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Condensed matter physics in the 21st century: The legacy of Jacques Friedel

The beauty of impurities: Two revivals of Friedel's virtual bound-state concept

La beauté des impuretés : nouveaux contextes pour le concept d'état lié virtuel

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A B S T R A C T

Jacques Friedel pioneered the theoretical study of impurities and magnetic impurities in metals. He discovered Friedel oscillations, introduced the concept of virtual bound-state, and demonstrated that the charge on the impurity is related to the scattering phase-shift at the Fermi level (Friedel sum-rule). After a brief review of some of these concepts, I describe how they proved useful in two new contexts. The first one concerns the Coulomb blockade in quantum dots, and its suppression by the Kondo effect. The second one is the dynamical mean-field theory of strong electronic correlations.

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R É S U M É

Jacques Friedel est l'auteur de travaux pionniers sur la physique des impuretés dans les métaux. On lui doit, outre la découverte des oscillations de Friedel, le concept d'état lié virtuel et la découverte du lien entre déphasage et charge sur l'impureté (règle de somme de Friedel). Après avoir brièvement décrit certaines de ces notions, je présente leur utilisation fructueuse dans deux contextes récents : le blocage de Coulomb dans un point quantique et sa suppression par l'effet Kondo, ainsi que la théorie de champ moyen dynamique des matériaux à fortes corrélations électroniques.

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1. Introduction

Jacques Friedel pioneered the theoretical study of impurities in metals. Encouraged by his thesis mentor (and future brother in law) Nevill Mott, he recognised the fundamental interest of the subject – not to mention its practical importance [1]. His own words, from a 2001 interview that I will quote several times in the following, make this very clear:

"Mott m'a mis sur un problème fondamental, celui des impuretés. Quand on change la nature d'un atome de métal, qu'on ajoute ou retire un atome de ce cristal, comment les électrons réagissent-ils?" [2]. In just a few years, during his PhD (Bristol, 1952) [3] and in the years immediately following [4–6], he unraveled physical effects and forged key concepts which were to become classic in solid-state physics, among which: Friedel's oscillations of the charge density around an impurity, the analysis of these problems in terms of phase-shifts, and the concept of virtual bound-state. Between 1952 and 1961, the vast majority of theoretical studies devoted to impurities in metals was performed by Friedel and his first students (André Blandin, Émile Daniel, Paul de Faget de Casteljau) [7–10], first at the "École des mines" and then at the Laboratoire de physique des solides in Orsay, which Friedel co-founded. For an attempt to analyze some of the developments of this field of theoretical solid-state physics between 1952 and 1973 with a "sociology of science" perspective, the reader may want to consult the curious and rarely quoted book and article by Giuseppe Morandi and coworkers [11,12].

By the early 1960s, it had become clear that the observation of a minimum in the temperature dependence of the resistivity of some metals containing magnetic impurities ("Kondo effect") was a puzzle of far-reaching fundamental significance – for a review of the early experiments, see, e.g., Ref. [13]. Following Kondo's pioneering theoretical work in 1964 [14], the topic attracted enormous international attention and became a central topic of condensed matter physics all over the world. Research on this topic at the Orsay laboratory remained very active well into the 1960s and early 1970s, with important contributions on the theory side by André Blandin, Bernard Coqblin [15,16] and Marie-Thérèse Béal-Monod [17] and, on the experimental side, by Philippe Monod [18] and Henri Alloul [19].

In this article, I would like to describe two recent revivals of magnetic impurity physics in general, and especially of the virtual bound-state concept. For an early review of this concept and of its applications by Friedel himself, see Ref. [20].

The first one is about small electronic devices known as quantum dots, in which it has been realized that an odd number of electrons trapped on the dot can act as an "artificial magnetic impurity", and that the resulting many-body effects deeply affect the physics of the Coulomb blockade in these devices.

The second one – the dynamical mean-field theory of strong electronic correlations – carries the concepts developed for magnetic impurities outside of the context of impurity physics proper. Impurity problems are viewed there as the essential building block for constructing a mean-field theory of solids with strongly interacting electrons.

One of the distinguished strengths of deep and lasting concepts is that they can be exported well beyond the realm in which they were originally invented. I hope that these two examples will illustrate the lasting beauty of impurity physics in a manner of which Friedel would not disapprove.

2. Virtual bound states and phase shifts

Consider an impurity atom embedded in a metallic host, with some of its energy levels not deep enough to trap electrons into a bound state. This happens in particular when the impurity level is located within the conduction band of the host. In this case, the conduction electrons spend an appreciable time near the impurity, but the wave function is still a Bloch wave far from the impurity instead of being localized as in a bound state. Friedel recognized the importance of this situation for impurities in metals and introduced the concept of "virtual bound state" to describe it.

The simplest model for this is the following. Let us denote by ε_d the impurity level (relative to the Fermi level of a conduction electron gas, which in the following will always be taken at energy $\varepsilon = 0$), by V a typical matrix element for the transfer of electrons between the conduction band and the impurity level and by ρ_c^0 the conduction density of states (assumed here to be constant, corresponding to a wide band). The impurity level acquires a width given by Fermi's golden rule:

$$\Gamma = \pi V^2 \rho_c^0 \tag{1}$$

The typical time spent by an electron on the impurity level is of order $\hbar/\Gamma$. The additional density of states created by the impurity at energy ω (we set $\hbar = 1$ in the following) counted from the Fermi level has a simple Lorentzian form:

$$A_d^0(\omega) = \frac{1}{\pi} \frac{\Gamma}{(\omega - \varepsilon_d)^2 + \Gamma^2} \tag{2}$$

The average number of electrons on the impurity level (which is not an integer, since electrons come and go from the level into the metallic host) is therefore:

$$n_d = \int_{-\infty}^{0} d\omega A_d(\omega) = 1 - \frac{2}{\pi} \tan^{-1} \frac{\varepsilon_d}{\Gamma} \tag{3}$$

As expected, it varies from $n_d = 2$ for an impurity level well below the Fermi level ($\varepsilon_d \ll 0$) to $n_d = 0$ when the level is well above. The special case $\varepsilon_d = 0$ of an impurity level exactly at the Fermi level has an additional "particle–hole" symmetry and in this case $n_d = 1$ (half-filled impurity level $n_d = 1$).

Conduction electrons states are affected by the impurity: their wave-function suffers a phase-shift (see Appendix A for a derivation):

$$\delta(\omega) = \frac{\pi}{2} - \tan^{-1}\frac{\varepsilon_d - \omega}{\Gamma} \tag{4}$$

Conduction electrons with an energy well below that of the impurity level ($\omega \ll \varepsilon_d$) have $\delta = 0$, while those well above ($\omega \gg \varepsilon_d$) have $\delta = \pi$. Electrons with an energy equal to that of the impurity level suffer maximal (unitary) scattering $\delta = \pi/2$. In particular, the conduction electrons at the Fermi level are resonantly scattered at the unitary limit by an impurity level resonant with the Fermi level.

From (3) and (4), it is seen that the average number of electrons *on the impurity level* and the phase-shift of conduction electrons *at the Fermi level* are related by:

$$n_d = \frac{2}{\pi}\delta(\omega = 0) \tag{5}$$

This is the celebrated *Friedel sum-rule*. A remarkable aspect of this relation is that it relates a low-energy property (phase-shift on the Fermi surface) to an equilibrium mean value, the total charge on the impurity, which involves an integration of the density of states over all occupied states (hence, the term "sum-rule").

Until the early 1960s, all the pioneering work of the "Orsay school" on non-magnetic and transition-metal impurities in metals was formulated in terms of these concepts – phase shifts and virtual bound states – using a first quantized language – wave-functions and scattering theory. Naturally, in a form much more detailed and sophisticated than the simple toy-model used here to illustrate these concepts. In particular, the different angular momentum channels of the scattered conduction electron waves, as well as the orbital degeneracy of the impurity (neglected above) were taken into account. For transition-metal impurities, the key question was: "Under which condition does the impurity possess a magnetic moment, depending on the transition-metal element under consideration and on the nature of the host metal?". To address this question, the effect of intra-atomic interactions as well as inter-impurity exchange interactions were considered at the Hartree–Fock level, in particular by Blandin [8] (see also [21]).

In the characteristic style of the Friedel school, certainly influenced by the British tradition in solid-state physics, these articles were also rich with phenomenological considerations and connections to experimental observations, often intermingled with the theory itself. Most of these papers were written in French and appeared in *Le Journal de physique et le radium* (see, e.g., Refs. [5,7–10], among several others).

3. The Anderson impurity model

3.1. The model

In 1961 (incidentally, a year of special significance to me!), Philip W. Anderson published in *Physical Review* an article entitled "Localized Magnetic States in Metals" [22], in which he introduced the following model Hamiltonian:

$$H_{\text{imp}} = \sum_{\mathbf{p},\sigma=\uparrow,\downarrow} E_{\mathbf{p}}\, c^{\dagger}_{\mathbf{p}\sigma} c_{\mathbf{p}\sigma} + \sum_{\mathbf{p}\sigma}\left(V_{\mathbf{p}}\, c^{\dagger}_{\mathbf{p}\sigma} d_{\sigma} + V^{*}_{\mathbf{p}}\, d^{\dagger}_{\sigma} c_{\mathbf{p}\sigma}\right) + H_{\text{at}} \tag{6}$$

The model describes, in an extraordinarily simplified way, a metallic host (first term) coupled by the second ('hybridization') term to a single-level atom (last term). The atomic level is devoid of any orbital degrees of freedom and is described by the following expression:

$$H_{\text{at}} = \varepsilon_d(\hat{n}_{d\uparrow} + \hat{n}_{d\downarrow}) + U\,\hat{n}_{d\uparrow}\hat{n}_{d\downarrow} \ , \ \ \hat{n}_{d\sigma} \equiv d^{\dagger}_{\sigma} d_{\sigma} \tag{7}$$

in which ε_d is, as above, the atomic level position, and U is a repulsive interaction originating from the Coulomb repulsion between two electrons (necessarily with opposite spins) occupying the level simultaneously.

3.2. Some considerations on influence and style in science

One immediately recognizes in this model the influence of Friedel's thinking. The first two terms describe the coupling of the impurity level with the Fermi sea, leading to the formation of a virtual bound state as described in the previous section. This influence is actually acknowledged by Anderson in the introduction of his article, where he emphasizes that the key question is the formation (or not) of a local moment and that "Friedel and collaborators have made a start at an understanding of this phenomenon." This is formulated with more generosity in his Nobel lecture[1]: "More important was my first exposure to Friedel's and Blandin's ideas on resonant or virtual states at that conference[2]" [23]

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[1] I remind the reader that the citation for the 1977 Nobel prize to Anderson, Mott and van Vleck mentions both Anderson's work on magnetic impurities and his work on localization.

[2] The conference mentioned here is a 1959 Brasenose College, Oxford conference on magnetism in metals, see the article by Emile Daniel in this volume who refers to a 1957 conference also organized by the Oxford–Harwell group.

Anderson's 1961 article brought in several key innovations. It introduced in a crystal-clear manner the simplest possible model in which the physics of a single transition-metal impurity atom can be studied. It recognized the key importance of the intra-atomic Coulomb repulsion in the formation of a local moment.[3] And, most importantly, it formulated this model in the modern language of second-quantization, opening the road to the use of many-body theory techniques which blossomed in the 1960s and 1970s and for which the magnetic impurity problem proved to be an invaluable testing-ground.

In fact, the extent to which this problem has been a laboratory for the development of theoretical physics is simply amazing. The very elaboration of the renormalization group concept, the first elaboration of a numerical non-perturbative renormalization group, the Bethe Ansatz and integrability, boundary conformal field-theory and – last but not least – the development of powerful computational algorithms for the many-body problem were either born in the study of the magnetic impurity problem, or underwent significant new developments when applied to it (see [24] for a review). Quoting again Anderson's Nobel lecture [23]: "H. Suhl has been heard to say that no Hamiltonian so incredibly simple has ever previously done such violence to the literature and to national science budgets."

Both Friedel and Anderson certainly share a taste for simple formulations, although with very different stylistic choices. Two quotations echo each other in this respect in an interesting manner:

- Friedel [2]: "Depuis Bristol, j'ai toujours eu une activité de recherche sur l'électronique des solides. J'ai essayé de développer des modèles approximatifs mais simples, compréhensibles et même utilisables par des non-spécialistes. Je suis un vrai élève de N.F. Mott en ce que, dans un domaine complexe où 10^{24} particules sont en interactions fortes, des caricatures faites sur un dos d'enveloppe me semblent pouvoir être plus pertinentes que des collections de papillons, photos léchées à l'ordinateur mais souvent peu généralisables."
- Anderson [23]: "One of my strongest stylistic prejudices in science is that many of the facts Nature confronts us with are so implausible given the simplicities of non-relativistic quantum mechanics and statistical mechanics, that the mere demonstration of a reasonable mechanism leaves no doubt of the correct explanation. Very often, such a simplified model throws more light on the real workings of nature than any number of "ab initio" calculations of individual situations, which even when correct often contain so much detail as to conceal rather than reveal reality. It can be a disadvantage rather than an advantage to be able to compute or to measure too accurately, since often what one measures or computes is irrelevant in terms of mechanism. After all, the perfect computation simply reproduces Nature, does not explain her."

3.3. Physics of the Anderson impurity model, in a nutshell

Let me know briefly review the physics of model (6) – covering only a very limited number of key things and not following historical order.

Consider first the isolated "atom" $V = 0$. H_{at} has four eigenstates: $|0\rangle$ with energy 0, the two degenerate states $|\uparrow\rangle$ and $|\downarrow\rangle$ with energy ε_d and the doubly occupied state $|\uparrow\downarrow\rangle$ with energy $2\varepsilon_d + U$. Level-crossings occur for $\varepsilon_d = -U$ and $\varepsilon_d = 0$. The ground state is $|\uparrow\downarrow\rangle$ for $\varepsilon_d < -U$, the degenerate doublet $|\uparrow\rangle$, $|\downarrow\rangle$ for $-U < \varepsilon_d < 0$ and the empty state $|0\rangle$ for $\varepsilon_d > 0$. Hence, the occupancy of the impurity level as a function of ε_d has the shape of a "staircase", see Fig. 1 (which in the mesoscopic context of next section can be called the 'Coulomb blockade' staircase). At finite temperature, the staircase is rounded off, according to the expression ($\beta = 1/kT$): $n_d/2 = (e^{-\beta\varepsilon_d} + e^{-\beta(2\varepsilon_d+U)})/(1 + 2e^{-\beta\varepsilon_d} + e^{-\beta(2\varepsilon_d+U)})$.

In the local moment (LM) regime $-U < \varepsilon_d < 0$, the ground state is degenerate and we are really dealing with an atom carrying an $S = 1/2$ local moment, which will then be coupled to the conduction electron 'bath'. Note that $\varepsilon_d = -U/2$ has an enlarged, particle–hole, symmetry (a property of the model for arbitrary V). Extra degeneracies appear for $\varepsilon_d \approx -U$ and $\varepsilon_d \approx 0$ in which the occupancy of the level, i.e. the valence of the atom, can easily fluctuate (mixed valent regimes).

The density of states, or more precisely the spectral function that measures transitions corresponding to adding or removing a particle on the atomic level reads, for the isolated atom:

$$A_d^{at}(\omega) = (1 - \frac{n_d}{2})\, \delta[\omega - \varepsilon_d] + \frac{n_d}{2}\, \delta[\omega - (\varepsilon_d + U)] \tag{8}$$

This simply corresponds to sharp atomic transitions between two valence states.

The opposite limit of an atomic level coupled to the conduction electron band, but without interactions ($U = 0$, arbitrary V) has been described in the previous section. The ground-state is a non-degenerate singlet, and the level hybridizes with the conduction electrons to form a virtual bound-state (VBS) of width Γ. No local moment appears on the impurity site.

The whole problem is to understand what happens in between these two extreme limits, either by turning on a weak hybridization V starting from the atomic limit, or by turning on interactions starting from the non-interacting VBS. In

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[3] Blandin and Friedel had considered early on the effect of interactions [8], mostly in the form of the intra-atomic Hund's exchange, and derived already in 1959 a Hartree–Fock criterion for the appearance of a local moment. A detailed discussion of the similarities and differences regarding the role of interactions in Blandin–Friedel and Anderson's points of views would take us too far from the main topic of this article. For an account by Friedel himself, see his 1962 review [20]. The opening sentence of this review is: "Le concept de niveau lié virtuel semble s'être imposé dans l'étude des impuretés de transition dissoutes dans les métaux normaux."
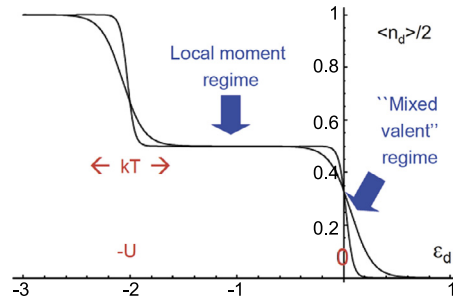
Fig. 1. Occupancy of the isolated single-level atom, as a function of level position ε_d, at two temperatures.

particular, can a local moment (with, e.g., a Curie susceptibility of the impurity atom) be sustained despite the coupling to the conduction electrons? This question is of course especially relevant in the LM regime.

Anderson [22] (and Blandin–Friedel [8] with their own perspective) made a Hartree treatment of interactions,[4] and concluded that a LM is formed when the ratio of interactions to the VBS width Γ reaches a critical value. However, it became clear (I think Schrieffer in particular pointed this out early on) that this phase transition is actually an artefact of the mean-field treatment, and that the behaviour is actually entirely smooth when increasing U, starting from the non-interacting limit. The ground state is a singlet for all finite values of U/Γ, with no LM remaining at $T = 0$. This statement is valid provided that the conduction band has finite density of states at the Fermi level, i.e. for an impurity atom inserted into a metallic host.

Interestingly, this continuity as a function of U took quite a while to be unravelled [25,26], and the focus of attention in the early 1960s was more on the physics for $\Gamma \ll U$, i.e. small V (which is of course the realistic limit from an experimental point of view). However, this is a singular limit: the double degeneracy of the LM ground state is lifted as soon as $V \neq 0$.

Of course, the Kondo calculation [14] played an essential role in focusing the attention on that limit. Schrieffer and Wolff [27] realised that in the LM regime and at strong coupling $U \gg \Gamma$, the low-energy sector of the Anderson model can be described by an exchange model of the type used by Kondo and many other researchers at the time:

$$H_K = \sum_{\mathbf{p},\sigma=\uparrow,\downarrow} E_{\mathbf{p}} c^\dagger_{\mathbf{p}\sigma} c_{\mathbf{p}\sigma} + J_K \vec{S}_d \cdot \vec{s}_c(0) + \text{potential scattering} \tag{9}$$

with:

$$J_K = 2V^2 \left[\frac{1}{\varepsilon_d + U} - \frac{1}{\varepsilon_d} \right] \tag{10}$$

Note that strong coupling for the Anderson model corresponds to weak coupling $J_K \rho_c^0 \ll 1$ in the Kondo model. Understanding the Kondo model in this limit required nothing less than forging the concepts of renormalization group theory, providing the first example of its use in condensed matter physics. It became apparent that the singularities discovered by Kondo and resulting in the generation of the low-energy scale (with $\Lambda \sim \sqrt{U\Gamma}$ a high-energy cutoff):

$$T_K = \Lambda \, e^{-1/J_K \rho_c^0} \tag{11}$$

actually signal the RG flow to strong coupling and the formation of the singlet state for $T \ll T_K$. In this regime, one can consider that the impurity LM has been 'swallowed' or screened out by the metallic host.

The formation of the singlet state has a direct consequence for the conduction electrons: the wave-functions are phase-shifted. Remarkably, the phase-shift at the Fermi level is related to the occupancy of the impurity level by exactly the same expression (5) that applies to the non-interacting case[5]:

$$\delta(0) = \frac{2}{\pi} n_d \tag{12}$$

The Friedel sum-rule is thus valid and unchanged for arbitrary interaction strength U/Γ. A formal proof, due to Langreth [28], is given in Appendix B.

Let us come back to the adiabatic continuity as a function of U/Γ, explored in great details in particular in a series of remarkable papers by Yosida and Yamada (see, e.g., [26] and references therein). First, we note that the physics of the electrons on the impurity level does not depend on all details of the conduction electron band (such as the precise form of the dispersion $E_{\mathbf{p}}$ and hybridisations $V_{\mathbf{p}}$), but actually only on the hybridisation function (or energy-dependent VBS width):

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[4] Hartree–Fock is often invoked here, but there is actually no Fock term in this model.

[5] While of course the specific relation between ε_d and n_d is U-dependent, and (3), (4) do not apply to the interacting system.

$$\Gamma(\omega) \equiv \pi \sum_{\mathbf{p}} |V_{\mathbf{p}}|^2 \delta(\omega - E_{\mathbf{p}}) \tag{13}$$

This can be seen formally by "integrating out" the conduction electron environment, leading to the following effective action for the dynamics of the impurity level:

$$S_{\text{eff}} = -\int_0^\beta \mathrm{d}\tau \int_0^\beta \mathrm{d}\tau' \sum_\sigma d_\sigma^\dagger(\tau) \mathcal{G}_0^{-1}(\tau - \tau') d_\sigma(\tau') + U \int_0^\beta \mathrm{d}\tau\, n_{\mathrm{d}\uparrow}(\tau) n_{\mathrm{d}\downarrow}(\tau) \tag{14}$$

with:

$$\mathcal{G}_0^{-1}(\mathrm{i}\omega_n) = \mathrm{i}\omega_n - \varepsilon_{\mathrm{d}} - \Delta(\mathrm{i}\omega_n) \;,\;\; \Delta(\mathrm{i}\omega_n) = \frac{1}{\pi} \int \mathrm{d}\varepsilon \frac{\Gamma(\varepsilon)}{\mathrm{i}\omega_n - \varepsilon} \tag{15}$$

Physically, (14) describes electrons that are transferred from the conduction electron bath onto the impurity level at a certain time τ and leave it at another time τ' to return to the bath. While residing on the impurity, they feel the local interaction U. The hybridization function $\Gamma(\omega)$ is the frequency of these processes in the absence of interactions as dictated by Fermi's golden rule.

The action (14) is that of a local theory with interaction and retardation, in which no spatial degrees of freedom are involved. $\mathcal{G}_0(\tau)$ plays the role of the non-interacting propagator of this theory. For a metallic host (gapless $\Gamma(\omega)$), it decays at long-time as $1/\tau$, exactly as any local propagator in a free electron gas. Therefore, this theory obeys all the requirements for *Fermi-liquid behaviour* at low-energy, only in local form. Philippe Nozières pioneered the use of such local Fermi liquid concepts in his analysis of the Kondo problem, in which the phase-shift concept plays an essential role [25]. Here, we rather look at it from the point of view of the impurity level.[6] We can encode the effects of interactions into a local d-level self-energy related to the d-level Green's function by Dyson's equation:

$$G_{\mathrm{d}}(\mathrm{i}\omega_n)^{-1} = \mathcal{G}_0^{-1}(\mathrm{i}\omega_n) - \Sigma_{\mathrm{d}}(\mathrm{i}\omega_n) = \mathrm{i}\omega_n - \varepsilon_{\mathrm{d}} - \Delta(\mathrm{i}\omega_n) - \Sigma_{\mathrm{d}}(\mathrm{i}\omega_n) \tag{16}$$

And the retarded self-energy $\Sigma_{\mathrm{d}}(\omega + \mathrm{i}0^+) = \Sigma_{\mathrm{d}}'(\omega) + \mathrm{i}\Sigma_{\mathrm{d}}''(\omega)$ obeys at $T = 0$ the following low-frequency behaviour characteristic of a local Fermi liquid:

$$\Sigma_{\mathrm{d}}'(\omega) = \Sigma_{\mathrm{d}}(0) + \omega\left(1 - \frac{1}{Z}\right) + \cdots \;,\;\; \Sigma_{\mathrm{d}}''(\omega) = -A\omega^2 + \cdots \tag{17}$$

When the conduction electron band density of states is very wide and featureless (a convenient approximation often justified for magnetic impurities in metals but *not* in the novel context of Sec. 5), $\mathrm{Re}\Delta(\omega + \mathrm{i}0^+) \simeq 0$ vanishes and $\mathrm{Im}\Delta(\omega + \mathrm{i}0^+) \simeq -\Gamma$ is a constant, so that the d-level density of states (spectral function) reads:

$$A_{\mathrm{d}}(\omega) \equiv -\frac{1}{\pi} \mathrm{Im} G_{\mathrm{d}}(\omega + \mathrm{i}0^+) = \frac{1}{\pi} \frac{\Gamma - \Sigma_{\mathrm{d}}''(\omega)}{\left[\omega - \varepsilon_{\mathrm{d}} - \Sigma_{\mathrm{d}}'(\omega)\right]^2 + \left[\Gamma - \Sigma_{\mathrm{d}}''(\omega)\right]^2} \tag{18}$$

The shape of this spectral density (Fig. 2) can be discussed qualitatively using the simple considerations on the atomic limit (8) as well as the Fermi liquid considerations (17) and the Friedel sum-rule. We focus on the regime where U/Γ is large enough for the different spectral features to be well separated.

At high energies (far from the Fermi level), we expect the atomic transitions in (8) at ε_{d} and $\varepsilon_{\mathrm{d}} + U$ to be simply broadened, by a width of order Γ (the width is actually $\simeq 2\Gamma$ [31]). From a high-energy perspective, the effect of interactions is to split the spectral signature of the VBS at ε_{d} into a hole-like spectral peak corresponding to transitions $|\sigma\rangle \to |0\rangle$ (transition energy ε_{d}) and an electron-like one corresponding to $|\sigma\rangle \to |\uparrow\downarrow\rangle$ (transition energy $\varepsilon_{\mathrm{d}} + U$). Properly interpreted, the Hartree approximation precisely does this. For temperatures $T \gg T_{\mathrm{K}}$, when the system is in a LM state, we do not expect any other spectral feature and this two-peak structure is a reasonable description of the spectral function (Fig. 2).

In contrast, for $T \ll T_{\mathrm{K}}$, the formation of the singlet ground state has a dramatic consequence on the low-energy spectral features in A_{d} (Fig. 2). Indeed, from the Fermi-liquid form (17), we see that the $T = 0$ spectral function can be approximated by (neglecting all higher order terms including the ω^2 one):

$$A_{\mathrm{d}}(\omega \simeq 0) \simeq \frac{Z}{\pi} \frac{\widetilde{\Gamma}}{(\omega - \widetilde{\varepsilon}_{\mathrm{d}})^2 + \widetilde{\Gamma}^2} \tag{19}$$

where Z has been defined above in Eq. (17), and:

$$\widetilde{\Gamma} = Z\Gamma \;,\;\; \widetilde{\varepsilon}_{\mathrm{d}} = Z\left[\varepsilon_{\mathrm{d}} + \Sigma_{\mathrm{d}}'(0)\right] \tag{20}$$

Hence at low energy, the formation of the Kondo singlet leads to the appearance of a narrow resonance (Fig. 2), of spectral weight Z, with a renormalized width $\widetilde{\Gamma}$ and position $\widetilde{\varepsilon}_{\mathrm{d}}$. Often called the 'Kondo' resonance, the fact that the true low-energy

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[6] For a detailed Fermi liquid analysis of the Anderson impurity model, see the recent work by C. Mora et al. [29].
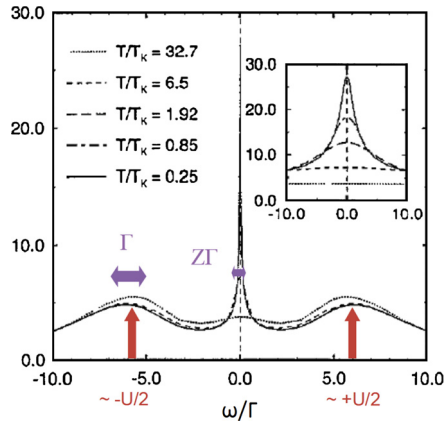
Fig. 2. Spectral function of the Anderson impurity model (symmetric case $\varepsilon_d = -U/2$, $n_d = 1$) showing the gradual formation of the low-energy 'Kondo' resonance as temperature is lowered below T_K, as obtained from Numerical renormalisation-group calculations. Reproduced from Ref. [30].

behaviour corresponds to resonant scattering at the Fermi level (in the symmetric case), and that the singularities discovered by Kondo are not really physical, was first pointed out, I believe, by Abrikosov [32], Suhl [33] and Nagaoka [34]. The spectral weight Z of this resonance in A_d can be shown to be of order T_K/Γ, a very small number at strong coupling deep into the LM regime.

For $\varepsilon_d = -U/2$ (center of the Coulomb blockade plateau with $n_d = 1$), the additional particle–hole symmetry makes the discussion easier. Because of this symmetry, $A_d(\omega) = A_d(-\omega)$, implying that $\Sigma'_d(0) = U/2$ and $\widetilde{\varepsilon}_d = 0$: the resonance sits right at the Fermi level. Hence, the height of the resonance is:

$$A_d(0) = \frac{1}{\pi\Gamma} \tag{21}$$

Note that the spectral weight Z drops out of this expression, and that the height appears as that of an unrenormalized VBS now sitting at the Fermi level, i.e. the resonant condition. As expected in that case, the conduction electrons undergo resonant scattering from the Kondo singlet, corresponding to a phase shift:

$$\delta = \frac{\pi}{2} \tag{22}$$

This phase-shift simply means that the formation of the Kondo singlet makes the impurity site inaccessible to other conduction electrons: their wave functions vanishes on the impurity site thanks to the $\pi/2$ phase-shift, which is such that a maximum of the Bloch wave becomes a node – a picture that becomes transparent in the strong Kondo coupling limit.

The analysis of the general case requires to relate the Fermi liquid renormalization of the d-level to the phase-shift. This is done in Appendix A, in which it is shown that the scattering T-matrix of the conduction electrons is given by $T_{\mathbf{p}\mathbf{p}'}(\omega) = V_{\mathbf{p}}^* G_d(\omega) V_{\mathbf{p}'}$, so that the phase-shift at energy ω reads:

$$\tan\delta(\omega) = \frac{\Gamma - \Sigma''_d(\omega)}{\varepsilon_d + \Sigma'_d(\omega) - \omega} \tag{23}$$

Hence, at $T = \omega = 0$, we obtain (we use $\delta \equiv \delta(0)$ for simplicity):

$$\varepsilon_d + \Sigma'_d(0) = \frac{\Gamma}{\tan\delta} \tag{24}$$

and using the Friedel sum-rule, one finally obtains the position of the resonance and height at the Fermi level, in the general case, as:

$$\widetilde{\varepsilon}_d \equiv Z\left[\varepsilon_d + \Sigma'_d(0)\right] = \frac{Z\Gamma}{\tan\delta} = \frac{Z\Gamma}{\tan(\pi n_d/2)} \tag{25}$$

$$A_d(0) = \frac{\sin^2\delta}{\pi\Gamma} = \frac{\sin^2(\pi n_d/2)}{\pi\Gamma} \tag{26}$$

Comparing to Sec. 2, we see that the value of A_d at the Fermi level is *unrenormalized by interactions*. The resonance is in general not peaked at the Fermi level however, but the above expression shows that $\widetilde{\varepsilon}_d$ is quite close to $\omega = 0$ as long as $T_K \ll \Gamma$.
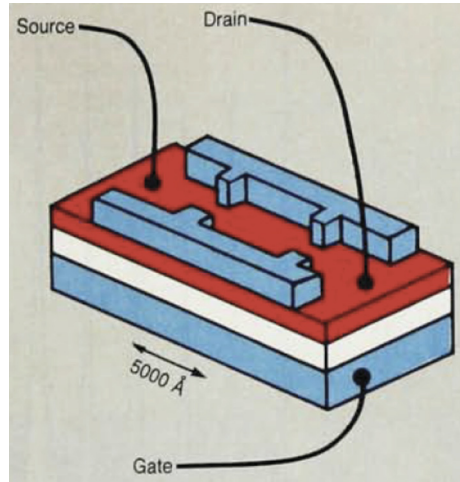
Fig. 3. Schematic view of a quantum dot (from [35]). A two-dimensional electron gas is created at the interface between a semiconducting layer (in red) and an insulating layer (in white). A metallic backgate, as well as top gates, (in blue) allow for the control of the device.

Using the quoted expression of the T-matrix, one can deduce (Appendix A) the expression of the local conduction electron density of states at the Fermi energy:

$$A_c(0) = \rho_c^0 \cos^2 \delta = \rho_c^0 \cos^2 \frac{\pi n_d}{2} \tag{27}$$

and derive an expression for the contribution of the impurity to the conductivity:

$$\frac{\sigma_{imp}(T)}{\sigma_u} = \int d\omega \left(-\frac{\partial f}{\partial \omega} \right) \frac{1}{\pi \Gamma A_d(\omega)} \tag{28}$$

Hence, at $T = 0$, the contribution to the *resistivity* is:

$$\frac{R_{imp}}{R_u} = \pi \Gamma A_d(0) = \sin^2 \delta = \sin^2 \frac{\pi n_d}{2} \tag{29}$$

In these expressions, R_u (σ_u) denote the unitary limit (maximal scattering). Hence, we see that in the LM regime with $\delta = \pi/2$ ($n_d = 1$), the Kondo effect leads to a vanishing of the local conduction electron d.o.s (Kondo hole) and to scattering at the unitary limit.

4. Quantum dots, Coulomb blockade, and the Kondo effect

Fig. 3 displays a schematic representation of a "quantum dot". Modern semiconductor elaboration techniques allow one to isolate a small region of an electron gas, containing only a few electrons (the dot) connected to reservoirs by tunable tunnel barriers [35]. Quantum dots are often referred to as "artificial atoms", since they have a discrete spectrum of energy levels which is determined by the geometric properties of the confining potential (as well as the interaction between electrons on the dot).

For small dots in semiconductors, the level spacing can be quite large, and focusing on a single energy-level may be a reasonable approximation in a range of gate voltage. In this case, the device is well described by a slight generalization of the Anderson impurity model, in which the 'impurity level' is connected not to a single but to two (left and right) reservoirs. The position of the energy level (ε_d) can be tuned by the overall gate voltage, while the tunnel matrix elements V_L, V_R connecting the dot to the left and right reservoirs are also tunable by applying appropriate tensions. The interaction U is determined by the capacitive energy of the dot $U \sim e^2/C$.

In this model, the occupancy (charge) of the dot depends on the "gate voltage" ε_d according to the staircase of Fig. 1. The addition of a new electron costs an energy U, and for a range of gate voltages the charge on the dot may correspond to an odd number of electrons (a single one in this simple model), and hence to a half-integer spin. In this situation, the device can be seen as an artificial magnetic impurity connected to two conduction electron reservoirs.

One of the simplest measurement on such a device is to apply a small voltage between the drain (R reservoir) and the source (L reservoir) and to measure the linear-response conductance G of the device. I emphasize that this is the conductance corresponding to *transmission* of electrons through the dot. Hence, a Landauer-like formula should be applicable to describe this process. However, the Landauer approach must be significantly generalized here, since electrons interact on the dot. This problem was considered by Meir and Wingreen [36], who established a remarkably simple formula for the equilibrium conductance:
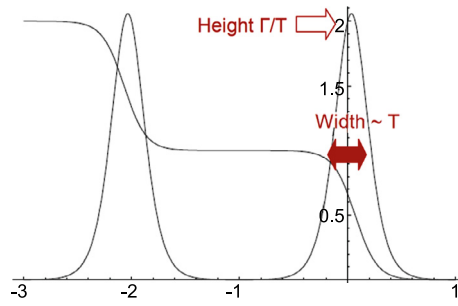
Fig. 4. Coulomb blockade staircase and conductance peaks (in units of $2e^2/h$), as a function of 'gate voltage' ε_d, according to the approximate high-temperature expression (32).

$$G = \frac{2e^2}{h} \frac{4\Gamma_L\Gamma_R}{(\Gamma_L + \Gamma_R)^2} \int d\omega \left(-\frac{\partial f}{\partial \omega}\right) \pi \Gamma A_d(\omega) \tag{30}$$

$$= \frac{2e^2}{h} \int d\omega \left(-\frac{\partial f}{\partial \omega}\right) \pi \Gamma A_d(\omega) \ , \ (\Gamma_L = \Gamma_R) \tag{31}$$

In this expression, $A_d(\omega)$ is the equilibrium spectral function introduced above, $\Gamma_{R,L}$ are the Fermi golden rules level widths associated with tunneling to the two reservoirs, $\Gamma = \Gamma_L + \Gamma_R$, and $f(\omega) = 1/(1 + e^{\omega/kT})$ is the Fermi function. The second expression is obtained for a dot with equal L- and R-coupling, a simplification that we shall make in the following.

This expression is obviously very analogous to Landauer's formula, with $\pi \Gamma A_d(\omega)$ playing the role of the transmission coefficient through the device. It is remarkable that such a simple formula applies to an interacting system, and especially that the conductance can be expressed in terms of a *single-particle* spectral function. This is due to the fact that the current operator couples one electron on the dot to a *non-interacting* state in the reservoir, e.g., $iV_L(d_\sigma^\dagger c_{\mathbf{p}\sigma} - c_{\mathbf{p}\sigma}^\dagger d_\sigma)$. It is also interesting to contrast this formula to expression (28), which described the contribution σ_{imp} of magnetic impurities to the conductivity of a metallic host. It is the *inverse* of $\pi \Gamma A_d$ that appears in this formula, so that $\pi \Gamma A_d$ plays there the role of a *scattering rate*. In contrast, when considering the *transmission* conductance, the same quantity plays the role of a transmission coefficient!

At high temperature, the spectral function (transparency) has the two-peak structure described in the previous section. As a first approximation, one may use in (31) the expression (8) for an isolated dot. One then obtains:

$$G(T \gg \Gamma) \simeq \frac{2e^2}{h} \frac{\pi \Gamma}{kT} \left[(1 - \frac{n_d}{2}) \frac{1}{4 \cosh^2 \frac{\varepsilon_d}{2kT}} + \frac{n_d}{2} \frac{1}{4 \cosh^2 \frac{\varepsilon_d + U}{2kT}} \right] \tag{32}$$

A plot of this expression is displayed in Fig. 4, together with the Coulomb blockade staircase itself. As a function of the gate voltage ε_d, the conductance displays two peaks that are located at the "mixed valent" points where two atomic states with charge differing by one unit are degenerate. At these points, the charge easily fluctuates and transmission of an electron through the dot is possible. In between (LM regime), the charge is blocked by the Coulomb repulsion and the transmission conductance is therefore very small (Coulomb blockade).

This is not the whole story however. It was realized in 1988 by Glazman and Raikh [37] and Ng and Lee [38] that the Kondo effect should profoundly modify this picture at low temperature. Indeed, at $T = 0$, it should *entirely suppress the Coulomb blockade* throughout the LM regime and restore transmission conductance to its maximal possible value $2e^2/h$! This remarkable many-body effect can be viewed as a direct consequence of the appearance of the 'Kondo' resonance in the density of states, and of the Friedel sum-rule. Indeed, at zero temperature, expression (31) reduces to:

$$G = \frac{2e^2}{h} \pi \Gamma A_d(0) = \frac{2e^2}{h} \sin^2 \frac{\pi n_d}{2} \quad (T = 0) \tag{33}$$

in which the second expression makes use of the value of $A_d(0)$ derived above, based on the Friedel sum-rule. This expression shows that the Coulomb blockade should be essentially suppressed at $T = 0$, and in particular that maximum conductance is restored for $n_d = 1$, precisely in the middle of the valley where the conductance was at a minimum in the high-temperature regime! Note that the tiny spectral weight $Z \sim T_K/\Gamma$ of the Kondo resonance does not limit the conductance: it is the value of the density of states that matter. Indeed, we know from Landauer that a single resonant state with perfect transmission ($\pi \Gamma A_d(0) = 1$ for $n_d = 1$) carries a full quantum of conductance.

Another, more physical, point of view on this somewhat "magic" derivation is to think in terms of wave functions. This is most easily understood in a simple version of the problem in which a single state would be present on the leads (strong Kondo coupling limit), or if one wants more sophistication using a variational wave-function à la Gunnarsson–Schönhammer [39]. One immediately sees then that the formation of the Kondo singlet is possible because the ground-state wave-function contains a small contribution involving both the empty and doubly-occupied configurations (in the single-level description, this contribution is proportional to V/U). It is these virtual contributions to other charge states that allow
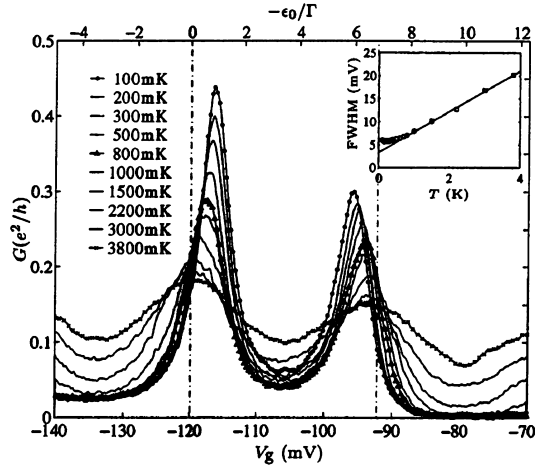
Fig. 5. Conductance through a quantum dot in the Kondo regime: early experiments. Reproduced from Ref. [41].
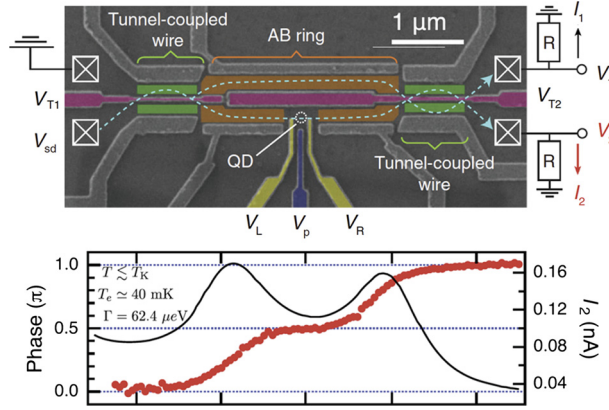

Fig. 6. Top: Interferometric device recently used in [46] to measure the phase-shift through a quantum dot (QD). Bottom: Gate-voltage dependence of the measured phase-shift, evidencing in particular the value $\delta = \pi/2$ at the center of the Coulomb blockade valley. Reproduced from Ref. [46].

for perfect transmission (no matter how small their contribution to the wave-function). At low T, the dot and the reservoirs are strongly entangled by the formation of the singlet, and this coupling must be taken into account from the start when considering the conductance.

Experimental observation of this remarkable effect had to wait for the ability to fabricate dots in which the appropriate regime can be reached ($T \lesssim T_K < \Gamma < U$). Note that expressions (10) and (11) show that the Kondo temperature $T_K \simeq \sqrt{\Gamma U} e^{\pi \varepsilon_d (\varepsilon_d + U)/2U\Gamma}$ depends exponentially on the Coulomb repulsion U and on ε_d, and that the Kondo temperature is smallest at the center of the valley ($\varepsilon_d = -U/2$, $n_d = 1$). Experimental observation came in 1998, ten years after the theoretical prediction, in the remarkable doctoral work of David Goldhaber-Gordon [40,41] and also in the group of Leo Kouwenhoven [42] (see also [43]). Measurements from these early experiments [40,41] are reproduced in Fig. 5. The *increase* of conductance in the middle of the valley as temperature is lowered is clearly seen there.

What about the phase shift? As suggested in [44], this should be observable by placing a quantum dot in an Aharonov–Bohm interferometer. Such delicate experiments were soon performed (see, e.g., [45] and additional references in [46]), but their interpretation was complicated by the contribution of multiple paths traversing the interferometer. Very recently, however [46], a collaboration between the groups of C. Bäuerle in Grenoble and S. Tarucha in Tokyo (as well as that of J. von Delft in Munich on the theory side) managed to setup a remarkable experiment and device which overcomes this difficulty. Their results, reproduced in Fig. 6, provide a clear observation of the gate-voltage dependence of the phase-shift, and in particular of the unitary value $\delta = \pi/2$ at the center of the valley, in agreement with Friedel's sum-rule.

5. Dynamical mean field theory

5.1. Atom matter

The second 'revival' that I want to briefly discuss here will take us outside of the field of impurity physics properly speaking. Here, an impurity atom coupled with an environment will be viewed as a building block for constructing a

mean-field theory of quantum many-body problems and materials with strong electronic correlations. This is the basic concept underlying dynamical mean-field theory (DMFT).

This is best illustrated by considering a 'simple' model, such as the Hubbard model:

$$H = \sum_i H_{\mathrm{at}}(i) - \sum_{ij} t_{ij} \left(d^\dagger_{i\sigma} d_{j\sigma} + d^\dagger_{j\sigma} d_{i\sigma} \right) \tag{34}$$

The hamiltonian H_{at} on each lattice site i, given by (7), is that of the single-level 'atom' introduced above.[7] Often this model is referred to as a single band of electrons, subject to a local interaction U between electrons of opposite spins on the same site. In this perspective, the first thing one does is to diagonalize the non-interacting (one-body) part of the hamiltonian in the form $\sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} d^\dagger_{\mathbf{k}\sigma} d_{\mathbf{k}\sigma}$, with $\mathbf{k}$ the quasi-momentum and $\varepsilon_{\mathbf{k}}$ the tight-binding energy band:

$$\varepsilon_{\mathbf{k}} = - \sum_j t_{ij} e^{i\mathbf{k}\cdot(\mathbf{R}_i - \mathbf{R}_j)} \tag{35}$$

Here, I want to adopt a different perspective, and view this model as describing a periodic lattice of atoms[8] that can exchange electrons through the hopping term t_{ij} due to the overlap of atomic orbitals on different sites. These two points of views are equally valid, but they naturally lead to different kinds of approaches. In the former, one will start from an electron gas and Bloch waves in momentum-space, and consider what happens when interactions are turned on. The latter, in contrast, emphasizes that *atoms are the basic building blocks of a solid*. Each atom is a small many-body problem in itself: one will insist on a correct description of the atomic eigenstates (multiplets), and only then consider the transfer of electrons between atoms. The first point of view is mostly a wave-like picture in momentum-space, while the second is a real-space (particle-like) picture closer to the intuition of the chemist.

Most physics textbooks adopt the first point of view: solid-state physics is presented as the science of the electron gas. However, for materials with strong electron correlations, an atomic viewpoint is actually essential.[9] More precisely, we need for these materials a theoretical description which is able to describe *both* atomic-like excitations at high energy (such as the valence-changing transitions when adding or removing an electron), as well as the low energy excitations which in a metallic system will be wave-like quasiparticles (possibly strongly renormalized in comparison to free electrons). Only a theory which can *adapt to the energy scale under consideration* is able to do this.

5.2. Dynamical Mean-Field Theory in a nutshell

The basic concept of DMFT is illustrated in Fig. 7. The idea is to start from the atom and embed it into an effective medium with which it can exchange electrons [48] (for a review, see, e.g., [49]). For our simple single-level atom, this is precisely the single-impurity Anderson model! The "impurity" here is really no impurity at all: it is just one of the atoms of the translationally-invariant system that we have decided to focus on.

The effective medium (or 'reservoir', or 'bath'), in turn, is just a representation of the whole system (minus the atom we are focusing on). Hence, it should be *self-consistently determined*, in order to reflect the fact that all atoms are actually equivalent. What is this self-consistency condition?

The answer to this question relies on two elements: a choice of observable and an approximation. The observable we shall focus on is the on-site (local) Green's function of our solid. It is given by:

$$G_{\mathrm{loc}}(i\omega_n) \equiv \sum_{\mathbf{k}} \frac{1}{i\omega_n + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, i\omega_n)} \tag{36}$$

Here, $\Sigma(\mathbf{k}, i\omega_n)$ is the (unknown) self-energy of our lattice model, encoding the effects of interactions, and the normalized momentum-sum is over the Brillouin zone. We recall that, given a value of U and $\varepsilon_{\mathrm{d}} = -\mu$, the Green's function of the impurity level in the Anderson impurity model is entirely determined by the hybridisation function $\Delta(i\omega_n)$. Assuming that one knows the local Green's function G_{loc} of our solid, let us find a hybridisation function such that the Green's function of the impurity problem coincides with it, namely demand that:

$$G_{\mathrm{loc}}(i\omega_n) = G_{\mathrm{imp}}[i\omega_n, \Delta] \tag{37}$$

At this point, all that we have done is to introduce a representation of the exact local Green's function by that of an embedded atom (effective impurity).

We now introduce an approximation, which allows one to obtain a closed set of equations determining both Δ and G_{loc}. This approximation is to neglect the momentum-dependence of the self-energy (i.e. in real-space ignoring all non-local

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[7] The atomic level can be taken in this context to be (minus) the chemical potential, used to fix the average number of electrons per site n: $\varepsilon_{\mathrm{d}} = -\mu$.

[8] The "atoms" are caricatured here as single-level ones, but more realistic aspects of true atomic shells can be included such as orbital degrees of freedom and intra-atomic exchange.

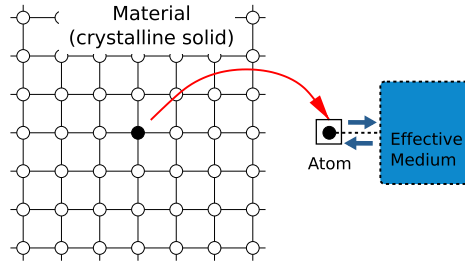[9] For additional qualitative considerations along these lines, see Ref. [47].

Fig. 7. The Dynamical Mean-Field Theory (DMFT) concept. A solid is viewed as an array of atoms exchanging electrons, rather than as a gas of interacting electrons moving in a periodic potential. DMFT replaces the solid by a single atom exchanging electrons with a self-consistent medium and takes into account local many-body correlations on each site.

terms of the self-energy, keeping the on-site term only) and to approximate the local component by that of the effective impurity: $\Sigma_{ij}(i\omega_n) \simeq \Sigma_{\mathrm{imp}}(i\omega_n)\,\delta_{ij}$. Recalling that for the impurity model, the self-energy is related to G_{imp} and $\Delta(i\omega_n)$ by the local Dyson equation $\Sigma_{\mathrm{imp}}(i\omega_n) = \mathcal{G}_0^{-1}(i\omega_n) - G_{\mathrm{imp}}^{-1}(i\omega_n) = i\omega_n + \mu - \Delta(i\omega_n) - G_{\mathrm{imp}}^{-1}(i\omega_n)$, and inserting this expression into (36), we obtain:

$$G_{\mathrm{imp}}[i\omega_n, \Delta] = \sum_{\mathbf{k}} \frac{1}{G_{\mathrm{imp}}[i\omega_n; \Delta]^{-1} + \Delta(i\omega_n) - \varepsilon_{\mathbf{k}}} \tag{38}$$

This is a self-consistent equation for the hybridisation function $\Delta(i\omega_n)$. The geometry of the lattice and dispersion of the band enters this equation[10] through $\varepsilon_{\mathbf{k}}$. A more explicit form of this equation can be given if we specialize the density of states associated with $\varepsilon_{\mathbf{k}}$ to a semi-circular one with half-width D (corresponding to an infinite-connectivity Bethe lattice), in which case (38) reads:

$$\Delta(i\omega_n) = \frac{D^2}{4}\, G_{\mathrm{imp}}(i\omega_n) \tag{39}$$

The prescription is to find Δ in such a way that the d-level Green's function associated with the impurity model, specified by $\Delta(i\omega_n)$, U and μ (cf. Eq. (14)) also satisfies (38) and can hence be identified with the local Green's function of our solid. Once this is done, the self-energy (approximated by its local component) is obtained as $\Sigma(i\omega_n) = i\omega_n + \mu - \Delta(i\omega_n) - G_{\mathrm{imp}}^{-1}(i\omega_n)$, and many other physical quantities and response functions can be calculated. In practice this is done by following an iterative scheme, which requires repeatedly solving the impurity model in conjunction with (38) until convergence is reached. For this purpose, efficient algorithms must be used to calculate the impurity Green's function, self-energy and possibly two-particle response functions of the embedded atomic shell ('impurity solvers'). Remarkable progress on this front, especially for atomic shell with orbital degeneracies or cluster extensions of DMFT, has been achieved in the past few years, thanks to continuous time quantum Monte-Carlo techniques (see [50] for a review). Several software libraries have been developed and made available on the web [51,52]. For small orbital degeneracy, accurate real-frequency algorithms such as the numerical renormalisation group can also be used.

In a nutshell, the DMFT construction involves the solution of a self-consistent 'impurity' problem: an atomic shell embedded in a self-consistent medium with which it exchanges electrons [48,49].

5.3. When is DMFT accurate?

The single-site DMFT construction becomes exact in the following limits.

- In the atomic limit $t_{ij} = 0$, by construction (then, $\Delta = 0$).
- In the non-interacting limit $U = 0$. Indeed, in this case the self-energy $\Sigma = 0$, so that it is trivially **k**-independent.
- Hence, both the limit of a non-interacting band and that of isolated atoms are correctly reproduced by DMFT, which provides an interpolating scheme between these extreme cases.
- In the limit of infinite lattice coordination (infinite number of spatial dimensions), first introduced for fermions in the pioneering work of Metzner and Vollhardt [53]. The hopping must be scaled as $t_{ij} = t/\sqrt{d}$ for this limit to be properly defined and non-trivial.
- Being an exact solution of Hubbard-like models in the limit of infinite dimensions, it is thus guaranteed that DMFT preserves all sum-rules and conservation laws.

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[10] The DMFT equations have been written here in the paramagnetic phase with no symmetry breaking whatsoever. These equations are easily generalized to phases with long-range order [49], such as an antiferromagnet or an s-wave superconductor. In those phases, and for two-particle response functions as well, the DMFT equations depend on additional details of the lattice structure.

Besides these formal considerations, it is important to emphasize when single-site DMFT is accurate and physically mean-ingful. Obviously, this is the case when inter-site correlations do not strongly affect single-particle properties. This is true when the correlation lengths for any kind of incipient ordering are small, i.e. sufficiently far away from critical boundaries. The local approximation (single-site DMFT) is a good starting point when spatial correlations are short-range, which is the case in any of the following regimes: high-temperature, high-energy, high doping, large number of fluctuating degrees of freedom competing with each other, large orbital degeneracy, large degree of frustration. For further considerations along these lines, especially in relation to the high-temperature regime, see [54].

5.4. The Mott transition: from waves to particles

One of the early successes of DMFT is to provide a detailed and consistent theory of the Mott transition. A schematic 'generic' phase diagram of the half-filled Hubbard model as obtained in this approach is depicted in Fig. 8. The corresponding evolution of the local spectral function $A_{loc}(\omega)$ as the coupling is increased is also schematically depicted in Fig. 9. In the Mott insulating phase, we see that the excitations consist of two broadened versions of the atomic transitions $|\sigma\rangle \rightarrow |0\rangle$ (around energy $\varepsilon_d = -\mu$) and $|\sigma\rangle \rightarrow |\uparrow\downarrow\rangle$ (around energy $U - \mu$), colloquially called (somewhat improperly) the lower and upper Hubbard 'bands'. In the strongly correlated metal, besides these high-energy excitations we also have low-energy quasiparticle excitations. Their density of states has a spectral weight and bandwidth reduced by a factor $Z < 1$ (from Eq. (17)) in comparison to the non-interacting system. This corresponds to Brinkman–Rice physics [55]: enhancement of the quasiparticle effective mass $m^*/m \sim 1/Z$ and reduction of the quasiparticle kinetic energy. At the Mott transition, Z vanishes and m^*/m diverges in the single-site DMFT (as observed, e.g., in $La_{1-x}Sr_xTiO_3$ [56]).

This illustrates the point made above: in strongly-correlated materials, electrons are 'hesitant' entities with a dual charac-ter. At high-energy they behave as localized, and the relevant excitations are atomic (particle-like) excitations. At low-energy in metallic compounds they eventually form wave-like itinerant quasiparticles.

The key features which allow DMFT to handle both types of excitations are that (i) it has by construction a correct description of atomic eigenstates and that (ii) the hybridisation function $\Gamma(\omega) = -\text{Im}\Delta(\omega + i0^+)$ is energy dependent, and changes in a self-consistent manner as the coupling, electron density and temperature[11] are varied. This hybridisation function, or dynamical mean-field, can be viewed as the quantum generalisation of Pierre Weiss mean field in the context of classical systems.

In the Mott insulating phase, $\Gamma(\omega)$ has a gap, identical to that of $A_{loc}(\omega)$, as clear, e.g., from (39). The shape of $\Gamma(\omega)$ for $\omega \sim -\mu$ and $\omega \sim U - \mu$ determine the broadening and lineshape of the lower and upper Hubbard bands (cf. broadening of a VBS). At low-energy however, $\Gamma(\omega)$ has no spectral weight: the Kondo effect cannot take place and the degeneracy of the spin-1/2 doublet is not lifted. Hence, the Mott insulator is a paramagnet with fluctuating local moments.

In contrast, in the metal, $\Gamma(\omega)$ has low-energy spectral weight and the Kondo effect screens out this local moment. The excitations are quasiparticles described in the single-site DMFT by a local Fermi-liquid theory (17). It is important to realize however that this is a self-consistent Kondo effect, in which $\Gamma(\omega)$ is constrained to have a similar shape than $A_{loc}(\omega)$. In particular, the widths of these functions are both of order ZD (the Brinkman–Rice scale). The kind of Kondo problem we get close to the Brinkman–Rice transition is thus in a very different regime (intermediate coupling) than the weak-coupling Kondo problem of a physical magnetic impurity in a very broad band discussed in the previous sections [57,58]. Hence, Z no longer depends exponentially on U has above, but actually vanishes as $U_c - U$. Furthermore, in this situation, the true Kondo scale cannot be identified with ZD: it is actually a smaller scale [59] (not parametrically, but the renormalisation factor is small). This explains why the Fermi-liquid scale T_{FL} (below which, e.g., T^2 resistivity is observed) is actually much smaller than the Brinkman–Rice scale (at which quasiparticles excitations are fully incoherent and the Mott–Ioffe–Regel value is reached [60]).

Hence, in a strongly correlated system, the formation of quasiparticles as temperature or energy are reduced, and most importantly the scale below which they form, depend on how the local atomic multiplets are screened in the solid-state environment. This is why starting from a proper description of the atomic physics and describing how this gradual screening process takes place is essential. Remarkably, it recently became apparent that this is not only true for metals very close to the Mott transition, but also for quite itinerant metals which are far from this transition, in which the physics is dominated by effects due to the Hund's coupling, such as iron-based superconductors and transition-metal oxides of the 4d series like ruthenates (for a review, see, e.g., [61]). In a nutshell: atoms matter.

6. Conclusion

In this note, I have tried to illustrate how the physics of impurities and the concepts introduced by Friedel have proven fertile in other contexts. In his scientific work, Friedel harmoniously combined the two ways of looking at a solid emphasized above: the atomic picture and the electron-gas picture. He mentioned this, for example, in his 2001 interview [2]: "On reconnaît là la dichotomie atomes-électrons qu'il est d'ailleurs difficile de maîtriser dans un domaine aussi étendu." I hope to have convinced the reader that the concept of an effective impurity or self-consistently embedded atom, which is at the

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[11] The change as a function of temperature is crucial for example to describe the transfers of spectral weights observed in strongly correlated materials.
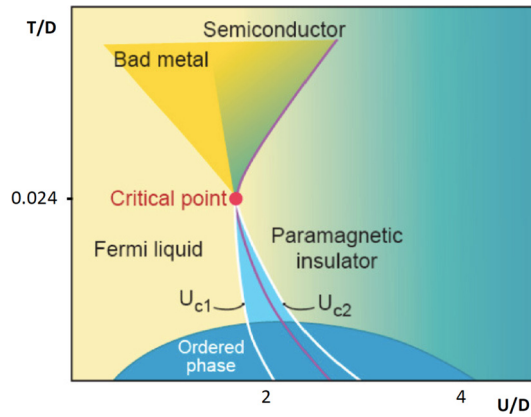
Fig. 8. Generic (schematic) phase diagram of the half-filled Hubbard model, obtained from DMFT, as a function of interaction strength and temperature normalized to the half-bandwidth *D*. For a lattice with frustration (e.g., with next-nearest neighbour hopping), the transition temperature into phases with long-range spin ordering is reduced. Then, a first-order transition from a metal to a paramagnetic Mott insulator becomes apparent. Adapted from Ref. [62].
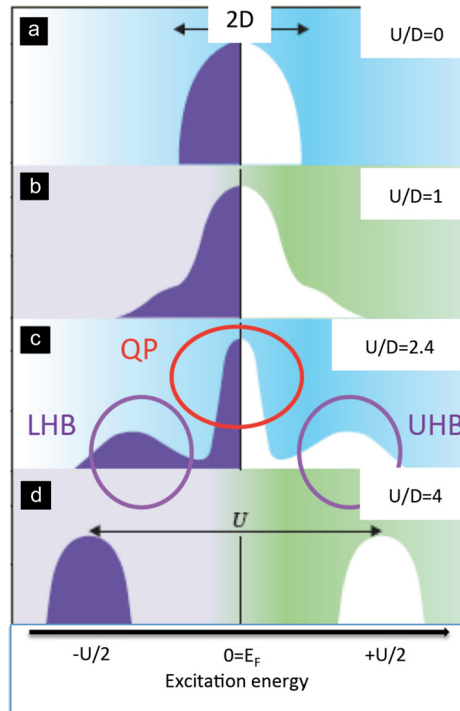

Fig. 9. Schematic evolution of the momentum-integrated spectral function (total density of states) as the coupling is increased, for the half-filled Hubbard model in its paramagnetic phase, according to single-site DMFT. The low-energy (quasiparticle) part of the spectrum gradually narrows down, while the corresponding spectral weight is transferred to the lower and upper Hubbard bands (atomic-like excitations). Adapted from Ref. [63].

heart of the dynamical mean-field theory of materials with strong electronic correlations, can be viewed as an attempt to reconcile and combine these points of view.

Acknowledgements

Appendix A. Conduction electrons T-matrix and phase-shift

A direct examination of perturbation theory, or the functional integral technique, leads to the following expression for the conduction electron Green's function (which depends on two momenta, since the system is not translationally invariant!):

$$G^c_{\mathbf{pp'}} = G^{c0}_{\mathbf{p}} \delta_{\mathbf{pp'}} + G^{c0}_{\mathbf{p}} T_{\mathbf{pp'}}(\omega) G^{c0}_{\mathbf{p'}} \tag{40}$$

in which the scattering T-matrix reads:

$$T_{\mathbf{pp'}}(\omega) = V^*_{\mathbf{p}} G_d(\omega) V_{\mathbf{p'}} \tag{41}$$

Defining the phase-shift in the standard manner:

$$T_{\mathbf{pp'}}(\omega) = -|T_{\mathbf{pp'}}(\omega)| e^{i\delta_{\mathbf{pp'}}(\omega)} \tag{42}$$

we obtain (considering for simplicity, real $V_{\mathbf{p}}$'s):

$$\tan\delta(\omega) = \frac{\mathrm{Im}G_d(\omega+i0^+)}{\mathrm{Re}G_d(\omega+i0^+)} = \frac{\Gamma - \Sigma''_d(\omega)}{\varepsilon_d + \Sigma'_d(\omega) - \omega} \tag{43}$$

Using the Fermi-liquid expansion of Σ_d, we thus obtain the $T=0$ expression of the phase-shift at the Fermi surface as:

$$\tan\delta(0) = \frac{\Gamma}{\varepsilon_d + \Sigma'_d(0)} \tag{44}$$

Hence leading to the expression of the d-level spectral function at the Fermi level:

$$A_d(0) = \frac{1}{\pi} \frac{\Gamma}{\left[\varepsilon_d + \Sigma'_d(0)\right]^2 + \Gamma^2} = \frac{1}{\pi\Gamma} \sin^2\delta \tag{45}$$

and, by summing the above expression for G_c over momenta, to the local conduction electron density of states at the Fermi level:

$$A_c(0) = \rho^0_c \cos^2\delta \tag{46}$$

Appendix B. Langreth's proof of Friedel's sum rule for the Anderson impurity model

Langreth's proof [28] is non-perturbative, applies at $T=0$, and uses the formalism of Luttinger and Ward. It is in the same spirit than the derivation of Luttinger's theorem based on this formalism. See also the earlier work of Langer and Ambegaokar [64] devoted to the Friedel sum-rule in an interacting system.

The case of an infinite bandwidth is considered. This is important, actually: for a finite bandwidth and a hybridization function $\Delta(\omega+i0^+)$ having a non-vanishing real part, additional terms are involved and the displaced (screening) charge no longer coincides with the occupancy of the impurity level.

Let us consider the retarded impurity's Green's function: $G^{-1}_d(\omega+i0^+) = \omega - \varepsilon_d + i\Gamma - \Sigma_d(\omega+i0^+)$ and take the frequency derivative of its logarithm:

$$-\frac{\partial}{\partial\omega} \ln G_d(\omega) = \left[1 - \frac{\partial\Sigma_d}{\partial\omega}\right] G_d(\omega) \tag{47}$$

Noting that:

$$-\frac{1}{\pi} \int_{-\infty}^{0} d\omega\, \mathrm{Im}\, G_d = \frac{n_d}{2} \tag{48}$$

we integrate the above relation to obtain:

$$\frac{\pi n_d}{2} = \mathrm{Im}\ln G_d(\omega=0) - \mathrm{Im}\ln G_d(\omega\to-\infty) + \mathrm{Im}\int d\omega\, \frac{\partial\Sigma_d}{\partial\omega} G_d(\omega) \tag{49}$$

The last term will be shown to vanish, so that we get (we use $\mathrm{Im}G_d < 0$, the above definition of the phase shift, and a branch-cut of the ln on the negative real axis):

$$\frac{\pi n_d}{2} = (\delta - \pi) - (-\pi) = \delta \tag{50}$$

To prove that the last term vanishes, we integrate it by part:

$$\int d\omega \, \frac{\partial G_d}{\partial \omega} \, \Sigma_d(\omega) \tag{51}$$

and observe that the self-energy is obtained from the Luttinger–Ward functional as:

$$\Sigma_d(\omega) = \frac{\delta \Phi[G_d]}{\delta G_d(\omega)} \tag{52}$$

so that the above reads:

$$\int d\omega \, \frac{\partial G_d}{\partial \omega} \, \frac{\delta \Phi[G_d]}{\delta G_d(\omega)} = \delta \Phi \tag{53}$$

This is the change of the Luttinger–Ward functional when all frequencies are shifted. Being a scalar functional with no external frequency, and leaving aside the (exotic but documented in other systems [65]) possibility of an anomaly, we conclude that $\delta \Phi = 0$.

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