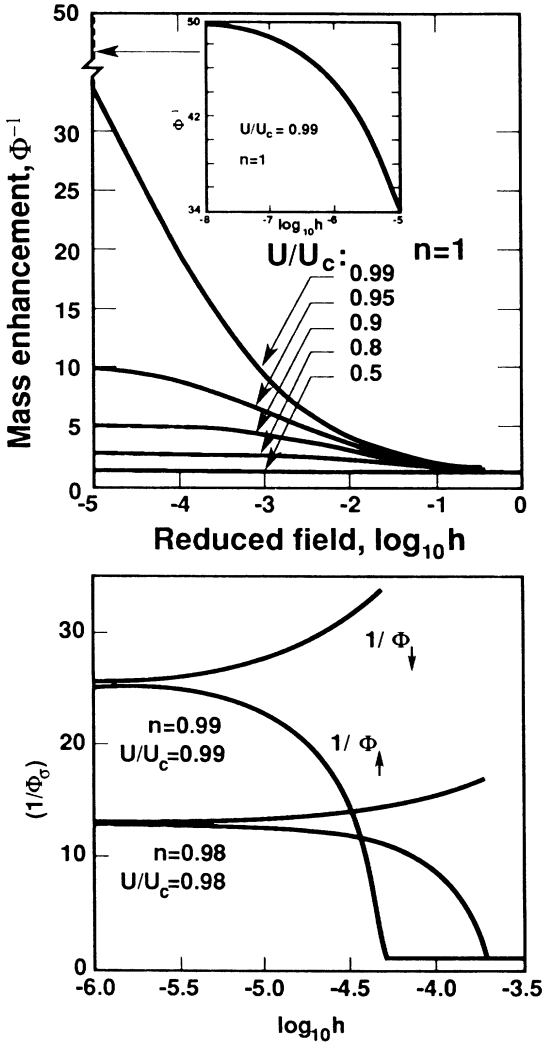

FIG. 2. Effective-mass enhancement vs $h = \frac{1}{2} g\mu_B H_a/W$ for $n=1$ (top) and $n < 1$ (bottom). The x axis is the same on both figures. Inset: The behavior of m^* as $h \to 0$.

calculations of the spin-split masses postulated by Stamp[7] in his approach to the de Haas-van Alphen (dHvA) effect in strongly interacting electron systems. Some specific features of the dHvA data,[1,8] such as the absence of the spin splitting and the observation of a single heavy mass, are understandable only within an explicit model such as that provided here, as discussed below for the system CeB_6.

To obtain the value of the magnetic moment $\bar{m}$ for a given value of reduced field $h \equiv \frac{1}{2} g\mu_B H_a/W$, we calculated the value η for given m from the condition $\partial E/\partial \eta = 0$, which leads to the relation

$$\eta = \frac{n^2-m^2}{4}\left\{1 - \frac{U}{U_c}\frac{(n^2-m^2)[(1-n/2)^2-m^2/4]}{[n(1-n/2)-m^2/2]^2-m^2(1-n)}\right\}. \quad (4)$$

The obtained rapid increase of m with the field can be understood physically by noting that the effective magnetic field

acting on particles of spin σ is[2] $H_\sigma \equiv H_a/\Phi_\sigma$ and with growing H_a we reach the limit $H_\downarrow \gg H_\uparrow$, i.e., strong asymmetric shift of the spin subbands takes place, in addition to their distortion (cf. Fig. 1). In effect, the moment saturates in physically accessible fields. Also, the magnetization curves are nonlinear for $n=1$ while for $n<1$, $m=2h/(1-U/U_c)^2$. The difference between $n=1$ and $n<1$ in this respect can be understood by noting that for $n=1$ and $\eta \to 0$, the system transforms into a lattice of localized moments with $U \to U_c$, while for $n<1$, the metallic phase is always stable because the number of holes is nonzero even for $n \to 0$. Only the latter case will resemble the linear, Fermi-liquid-like dependence of $m(H_a)$ which saturates in the field $h_c=(1-U/U_c)^2/2 \ll 1$. Also, in the $n=1$ case we observe an upturn in $m(H_a)$ dependence, not encountered for strictly localized moments. Such behavior mimics a metamagnetic behavior and has been obtained before.[7]

We apply our theory to the heavy-fermion system CeB_6 for which the number of carriers is $n=1$ per unit cell containing one Ce atom.[8] In our view the carriers cannot be attributed to B atoms since each of them would contribute at least one $2p$ electron to the conduction band resulting in even n. Consequently, we assume that in the low-temperature limit $k_BT \ll W^* = W\Phi_0$, the $4f$ electrons form a narrow band which is almost half filled ($n_f \to 1$). Thus, only quasiparticles with one (large) mass will be present for $H_a=0$. Similar properties are observed in $CeCu_6$ (Ref. 9), though the explicit form of $m^*(H_a)$ dependence has been determined only in the CeB_6 case. On the other hand, if the magnetization is substantial, almost all electrons will occupy the majority-spin subband. For CeB_6, the dHvA measurements of m^* were reported for the applied field in the range 13–30 T. For $H_a=13$ T, $\bar{m} \simeq 0.5$, and $n_\uparrow = 0.75$ and $n_\downarrow = 0.25$; under these conditions $m_\downarrow^* \gg m_\uparrow^*$. Also, the short lifetime of the minority-spin quasiparticles mentioned above will hamper their detection. This is because for $\bar{m} \to 1$ we have few electrons in the minority-spin subband, each encountering many $\sigma=\uparrow$ electrons and hence scattering strongly. The resulting strongly peaked DOS structure for $\sigma=\downarrow$ electrons drawn in Fig. 1 may be influenced by finite lifetime, the effect missing in the present scheme.[2,3] What is most important is that the collection of heavy carriers may be regarded as constituting two different types of electron liquids, one with $\sigma=\uparrow$ and one with $\sigma=\downarrow$. These two subsystems share the common Fermi-level position; therefore, the total number of electrons will determine the period of dHvA oscillations. Furthermore, no spin splitting and no beats can be observed here, whereas in ordinary metals they are always there.[10,11] This explains in a natural way the mysterious absence of such features in the experiments.[1] Nonetheless, it would be important to search for a presence of electrons with the mass $m_\uparrow^* \gg m_\uparrow^*$ at still lower temperatures.

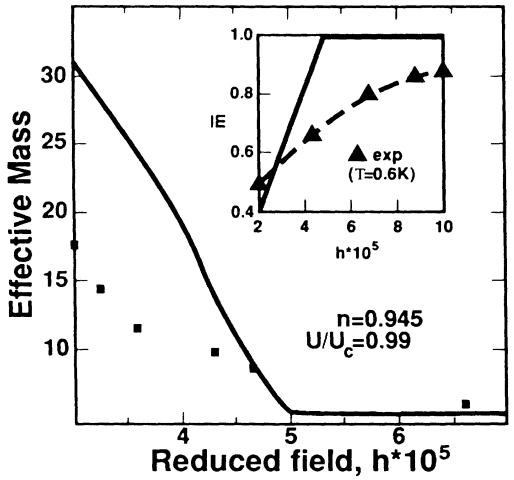

FIG. 3. Field dependence of the effective mass for CeB_6: theory, continuous line; the points represent the data for CeB_6 (cf. Ref. 1). Inset: The corresponding magnetization curves.

In Fig. 3 we have plotted the field dependence of the effective mass $m_\uparrow^*$, and compared the results with the experimental data.[1] The bare bandwidth was chosen to match the observed value of the linear specific-heat coefficient in the zero field, i.e., $\gamma \equiv \frac{2}{3}\pi^2 k_B^2 N_{Av}/W\Phi_0 = 260$ mJ/mol K^2, which, in combination with the result for LaB_6, $\gamma \simeq 2.6$ mJ/mol K^2 and band mass 0.61, yields the band enhancement for CeB_6, $A=61\Phi_0$. The values of δ and U/U_c have been chosen to meet the relation at $H_a=0$, i.e., $\Phi_0=2(\delta+1-U/U_c)$. The field axis has been rescaled according to $h=BH_a$, with $B=2.3\times10^{-6}$ T^{-1}. While the agreement between theory and experiment is not quantitative, our model does properly reproduce the principal features of the data. A more detailed approach would require use of realistic DOS and inclusion of antiferromagnetic ordering.[12] The inset in Fig. 3 illustrates the difference between the calculated and observed[12] $m(H_a)$ curves. One should also notice that the field-dependent mass m^* determined[1] from $\gamma(H_a)$ exceeds that from dHvA measurements. This situation arises because only the electrons from the majority-spin subband contribute to the observed mass in dHvA effect, whereas both spin subbands contribute to γ. Hence, $m^*(H_a)$ as determined from γ is expressed as $(\Phi_\uparrow^{-1}+\Phi_\downarrow^{-1})/2$ and in view of inequality $\Phi_\uparrow \gg \Phi_\downarrow$ the specific-heat data yield m^* values greater than those shown in Fig. 3. Our detailed numerical calculations indeed support the observed[1] behavior: The $m^*(H_a)$ determined from γ are systematically higher than the corresponding dHvA values, and coalesce as $\bar{m} \to 1$. Hence, our theory provides a complete rationalization of the CeB_6 data provided in Ref. 1.

Finally, we discuss the properties in the strong-correlation limit $U \gg U_c$. As the double occupancy $\eta \to 0$, the band narrowing factor (2) approaches the

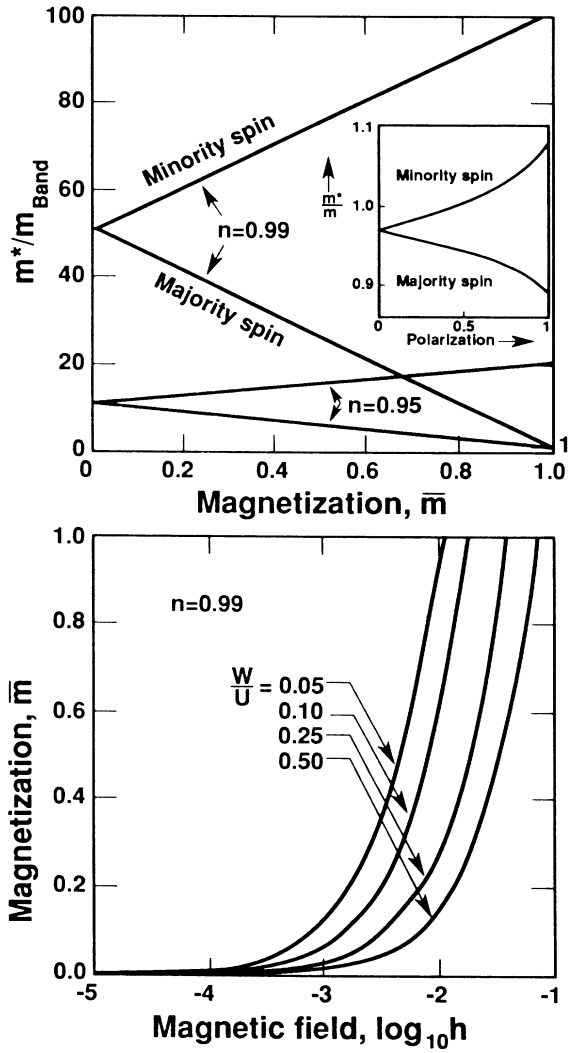

FIG. 4. Effective mass as a function of magnetic polarization $\bar{m}$ (top) and the dependence $\bar{m}$ vs h for the strongly correlated electrons ($\eta=0$). Inset: The $m^*(\bar{m})$ dependence for the interacting gas (taken from Ref. 6).

limit $\tilde{\Phi}_\sigma \simeq (1-n)/(1-n_\sigma)$; the quantity $\tilde{\Phi}_\uparrow^{-1}+\tilde{\Phi}_\downarrow^{-1}$ then does not depend on magnetic field. Therefore, for the systems with $U \gg U_c$, the specific-heat coefficient γ also should not depend on the magnetic field for electrons with the value of U well above U_c. This type of behavior is encountered in UPt_3.[8(b)] In order to calculate m^* and polarization $\bar{m}$ as a function of H_a, we start from the effective Hubbard Hamiltonian in the $U \gg U_c$ limit, with the kinetic exchange interactions included[2,13] and treated in the mean-field approximation. The results for Φ_σ^{-1} and $\bar{m}$ are plotted in Fig. 4. For comparison, the value of mass enhancement for the interacting gas[6] has been provided in the inset. $\bar{m}(H_a)$ changes very rapidly as the system approaches saturation; the antiferromagnetic exchange interaction with the coupling constant $J \sim W^2/U$ shifts the saturation towards fields much higher than h_c specified above for $U < U_c$.

In summary, we have analyzed the magnetic-field dependence of the spin-split effective masses of almost-localized electrons. We have also calculated the magnetization curves and have shown that they saturate in physically accessible fields. The results obtained here rationalize the experimental observations for the heavy-fermion systems at low temperature.

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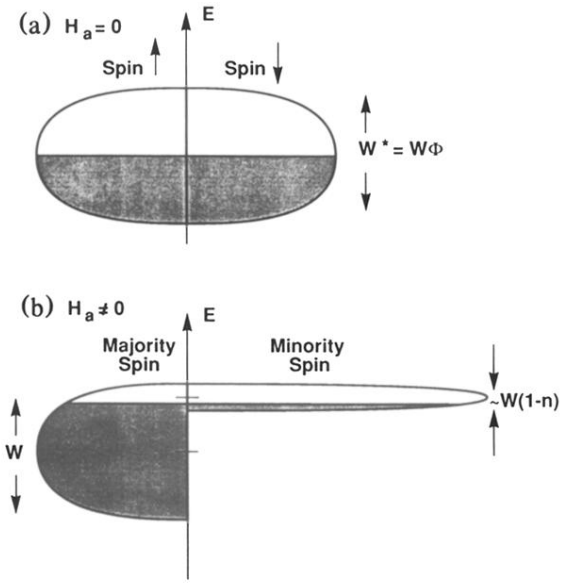

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