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DYNAMICAL AND STATIC SUSCEPTIBILITY OF ANOMALOUS RARE-EARTH SYSTEMS

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Dynamical and static susceptibility of anomalous rare-earth systems are computed within the atomic limit of the periodic Anderson Hamiltonian in both Kondo and intermediate valence regimes. The model can give an explanation of the inelastic peaks observed by neutron scattering measurements in $CeSn_3$ or $CePd_3$.

Anomalous rare-earth systems have been extensively studied in recent years by neutron scattering measurements. A broad quasi-elastic line is observed in all these systems and the linewidth increases with increasing temperature [1,2]. On the other hand inelastic peaks have been observed in some cerium compounds. In the case of cerium Kondo compounds, these inelastic peaks originate from crystal field effects within the $4f^1$ configuration of cerium [3] and they have been previously accounted for by a model based on the effective exchange Hamiltonian describing both Kondo and crystalline field effects [4]. On the contrary, broad inelastic humps have been observed in intermediate valence (IV) cerium compounds $CeSn_3$ [5] or $CePd_3$ [6,7] at, respectively, 40 or 60 meV and the origin of these peaks is attributed to a valence transition, as proposed by several authors [8–10].

In order to study preferentially the inelastic peaks of the dynamical susceptibility, we utilise here a localised description of valence fluctuations previously developed [11] which exhibits clearly the different effects of hybridization and Coulomb repulsion, and we compute the static and dynamical susceptibilities.

The Hamiltonian of the system is:

$$H = E\sum_\sigma f_\sigma^+ f_\sigma + U_{n_{f\uparrow} n_{f\downarrow}} + E_B\sum_\sigma c_\sigma^+ c_\sigma + V\sum_\sigma (c_\sigma^+ f_\sigma + \text{h.c.}) \quad (1)$$

where E (E_B) is the energy of the f- (c-)level, V the hybridization between f- and conduction elec-

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Table 1
The eigenvalves and eigenfunctions of H, with $\tan\phi = V/\{\frac{1}{2}(E_B - E) + [(\frac{1}{2}(E_B - E))^2 + V^2]^{1/2}\} = V/(-\alpha + \delta)$ and $\tan\lambda = 2^{1/2}V/\{\frac{1}{2}(E_B - E) + [(\frac{1}{2}(E_B - E))^2 + 2V^2]^{1/2}\} = 2^{1/2}V/(-\alpha + \gamma)$

Eigenstate	Energy	Number of electrons
$\|1\rangle = \|0\rangle$	0	0
$\|2\rangle = \cos\phi\, f_\downarrow^+ \|0\rangle - \sin\phi\, c_\downarrow^+ \|0\rangle$	$\frac{1}{2}(E_B + E) - [(\frac{1}{2}(E_B - E))^2 + V^2]^{1/2}$	1
$\|3\rangle = \cos\phi\, f_\uparrow^+ \|0\rangle - \sin\phi\, c_\uparrow^+ \|0\rangle$	$\frac{1}{2}(E_B + E) - [(\frac{1}{2}(E_B - E))^2 + V^2]^{1/2}$	1
$\|4\rangle = \sin\phi\, f_\downarrow^+ \|0\rangle + \cos\phi\, c_\downarrow^+ \|0\rangle$	$\frac{1}{2}(E_B + E) + [(\frac{1}{2}E_B - E))^2 + V^2]^{1/2}$	1
$\|5\rangle = \sin\phi\, f_\uparrow^+ \|0\rangle + \cos\phi\, c_\uparrow^+ \|0\rangle$	$\frac{1}{2}(E_B + E) + [(\frac{1}{2}(E_B - E))^2 + V^2]^{1/2}$	1
$\|6\rangle = c_\downarrow^+ f_\downarrow^+ \|0\rangle$	$E_B + E$	2
$\|7\rangle = (1/2^{1/2})(c_\downarrow^+ f_\uparrow^+ + c_\uparrow^+ f_\downarrow^+)\|0\rangle$	$E_B + E$	2
$\|8\rangle = c_\uparrow^+ f_\uparrow^+ \|0\rangle$	$E_B + E$	2
$\|9\rangle = [(\cos\lambda/2^{1/2})(c_\downarrow^+ f_\uparrow^+ - c_\uparrow^+ f_\downarrow^+) - \sin\lambda c_\downarrow^+ c_\uparrow^+]\|0\rangle$	$\frac{1}{2}(E + 3E_B) - [(\frac{1}{2}(E_B - E))^2 + 2V^2]^{1/2}$	2
$\|10\rangle = [(\sin\lambda/2^{1/2})(c_\downarrow^+ f_\uparrow^+ - c_\uparrow^+ f_\downarrow^+) + \cos\lambda c_\downarrow^+ c_\uparrow^+]\|0\rangle$	$\frac{1}{2}(E + 3E_B) + [(\frac{1}{2}(E_B - E))^2 + 2V^2]^{1/2}$	2
$\|11\rangle = c_\downarrow^+ c_\uparrow^+ f_\downarrow^+ \|0\rangle$	$E + 2E_B$	3
$\|12\rangle = c_\downarrow^+ c_\uparrow^+ f_\uparrow^+ \|0\rangle$	$E + 2E_B$	3

trons, and U the Coulomb repulsion between f-electrons.

We study the case of an aggregate of independent ions, i.e. there is no band terms; and the limit $U \to \infty$ is considered.

The eigenfunctions and eigenvalves of the Hamiltonian are reproduced in table 1 [11].

The f-susceptibility is calculated from the correlation function:

$$\chi^{ff}_{+-}(\omega) = \langle\langle S^+_{f;} S^-_f \rangle\rangle_\omega \tag{2}$$

that can be evaluated from the spectral representation of the Green functions [12], i.e.:

$$\langle\langle S^+_{f;} S^-_f \rangle\rangle = Q^{-1} \sum_{m,n} [e^{-\beta E_m} - e^{-\beta E_n}] \times \frac{|\langle m| S^+_f |n\rangle|^2}{\omega - (E_n - E_m)} \tag{3}$$

where E_n and $|n\rangle$ are the eigenstates and eigenfunctions of H, $\beta = (k_B T)^{-1}$, Q is the partition function and E_B is the zero of energy.

From eq. (3) we have computed the static and dynamical susceptibility. The spin operator S^+_f has non-zero matrix elements between states with the same number of particles.

If $E_n = E_m$ (degenerate states), the pole of Im $\chi(\omega)$ at $\omega = 0$ corresponds to an elastic transition. For $E_n \neq E_m$ one finds the inelastic peaks of the imaginary part of the susceptibility.

The ground state of the system is a Kondo singlet (state $|9\rangle$), while the excited states of two particles are a triplet ($|6\rangle$, $|7\rangle$, $|8\rangle$) and an extra singlet ($|10\rangle$).

In the Kondo limit, $|E| \gg V$, one has:

$$E_9 - E_7 \cong -2V^2/|E| \tag{4}$$

which is much less than the differences between one-particle and other two-particle states: thus one finds only one inelastic peak of the susceptibility in the Kondo limit.

In the IV case, for example with $|E| = V$, one has:

$$E_9 - E_7 = -V, \tag{5a}$$

$$E_7 - E_{10} = -2V, \tag{5b}$$

$$E_3 - E_4 = -\sqrt{5}\,V, \tag{5c}$$

the three energies are of the same order of magnitude and the three peaks are observed in the imaginary part of the dynamical susceptibility.

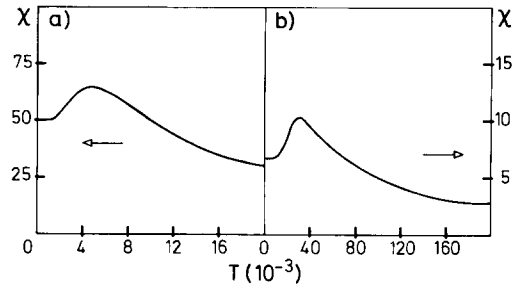

Fig. 1. Plot of the static susceptibility χ vs. temperature T in the Kondo regime (case a) with $E = -1.0$ and $V = 0.1$, and in the IV regime (case b) with $E = -0.1$ and $V = 0.1$.

We present now numerical calculations characteristic of the two physical cases and we have chosen the following parameters:

a) $E = -1.$, $V = 0.1$ for the Kondo case and

b) $E = -0.1$, $V = 0.1$ for the IV case

Energies E and V, temperature T and frequency ω are measured here in units of a typical conduction band width.

Fig. 1 shows the static susceptibility χ vs. T for the two preceding cases. χ exhibits a peak at a temperature T_m of order $2V^2/|E|$, as previously shown. When going from the Kondo limit to the IV case, T_m increases and the value of χ decreases,

Fig. 2 shows the imaginary part Im $\chi(\omega)$ of the dynamical susceptibility vs ω at different temperatures for the two preceding cases. One observes in

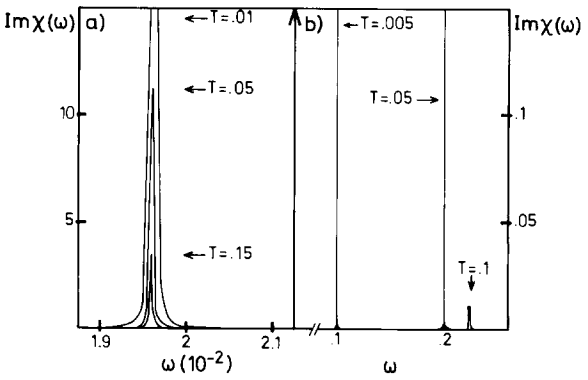

Fig. 2. Plot of the imaginary part Im $\chi(\omega)$ of the dynamical susceptibility vs. ω, in the Kondo regime(case a) and in the IV regime(case b) with the same parameters of fig. 1. Units are explained in the text.

the Kondo limit (case a) one inelastic peak at ω of the order of $2\ V^2/|E|$ corresponding to the Kondo temperature, and the intensity decreases with temperature. The situation is different in the IV case: at very low temperatures ($T = 0.005$) we observe only one peak at $\omega = V$; then, when temperature increases ($T > 0.05$, see fig. 2) a second peak occurs at $\omega = 2V$, and finally at still higher temperature ($T > 0.1$) a third peak appears at $\omega = \sqrt{5}\ V$ with a very small intensity, corresponding to a transition between one-particle states.

The inelastic peaks so obtained are extremely narrow indeed, and almost δ-like. This is certainly due to the neglect of the conduction band width; calculations taking into account finite band width as in ref. [13] are in progress. However, we can conclude that our model yields an explanation for the inelastic peaks corresponding to valence transitions in IV cerium compounds such as $CeSn_3$ or $CePd_3$.

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