Impurity scattering in strongly correlated metals close to the Mott transition

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This work explores a simple approximation to describe isolated impurity scattering in a strongly correlated metal. The approximation combines conventional one electron scattering theory and the Dynamic Mean Field Theory to describe strong correlations in the host. It becomes exact in several limits, including those of very weak and very strong impurity potentials. Original electronic structure appears at the impurity site when the impurity potential strength is moderate and the host is close to the Mott transition. Our results may provide useful guidance for interpretation of scanning tunneling microscopy experiments in strongly correlated systems.

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\textit{Introduction.} The physics of dilute impurities in broad band metals such as Cu or Al is one of the success stories of quantum mechanics in the fifties and sixties \cite{1}. By dilute we mean that physical effect due to interactions between impurities can be neglected, and that the properties described vary linearly with the concentration of impurities. For broad band metals, electron-electron interactions may be treated as perturbations, because the ratio $E_{\text{el-el}}/E_K$ is small compared to unity. $E_{\text{el-el}} \approx 1-2$ eV is the screened Coulomb interaction between electrons, and $E_K \approx 10$ eV is the electronic kinetic energy, of the order of $\epsilon_F$, the Fermi energy measured from the bottom of the conduction band. Historically, the treatment of electron-electron interactions in impure metals has concentrated for a long time on the Kondo problem, for which electron-electron interactions in the host where irrelevant \cite{2}. More recently, renormalizations of the Kondo screening due to correlation effects in the host where also considered \cite{3,4}. On the other hand, impurity scattering with electron-electron interactions on the impurity site and in the host, have been treated within weak coupling methods in a few simple cases, such as the PdNi system \cite{5}. In recent years, improvements in local probe techniques such as nuclear magnetic resonance and scanning tunneling microscopy shed new light on the problem of impurity effects in strongly correlated metallic materials, in particular with connection to High Tc superconductivity. For instance, in underdoped superconducting cuprates, non magnetic impurities such as Zn or Li trigger the appearance of magnetic moments on neighbouring sites \cite{6,7}. This is a natural phenomenon if the two dimensional CuO plane at metallic doping level is reasonably well described by some variant of the RVB theory \cite{8}.

The present paper is dedicated to introduce and discuss the results of a simple approach to the problem of isolated impurity scattering in a strongly correlated metallic system close to the Mott transition.

The understanding of the Mott metal-insulator transition in a half filled band has evolved, since the early papers by Hubbard \cite{9}, with Gutzwiller’s treatment of strong correlations in narrow band metal \cite{10}, and the application of that theory to the half filled band by Brinkman and Rice \cite{11}. The latter paper showed that close to the critical parameter $U_c$ governing the metal-insulator transition, both the Pauli susceptibility $\chi$ and the effective electronic mass $m^*$ diverge with a constant ratio. This behaviour became understandable thanks to the Dynamic Mean Field Theory (DMFT) \cite{12}. In the simple Hubbard picture of the metal-insulator transition, the upper band, with doubly occupied sites, separates at the transition from the lower singly-occupied band. Following the DMFT picture, a central peak appears between two well separated “Hubbard” bands in the metallic phase. The integrated density $Z$ of the central peak goes to zero with the peak width at the transition. The peak is analogous to the Kondo resonance for the single magnetic impurity problem, except that it is a homogeneous property of the system. In addition, this narrow feature at the Fermi energy sets a scale for a characteristic temperature $T^*$, which goes to zero at the metal-insulator transition. Approaching the transition, the electronic specific heat coefficient and the Pauli susceptibility diverge at zero $T$ as $1/T^*$ \cite{12}.

A number of important properties of the impurity in the broad band case are given by the impurity site Green’s function $G_{\omega_0}(\omega) = \sum_{k,k'} G(k,k',\omega)$. All electronic properties of the dilute metal are contained in $G(k,k',\omega)$, which is known exactly for the case when electron-electron interactions may be neglected altogether. $G_{\omega_0}(\omega)$ is entirely computed in terms of the scattering potential $V$, which we will assume for simplicity to be a point scattering one, and the site diagonal host Green’s function $G_{\omega_0}^{\text{dd}}(\omega)$. On the other hand, if one considers the case of a narrow band system, the solution of the DMFT equations provides the site diagonal Green’s
function $G_{oo}^{0,U}$ of a strongly interacting host, where $U$ denotes the electron-electron intra atomic “Hubbard” interaction potential. This Green’s function depends solely on the momentum independent self energy $\Sigma(U, \omega)$ and on the non-interacting band structure [12]. While the DMFT is exact in the limit of large spatial dimensions (or lattice connectivity), it can be considered as an approximation for a finite dimensional lattice, which is the view that we adopt here to describe the host.

This paper explores the following simple idea: in a certain limit, the impurity properties when the host system is close to the metal-insulator transition may be approximated by the substitution of the DMFT host site diagonal Green’s function in the expression for the impurity site Green’s function. This may be considered as a (dynamical) mean field description of the impurity electronic properties. This simple approach has several correct limits, including the case of $V \to 0$ (and any $U$) and the case $U \to 0$ (and any $V$), so one may expect that it may be reasonably accurate for the general case.

**The model.** We assume that the host is close to the Mott transition and has a number of equivalent degenerate bands, such as can be found in a cubic crystal. We neglect interband electron-electron interactions.

The Hamiltonian for each band is then:

$$H = H_0 + H_{imp}$$

$$H_0 = \sum_{i,j} t_{ij} c_{i,\sigma}^\dagger c_{j,\sigma} + h.c. + U \sum_i n_{i,\uparrow} n_{i,\downarrow}$$

$$H_{imp} = V(n_{o,\uparrow} + n_{o,\downarrow})$$

The site diagonal pure host Green’s function is given in the DMFT approximation as [12]:

$$G_{oo}^{0,U}(\omega) = \sum_k G_{oo}^{0,U}(k, \omega) = \sum_k \frac{1}{\epsilon_k - \omega - \Sigma(U, \omega)}$$

where $\epsilon_k$ is the host electronic dispersion relation in the limit $U = 0$.

From standard isolated impurity scattering theory [13], the impurity Green’s function for $U = 0$ is:

$$G_{oo}^{V,0}(k, k') = G_{oo}^{0}(k) \delta_{k,k'} + V \sum_k G_{oo}^{0}(k) \frac{G_{oo}^{0}(k')}{1 - V \sum_k G_{oo}^{0}(k)}$$

The site diagonal impurity Green’s function for $U = 0$ is given by $G_{oo}^{V,0} = \sum_{k,k'} G(k, k')$:

$$G_{oo}^{V,0}(\omega) = \frac{G_{oo}^{0}(\omega)}{1 - VG_{oo}^{0}(\omega)}$$

*Scattering potential model.* In the scheme that we propose here, host correlations are taken into account by simply replacing the uncorrelated Green’s function in Eq (6) by the DMFT expression of the correlated one. The local Green’s function at an impurity site is thus described by

$$G_{oo}^{V,U}(\omega) = \frac{G_{oo}^{0,U}(\omega)}{1 - VG_{oo}^{0,U}(\omega)}.$$  \hspace{1cm} (7)

It is important to realize that this expression has the correct limiting behaviours that we mentioned before. In fact, one easily checks that in Eq (7) the Green’s function $G_{oo}^{V,0}$ becomes the homogeneous system $G_{oo}^{0}$ for $V = 0$, and the impurity Green’s function in a normal (ie, non-correlated) host $G_{oo}^{V,0}$ when $U = 0$.

It is also worth pointing out that the model [13] is not the exact site diagonal impurity Green’s function in DMFT scheme [1]. In fact, the derivation of the impurity Green’s function in a correlated host fully within DMFT (ie, in the limit of large dimensions), can be obtained in a straightforward manner using the “cavity construction” [12]. However, in that case one realizes that the cavity (ie, the Weiss field) of the impurity site exactly coincides with the cavity of the clean homogeneous system. Therefore, there is no renormalization of the impurity environment due to the presence of the scattering potential $V$, or, in other words, the Friedel oscillations on neighboring sites are suppressed in the limit of infinite dimensions. Thus, that approach is unlikely to provide a correct physical description of finite dimensional systems.

Within our scheme, the poles of $G_{oo}^{V,U}(\omega)$ contain all the information about the perturbed local density, the local change of the density of states (DOS), and the possible occurrence of bound states. In particular, a bound state at energy $\omega_b$ is determined by the simultaneous equations:

$$1 - V \text{Re}[G_{oo}^{0,U}(\omega_b)] = 0$$

$$\text{Im}[G_{oo}^{0,U}(\omega_b)] = 0$$

When the first equation is satisfied for $\omega_b$, and $\text{Im}[G_{oo}^{0,U}(\omega_b)]$ is small compared to its value in the bulk of the pure metal bands, a resonant state is formed, with

$$\text{width} \propto \frac{|\text{Im}[G_{oo}^{0,U}(\omega_b)]|}{\partial \text{Re}[G_{oo}^{0,U}(\omega)] / \partial \omega|_{\omega = \omega_b}}.$$  \hspace{1cm} (9)

In principle $V$ is a free parameter. However when describing actual impurities, the effective scattering potential results from the perfect screening constraint. In the language of scattering theory, this constraint results in the well known Friedel sum rule [1], which connects the spherical harmonics phase shifts of the scattered wave function to the charge which has to be screened locally in order to ensure electrostatic equilibrium of the host metal. In the present formulation, the effective scattering potential is such that the integrated displaced density below the Fermi level must counterbalance the difference between the impurity nuclear potential and the host one. If the series of 3d transition elements Ti, Cr, Mn, Fe, Co, Ni are dissolved as dilute impurities in, say, $V_2O_3$, each impurity will be described by a potential which attracts below or above the Fermi level the number of states required for electric neutrality of the alloy.
Within the present approach, the electronic structure around the impurity site in real space can be simply obtained by Fourier transformation of an expression similar to Eq[5] where the Fourier transform of $\tilde{G}^0(i-j)$, $G^0(k)$, is replaced by the interacting host Green’s function $G^0,U(k,\omega)$ (cf Eq[4]).

Results. It is now straightforward, using the known expression for the host correlated site diagonal Green’s function provided by the DMFT approach, to find the impurity electronic structure in the whole range of $V,U$ values. We use the Iterated Perturbation Theory to solve for the host Green’s function [12], adopting a semi-circular non-interacting DOS. We begin our discussion of our results with the systematic study of the local DOS $\rho(\omega)$ as a function of the scattering potential $V$ in two extreme cases. In the upper panel of Fig. 1 we show $\rho(\omega)$ for the case of a metallic non-interacting host (standard Wolf model). In contrast, in the lower panel we show the results for the case of an insulating host which is well into the Mott state due to strong correlation effects. The local DOS of the Mott insulator corresponds to the $V=0$ case (continuous line), where symmetric lower and upper Hubbard bands split by a gap $\approx U - 2D$ can be observed. The former case describes the well known results for a non-interacting metallic host, where the effect of the potential is to initially deform the local DOS of the host, shifting spectral weight towards lower energies (since $V < 0$), until it becomes strong enough to create a bound state out of the conducting band. The condition for the formation of the bound state is $|V| > D$ for a reasonably regular DOS, where $D$ is the half-bandwidth of the metal host and is taken equal to one, to set the units of the problem. For a Mott insulating host, as $V$ increases, spectral weight is also shifted to lower energies in both, the upper and the lower Hubbard bands. In addition, there is also a transfer of spectral weight from the upper Hubbard band to the lower one. Interestingly, as the strength of the scattering potential continues to increase, an impurity resonant state forms simultaneously at the bottom of the upper and the lower Hubbard bands. The threshold value for the appearance of the bound states is not as clear cut as in the non-interacting metallic case, but remains of the same order of magnitude. It is also worth noting that the width of the bound state is very narrow for the upper Hubbard band, while substantially broader (and more intense) for the lower Hubbard band.

A correlated metal host is well described with DMFT, and the clean (ie $V=0$) case is shown in Fig. 2 (top left panel). The local DOS shows the characteristic “Kondo” peak at $\omega = 0$ that corresponds to a renormalized band of heavy mass quasiparticles. The effective bandwidth $ZD$ of this feature is proportional to its spectral intensity (ie the quasiparticle residue $Z$) and scales with $U_c - U$, where $U_c \approx 3D$ is the critical value of the interaction where a metal-insulator occurs. Spectral weight $1-Z$ is thus shifted from low energies to frequencies of order $U$, to form the broad lower and upper Hubbard bands. At the transition, the quasiparticle peak becomes infinitely narrow and then disappears, leaving behind just the higher energy Hubbard bands (cf Fig[1] lower panel). It is interesting to observe the systematic effect of the strength of the scattering potential on the local DOS for this strongly correlated metallic host. As $V$ is stepped up in absolute value, the DOS is shifted to lower energies in both Hubbard bands, as well as in the central peak. At the same time a finite amount of spectral weight is transferred from the upper Hubbard band to the lower one, below the Fermi level, allowing a local screening at the impurity site. For $|V|$ larger than about $1.5D$, a very sharp resonance appears at the impurity site. This sharp resonance carries a spectral intensity $\sim Z$, ie the value of the quasiparticle residue, and thus can be interpreted as a bound state pulled out of the narrow renormalized

![FIG. 1: Top panel: Evolution of the impurity density of states $\rho(\omega)$ for increasing $V$ in a non-correlated host (ie, $U = 0$). Bottom panel: Same quantity when the host is in the Mott insulator state with $U = 4$.](image1)

![FIG. 2: The evolution of the impurity site $\rho(\omega)$ when the host is close to the Mott transition with $U = 2.9$. The panel series show the effect of increasing the strength of the scattering potential $V$. The bottom panel inset shows the detail of the evolution of the mid gap resonance for $V = 0, 0.5$ and 2.](image2)
band. This interpretation is further substantiated by the results shown in the inset of the lower right panel, that shows a detail of the evolution of the DOS near the Fermi energy. We observe that the evolution of $\rho(\omega)$ is qualitatively similar as that of the non-interacting metal (Fig.1 top panel), but with the frequency scale renormalized down by a factor of $Z$. Eventually, when the strength of $V$ is large enough, the quasiparticle band picture near the Fermi energy breaks down and a sole resonant state at a frequency $\omega \approx V + U/2$ is left.

In Fig.3 we now consider the case of a strongly attractive impurity potential $|V| \geq D$, and study the systematic evolution of the local DOS as a function of the electron-electron interaction parameter $U$, as the host evolves through a metal-insulator transition. For weak $U$, the main effect of el-el interactions is to transform the impurity bound state to a resonant state, due to the tails of the single correlated band. As $U$ is stepped up, the two host Hubbard bands form, and the impurity site develops a multiple-peak structure. There are two resonances associated with the upper and lower Hubbard bands (with the latter being more prominent as $V$ is assumed attractive), and a central one that is associated to the central quasiparticle narrow band. This central feature occurs close to the Fermi energy and becomes a very sharp resonance at some particular value of $U$, and then broadens again before disapearing as the host becomes a Mott insulator. We find that the spectral strength of the mid gap resonance at the impurity site scales roughly like the host mid gap quasiparticle peak, and both mid gap features vanish together at the Mott transition.

Finally, Fig.4 exhibits the increase of electronic density at the impurity site, as a function of $|V|$ and $U$. As $U$ increases, it requires a stronger attractive potential $V$ to screen the same amount of impurity nuclear charge. This simply reflects the fact that a large amount of states in Hilbert space are pushed up in energy away from the Fermi level as $U$ increases, so the amount of states available to screen he impurity nuclear potential decreases.

**Conclusion.** We believe that the simple approximation described in this work should be confronted with some experimental testing, so as to check its relevance to actual strongly correlated impure metals, close to the Mott transition. A candidate system to look at would be the V$_2$O$_3$ system, with a small concentration $c (\approx 10^{-2})$ of Sc, Ti, Cr, Mn, Fe, Co, Ni, or Cu. The measurements of interest are the transport ones, such as residual resistivity, temperature dependence of the resistivity, thermoelectric power, etc. and the NMR properties, such as Knight shift, $T_1$. However, perhaps the most clear cut validation may be obtained through the direct observation of the local electronic density by Scanning Tunneling Spectroscopy or precision photoemission measurements.

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![FIG. 3: Evolution of the impurity density of states $\rho(\omega)$, for a fixed strong attractive impurity potential $V = 1.5$. The panel series show the effect of increasing the strength of correlations $U$, as the host is driven across the Mott transition.](image1)

![FIG. 4: The variation of the electronic density at the impurity site as a function of $|V|$ for various $U$ values (left), and as a function of $U$ for various values of $V$ (right).](image2)

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