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Impurity scattering in a strongly correlated host

P. Lederer[1,2] and M. J. Rozenberg[1,3]

[1] *Laboratoire de Physique des Solides, CNRS et Université Paris-Sud - Bâtiment 510, F-91405 Orsay, France*
[2] *Departamento de Fisica, UFPE - 50670-901 Recife, Brasil*
[3] *Departamento de Física, FCEN, Universidad de Buenos Aires - Ciudad Universitaria Pab.I, (1428) Buenos Aires, Argentina*

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Abstract – This work explores a simple approximation to describe isolated impurity scattering in a strongly correlated host. 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 strong interactions. We study the problem for a large range of parameter strengths and focus on the case of a strongly correlated metal host near the Mott metal-insulator transition. We find interesting effects on the electronic structure at the impurity site with the appearence of bound states at frequencies that are strongly renormalized from the bare impurity potential value. However, the strength of the threshold potential for the onset of the bound states remains of the order of the bare host bandwidth, *i.e.* essentially unrenormalized with respect to the non-interacting case. Our results may provide useful guidance for interpretation of scanning tunneling microscopy experiments in strongly correlated systems.

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 [1]. By dilute we mean that the 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 its ratio to the kinetic energy is small compared to unity. $U \approx 1$–$2\,\mathrm{eV}$ is the screened Coulomb interaction between electrons, and the electronic kinetic energy $\approx 10\,\mathrm{eV}$, is of the order of ϵ_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 are usually irrelevant [2]. Nevertheless, the renormalizations of the Kondo screening due to correlation effects in the host have also been investigated [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 [5]. In addition, motivated by experiments in impure cuprate superconductors, Ziegler *et al.* introduced a T-matrix formulation for the strong-coupling case, and focus their study on the problem of impurity defects (vacancies) in a correlated 2D antiferromagnetic Mott insulator host [6]. 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, underlying the need for improved theoretical methods [7]. This work is a contribution towards that goal. We shall introduce and discuss a simple approach to the problem of isolated impurity scattering in a strongly correlated metallic host near the Mott metal-insulator transition. Our method combines the familiar expressions for potential impurity scattering [8] with the propagators of a strongly correlated electron host system that is driven across a paramagnetic Mott transition, obtained within the Dynamical Mean-Field Theory (DMFT) [9].

Though we address a different physical question, our approach can be cast within the general T-matrix formulation of Ziegler *et al.* [6]. In that approach the correlation effects of the impurity and the host where encoded in two self-energies: one containing many-body effects only, and another with correlation diagrams with at least one scattering interaction. The approach that we shall

describe here can be viewed as an approximation where a very good estimate for the first self-energy is adopted, while the second self-energy is neglected. We shall show that in several limits the approximation becomes asymptotically exact, however, the accuracy at intermediate couplings is harder to assess. Thus, our approach provides an interpolation between various limiting cases, with physical sensible results in all parameter regimes.

A key ingredient of our approach is the use of propagators that can properly describe the strong correlations in the bulk, as it is driven across a Mott-Hubbard metal-insulator transition. The pioneer works on the Mott transition by Hubbard [10] and Brinkman and Rice [11], predicted the existence of a correlation driven metal-insulator transition at a critical value of the ratio of the Coulomb repulsion to the conduction electron bandwidth U_c/W. These early approaches became embodied within the DMFT for strongly correlated systems. DMFT predicted a scenario where a heavy electron metal with a narrow quasiparticle band of spectral intensity Z is formed as the interaction U is increased. In addition, at higher energies ($\sim U$) two Hubbard bands are formed describing the localization effects. At a critical value U_c, the effective mass of the carriers diverge and their spectral intensity goes to zero $Z \sim (U_c - U)$. Beyond U_c the electrons become localized, and one obtains a Mott insulator state with a large charge gap ($\sim U$). Recently, predictions of DMFT for the detailed nature of the metal-insulator transition were verified in experiments on V_2O_3 [12].

A number of important properties of the impurity in a broad-band metal host are given by the impurity site Green's function. If electron-electron interactions may be neglected altogether, then the propagator of the system can be exactly computed. This is done by using standard potential scattering theory [8], where the isolated impurity Green's function is computed in terms of the potential V, which we will assume for simplicity to be a point scattering one, and of the pure host (non-interacting) Green's function $G^0(k,\omega) = 1/(\omega - \epsilon_k)$, where ϵ_k is the lattice dispersion.

On the other hand, if one considers the case of an impurity in a narrow-band metal, the correlations effects in the host cannot be neglected and scattering theory in terms of $G^0(k,\omega)$ is no longer applicable. Nevertheless, DMFT has been shown to account for the main correlation effects in pure correlated metals through the corrections to $G^0(k,\omega)$ introduced by local self-energy $\Sigma_{\rm DMFT}(\omega)$. Therefore, one may expect the DMFT pure host Green's function to provide a good starting point for the study of the potential scattering effects.

In fact, while the DMFT is formally exact in the limit of large lattice connectivity (or large spatial dimensions), it may nevertheless provide a very good approximation for finite-dimensional lattices. This is at the core of the recent efforts to develop *ab initio* realistic band-structure methods for strongly correlated systems known as LDA+DMFT [13]. Thus, here we shall assume that the DMFT can provide a good description of the narrow-band host of the impurity.

We consider a model for an impurity embedded in a correlated system which exhibits a Mott metal-insulator transition. It reads,

$$H = H_H + H_{imp}, \tag{1}$$

$$H_H = \sum_{i,j} t_{ij} c^\dagger_{i,\sigma} c_{j,\sigma} + \text{h.c.} + U \sum_i n_{i,\uparrow} n_{i,\downarrow}, \tag{2}$$

$$H_{imp} = -V(n_{o,\uparrow} + n_{o,\downarrow}), \tag{3}$$

where H_H denotes the host's Hubbard Hamiltonian with hopping t. The minus sign in front of the impurity potential is to consider an attractive potential with $V > 0$. To capture a Mott-Hubbard transition in the pure host, we consider it to be half-filled and in the paramagnetic phase.

The site diagonal pure host Green's function is computed within the DMFT as [9]:

$$G^{0,U}_{oo}(\omega) = \sum_k G^{0,U}(k,\omega) = \sum_k \frac{1}{\omega - \epsilon_k - \Sigma_{\rm DMFT}(\omega)}, \tag{4}$$

where ϵ_k is the host lattice electronic dispersion relation and the local (*i.e.* k-independent) self-energy is obtained from the solution of the DMFT self-consistent equations [9]. For simplicity we consider the well studied case of a Bethe lattice that has a semicircular non-interacting density of states (DOS) of bandwidth $W{=}4t$. In this case, the DMFT equations can be efficiently solved at $T = 0$ using the Iterative Perturbation Theory (IPT) method which produce very good approximate solutions in the whole range of U/W. In particular, close to U_c, it also captures all the features of the Mott-Hubbard transition scenario that we described in the introduction [9]. We set the half-bandwidth $W/2$ as the unit of energy, thus $U_c \approx 3.3$ within IPT.

From standard isolated impurity scattering theory [8], the electron Green's function for the case $U{=}0$ is exactly computed as

$$G^{V,0}(k,k',\omega) = G^0(k,\omega)\delta_{k,k'} + V \frac{G^0(k,\omega) G^0(k',\omega)}{1 - V \sum_k G^0(k,\omega)}. \tag{5}$$

Then, the local impurity-site diagonal Green's function $G^{V,0}_{oo}(\omega) = \sum_{k,k'} G(k,k',\omega)$ is given by

$$G^{V,0}_{oo}(\omega) = \frac{G^0_{oo}(\omega)}{1 - V G^0_{oo}(\omega)}, \tag{6}$$

where $G^0_{oo}(\omega) = \sum_k G^0(k,\omega)$.

The scheme that we propose amounts to replacing the uncorrelated Green's function of eqs. (5) and (6) by the DMFT expression of the correlated one. Explicitly, the lattice Green's function becomes

$$G^{V,U}(k,k',\omega) = G^{0,U}(k,\omega)\delta_{k,k'} + V \frac{G^{0,U}(k,\omega) G^{0,U}(k',\omega)}{1 - V \sum_k G^{0,U}(k,\omega)} \tag{7}$$

and the impurity site Green's function is given by

$$G_{oo}^{V,U}(\omega) = \frac{G_{oo}^{0,U}(\omega)}{1 - V G_{oo}^{0,U}(\omega)}. \quad (8)$$

This equation is similar to the final T-matrix expression of ref. [6], but already contains the DMFT self-energy $\Sigma_{\rm DMFT}(\omega)$ to describe the pure many-body correlation effects. Due to the locality of this self-energy, our approach only describes scattering in the s-symmery channel. Near the paramagnetic Mott metal-insualtor transition the physics becomes dominated by local effects, therefore one may expect this approximation to be appropriate for the correlated metal near the transition and also on the paramagnetic insulator side.

Another physical motivation for our approach follows from the observation that within DMFT, the main effect of correlations is to merely "shrink" the non-interacting electron dispersion by a renormalization factor Z (the quasiparticle residue). In fact, near the Fermi energy, the electronic states form a quasiparticle band approximately given by $Z\epsilon_k$ (*i.e.* the effective mass is increased in $1/Z$). Therefore, besides this renormalization effect, the electronic structure of the metallic states of the host remains qualitatively similar to the non-interacting one. Of course, in addition to the quasiparticle band, the DMFT solution also produces the incoherent Hubbard bands; but those are higher-energy features that do not affect the electronic structure near the Fermi energy.

Equation (8) is obviously exact for both, the $U = 0$ and $V = 0$ cases. Less evident is that it also asymptotically captures the strong-coupling limits of large U/W or large V/W. This is checked by replacing into eqs. (4) and (8) the large-U expression of $\Sigma_{\rm DMFT}(\omega) \approx (U/2)^2/\omega$ [9], and verifying that the correct limits are obtained (essentially given by atomic-like expressions).

Therefore, as several weak- and strong-coupling limits are correctly captured, one may expect that eq. (8) may provide a good approximation in the challenging intermediate case. Our approach does not include the vertex corrections; though it is not an easy task to evaluate their contribution near the metal-insulator transition, the sensible behavior of the solutions in the whole parameter regime implies that our method, at least, provides a successful interpolative scheme.

We should also mention that the model (1-3) can be treated fully within DMFT (*i.e.* following [4] and setting the impurity site $U = 0$). However, within DMFT, spatial fluctuations (*i.e.* Friedel oscillations) are suppressed as $1/d$. In contrast, in the present approach the electronic structure around the impurity site in real space can be simply obtained by Fourier transformation of $G^{V,U}(k, k'\omega)$.

In our study we treat V as a free parameter. However, when describing actual impurities, the strength of the effective scattering potential is set from the perfect screening constraint. In the language of scattering theory, this constraint results in the well-known Friedel sum rule [1],

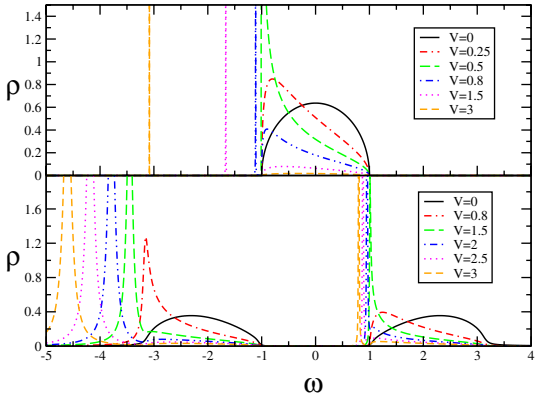

Fig. 1: Top panel: evolution of the impurity density of states $\rho(\omega)$ for increasing V in a non-correlated host (*i.e.* $U = 0$). Bottom panel: same quantity when the host is in the Mott insulator state with $U = 4$.

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.

We now turn to the discussion of the results of our model equations for the impurity site density of states. We begin by considering the systematic evolution of the DOS as a function of the scattering potential, when the host is in two extreme cases, either a non-correlated metal or a Mott insulator. The first case corresponds to standard impurity scattering theory (Wolf model). As is well established, the effect of the potential is to initially deform the local DOS of the host by shifting spectral weight towards lower energies (since we assume an attractive potential). This continues until V becomes strong enough ($V \lesssim W/2$) to create a bound state out of the conduction band [8]. The other extreme case, that of a Mott insulating host is shown in the bottom panel of fig. 1. The local DOS of the Mott insulator corresponds to the $V = 0$ case (continuous line), where symmetric lower and upper Hubbard bands are split by a gap $\approx U - W$. In this case, as V increases, both Hubbard bands get their spectral weight shifted to lower energies. In addition, there is an expected interband transfer of spectral weight, from the upper Hubbard band to the lower one. Interestingly, as the strength of the scattering potential continues to increase, impurity resonant states form simultaneously at the bottom of both Hubbard bands. The threshold value for the appearance of the bound states remains of the order of $W/2$. It is also worth noting that, unlike the $U = 0$ case, the states

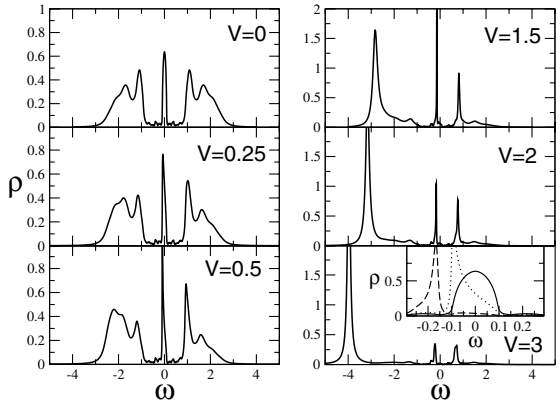

Fig. 2: The evolution of the impurity site $\rho(\omega)$ when the host is close to the Mott transition with $U = 2.9$. The panel series show the effect of increasing the strength of the scattering potential V. The bottom panel inset shows the detail of the evolution of the mid gap resonance for $V = 0$, 0.5 and 2 (solid, dotted, dashed line, respectively).

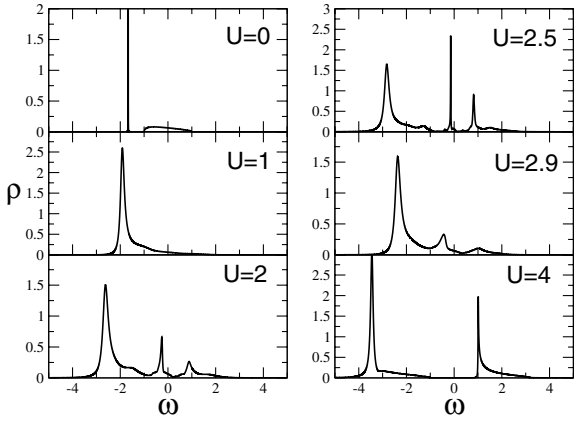

Fig. 3: Evolution of the impurity density of states $\rho(\omega)$, for a fixed strong attractive impurity potential $V = 1.5$. The panel series show the effect of increasing the strength of correlations U, as the host is driven across the Mott transition ($U_c \approx 3.3$).

that are pulled down from the Hubbard bands produce a resonance of a finite width. This is understood from the fact that they are not band-like quasiparticle states, but belonged to incoherent Hubbard bands, which already have rather short lifetimes [9].

We now turn to the interesting case of a strongly correlated metallic host. The results are shown in fig. 2. We set the interaction $U = 2.9 < U_c$ which places the host near the correlation driven Mott transition [9]. The clean host DOS corresponds to the case $V = 0$ (top panel). As discussed before, the DOS shows a narrow quasiparticle band at low frequencies with an effective bandwidth ZW, where $Z \ll 1$ is the quasiparticle residue. The semi-elliptical shape reflects that its band-structure simply gets "shrunk" with respect to the non-interacting one. The remaining spectral weight $1 - Z$ goes to frequencies of order $\pm U/2$, forming broad lower and upper Hubbard bands.

As V is stepped up, all three features, the two Hubbard bands and the central quasiparticle peak in the DOS at the impurity site are affected. On the one hand, the evolution of the Hubbard bands towards resonances with finite width is similar as in the Mott insulator host case discussed before (fig. 1, bottom). On the other, in contrast, the changes induced by V on the DOS of the central quasiparticle peak are more subtle. As can be seen in the inset to the bottom panel of fig. 2, the narrow semi-elliptical resonance gets deformed towards lower energies as the attractive potential is increased. When V is sufficiently strong, a resonance is fully pulled down from the narrow quasiparticle band (dashed line in the inset). This behavior is indeed qualitatively similar to the one that we discussed before for the U=0 case (fig. 1, top), but with the exception that the changes occur within the renormalized small energy scale ZW. We note, however, that the threshold value of V to pull down the resonance does not get renormalized by Z and remains $\sim W/2$.

Eventually, when the scattering strength V is large enough, the whole band-structure of the host at the impurity site breaks down, and a sole resonant state at a frequency $\omega \approx -V$ is left (which is the correct asymptotic limit for large V).

To complete our study, in fig. 3 we consider the case of a strongly attractive impurity potential $V > W/2$, and obtain the systematic evolution of the local DOS at the impurity site, as the electron-electron interactions in the host are increased. As we discussed already, the clean host undergoes a Mott metal-insulator transition at $U_c = 3W/2$ [9]. For weak U, the main effect of el-el interactions is to transform the impurity bound state to a resonant state. This is because the interactions provide a self-energy to the electronic states of the metallic host, and their finite life-times are then reflected as a width for the resonant states pulled down by V. As U is stepped up, the Hubbard bands form in the host, and they are reflected as a multi-resonant peak structure in the impurity site DOS. These peaks occur at energies $\sim \pm U - W/2$ that correspond to the lower energy edge of the Hubbard bands. The central feature near the Fermi energy emerges only when the host is in a strongly correlated metallic state, and disappears immediately after U becomes greater than U_c, *i.e.* when the host undergoes the Mott transition and its DOS acquires a gap. Thus, our results predict the presence of spectral weight at low frequencies only in a rather narrow range of interaction U, just below the critical U_c. This prediction may provide useful guidance for the interpretation of STM experiments of impurities in correlated metals.

To conclude we mention some experimental studies that may test the predictions that may be computed within the present approach and which will be worked out in detail elsewhere. A strongly correlated candidate system which should exhibit the effects described here would be V_2O_3, with a small concentration ($\sim 10^{-2}$) of

Sc, Ti, Cr, Mn, Fe, Co, Ni, or Cu. Measurements of interest are, for instance, residual resistivity, temperature dependence of the resistivity, and thermoelectric power, for the transport properties, and also the Knight shift and T_1, for the NMR. However, the most clear cut predictions of the present approach may be more directly tested by comparison to the local electronic density measured by scanning tunneling spectroscopy.

One final remark is that our approach is quite general. In fact, one may easily extend eqs. (7) and (8) for the case of a host with a symmetry-broken phase, such as in a Néel state. These extensions open new exciting perspectives for the study of impurities within strongly correlated hosts.

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