Characteristic time scales of the local moment dynamics in Hund’s-metals

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We study the characteristic time scales of the fluctuating local moments in Hund’s metal systems for different degrees of correlation. By analyzing the dynamical spin susceptibility in the real-time domain via the fluctuation-dissipation theorem, we determine the time scales controlling oscillation and damping of on-site fluctuations - a crucial factor for the detection of local moments with different experimental probes. We apply this procedure to realistic many-body calculations of different families of iron-pnictides and chalcogenides, explaining the material-specific trend in the discrepancies reported between experimental and theoretical estimates of the magnetic moments.

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Introduction – Our perception of the natural world is significantly shaped by the properties of the detection process considered. One crucial aspect is the time scale of the probing mechanism: If this is larger than the typical time scale of the phenomenon under investigation, only averaged information will be gained. This general statement applies to a very broad class of detectors, ranging, e.g. from the vision process in our eyes to the case of interest for this work: the measurement of magnetic properties in correlated materials.

In particular, we will focus on the detection of the local magnetic moments in correlated metallic systems. Their proper description is indeed a key to understanding many-electron systems beyond the conventional band-theory framework. The formation of sizable local moments and their screening-processes is central to: The Kondo physics\cite{1, 2}, Mott-Hubbard\cite{3-5} or Mott-Hund’s\cite{6-10} metal-insulator transitions, quantum-criticality of heavy fermion systems\cite{11, 12}, magnetic and spectroscopic properties of Ni and Fe\cite{13-15} and of unconventional superconductors\cite{16}.

Reflecting the high physical interest, several experimental procedures are used to detect the local magnetic moments and their manifestations\cite{17}: measurements of static susceptibilities (e.g. in the presence of a Curie-Weiss behavior)\cite{13, 17}, inelastic neutron spectroscopy (INS)\cite{18}, by integrating over the Brillouin zone\cite{19}, X-ray absorption spectroscopy (XAS), etc.

Whether it is possible to obtain an accurate description of the local moments strongly depends on the relation between the intrinsic time scales of the different experimental probes and those characterizing the dynamical screening mechanisms at work. The emerging picture is typically clear-cut in the cases where the screening processes are strongly suppressed, such as in Mott or Hund’s-Mott insulating phases: Here a coherent description of the magnetic moment properties (including its size) can be easily obtained in all experimental setups. On the opposite side, a much more complex, multifaceted situation characterizes systems where well preformed magnetic moments present a rich dynamic. Good examples are the strongly correlated metallic regimes adjacent to a Mott metal-insulator transition, or even better, compounds displaying a Hund’s metal behavior\cite{6, 20}, such as iron pnictides and chalcogenides\cite{21}.

An intuitive picture – In order to provide a direct interpretation for the physics of our realistic material calculations, we start from some heuristic considerations on the dynamics of the local magnetic moment $\vec{\mu} = g \mu_B \vec{S}$ in a correlated metal. The relevant information is encoded in the time-dependence of its correlation function:

\[ F(t) \equiv \frac{1}{2} g^2 \frac{\mu_B^2}{\hbar^2} \langle \{ \hat{S}_z(t), \hat{S}_z(0) \} \rangle \]

(1)

where $g = 2$ is the Landé factor, $\mu_B$ the Bohr magneton and $\hat{S}_z = \sum_i \hat{s}_z^i$ the $z$-component of the total spin hosted by the correlated atom (e.g. a transition metal element), built up by the unpaired electronic spins $s_z$ of its partially filled $d$ or $f$ shells\cite{17}. The function $F(t)$ describes both the static (thermal) and dynamic (Kubo) part of the response\cite{22}, which is needed for our study. In general, one expects the maximum values $F(t)$ at $t = 0$: This describes the instantaneous spin configuration of the system, which is often quite large in a multi-orbital open shell due to the Hund’s rule. As a result of the electronic fluctuations, the amplitude probability to find a magnetic moment of the same size and with the same orientation will be decreasing with increasing time. At a first approximation, one can identify two distinct patterns for this process: (i) a gradual rotation (with constant amplitude) and (ii) a progressive reduction of the size of the local moment. Within this simple picture, these effects naturally define two characteristic time-(and energy) scales for the local moment dynamics: (i) the period of the rotation ($t_{\omega} \propto \frac{1}{B}$) and (ii) the characteristic time ($t_{\gamma} \propto \frac{1}{\hbar}$) for the amplitude damping.

The impact of the electronic mobility on the local moment dynamics will reflect the degree of metallicity of the different materials and its interplay with the stabilization...
of high-spin states driven, e.g. by the Hund’s exchange $J$. Hence, the values of these characteristic scales may vary considerably from one material to another, with overall larger values associated to a suppressed electronic mobility. In the extreme case of a Mott insulator, one expects to observe long-living magnetic moments, consistent with the analytic divergence of the time scales found in the fully localized (atomic) limit ($t_ω, t_γ \to \infty$). On the opposite side, in a conventional (weakly correlated) metal both scales will be extremely short, roughly of the order of the inverse of the bandwidth $W$ of the conducting electrons ($t_ω \sim t_γ \propto 1/\hbar \nu$). The most interesting situation is realized in a correlated metallic context. Here, the slowing down of the electronic motion, induced by the electronic scattering, increases the values of both time scales that remain nonetheless finite. The enhancement will depend on specific aspects of the many-electron problem considered, possibly affecting the two time scales in a different fashion: This leads to the distinct regimes of underdamped ($t_γ \gg t_ω$) and overdamped ($t_γ \ll t_ω$) local moment fluctuations, schematically depicted in Fig. 1. As we will discuss below, the actual hierarchy of the time scales will strongly impact the outcome of different spectroscopic experiments. Further, quantitative information about the dynamics of the magnetic fluctuations in systems at equilibrium may also provide important information for the applicability of the adiabatic spin dynamics\cite{23,24} and, on a broader perspective, crucial insights for the highly non-trivial interpretation of the out-of-equilibrium spectroscopies.

Quantification of time scales – The procedure to quantitatively estimate the characteristic time scales from many-electron calculations and/or experimental measurements relies on the Kubo-Nakano formalism for linear response. In this respect, we recall that the dynamical susceptibility is defined as

$$\chi(\tau) \equiv \langle T_\tau \hat{S}_z(\tau)\hat{S}_z(0)\rangle$$ \hspace{1cm} (2)

in imaginary Matsubara time (where $T_\tau$ is the imaginary time-ordering operator). The corresponding (retarded) spectral functions $\chi^R(\omega)$ are obtained analytic continuation of Eq. (2). The absorption component of the spectra, $\text{Im}\chi^R(\omega)$, which is directly measurable (e.g. in INS), provides a simple means to precisely estimate time scales. In particular, simple analytic expressions, directly derived for damped harmonic oscillators, can be exploited for fitting the (one or more) predominant absorption peak(s) of $\text{Im}\chi^R(\omega)$. In the illustrative case discussed above, one has

$$\text{Im}\chi^R(\omega) = A \frac{2\gamma\omega}{(\omega_0^2 - \omega^2)^2 + 4\omega^2\gamma^2},$$ \hspace{1cm} (3)

where $\gamma$ and $\omega_0$ are the scales associated to the major absorption processes active in the system under consideration, and the constant $A$ reflects the size of the instantaneous magnetic moment. The expression is clearly generalizable to all cases, where more absorption peaks are visible in the spectra, as a sum of the corresponding contributions. See supplemental material for a more detailed discussion of Eq. (3).

The full time-dependence of the fluctuating local moment, which will be mostly shaped by the interplay of the time scales defined above, is eventually obtained via the fluctuation-dissipation theorem:

$$\mathcal{F}(t) = \frac{1}{T_0} \int_0^\infty d\omega \cos(\omega t) \coth(\beta/2\omega) \text{Im}\chi^R(\omega),$$ \hspace{1cm} (4)

where $\beta = \frac{1}{k_B T}$ is related to the inverse temperature by the Boltzmann constant $k_B$.

The case of the Hund’s-metals – While the procedure illustrated above is applicable to all the spectroscopy experiments of all condensed matter systems, we will demonstrate its advantages by applying it to Hund’s metals\cite{6,20}, where the dynamics of fluctuating moments is of particular interest\cite{26}. These systems can be viewed as a new “crossover”-state of matter, emerging from the competition of the local Hubbard repulsion ($U$) and Hund’s rule coupling ($J$), when the corresponding atomic shell is (about) one electron away from a half-filled multi-orbital configuration. In fact, increasing $U$ suppresses the double occupancy in the correlated orbitals, driving the system towards a Mott insulating state, while an increase in $J$ favors the onset of local high-spin configurations, possibly inducing charge disproportionation\cite{9}. Out of half-filling, the competition between these two tendencies stabilizes a metallic ground state also in the presence of high values of the electronic interaction\cite{6,9,27}. The emerging physics of a large local magnetic moment fluctuating in a strongly correlated metallic surrounding evidently represents one of the best playgrounds to apply our time-resolved procedure.

The prototypical class of materials displaying Hund’s metal physics is represented by the iron pnictides or chalcogenides. These compounds, which often display unconventional superconducting phases upon doping, are also characterized by interesting magnetic properties\cite{19,21,28}. Both, the ordered magnetic moments (measured by neutron diffraction in the magnetically ordered phase) and the fluctuating moments (measured by
INS in the paramagnetic high-$T$ phase) are reported to be systematically lower\cite{29} in experiment than in (static) LSDA calculations (predicting a large ordered moment of about $2\mu_B$ for almost all compounds of this class). It was also noted that, surprisingly, the larger discrepancies are found for the “less correlated” families 1111 (e.g. LaFeAsO) and 122 (e.g. BaAs$_2$O$_3$), which display milder quasiparticle renormalization effects and are characterized by lower values of the screened Coulomb interaction estimated in cRPA\cite{30}. Significantly smaller (or almost no) deviations are reported, instead, for the most correlated families such as the 11 subclass (e.g. FeTe), where relatively large local moments are found both in neutron experiments and theory. Previous dynamical mean-field theory (DMFT) studies of the inelastic neutron spectra and correlation function in real time (third row), computed for different families of iron- pnictides/chalcogenides in the DFT+DMFT (third column), compared with the corresponding results of the bare (first column) and the DMFT (second column) bubble calculations.

We report here on our \textit{ab-initio} + DMFT calculations – We report here on our density functional theory (DFT) + DMFT calculations\cite{35,36} of the local spin susceptibilities in the iron pnictides/chalcogenides. Different from preceding works, we computed the spin-spin response functions on equal footing for several different compounds, chosen as representative of the most relevant families (1111, 122, 111, 11). As a step forward in the theoretical description, we put emphasis on a quantitative time-resolved analysis of the results, eventually allowing for a precise interpretation of the physics at play and of the spectroscopic results.

For our DMFT calculations\cite{37,38}, we considered the following low-energy Hamiltonian obtained via a projection on the Fe-3$d$ (maximally localized) Wannier-orbital manifold:

\[ H = \sum_{k \alpha \mu} H_{\alpha \mu} (k) \, \tilde{c}_{k \alpha \mu}^{\dagger} \, c_{k \alpha \mu} + \sum_{\mathbf{r} \sigma \sigma'} \sum_{l m n} U_{l m n} \, \tilde{c}_{\mathbf{r} \sigma \alpha}^{\dagger} \, c_{\mathbf{r} \sigma' \alpha'} \, c_{l m n} \, \tilde{c}_{\mathbf{r} \sigma' \alpha'}^{\dagger}, \]  

where $l, m, n, \sigma$ are orbital indices, $\mathbf{k}$ denotes the fermionic momentum, $\mathbf{r}$ the lattice site and $\sigma, \sigma'$ the spin,
and $U_{\text{Im}}$ is the (static) screened full-Coulomb local interaction estimated in cRPA\cite{37}.

Our DMFT results are summarized in Fig. 2, where, we show the dynamical spin susceptibility on the Fe atoms of all compounds considered in its different representations: imaginary time in the first panels [cf. Eq. (2), direct output of the QMC solver], real-frequency in the second row [from analytic continuations], real-time in the third row [Eq. (4), via Eq. (1)]. In all cases, we performed our analysis not only for the full DMFT calculation (third column panels), but also for the corresponding "bubble" terms (i.e., $\chi_0 = -\beta GG$) either computed with the non-interacting Green’s function ($G = G_0$, first column) or with the DMFT one ($G = G_{\text{DMFT}}$, second column). This allows to disentangle the roles played by band-structure, self-energy and vertex-corrections in controlling the dynamics of the fluctuating moments.

A quick glance at $\chi(\tau)$ already illustrates an important finding of our work: The different bandstructure of the materials does not generate by itself any distinguishable effects in the local moment dynamics (first two columns in Fig. 2). Instead, the definite material dependence observed is almost totally originated by vertex corrections (third column).

One can understand the overall trend as follows: Instantaneous ($\tau = 0$) magnetic moments of similar (and large) sizes but subjected to quite different screening effects ($\tau \rightarrow \frac{\tau}{\hbar}$). However, only the corresponding analysis of $\text{Im} \chi_R(\omega)$ and $\mathcal{F}(t)$ allows to extract clear-cut physical information. By looking at the data for $\mathcal{F}(t)$, we easily note that the moment dynamics described by the "bubble terms" (with/without $\Sigma_{\text{DMFT}}$) is controlled by very short time scales for oscillation and damping ($\sim 0.5$ fs), roughly corresponding to $\sim \hbar / \Omega$.

The scenario emerging from the visual inspection of $\mathcal{F}(t)$ is supported, at a more quantitative level, by the fit of the main absorption peaks of $\text{Im} \chi_R(\omega)$, as we get, e.g., $t_{\omega} \approx 5$ fs and $t_{\gamma} = 2$–3 fs for LaFeAsO and about 10 times larger values for FeTe (see Tab. I for details).

**Spectroscopic measurements** – The considerably different values of the time scales, determined above, will evidently affect the detectability of the local magnetic moments ($m_{\text{loc}}$) in the iron-pnictides. While fast probes (e.g. XAS) are able to detect the high-spin instantaneous configuration of these Hund’s metal, the characteristic time scale of the INS ($t_{\text{INS}} \simeq 5$–10 fs $\simeq \hbar / 100$ meV) are of the same order as those in Tab. I: time-averaging effects will, thus, lead to underestimate the local magnetic moment:

$$m_{\text{loc}}^2 = \frac{3}{4} \lim_{\Omega \rightarrow \infty} \int_{\Omega} \int_{\Delta \Omega} \text{Im} \chi_R^{\hbar}(\omega,\bar{\omega}) b(\omega) d\bar{\omega} d\omega$$

where $b(\omega) = 1/(e^{\hbar \omega} - 1)$ it the Bose-Einstein distribution function. This is especially relevant for the “less correlated” compounds (LaFeAsO and BaFe$_2$As$_2$), where $t_{\gamma}$, $t_{\omega} < t_{\text{INS}}$. In families with higher degrees of (e.g. for FeTe, where $t_{\gamma}$, $t_{\omega} > t_{\text{INS}}$) the averaging effect gets “mitigated”, allowing the detection of larger magnetic moment sizes, consistent with fast probe XAS experiments. The material dependence of local moment dynamics is directly mirrored in the progressive red-shift of the first-absorption peak in $\text{Im} \chi_R(\omega)$, as shown in Fig. 3. Here, one can appreciate how an increasing part of the spin absorption-spectra gradually enters the typical experimentally accessible window of the INS (main panel). This explains the progressively reduced discrep-

| $\omega_0$ [eV] | $\gamma$ [eV] | $t_{\gamma}$ [fs] | $t_{\omega}$ [fs] |
|---------------|--------------|-----------------|-----------------|
| LaFeAsO       | 0.3406  | 0.2787 | 2.4  | 3.4  |
| BaFe$_2$As$_2$ | 0.2733 | 0.2743 | 2.6  | -    |
| LiFeAs        | 0.2607  | 0.5191 | 9.4  | -    |
| KFe$_2$As$_2$  | 0.3369  | 1.0657 | 12.0 | -    |
| FeTe          | 0.0346  | 0.0273 | 24.1 | 30.9 |

TABLE I. Fitting parameters $\omega_0$ and $\gamma$ of the absorption peak(s) computed in DMFT with Eq. (3) (second and third column, where the largest energy scale is marked in bold); effective lifetime $\chi(t \rightarrow \infty) \propto e^{-t/t_{\gamma}}$ (third column) and effective oscillation period $t_{\omega} = \hbar / \sqrt{\omega_0^2 - \gamma^2}$ (fourth column) for the different material considered, see [37] for further details.
ancies in the size of the magnetic moment (see inset) observed in the more correlated families of the iron pnictides/chalcogenides.

Conclusions – We illustrated how to quantitatively investigate, in the real time domain, the dynamics of the local magnetic moments of correlated systems, and how to physically interpret the obtained results in terms of their characteristic time scales. Our procedure, exploiting the fluctuation-dissipation theorem, is directly applied to clarify the results of INS experiments in several families of iron-pnictides and chalcogenides. In particular, the different degrees of discrepancies w.r.t. the standard ab-initio calculations is rigorously explained by comparing the time scales of the fluctuating moments to the characteristic time scale of the INS probe. Remarkably, the strong differentiation between the time scales of the materials considered, crucial for a correct understanding of the underlying physics, is almost entirely due to vertex corrections.

While the dynamics of the magnetic moments is particularly intriguing in the Hund’s metal materials considered here, the same procedure is directly applicable to all many-electron systems and to fluctuations of different kinds[2]. A precise quantification of the characteristic time scales may provide new keys to connect the findings of equilibrium and out-of-equilibrium spectroscopies, as well as crucial information on the applicability of adiabatic spin dynamics approaches[25].

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