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ABSTRACT SHEET

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ABSTRACT

We have used the Mori-Zwanzig memory function formalism to analyze the observed excited state EPR mode in CeP. The mixing of the Zeeman-split crystal-field excitations by the exchange, particularly among those with degenerate frequencies, yields a normal mode determining the observed low-frequency spectrum. This is illustrated by our calculation with Heisenberg exchange which yields a single peak in qualitative agreement with the experiment.

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INTRODUCTION

The cerium mononictides have NaCl structure. In the paramagnetic phase, the Ce^{3+} ($J = 5/2$) ion has a Γ_7 crystal-field ground state doublet, and an excited Γ_8 quartet. In EPR experiments,¹ at 9.2 GHz we observed a single resonance peak for polycrystalline CeP ($T_N = 10$ K) from 77 to 300 K. The intensity indicates that the mode originates from within the excited Γ_8 quartet. The width is about 300 G, increasing with temperature. Here, we apply theory based on the memory function formalism of Mori and Zwanzig²⁻⁴ to analyze the behavior of these low lying quasielastic modes, and compare the results with the experimental findings.

Because of time reversal symmetry, before introducing exchange interaction there are pairs of equal energy differences within the Zeeman-split Γ_8 quartet.⁵ When exchange is introduced, there is very significant mixing of these pairs of excitations, resulting in

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modes of quite different character from the Zeeman-split crystal-field-only modes. Notably, if the operator generating a mode almost commutes with the exchange interaction, then the corresponding 'current' term cancels out the main contribution to the damping function, and the width becomes small. (In the absence of exchange, the excitations within the Zeeman-split f_8 quartet have no width. Exchange introduces a damping width to all transitions between crystal-field states. However, for the pairs of modes that are degenerate in the absence of exchange, the mixing by exchange can largely cancel the damping for one of the resultant modes. As discussed below, it happens that this effect is most significant for H close to $\langle 111 \rangle$ alignment.)

The mixing of the modes, and the damping mechanism, depend on the exchange interaction in the system. Here, we examine two simple cases, one with the isotropic Heisenberg exchange, and one with the s-f interaction where we explicitly include the mixing of the localized and conduction electron modes. (However, this is done only to second order in the s-f interaction, so that important mixing effects for the Heisenberg case are not included.)

For the Heisenberg case, we find that the mixed mode, as described above, gives behavior close to the experimentally observed peak, having low damping width in comparison to the other modes which are essentially crystal field excitations strongly damped by the exchange interaction. Due to the crudeness of the model exchange interaction, a completely quantitative fitting to the experimentally observed g-factor and damping width has not been possible. Further study with the Coqblin-Schrieffer interaction, which has been recently applied to explain the anisotropic magnetic ordering in the cerium mononictides,⁶ will be pursued.

MORI-ZWANZIG THEORY OF THE QUASIELASTIC MODE

The single-ion Hamiltonian, composed of the fourth order crystal field term and the resonant field Zeeman energy (including the exchange contribution to the effective field), can be simply diagonalized to give the crystal field states and the corresponding energies. The operators can then be projected into this manifold. For example, with field along the $\langle 111 \rangle$ direction, the J^+ operator gives

$$J^+ = 2\sqrt{2} (\nu_2 L_{35} - \mu_1 L_{46}) + \sqrt{5} (\nu_1 L_{43} + \mu_2 L_{65}) - 3 \mu_1 \nu_2 L_{54} \\ + 3 \nu_1 \mu_2 L_{21} + \dots \quad (1)$$

where the $L_{\ell m} \equiv |\ell \rangle \langle m|$ are the transition operators among the crystal field states; and the coefficients $\nu_2 \sim \mu_1$, $\nu_1 \sim \mu_2$

depend on the crystal field and the resonant field strength.

The matrix elements of the J^+ operator depend on the angle of the resonant field with crystal axis. The operators in Eq. (1) identify the dipolar transition operators, the first five being those within the Γ_8 excited state quartet, the L_{21} is within the Γ_7 ground state doublet, and the rest are the intermultiplet transitions with frequencies $\Delta \sim (200 \text{ K})$. These operators correspond to the observable EPR modes. We notice that the zero order (molecular field) frequencies of the excitations generated by L_{35} and L_{46} are almost degenerate because of time reversal symmetry. The effect of the mixing of these excitations by the exchange interaction will be of vital importance to the behavior of the system.

The set of dipolar excitation operators is taken as the set of dynamical variables $\{A_\mu\}$. The matrix of relaxation functions among the variables, $R_{\mu\nu}(\omega)$, in the Mori-Zwanzig theory is given by:

$$\sum_{\lambda} (i\omega\delta_{\mu\nu} - i\Omega_{\mu\nu} + \Gamma_{\mu\nu}(\omega)) R_{\nu\lambda}(\omega) = S_{\mu\nu} \quad (2)$$

where the frequency matrix $\Omega_{\mu\nu}$ is given by

$$\Omega_{\mu\nu} = \sum_{\lambda} (LA_{\mu}, A_{\lambda}^+) (S^{-1})_{\lambda\nu} \quad (3)$$

and the memory function $\Gamma_{\mu\nu}(\omega)$, from which we calculate the damping width is given by

$$\Gamma_{\mu\nu}(\omega) = -i \sum_{\lambda} \left(\frac{1}{\omega - (1-P)L(1-P)} (1-P) LA_{\mu}, (1-P)(LA_{\lambda}^+) \right) (S^{-1})_{\lambda\nu} \quad (4)$$

The definition of the inner product, the projection operator P , the Liouville operator L , and the static susceptibility matrix $S_{\mu\nu}$ are the usual ones.² Under the assumption that $\{A_\mu\}$ gives the complete set of relevant slow variables, Eq. (2-4) give the solution to the relaxation functions, which are related to the zone centre dynamic susceptibility $\chi''_{xx}(\omega) = \omega \text{Re} R_{xx}(\omega)$. Our aim here is to calculate the matrix elements in S , Ω , and $\Gamma(\omega)$. In the following, we will discuss the random phase approximation results for the system with isotropic exchange interaction, and with the s-f interaction.

HEISENBERG EXCHANGE CASE

Here, we assume an isotropic Heisenberg exchange interaction. Using the random phase approximation³, one can easily calculate the matrix elements of the static susceptibility S and the

frequency matrix Ω . The damping functions are calculated as the convolution of two relaxation functions derived from the equations of motion. This corresponds to the summation of the lowest order loop diagrams to the damping term.

The off diagonal elements of Ω are observed to be large between the operators of the same frequency, and in particular, for modes due to L_{35} and L_{46} for $H \parallel \langle 111 \rangle$. The mixing does not affect the frequencies, but is very important for the damping width of the modes. The mixing depends on the matrix elements of J^+ operator (thus the direction of the resonant field), the exchange integral, and the temperature through the static susceptibility.

For $H \parallel \langle 111 \rangle$, in the low temperature regions, one of the mode generating operators coming from the exchange mixing of L_{35} and L_{46} is almost the same as the first term in the expression for J^+ given in Eq. (1). Consequently this yields a damping width much smaller than those of the other modes. This is shown in Fig. 1. The damping is very anisotropic; on departing from $\langle 111 \rangle$ direction, it quickly becomes large. For field along $\langle 110 \rangle$, the modes due to L_{45} and L_{36} have width comparable to the frequencies, while for

Fig. 1. Damping width of the modes versus temperature for field along $\langle 111 \rangle$ in the Heisenberg case. Modes are labeled by the corresponding generating operators. (Modes corresponding to two different mixtures of L_{65} and L_{43} fall in the range shown.)

field along $\langle 100 \rangle$, all modes are strongly damped by the exchange interaction.

In Fig. 2, the temperature dependence of the g-factor and of the intensity for the $L_{46} + L_{35}$ mode are shown for applied field in the $\langle 111 \rangle$ direction. The g-factor obtained is too high, and its temperature dependence too weak comparing to experiment. (The experimental g-factor is 2.28 at 100 K and decreases to 2.16 at 300 K.) Probably the difference is due to the crudeness of the model exchange interaction.

The expected lineshape for the polycrystalline system, coming from superposition of lineshape from all field directions in the (110) plane is shown in Fig. 3. The narrow mode labeled L_{21} comes from within the Γ_7 doublet, and corresponds to a g-factor below the experimentally observed range. The mode of interest is that with peak near 3 KG. The width of the mode is in good agreement with experiment. We feel that we have a qualitative explanation for the single-peak lineshape seen in the EPR experiment.

s-f INTERACTION CASE

We have also calculated the case with s-f interaction. Here we introduce the conduction electron spin as an added dynamical variable, and treat the case for $H \parallel \langle 111 \rangle$ only. All the excita-

Fig. 2. g-factor and intensity of the mixed $L_{46} + L_{35}$ mode as functions of temperature for field along $\langle 111 \rangle$ in the Heisenberg case.

Fig. 3. Polycrystalline lineshape in the Heisenberg case versus field, at 9.2 GHz and versus mode frequency, at $\mu_B H = 0.268 K$.

tions for the localized moments mix with the conduction electron modes. The s-f interaction introduces the damping due to conduction electron relaxation (the bubble diagram contribution)⁴. Similarly to the Heisenberg case, we diagonalize the frequency matrix and calculate the damping to the normal modes. The result is shown in Fig. 4. The three modes labeled I, II and III are mixtures of the L_{35} and L_{46} modes of the localized moment system with the mode of the conduction electron moment system. Mode III is predominantly conduction electron excitation. The width of mode III is much smaller than those of I and II, but is still too large in comparison to the frequency for agreement with experiment. (The g-factor obtained for mode III, however, does fit well with the experimental values.¹) At present, these calculations include s-f interaction only to second order. To treat the important mode mixing effects, the s-f interaction must be treated to higher order.

CONCLUSION

We have identified one important aspect of the behavior of the quasielastic modes in a Kramers system. The exchange mixing of the modes for the Zeeman-split crystal-field only system is crucial. The mixing yields a normal mode determining the observed

Fig. 4. Damping width versus temperature in the s-f interaction case.

low frequency spectrum. This is illustrated by our calculation with Heisenberg exchange which yields behavior in qualitative agreement with experiment.¹

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