Strongly Correlated Materials: Insights from Dynamical Mean-Field Theory

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Abstract: *Strongly correlated materials are a wide class of electronic materials that show unusual electronic and magnetic properties, such as metal-insulator transitions or half- metallicity.They are the systems in which the potential energy due to electron – electron interactions are comparable to or is larger than the kinetic energy. Dynamical mean field theory is a method to determine the electronic structure of strongly correlated materials. Materials with correlated electrons exhibit some of the most intriguing phenomena in condensed matter physics. A new theoretical framework is now allowing theorists to calculate the electronic structure of these materials, which can exist in a rich variety of phases. Strongly correlated materials are a wide class of electronic materials that show unusual electronic and magnetic properties, such as metal-insulator transitions or half-elasticity. DMFT is exact in the limit of infinite dimensions, the Lattice problem which is a Single site impurity problem in a self- consistent bath. Most importantly, the kinetic energy term and the interaction term are treated on an equal footing.*
Key words: *strongly correlated system, metal – insulator transition, electron density, Fermi liquid, phase diagram, photo emission, phonon spectra.*

I. Introduction:

The physical properties of various materials are explained by solid -state physics. Materials with open d and f electron shells, where electrons occupy narrow orbitals, have properties that are harder to ex-plain. In transition metals, such as vanadium, iron, and their oxides, for example, electrons experience strong Coulombic repulsion because of their spatial confinement in those orbitals. Such strongly interacting or "correlated" electrons cannot be described as embedded in a static mean field generated by the other electrons. The influence of an electron on the others is simply too pronounced for each to be treated independently. Silent characteristics:

(a) Partially filled d and f –shells that either from very narrow bands (d, 5f) or are almost fully localized (4f).
(b) Non- monotonic resistivities, rich variety of phase transitions, extreme sensitivity to magnetic fields, temperature, doping etc.

The effect of correlations on materials properties is often profound. The interplay of the d and f electrons' internal degrees of freedom—spin, charge, and orbital moment—can exhibit a whole zoo of exotic ordering phenomena at low temperatures. That interplay makes strongly correlated electron systems extremely sensitive to small changes in external parameters, such as temperature, pressure, or doping.

In materials called heavy fermion systems, mobile electrons at low temperature behave as if their masses were a thousand times the mass of a free electron in a simple metal. Some strongly correlated materials display a very large thermoelectric response; others, a great sensitivity to changes in an applied magnetic field—an effect dubbed colossal magnetoresistance. Such properties make the prospects for developing applications from correlated-electron materials exciting.

The theoretical challenge:

To understand materials made up of weakly correlated electrons—silicon or aluminum, for example—band theory, which imagines electrons behaving like extended plane waves, is a good starting point. This theory helps to capture the delocalized nature of electrons in metals. Another theory called the Fermi liquid theory describes the transport of conduction electrons in momentum space and provides a simple but rigorous conceptual picture of the spectrum of excitations in a solid. It describes, excited states consist of independent quasiparticles that exist in a one-to-one correspondence to states in a reference system of non interacting Fermi particles plus some additional collective modes. To calculate the various microscopic properties of such solids, we have accurate quantitative techniques at our disposal. Density functional theory (DFT), for example, allows us to compute the total energy of some materials with remarkable accuracy, starting merely from the atomic positions and charges of the atoms. However, the independent-electron model and the DFT method are not

accurate enough when applied to strongly correlated materials.. Band theory incorrectly predicts them to be metallic when magnetic long-range order is absent .

The dual particle–wave character of the electron forces the adoption of components of the real-space and momentum space pictures.

Systems with strongly correlated electrons fall within that middle ground. Traditionally, such materials have been described using the model Hamiltonian approach. That is, the Hamiltonian is simplified to take into account only a few relevant degrees of freedom—typically, the valence electron orbitals near the Fermi level.

One of the simplest models of correlated electrons is the Hubbard Hamiltonian, defined in equation

$$H = \sum_{ij,\sigma} t_{ij} c_{i\sigma}^+ c_{J\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, .$$

This Hamiltonian describes electrons with spin directions $\sigma = \uparrow\ or\ \downarrow$ moving between localized states at lattice sites i and j. The electrons interact only when they meet on the same lattice site i. (The Pauli principle requires them to have opposite spin.) The kinetic energy and the interaction energy are characterized by the hopping term t_{ij} and the local Coulomb repulsion U, respectively. These two terms compete because the kinetic part favors the electrons' being as mobile as possible, while the interaction energy is minimal when electrons stay apart from each other—that is, localized on atomic different sites. The parameters that determine the properties described by the Hubbard model are the ratio of the Coulomb interaction U and the bandwidth W (W is determined by the hopping, t_{ij}), the temperature T, and the doping or number of electrons. The lattice structure and hopping terms influence the ability of the electrons to order magnetically, especially in the insulating state. If the magnetic ordering is impeded crucial for observing subtle transitions between nonmagnetic phases—the system is said to be "frustrated."

Dynamical mean-field theory (DMFT) : Dynamical mean field theory(DMFT)is a method to determine the electronic structure of strongly correlated materials. A wide variety of numerical techniques and analytical methods have been used to treat strongly correlated electron systems. A new limit has been introduced to the correlated electron problem, that of infinite lattice coordination: Each lattice site is imagined to have infinitely many neighbors, that approach retained the competition between kinetic energy and Coulomb interaction of electrons while simplifying the computation.

Dynamical mean- field theory of correlated – electron solids replaces the full lattice of atoms and electrons with a single impurity atom imagined to exist in a bath of electrons .The approximation captures the dynamics of electrons on a central atom as it fluctuates among different atomic configurations, In the simplest case of an s orbital occupying an atom, fluctuations could vary among $|0\rangle, |\uparrow\rangle, |\downarrow\rangle\ or\ |\uparrow\downarrow\rangle,$, which refer to an unoccupied state, a state with a single electron of spin-up, one with spin –down, and a doubly occupied state with opposite spins. In this illustration of one possible sequence involving two transitions, an atom in an empty state absorbs an electron from the surrounding reservoir in each transition. The hybridization V_v is the quantum mechanical amplitude that specifies how likely a state flips between two different configurations.

Consequently, a better understanding of various approximation schemes emerged along with an exact solution of some simpler models. A second advance came when the Hubbard model (a lattice model) is mapped onto a self-consistent quantum impurity model—a set of local quantum mechanical degrees of freedom that interacts with a bath or continuum of non interacting excitations.

The construction provides the basis of the dynamical mean-field theory of correlated electrons. Hamiltonians on the lattice using analytic and numerical techniques such as quantum Monte Carlo, previously developed to study impurity models. The DMFT solutions become exact as the number of neighbors increases.

A mean-field theory maps a many-body lattice problem to a single-site problem with effective parameters. In the fermionic case, the degrees of freedom at a single site are the quantum states of the atom inside a selected central unit cell of the crystal; the rest of the crystal is described as a reservoir of noninteracting electrons that can be emitted or absorbed in the atom. Figure 1 depicts that emission or absorption as mediated by a quantum mechanical amplitude V_v, and its mathematical description is as follows:

To treat strongly correlated electrons, one has to introduce a frequency resolution for the electron occupancy at a particular site. A Green function that specifies the probability amplitude required to create an electron with spin $\sigma(\uparrow\ or\ \downarrow)$ at a site I at time τ' and destroy it at the same site at a later time τ' and destroy it at the same site at a later time τ will do the job;

$$G_{i\bar{j}}(\tau - \tau') \equiv -\langle c_{i\bar{j}}(\tau) c_{i\bar{j}}^+(\tau') \rangle. \tag{1}$$

The Green function contains information about the local one – electron photoemission spectrum.The dynamical mean- field theory (DMFT) can be used to investigate the full many – body problem of interacting quantum mechanical particles or effective treatments such as the Hubbard model

$$H = \sum_{ij,\sigma} t_{ij} c_{i\sigma}^{+} c_{J\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}, \tag{2}$$

Which is the simplest model of interacting electrons on a lattice. The equation contains a matrix element t_{ij} that describes hopping of electrons with spin σ between orbitals at sites I and j , and a local Coulomb interaction U between two electrons occupying the same site I ; $n_{ij\tau} = c_{ij\tau}^{+} c_{ij\tau}$ is the density of electrons at site I with spin σ.

The Anderson impurity model

$$H_{AIM} = H_{atom} + \sum_{\nu,\sigma} \varepsilon_{\nu}^{bath} n_{\nu\sigma}^{bath} + \sum_{\nu,\sigma} \left(V_{\nu} c_{0,\sigma}^{+} a_{\nu,\sigma}^{bath} + h.c. \right) \tag{3}$$

(in which h.c. is the Hermitian conjugate) serves as a reference system for the Hubbard model because it yields the exact local Green function in DMFT when the V_{ν} fulfills a self- consistency condition. It provides the mathematical description of the physics which states that there is one possible sequence involving two transitions, an atom in an empty state absorbs an electron from the surrounding reservoir in each transition. The hybridization V_{ν} is the quantum mechanical amplitude that specifies how a state flips between two different configurations.

Starting from the general Hamiltonian, the lattice site's atomic degree of freedom is separated, described by H_{atom}, from the remaning degrees of freedom, which are treated as a bath of electrons with energy levels ε_{ν}^{bath}, electrons may hop in and out of that site via the hybridization V_{ν} between the atomic $(C_{O,\sigma})$ and the bath electrons $(a_{\nu,\sigma}^{bath})$. The parameters ε_{ν}^{bath}, and V_{ν} appear in a simple combination in the hybridization function

$$\Delta(\omega) = \sum_{\nu} \frac{|V_{\nu}|^{2}}{\omega - \varepsilon_{\nu}^{bath}}, \tag{4}$$

which here plays the role of a mean field. Its frequency dependence makes it a dynamic mean field. Because the bath describes the same electrons as those on the local site, Δ(ω) has to be determined from the self-consistency condition

$$G[\Delta(\omega)] = \sum_{k} \left\{ \omega - \sum [\Delta(\omega)] - t_{k} \right\}^{-1}, \tag{5}$$

where the self-energy term $\sum |\Delta(\omega)| \equiv \Delta(\omega) - 1/G|\Delta(\omega)| + \omega$ takes on the meaning of a frequency-dependent potential, and t_{k} is the Fourier transform of the hopping matrix elements ti j of the solid.

By analogy with density functional theory, an exact functional of both the charge density and the local Green function of the correlated orbital is introduced as

$$\Gamma[\rho(r), G] = T[\rho(r), G] + \int V_{ext}(r)\rho(r)d^{3}r + \frac{1}{2} \int \frac{\rho(r)\rho(r')}{|r - r'|} d^{3}r d^{3}r' + E_{XC}[\rho(r), G]. \tag{6}$$

The functional has a similar decomposition as in DFT. However, the kinetic energy is no longer that of a free electron system because T [ρ, G] is the kinetic energy of a system with given density ρ(r) and local Green function G. DMFT provides an explicit approximation for E_{XC} [ρ (r),G].

The local description of a correlated solid in terms of an atom embedded in a medium of non interacting electrons corresponds to the celebrated Anderson impurity model, but now with an additional self-consistency condition. The hybridization function plays the role of a mean field and describes the ability of electrons to hop in and out of a given atomic site. When the hybridization is very small, the electron is almost entirely localized at a lattice site and moves only virtually, at short durations compatible with the Heisenberg uncertainty principle. On the other hand, when it is large, the electron can move throughout the crystal.

We thus obtain a simple local picture for the competition between itinerant and localized tendencies underlying the rich phenomena that correlated materials exhibit. The mapping of the lattice model onto an impurity model—the basis of the dynamical mean-field theory—simplifies the spatial dependence of the correlations among electrons and yet accounts fully for their dynamics—that is, the local quantum fluctuations missed in static mean-field treatments like the Hartree–Fock approximation.

Besides its conceptual value of providing a quantum analog of the classical mean field, the mapping of a lattice model onto the Anderson impurity model has had great practical impact. Applications of DMFT have led to a lot of progress in solving many of the problems inherent to strongly correlated electron systems, such as the Mott metal–insulator transition, doping of the Mott insulator, phase separation, and the competition of spin,

charge, and orbital order.

The electrons in the material are divided into two sets: weakly correlated electrons, well described by a local-density approximation (LDA) that models the kinetic energy of electron hopping, and strongly correlated (or more localized) electrons—the titanium d orbitals—well described using DMFT. The one-body part of the Hamiltonian is derived from the Kohn–Sham Hamiltonian. The on-site Coulomb interaction U is then added onto the heavy d and f orbitals to obtain a model Hamiltonian. DMFT (or more precisely, "LDA + DMFT") is then used to solve that Hamiltonian.

Just as the Kohn–Sham equations serve as a reference system from which one can compute the exact density of a solid, the Anderson impurity model serves as a reference system from which one can extract the density of states of the strongly correlated electrons. The parallel between DFT and DMFT is best seen in the functional approach to DMFT, outlined from equations(1) to(6). It constructs the free energy of the system as a function of the total density and the local Green function. Finding the extreme of the functional leads to a self-consistent determination of the total energy and the spectra.

II.   Conclusion:

Mean-field methods clearly represent a new advances_ the strongly correlated electron regime of transition-metal oxides, for instance—in which electrons are neither fully itinerant nor localized—has simply not been accessible to other techniques. By extending DMFT from single sites to clusters, for example, capturing the effects of short-range correlations and combining DMFT with advanced electronic- structure methods should make it possible to evaluate the Hamiltonian and the frequency-dependent screened Coulomb interaction from first principles without having to first construct a local density functional.   As the number of DMFT implementations of new materials increases, more detailed comparisons with experiments are becoming possible. Such comparisons should help separate the effects that can be understood simply from the effect captured by DMFT and from the effects that require long-wavelength modes not captured by the DMFT approach.

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