Correlated electron theory of strongly anisotropic metamagnets

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I. INTRODUCTION

While ferromagnetism is known since antiquity, antiferromagnetism was only discovered in this century. It is not widely known that the concept of antiferromagnetic order was proposed independently by Neel [3] in 1932 and Landau [4] in 1933. Both sought to explain, at that time, puzzling low temperature behavior of the magnetic susceptibility of certain materials: of metals such as Cr and Mn in the case of Neel, and of insulators with layered structure such as the chlorides of Cr, Co and Ni in the case of Landau. While Neel correctly suggested the existence of interpenetrating sublattices in Cr and Mn with opposite magnetization [3], Landau equally correctly predicted the existence of stacks of ferromagnetically ordered layers whose magnetization alternates from layer to layer [4]. In both cases the total spontaneous magnetization adds up to zero. Assuming the interlayer coupling to be weak, Landau [4] argued that a relatively small magnetic field would suffice to modify the mutual orientation of the moments in each layer. This leads to deviations from the linear dependence of the total moment on the field, i.e., to an anomalous increase of the susceptibility, and finally (at high fields) to a saturation of the magnetization. Such a behavior was indeed observed by Becquerel and van der Handel in 1939 [3] in the manganite (carbonate of Fe and Mg) at low temperatures. Not being aware of Landau’s or Neel’s work they could not explain their observation in terms of ferro- and paramagnetism, and therefore suggested for it the name m metamagnetism [5].

Qualitatively similar, but even more drastic magnetization effects were later observed in many other systems of which FeCl₂ and Dy₃A₄O₁₂ (DAG) are well-studied prototypes [6]. These materials are insulators where the valence electrons are localized at the Fe and Dy ions, respectively. The resulting local order from antiferromagnetic planes is constrained to lie along an easy axis, implying a strong anisotropy such that a small spin-op transition cannot occur. Under the influence of a large magnetic field the staggered magnetization vanishes in a first or second order phase transition, the so-called metamagnetic transition. Apart from the thoroughly investigated m metamagnets mentioned above, there are also conducting systems in that class, e.g., Uramum (based on the well-ordered Cr₃As₂), Sm Mn₂Ge₂ [7], and TbRh₂₁₋ₓIrₓ [8].

Today the term ‘‘m metamagnetic transition’’ is used in a much wider sense [9, 10] namely whenever the magnetic susceptibility (χ) has a maximum at some value H_c, with m (H_c) being strongly enhanced for H > H_c. M metamagnetism is then found to be a rather common phenomenon which occurs also in spin-op antiferromagnets (e.g. the parent compound of high-T_c superconductivity, La₂CuO₄ [11]), strongly exchange-enhanced paramagnets (e.g. Tb₇Be₂YCo₂ [12]), heavy fermion and intercalation transition metal systems (e.g. CeRu₂Si₂, UPt₃ [13]).

In this paper we will be concerned only with m metamagnets in strongly anisotropic antiferromagnets, several of which are known to have a very interesting H-T phase diagram (H: internal magnetic field, T: temperature). In particular, in the insulating systems one often finds a tricritical point at which the first order phase transition becomes second order. Theoretical investigations of tricritical points began with the work of Landau, who described the critical behavior within his phenomenological theory of phase transitions [14]. Clearly, a genuine understanding of the origin of tricritical points etc. requires investigations on a more microscopic level.

In the case of strongly anisotropic metamagnets these investigations where to restricted to the insulating system, such as FeCl₂. They are usually based on the Ising model
where the summations extend over the nearest neighbors (NN) and next nearest neighbors (NNN) of every site. For \( J^2 > 0 \) one has an antiferromagnetic (AF) coupling between the \( Z \) nearest neighbor spins and a ferromagnetic coupling between the \( Z^0 \) next nearest neighbors \([8]\). It was pointed out by Kicinski and Cohen \([7]\) that in mean-field theory there exists only for \( R = (2 J)/3 = 5 \) a critical end point (CE) and a bicritical point (BCE) (see Fig. 1a). The latter behavior, especially the absolute angle between the two transition lines at CE and the pronounced minimum at the second order line, is qualitatively very similar to that observed in FeB \([2,3]\). However, the first order between CE and BCE has so far not been observed (not even theoretically when evaluating \([10]\) beyond mean-field theory \([11,12]\)). Most recently, with measurements of the excess magnetization and anomalous susceptibility \([13]\), the specific heat \([14]\), and with neutron scattering \([15]\) a stripe (shaped regime of strong non-critical fluctuations in the \( m \) plane) that is the applied field of FeB \([2,3]\) was reported which is at least reminiscent of the critical line \( CE \) \& BCE. This unusual behavior was then also found theoretically by Selke \([16]\) who evaluated \([10]\) and also a more detailed model \([17]\) for weak ferromagnetic coupling \((R = 0\)t) and large \( Z \) using Monte Carlo techniques. There does not yet exist a microscopic theory for the conducting systems such as the Uranium (based mixed systems \([17,18]\)), because this requires a fully quantum mechanical treatment of itinerant, correlated electrons. First steps in this direction, where genuine correlation effects were, however, neglected, are the semi-phenomenological theories of Wohlforth and Rhodes \([19]\) for magnetic phase transitions in paramagnetic and magnetic states of a systems. It is the purpose of this paper to examine the origin of the magnetic state of this model at half filling \((n = 1)\) to know if this state is the (paramagnetic) ground state of this model. To this end we will study the Hubbard model in the presence of a strong anisotropy direction, with a magnetic field along this direction, since it is the simplest microscopic model that describes insulating and metallic, spin-localized and bandlike antiferromagnets with easy axis in an external magnetic field.

In Sec. II the underlying model, the Hubbard model with easy axis, is introduced and its validity is discussed. Then the dynamical mean-field theory which is used to investigate the correlation problem all the way from weak to strong coupling, as well as the Quantum Monte Carlo techniques employed to solve the coupled self-consistency equations, are discussed in Sec. III. The results obtained for a half-filled band and for finite doping are presented in Sec. IV and Sec. V, respectively. A discussion of the results in Sec. V closes the presentation.

![FIG. 1. Schematic phase diagram, magnetic field \( H \) vs. temperature \( T \), for a) a typical Ising-type metamagnet (TCP: tricritical point), b) the Ising model (AF theory with \( R < 3 = 5 \) (CE: critical endpoint, BCE: bicritical endpoint). Solid lines: first order transition, broken lines: second order transition; P: antiferromagnetic phase, A: paramagnetic phase.](image-url)
by the broken rotational symmetry of the crystal lattice) into spin space, producing one, or more, easy axes which constrain the spins \( \mathbf{s} \). In an external magnetic field such a constraint leads to metastable magnetic transitions, with or without a spin flip depending on the strength of the spin-orbit interaction, as explained in Sec. I.

A microscopic theory of strongly anisotropic antiferromagnets should ultimately be able to take into account the orbital degeneracy of the electrons, and by including the relativistic spin-orbit interaction \( \mathbf{L} \cdot \mathbf{S} \) in the Hamiltonian, to generate an anisotropy axis within the model itself. At present, this is technically not possible [3]. Therefore we take the existence of the anisotropy axis for granted: we employ the Hubbard model [2] and constrain the magnetic moments to lie along \( \mathbf{k} \) (e). By this approach the kinetic energy and the Coulomb interaction are treated microscopically, whereas the relativistic corrections are not. We note that the relativistic corrections are of the order of \( 10^{-2} \) eV and are thus small compared to energies of the order of 1 eV for kinetic and Coulomb energy. Therefore the existence of the anisotropy and the correlation physics described by the Hubbard model are quite unrelated. This justifies our approach where the existence of the anisotropy is a priori assumed.

III. DYNAMICAL MEAN FIELD THEORY (ED 1)

For classical spin models (e.g., the Ising model) it is well known that the Weiss molecular field theory becomes exact in the limit of high spatial dimensions \( d = 1 \). For lattice electrons this limit was introduced only recently [31]. With the proper scaling of the hopping element in [3], \( t = t = Z \) (where \( Z \) = number of nearest neighbors), it leads to a quantum mechanical dynamic mean field theory (DFT); for a review see [32]. The interacting lattice model reduces to a self-consistent single site problem of electrons in an effective medium [3], which may be described by a complex, frequency dependent (i.e., dynamic) self energy \( \mathbf{i} \). This problem is, in fact, equivalent to an Anderson impurity model complemented by a self-consistency condition [33].

There are two limits in which the DFT recovers well-known static mean field theories:

1. Weak coupling: In this situation the effective medium may be approximated by a static field \( \mathbf{h} \), which is generated by the averaged densities of the electrons. This leads to the Hartree-Fock approximation, e.g., \( \mathbf{h} \) = \( \mathbf{U} \). In the homogeneous case, which is expected to give the qualitatively correct behavior at weak coupling, the averaged densities \( n \) have to be determined self consistently.

2. Strong coupling at half filling: Here the model [3] can be mapped onto the antiferromagnetic spin 1/2 Heisenberg model for which the limit \( d = 1 \) becomes equivalent to the Weiss molecular field theory.

The results obtained in these two limits will be presented, and compared to the results for interelectronic coupling, in Sec. IV.

The DFT has recently provided valuable insight into the physics of strongly correlated electron systems e.g., the Mott-Hubbard transition [33, 36] and transport properties [37]. The effect of the magnetic field on the model for \( n = 1 \) was also studied [40]. In particular, Laboux et al. [41] thoroughly investigated the magnetic behavior of the paramagnetic phase, assuming the AF order to be suppressed. For \( U = 3 \) \( Z \) t they found a first order magnetic transition between the strongly correlated metal and the Mott insulator at a critical field \( H \approx 0.2t \). Seekekus and Band et al. [42] took into account the AF order. They considered the isotropic case where the field orients the staggered magnetization perpendicular to itself, such that a magnetic transition cannot occur.

To investigate the magnetic phase transition of an antiferromagnet to a paramagnet we consider a bipartite (A-B) lattice and allow for symmetry breaking with respect to spin: \( Z f_{i}^{*} g = f_{i}^{*} + f_{j} \) and sublattice \( Z f_{i}^{*} B g = f_{i}^{*} - f_{j} \). The self energy \( G_{n} \) \((i_{1}, n)\), with Matsubara frequencies \( \Omega_{n} = (2 \pi n + 1)/\omega \), and the renewal function \( G_{n} \) are determined self consistently by two sets of coupled equations [33, 34]:

\[
G_{n} = \mathbf{d} Z n^{2} \mathbf{Z} n \mathbf{N}^{0}() \tag{3}
\]

\[
G_{n} = \mathbf{h}_{n} \mathbf{G}_{n} \mathbf{G}_{n} : \tag{4}
\]

Here \( Z_{n} = i_{1}^{*} n + n i_{1} \), and the thermal average of some operator \( \mathbf{O} (; \cdot ; \cdot) \) in [3] is defined as a functional integral over the Grassmann variables \( ; \mathbf{O} (; \cdot ; \cdot) \), with

\[
\mathbf{d} = \mathbf{Z} D \mathbf{D} \mathbf{D} \mathbf{D} : \mathbf{O} (; \cdot ; \cdot) ; \tag{5}
\]

in terms of the single site action

\[
A = \sum_{n} (\mathbf{G}_{n})^{-1} + \sum_{n} \mathbf{Z} U \mathbf{d} \mathbf{O} () \mathbf{d} () \mathbf{d} () : \tag{6}
\]

where \( Z \) is the partition function, and \( N^{0}() \) is the density of states (DOS) of the noninteracting electrons. As the results do not much depend on this precise form we choose a half-filled DOS \( N^{0}() = (2t)_{2}^{2} = 2 \mathbf{t} = (2 \mathbf{t})_{2}^{2} \). The constraint \( m, k \) \( H \) is enforced by setting the overlap diagonal (in spin space) elements of the Green function equal to zero: \( G_{0} = f_{i}^{*} = 0 \). From now on \( t = 1 \) will set our energy scale, i.e., the
total band width of $N^{-}\infty(\cdot)$ is equal to 4. The Dyson equation \[^\text{[3]}\] introduces the lattice into the problem. It couples $A$ and $B$ sublattices and can be solved by a simple integration, even analytically for the above DOS. The functional integral \[^\text{[4]}\] however is highly nontrivial since it couples all $M$ atomic frequencies. Georges and Kotliar \[^\text{[3]}\] and Jarrell \[^\text{[3]}\] realized that the action \[^\text{[3]}\] is equivalent to that of an Anderson impurity model, and can therefore be treated by standard techniques developed for this model. Here we employ a single temperature, auxiliary field Quantum Monte Carlo (QMC) method \[^\text{[3],[4]}\]. In this approach the electron-electron interaction is formally replaced by an interaction of independent electrons with a dynamical auxiliary field of Ising(type spins). To this end the integral $[D]$ is discretized into steps of size $\varnothing$. Equivalently, there is a high energy cut off of $M$ atomic frequencies, i.e. $\varnothing^m_j - T \varnothing^n + 1 < \varnothing, \varnothing = -2, \ldots, 2, 1$. All quantities have to be extrapolated to 0. The computer time grows like $3^2/3^2$, restricting to values below 150 and 500 on present supercomputers. For small $\varnothing$ one can perform a full enumeration (instead of the Monte Carlo sampling) of all possible configurations of the auxiliary field. We never encountered a minus sign problem hence no further approximations (like the $x$(node) method) were necessary.

The selfconsistency is obtained iteratively as follows: the Green function $G$ (omitting indices) is calculated from some initial self-energy, e.g. 0, by the Dyson equation \[^\text{[3]}\]. Now the new Green function $G_{\text{new}}$ is determined by solving \[^\text{[3]}\] with the QMC method. Finally, the calculation of the new self-energy $\varnothing_{\text{new}} = G_{\text{new}}^{-1} + G^{-1}$ completes one iteration. To prove convergence in the symmetry broken case $G_A$ and $G_B$ are updated by the Dyson equation \[^\text{[3]}\] after every QMC simulation for one sublattice (Eq. \[^\text{[8]}\]). In the symmetry broken phases, typically $10^2$ to $20^2$ iterations with 20000 M sweeps are necessary to obtain a convergence of $10^{-3}$. The calculation of an magnetization curve at $T = 50$ takes about 100 hours on a Cray-Y-MP. Close to a phase transition the convergence is much slower and the statistical errors are larger due to strong fluctuations, in particular in the case of a second order phase transition. These effects limit the accuracy in the determination of the critical values of the model parameters, e.g. the critical magnetic field (see Sec. \[^\text{IV}\]). At large $U$-values ($U > 4$) the Monte Carlo sampling becomes more and more ine cient due to a sticking problem, i.e. there are two (or more) $m$-independent solutions in the free energy and the single spin algorithm is no longer able to transfer between them.

From the resulting Green functions we calculate the densities and the homogeneous and antiferromagnetic magnetization:

\[
\begin{align*}
\rho &= \frac{1}{2} X \\
m &= \frac{1}{2} X \\
m_{\text{st}} &= \frac{1}{2} X \\
\end{align*}
\]

Since we are interested in the question whether the system is insulating or metallic we also determine the electronic compressibility \[^\text{[\varnothing]}\]. It was calculated both by numerical differentiation of $n(\cdot)$ and from the two-particle correlation functions (see \[^\text{[3],[4]}\] and Appendix \[^\text{A}\]). Both results agree with the statistical errors: however, the latter method is much more time consuming.

\section{Results for Half Filling}

\subsection{Weak coupling}

For weak coupling ($U < t$) we expect the Hartree-Fock approximation to give an, at least qualitatively \[^\text{[3]}\], correct picture of the metal magnetic phase transitions especially of their order. Within this static mean electron theory electronic correlations are neglected and the interaction is decoupled as

\[
\begin{align*}
\rho_i &= \rho_i^H \\
\rho_i^H &= \rho_i > + \rho_i < \\
\rho_i^H &= \rho_i > + \rho_i < \\
\end{align*}
\]

Note, that while for $k \neq \mathbf{0}$ the Fock term vanishes, $\chi_{ij}^F \chi_{kl} > 0$, its presence is essential in the case $m_{\text{st}} > m$ (see Appendix \[^\text{B}\]). We can use our investigations to states with homogeneous sublattice magnetization as described by the Ansatz

\[
\rho_i \rho_i^H > = \frac{1}{2} (\rho_i + m + m_{\text{st}}); \quad (9)
\]

Applying this Hartree-Fock decoupling scheme one obtains the effective one-particle Hamiltonian

\[X \begin{align*}
\rho_i &= \rho_i^H \\
\rho_i^H &= \rho_i > + \rho_i < \\
\rho_i^H &= \rho_i > + \rho_i < \\
\end{align*}
\]

This Hamiltonian is diagonalized and the one-particle energies $\epsilon$ are calculated as

\[
\epsilon = \text{sgn} (\cdot) \frac{2}{U} n_{\text{st}}^2 + \frac{U}{2} n_{\text{st}} + H; \quad (11)
\]

where $\text{sgn} (\cdot)$ denotes the sign of the noninteracting electron energy. In the antiferromagnetic phase the DOS
has a gap of width $U_{\text{st}}$, with square root singularities at its edge.

From the one-particle energies $\varepsilon_i$ the grand potential per lattice site ($L$ being the number of lattice sites) is obtained directly as

$$\gamma = \frac{1}{L} \ln Z = -\frac{1}{L} \sum_i N_i^0 \left( 1 + e^{-\varepsilon_i} \right)$$

$$+ \frac{U}{4} \left( \langle m^2 \rangle + m^2 \right) \frac{U n^2}{4}.$$ (12)

The potential has two shallow minima, one corresponding to the paramagnetic state ($m = 0; m_{\text{st}} = 0$) and the other to the antiferromagnetic state ($m = 0; m_{\text{st}} = 0$). By applying a sufficiently strong magnetic field to the antiferromagnetic state, the paramagnetic minimum becomes the lowest, such that a first order phase transition takes place. The reason for the occurrence of a first order phase transition is that, within the (static) Hartree-Fock approximation, the $U$ term becomes dominant for largest (static) local moments. Therefore, pure antiferromagnetic and ferromagnetic order are both energetically favored. Magnetic states with $m = 0$ and $m_{\text{st}} = 0$ (which would occur in the case of second order phase transitions) have a small local moment, i.e., a high Hartree-Fock energy, on every second site.

To calculate the magnetization curves, the self-consistent Hartree-Fock equations are obtained from the minimization conditions $\varepsilon = 0_{\text{st}} = 0$ and $\theta = 0 = 0_{\text{m}}$:

$$m_{\text{st}} = \frac{U}{Z} \sum_i N_i^0 \left( \frac{m_{\text{st}}^2}{1 + e^{-\varepsilon_i}} \right) \quad \text{(13)}$$

$$m = \frac{U}{Z} \sum_i N_i^0 \left( \frac{1}{1 + e^{-\varepsilon_i}} \right) \quad \text{(14)}$$

These equations are solved numerically by iteration and integration according to Newton-Cotes rules.

Within the Hartree-Fock approximation the metamagnetic phase transition is found to be of first order for all $T$, even for $U = 4$ (= band width); see Fig. 2 and 3. Hence a tricritical point never occurs. In this parameter range the Quantum Monte Carlo calculations, however, already show second order transitions in a broader range of temperatures (see Sec. IV C). Hence the Hartree-Fock solution can neither describe the experimental situation, where tricritical points are known to occur, nor the correct behavior of the model for intermediate values of $U$.

To estimate the anisotropy energy associated with the easy axis we compare the Hartree-Fock energies of the con gurations with $m_{\text{st}} = k m$ and $m_{\text{st}} = m$ (for details see Appendix E). At half filling and for $U$ equal to the band width, the difference between the free energy of these configurations does not exceed a few percent of the band width, i.e., $0(10^{-2} \text{eV})$. In this situation the spin-orbit interaction, which can be relatively strong, $0(10^{-1} \text{eV})$, indeed leads to a strong anisotropy, i.e., an easy axis $e$, along which $m_{\text{st}}$ is rigidly fixed.

In this system where hitherto described by the theory of itinerant electron metamagnetism (EEM). In the case of an antiferromagnetic system, a magnetic field $H$ and $U = 4$ were proposed a Landau theory with free energy

$$F (m; m_{\text{st}}) = \frac{1}{2} - m^2 + \frac{1}{2} m_{\text{st}}^2 + a m^4 + b m_{\text{st}}^2 + c m^2 m_{\text{st}}^2 + d m_{\text{st}}^4 + e m m_{\text{st}} H m; \quad \text{(15)}$$

where $m$ and $m_{\text{st}}$ are the homogeneous and staggered susceptibilities, respectively, and the coefficients $a$, $b$, $c$, and $d$ are the fourth-order derivatives of the non-interacting free energy. Within the EEM theory the Coulomb interaction is treated in random phase approximation. The corresponding susceptibilities are given as

$$\frac{1}{m} = \frac{1}{U_m} U; \quad \frac{1}{m_{\text{st}}} = \frac{1}{U_{\text{st}}} U.$$ (16)
where $\chi_m^0$ and $\chi_m^0$ are the respective susceptibilities of the non-interacting system. The random phase approximation for these susceptibilities is equivalent to the Hartree-Fock scheme described above. Therefore, we may ask whether we obtain the EEM in the limit $U \to 0$ (where $m$ and $m_{st}$ are assumed to be small). The answer is not straightforward and since the prefactors in the expansion depend, for example, on the lattice structure, on Bethe lattice electrons, as discussed here, they diverge for $T = 0$. Thus an expansion of the free energy in powers of $m_{st}$ and $m$ as assumed in the EEM Landau theory is not possible in general (for details see Appendix C).

In the limit $U \to 0$ the Hubbard model at half filling ($n = 1$) is equivalent to an effective Heisenberg spin model

$$H_{\text{Heis}} = \frac{J}{2} \sum_{i} S_i^x S_{i+1}^x + \sum_{i} 2H S_i^z;$$

where the antiferromagnetic exchange coupling is obtained in second order perturbation theory as $J = 4t^2 - U$. Spin operators are defined as $S_i^x = \frac{1}{2}(\sigma_i^x - \tau_i^y)$, $S_i^z = \frac{1}{2}(\sigma_i^x + \tau_i^y)$, and $S_i^y = \frac{1}{2}(\sigma_i^x - \tau_i^y)$. For this model the Wannier molecular field theory becomes exact in $d = 1$ yielding, under the constraint of uniaxial magnetization, the same results as for the Ising model. For Ising models in real space magnetization and second order transitions are well-studied. In the case of a purely antiferromagnetic next-neighbor coupling, the phase transitions are of first order only at $T = 0$, but of second order at all $T > 0$. The transition line in the $H - T$ phase diagram has indeed the form shown in Fig. 1a, but with a critical temperature of $T_c = 0$. This behavior can be understood already within Weiss molecular field theory, where the ground state energy per site is

$$E_{\text{m}}(\mathbf{n}_m, \mathbf{n}_{st}) = \frac{J}{8} (\mathbf{n}^2 - \mathbf{n}_{st}^2) H \mathbf{n} + \frac{U}{2} \mathbf{n}^2;$$

with

$$J = \frac{2t^2}{U}.$$

Magnetization with respect to $m$ and $m_{st}$ shows that the fully polarized antiferromagnet ($m_{st} = 1$) has lowest energy for $H < J = 4$, whereas the fully polarized ferromagnet ($m = 1$) is energetically favored for $H > J = 4$. Thus, by applying a magnetic field a first order transition is induced. At $H = J = 4$ the states are highly degenerated. A small magnetic phase with $m + m_{st} = 1$ have the same energy. For $T > 0$ this degeneracy is lifted by entropy which favors fully polarized phases. Therefore the first order transition at $T = 0$ in $m$ and $m_{st}$ become second order for $T > 0$, i.e., $T_c = 0$. Indeed, a tricritical point at a finite temperature is only obtained in the case of spin interactions which simultaneously favor both fully polarized antiferromagnetic and ferromagnetic configuration. In particular, adding a ferromagnetic interaction to between nearest neighbors (NNN) on a simple cubic lattice as in [4] stabilizes both ferromagnetic and antiferromagnetic order.

While in the case of effective spin models a ferromagnetic NNN coupling term is introduced ad hoc, simply to obtain the first order phase transition, this term naturally arises if we expand the strong coupling perturbation series of the Hubbard model to $O(U^3)$ in $\xi$. However, besides this $J^0$-Term there also appears a fourth order term $s$. For the hypercubic lattice the effective Hamiltonian $H_e$ reads

$$H_e = \frac{J}{2} \sum_{i,j} (\sigma_i^x \sigma_j^x + \sigma_i^y \tau_j^y) + \frac{t^4}{4} \sum_{i,j} Q_{12} Q_{23} + Q_{14} Q_{24};$$

$$J = \frac{4t^2}{U} + \frac{z t^3}{U^3};$$

$$J^0 = \frac{4t^4}{U^3} C;$$

Here $\xi$ and $\theta$ are lattice vectors connecting a site to its 2 neighbors, and $\tau$ represents a plaquette. Each plaquette is counted only once: the four sites $i; j; k; l$ represent its four corners in clockwise or counter-clockwise order. The constants $A, B$ and $C$ depend on the lattice, and the Hamiltonian operators $Q_{ij}$ are defined as

$$Q_{ij} = 2 \xi_i \xi_j \frac{1}{4}.$$

![Figure 3](image-url)
The plaquette contribution competes with the ferromagnetic NNN term ($J^0$) and drives the system to second order phase transitions. For the hypercubic lattice the plaquette contribution is stronger than the ferromagnetic NNN term, yielding second order phase transitions even for $T = 0$. The same is true for the Bethe lattice where, in fact, $g^0 < 0$ (for details see Appendix E). Thus in strong perturbation theory the metamagnetic phase transition is of second order even at $T = 0$.

C. Intermediate coupling

The perturbation analysis described above demonstrates that the order of the metamagnetic phase transition depends on the Coulomb interaction $U$ in a delicate way. For small $U$ the phase transition is purely of first order and for large $U$ of second order. Apparently, the tri- or multicritical point linking these two regimes must be found at intermediate coupling. In this important, non-perturbative regime quantum Monte Carlo techniques are employed to solve the problem numerically without any further approximation. The results for the magnetization $m$ ($H$) and the staggered magnetization $m_{st}$ ($H$) are shown in Fig. 3 for $U = 2$. Below the Neel temperature a metamagnetic behavior is clearly seen: for small $m$ the magnetization is exponentially suppressed with temperature. Then, towards the metamagnetic phase transition, the susceptibility increases drastically and becomes maximal at the critical field $H_c$. Second order phase transitions are observed for $14 \leq T \leq 0.10$, whereas the transition is of first order at lower temperatures, i.e. $T = 16$. At the phase transition the order parameter, i.e. the staggered magnetization, vanishes. From the curve $m_{st}$ ($H$) the critical $H_c$ and also the order of the phase transition is determined by a square root $t$ for second order transitions and by the mean of the hysteresis for first order transitions.

Using these values of $H_c$ the phase diagram $A$, Fig. 3, for different values of $U$ are constructed. The case $U = 4$ ($= $ band width) and half filling, Fig. 3, was already presented in Ref. [7]. This phase diagram shows both first order (for $T < 16$) and second order phase transitions (for $18 < T < T_N = 0.2$). The $H_c$ dependence at intermediate temperatures, i.e. $15 < T < 18$, is more complex: $m_{st}$ is almost $H_c$-independent in region $AF_1$ (where $m' > 0$), decreases sharply at the boundary to region $AF_2$ (where $m' > 0$) and, upon further increase of $H_c$, vanishes in a second order transition. A though the error bars do not permit an unambiguous interpretation it hence seems that the order parameter decreases by two consecutive transitions: the first one being of first order or corresponding to an anomalous, and the second one being of second order. Taken together the results seem to correspond to the scenario of Fig. 1b. For $U = 2$ phase transitions of first order are found for $T \geq 16$ and of second order for $T < 16$.
weak coupling with 1st order transitions only: as U decreases the regime with second order phase transitions ($T_c < T < T_H$) shrinks, while the temperature regime for 1st order transitions remains nearly unchanged up to $U=2$.

V. RESULTS AWAY FROM HALF FILLING

In the preceding sections metamagnetic transitions were investigated in the case of half filling. Beyond half filling the commensurate antiferromagnetic phase remains stable in the paramagnetic regime under consideration ($\frac{\Omega}{\Omega^*} > 0.875$, $T=16$). Incommensurate spin density waves become stable only in a small density regime at lower temperatures [44]. A further possible instability of the antiferromagnetic phase away from half filling is phase separation, found within the Hartree-Fock approximation and in second order perturbation theory at constant order parameter at $T=0$ [45]. However, at least for $T=16$ we do not observe phase separation since the electronic compressibility $\epsilon = \Omega = 0$ is finite and positive (see Fig. 7).

Upon doping the magnetization curve changes considerably and hardly indicates the existence of a metamagnetic phase transition (Fig. 9). This is due to the fact that in the metallic phase there is no longer a "Slater gap" at the Fermi energy; therefore the homogeneous susceptibility is not as strongly affected by the antiferromagnetic order as at half filling. The phase transition is, however, clearly seen in $m_{\text{sat}}(H)$.
From the \( m_{st} \) vs. \( H \) curve the phase diagram (Fig. 3) is constructed. The metamagnetic phase transition line is found at lower temperatures and fields compared to half filling.

Associated with the metamagnetic phase transition is a change of the electrical resistivity. To study this important effect we calculated the \( H \)-dependence of the electronic compressibility \( \epsilon \). This quantity indicates whether the system is metallic or insulating. For an insulator \( \epsilon \) vanishes for \( T = 0 \) and is exponentially small for temperatures lower than the antiferromagnetic gap. In a Fermi liquid, on the other hand, \( \epsilon (T = 0) \) is finite since it is proportional to the density of states at the Fermi level and hence proportional to the Drude conductivity.
The results for $\rho$ as a function of magnetic field $H$ at $U = 2$ are shown in Fig. 10. At half filling, $\rho = 0$, the compressibility is seen to increase with $H$. This effect is particularly pronounced at low temperatures ($T = 1/25$) where $\rho$ is essentially zero at low fields and rises to 0.3 above the critical field, indicated by an arrow. Hence the metamagnetic phase transition is a transition from an antiferromagnetic insulator to a metal with homogeneous magnetization. At higher temperatures, $T = 1/14$, the compressibility is always finite due to thermal excitations. We note that at $U = 4$, when the electrons are essentially localized, $\rho$ remains small ($0 < \rho < 0.03$) at $T = 1/8$; not shown in Fig. 10) even above the critical field, indicating an insulator-to-metal transition. The situation is very different at finite doping ($\delta = 0.05$). Here Fig. 10 shows that $\rho$ decreases with $H$ by approximately 50% as the system goes through the metamagnetic transition from an antiferromagnetic metal to a metal with homogeneous magnetization.

The field dependence of the electronic compressibility $\rho = \rho(T, H)$ for $U = 2$ at $T = 0$ ($T = 1/14$, $T = 1/25$) and $\delta = 0.05$ ($T = 1/14$). The arrows mark the respective critical fields for the metamagnetic phase transition.

**V. DISCUSSION**

In summary, we investigated the origin of metamagnetism in strongly anisotropic antiferromagnets starting from a microscopic model of strongly correlated electrons, the Hubbard model with easy axis, by employing a dynamical mean field theory. This approach is fundamentally different from previous investigations since we identified and explicitly evaluated the simplest electronic, i.e., fully quantum mechanical, correlation model that is able to explain the conditions for metamagnetism. For this electronic model, we show unambiguously that at intermediate coupling the phase transition is of first order at low temperatures and of second order near the Néel temperature, i.e., the order of the phase transition changes. This phenomenon, which has been of interest to various communities in classical statistical mechanics for a long time already, is extracted from a model of itinerant electrons.

Our approach allows us to describe a broad range of qualitatively different metamagnets within a single model. While at present this simple model does not permit a quantitative calculation of material properties it does describe itinerant and localized, metallic and insulating metamagnets and the crossover between them. This crossover is related to two fundamental experimental parameters, i.e., pressure (related to $U = t$ which decreases with pressure) and doping.

At half filling the Coulomb interaction leads to a crossover from a band insulator to an insulator with localized moments. Here the phase transition changes from first order for the bandlike metamagnet to second order for the localized one. Only at intermediate couplings are both first and second order phase transitions observed as found in experiment. The $H - T$ phase diagram obtained for an intermediate Coulomb interaction ($U = 4 = b$ and $b = 0.5$) is strikingly similar to that of FeB2 [23, 24] or the Ising model with $J = 1$ [4, 19].

We note that in these insulating systems the applicability of a theory which becomes exact for large coordination numbers is justified by the fact that the AF superexchange involves 20 equivalent sites in the two neighboring iron planes [51].

The calculations at half filling allow us to investigate the properties of itinerant metamagnets, such as the Uranium-based Mixed-System [44], for which a theory in terms of a correlated electron model is mandatory. In contrast to the insulating case, the metamagnetic phase transition in the metallic system is hardly visible in the magnetization curve. This is because there is no longer a gap at the Fermi energy. Quite generally, the critical temperatures and fields decrease upon doping.

The metamagnetic transition is accompanied by pronounced changes in the conductivity of the system. The Hubbard model with easy axis can qualitatively describe several scenarios:

(i) In the insulating, localized regime ($U = 4$ at half filling) a magnetic field causes a transition from an antiferromagnetic insulator to an insulator with homogeneous magnetization.

(ii) At lower $U$ values (e.g., $U = 2$) at half filling an insulator-to-metal transition occurs at a magnetic field where the AF order disappears. Such a phenomenon is observed, for example, in the AF phase of La$_{1-x}$Ca$_x$MnO$_3$, where the resistivity is found to change by several orders of magnitude [52]. This is referred to as \textit{colossal} magnetoresistance. We note that...
La₂₋₃CaₓMnO₃ shows no strong anisotropy. Therefore our approach can only describe the general features, in particular the existence of the insulator-to-metal transition.

(iii) Away from half filling a magnetic field induces a transition from a metallic antiferromagnet to a metal without staggered momentum. Here the corepressibility changes by less than an order of magnitude, e.g. about 50% at U = Z₂ = 0.05. A similar effect is found in several strongly anisotropic antiferromagnets, both in multilayers and bulk intermetallic compounds such as UPdGe₃. In these systems the origin of this "giant" magnetoresistance is attributed to band structure effects and spin scattering [3]. By contrast, our approach stresses the importance of genuine electronic correlation effects.

More detailed investigations, including band degeneracy and spin-orbit interaction, may eventually provide even quantitative insight into these interesting and important phenomena.

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Appendix A: Calculation of Susceptibilities from Correlation Functions

Quite generally susceptibilities can be obtained from the derivative of the order parameter w.r.t. the corresponding field:

\[ \frac{\partial \mu_x}{\partial x} = \frac{1}{2} \nabla \cdot \left( f_n \frac{\partial \mu_n}{\partial x} \right) \tag{A1} \]

\[ f_n = \frac{1}{2} \begin{cases} 1 & \text{for } x = \frac{1}{2} \\ 0 & \text{otherwise} \end{cases} \]

with \( f_x = \frac{1}{2} \begin{cases} 1 & \text{for } x = \frac{1}{2} \\ 0 & \text{otherwise} \end{cases} \) \constant{CDW}

Here \( x = H \) and \( x = H_{st} \) lead to the ferrimagnetic and antiferromagnetic susceptibilities, \( x = \) to the electronic compressibility, and \( x = \constant{CDW} \) to the charge density wave susceptibility. From the two self-consistency equations \( \constant{A} \) and \( \constant{B} \) one obtains two corresponding equations for the derivative of the Green function w.r.t. the variable \( x \). The derivative of the functional integral \( \constant{C} \) gives:

\[ \frac{\partial \mu_n}{\partial x} = T_n \begin{pmatrix} 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \end{pmatrix} \tag{A3} \]

\[ \frac{\partial \mu_n}{\partial x} = T_n \begin{pmatrix} 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \end{pmatrix} \tag{A4} \]

\[ \frac{\partial \mu_n}{\partial x} = T_n \begin{pmatrix} 0 & 0 \end{pmatrix} \begin{pmatrix} 1 \end{pmatrix} \tag{A5} \]

where is the local two-particle correlation function

\[ \begin{pmatrix} n_1 n_2 \end{pmatrix} \begin{pmatrix} m_1 m_2 \end{pmatrix} = n_1 n_2 \begin{pmatrix} m_1 m_2 \end{pmatrix} \begin{pmatrix} n_1 n_2 \end{pmatrix} \begin{pmatrix} m_1 m_2 \end{pmatrix} \begin{pmatrix} n_1 n_2 \end{pmatrix} \begin{pmatrix} m_1 m_2 \end{pmatrix} \]
Diagonalizing this Hamiltonian yields the one-particle energies
\[ q \sim \sgn \left( \frac{1}{m} + \frac{1}{m} \right) + \frac{1}{m} \] (A3)
From these energies the free energy is calculated, and the minimization with respect to m and m_{st} leads to the following Hartree-Fock self-consistency equations
\[ m_{st} = \frac{U}{2} X \sum_{mn} \left( \begin{array}{c} \pm \frac{1}{m_{st}} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
\[ m = \frac{U}{2} X \sum_{mn} \left( \begin{array}{c} \pm \frac{1}{m} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
As in Sec., these Hartree-Fock equations are solved numerically.

**APPENDIX B: HARTREE-FOCK THEORY FOR m_{st} ≠ m**

Similar to the derivation of the Hartree-Fock equations for m_{st} ≠ m (see Sec. IV A), we will now investigate the case with perpendicular orientation m_{st} ≠ m_{st}. The Ansatz for the one particle densities
\[ < n_{i2} > = \frac{1}{2} \left( n + m \right) \] (B1)
yields in addition to the Hartree term a Fock term in the coupling
\[ n_{i1} \frac{1}{m_{st}} \] (B2)
With this Ansatz one readily obtains the effective one-particle Hamiltonian
\[ H_{HF} = \sum_{i} \left( \frac{1}{2} \frac{U}{m} X \right) \sum_{n} \left( \begin{array}{c} n \mu \n \end{array} \right) \]
\[ \sum_{i} \frac{U}{2} m \rho_{i} \] (B3)
Diagonalizing this Hamiltonian yields the one-particle energies
\[ q \sim \sgn \left( \frac{1}{m_{st}} + \frac{1}{m_{st}} \right) \]

**APPENDIX C: SERIES EXPANSION OF THE HARTREE-FOCK FREE ENERGY**

The itinerant electron metamagnetic theory of Moriya and Uemura (21) can be derived from the Hartree-Fock approximation only if the free energy is analytic in m and m_{st}. Since in RPA the Hubbard interaction U contributes to the free energy analytically (see (15) and (16)) any non-analytic behavior must be due to the kinetic energy. Its expansion in the order parameter m_{st} at T = 0 is analyzed in this section.

To calculate the expansion in m_{st} a staggered magnetic field H_{st} is introduced:
\[ X \left( \begin{array}{c} \rho_{i} \end{array} \right) \]
\[ H_{st} = \frac{1}{2} \left( \begin{array}{c} \frac{1}{m_{st}} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
On a AB lattice the one-particle energies for this Hamiltonian show a gap at \[ \mu = 0 \] with square root singularities at its edge
\[ \sim \sgn \left( \frac{1}{m_{st}} + H_{st} \right) \]
We consider the half-filled band, where the staggered magnetization m_{st} is calculated from the one-particle energies. It shows the following asymptotic non-analytic behavior for H_{st} ≠ 0:
\[ m_{st} = \frac{1}{2} \left( \begin{array}{c} \frac{1}{m_{st}} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
\[ = 2N \sum_{i} \left( \begin{array}{c} \frac{1}{m_{st}} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
Similarly, the asymptotic behavior of the energy (C1) is obtained as
\[ E\left( m_{st} \right) = \sum_{i} \left( \begin{array}{c} \frac{1}{m_{st}} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
Subtracting the contribution due to H_{st}, E_{st} = H_{st}m_{st}, the asymptotic dependence of the kinetic energy on m_{st} reads
\[ E_{kin}\left( m_{st} \right) \sim \frac{1}{2} \left( \begin{array}{c} \frac{1}{m_{st}} \left( \begin{array}{c} n \mu \n \end{array} \right) \right) \]
This shows that E_{kin}(m_{st}) is non-analytic in m_{st}. Therefore the itinerant electron metamagnetic theory cannot be derived from the Hartree-Fock theory for the Hubbard model with an easy axis.

**APPENDIX D: THE METAMAGNETIC PHASE TRANSITION AT STRONG COUPLING**

In the limit of strong coupling and half-filling the O (U=U') perturbation theory yields the effective spin
Hamiltonian \( \mathcal{H} \). In the following, we study the metamagnetic phase transition, and especially the order of the transition, for this effective Hamiltonian. Restricting ourselves to solutions with \( m \) latitudes of ferrimagnetic (\( m \)) and antiferromagnetic (\( m_{\text{st}} \)) order, the ground state energy is a polynomial in \( m \) and \( m_{\text{st}} \) (\( t < 1 \)):

\[
E = \frac{1}{2U} m^2 + \frac{1}{2U} m_{\text{st}}^2 + \frac{1}{2U} \sum_{ij} 16B m^2 + 16B m_{\text{st}}^2 + A m^4 + 2A m^2 m_{\text{st}}^2 + 2A m^2 + A m_{\text{st}}^4 + 6A m_{\text{st}}^2 + 16C m^2 + 16C m_{\text{st}}^2 \quad \mathrm{H} + \text{const.} \quad (\text{D 1})
\]

One can see that the ferrimagnetic next nearest neighbor interaction \( C \) favors both saturated antiferromagnetism and saturated ferrimagnetism rather than ferrimagnetic phases. By contrast the plauette term \( A \) has contributions that support the formation of a ferrimagnetic state. To obtain the ground state the energy must be minimized with respect to \( m \) and \( m_{\text{st}} \) under the constraints \( m_{\text{st}}^2 = 1 \) \( m_{\text{st}} \) \( j \in j \). Differentiation of \( E \) with respect to \( m_{\text{st}} \) shows that, for \( x = m \), \( E \) has one maximum at \( m_{\text{st}} = 0 \) and two minima at

\[
m_{\text{st}} = \frac{8U^2 + A m^2}{3A + 8B + 8C} \quad (\text{D 2})
\]

For sufficiently strong coupling \( U \) (e.g., \( U > 2 \) in the case of the hypercubic lattice with \( A = 20, C = 2 \) and \( B = 4 \)) these minima are outside the constraint \( m_{\text{st}}^2 = 1 \). Therefore \( E \) becomes minimized at the border of the constraint, i.e., for \( m_{\text{st}} = 1 \). Replacing \( m_{\text{st}} \) by \( m \) in minimization with respect to \( m \) readily yields for the ground state

\[
m = \begin{cases} 0 & \text{for } H < A \frac{2}{3} + 2U^2 + 2B \frac{U^2}{A} \\ 1 & \text{for } H > A \frac{2}{3} + 2U^2 + 2B \frac{U^2}{A} \\ \frac{2}{3} + 2U^2 + 2B \frac{U^2}{A} & \text{else} \end{cases}
\]

\[
m_{\text{st}} = 1 \quad (\text{D 3})
\]

This ground state solution for the effective spin Hamiltonian shows a second order metamagnetic phase transition for \( A > 4C \). This is the case for the hypercubic lattice (\( A = 20; C = 2 \)) and for the Bethe lattice, where \( A = 0 \) but \( C = 1 \), i.e., the next nearest neighbor coupling is antiferromagnetic. In conclusion, the strong coupling theory shows a second order phase transition for all temperatures, even at \( T = 0 \).

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