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Magnetic Response Functions in Actinide Compounds

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Abstract

A brief review is given of neutron inelastic scattering experiments on actinide (5f) systems. Only a few compounds exhibit sharp crystal-field levels so that analogies with 4f compounds are rarely straight forward. In the systems that order magnetically, we do find well-defined magnetic excitations, although in many cases the strong interaction with the conduction electrons gives rise to overdamped modes. Finally in the strongly enhanced paramagnets and heavy-fermion systems the response is essentially featureless in $\vec{Q}$ space and can be characterized by a single Lorentzian with full-width at half-maximum of Γ . Attempts to relate this quantity Γ to other physical properties, particularly the coefficient of the electronic specific heat γ , have not been too successful.

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1. Introduction and neutron cross section

The neutron cross section for magnetic inelastic scattering may be written[1]:

$$\frac{d^2\sigma}{d\Omega d\nu} = \left\{ \left(\frac{r_e^2}{m_e c^2} \right)^2 \left[\frac{1}{2} g f(\vec{Q}) \right]^2 \frac{k_f}{k_i} e^{-2W(\vec{Q})} \right\} \times S(\vec{Q}, \omega) \quad (1)$$

where $(r_e^2/m_e c^2)$ is related to the neutron-electron interaction, g is the Lande factor, $f(\vec{Q})$ is the form factor of the electrons, k_f and k_i are final and initial wavevectors of the scattered radiation, $W(\vec{Q})$ is the Debye-Waller factor, and $S(\vec{Q})$ is the magnetic response function which is a function of both the momentum transfer $\vec{Q}$ and the energy transfer $\hbar\omega$.

The magnetic response function (or dynamical structure factor) is related directly to the imaginary component of the susceptibility, thus

$$S(\vec{Q}, \omega) = \frac{N}{\pi(g\mu_B)^2} [n(\omega) + 1] \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) \times \text{Im } \chi^{\alpha\beta}(\vec{Q}, \omega) \quad (2)$$

where $n(\omega) = (e^{\hbar\omega/kT} - 1)^{-1}$ and gives the so-called detailed balance condition, the sum is over all components α, β in the spin system, and the term $(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)$ ensures that scattering occurs from components perpendicular to $\vec{Q}$.

Normally we think of $\text{Im } \chi^{\alpha\beta}(\vec{Q}, \omega)$ [= $\chi''(\vec{Q}, \omega)$, where we drop the component notation] as consisting of discrete poles (or maxima) in $(\vec{Q}, \omega)$ space corresponding to either crystal-field or exciton behavior, in which case there is little $\vec{Q}$ dependence, or magnetic excitations which, like phonons, are discrete in $(\vec{Q}, \omega)$ space. Another example is when we find a relaxational spectrum. We then make use of first the Kramers-Kronig relation to give

$$\chi''(\vec{Q}, \omega) = \omega \chi'(\vec{Q}) F(\vec{Q}, \omega) \quad (3)$$

where $\chi'(\vec{Q})$ is the real component of the susceptibility, and $F(\vec{Q}, \omega)$ is the spectral weight function, which by the fluxation dissipation theorem is defined as

$$\int F(\vec{Q}, \omega) d\omega = 1 \quad (4)$$

A consequence of Eq. (3) is that the bulk susceptibility at a given $\vec{Q}$ can be found by

$$\chi(T) = \int \chi''(\omega)/\omega d\omega \quad (5)$$

and a consequence of Eq. (4) is that if the dynamics are purely relaxational then

$$F(\vec{Q}, t) = \exp [-\Gamma(\vec{Q})t] \quad (6)$$

and

$$F(\vec{Q}, \omega) = \pi^{-1} \frac{\Gamma(\vec{Q})}{\Gamma^2(\vec{Q}) + \omega^2} \quad (7)$$

If we substitute back into Eq. (2) we find that the intensity $I(\vec{Q}, \omega)$ is given by

$$I(\vec{Q}, \omega) \propto (k_f/k_i) \frac{\omega}{1 - e^{-\hbar\omega/kT}} \cdot \frac{\Gamma(\vec{Q})}{\Gamma^2(\vec{Q}) + \omega^2} \quad (8)$$

This gives a Lorentzian-type behavior at high temperature, with a FWHM of $\Gamma(\vec{Q})$, and at low T , where $e^{-\hbar\omega/kT} \approx 0$, the intensity is proportional to frequency $\hbar\omega$ and peaks roughly at $\Gamma(\vec{Q})/2$. We shall defer until later a discussion of what these $\Gamma(\vec{Q})$ parameters might mean.

2. Crystal-field states

For 4f systems one signature of localized behavior is the presence of well defined crystal-field (CF) levels that can be observed with neutron spectroscopy.[2] Such levels exist in all localized 4f systems, and only if the CF potential is smaller than the exchange interactions (e.g. in the rare-

earth metals) are the CF levels difficult to see in $4f$ systems. CF levels are observed in both ionic and conducting $4f$ compounds. In $5f$ systems the CF potential is known from optical work on ionic systems to be larger than that in corresponding $4f$ systems, but strength alone simply means that it is more difficult to perform the neutron experiment. A case in point are the CF levels in UO_2 , which have recently been observed[3] by using the large epithermal flux available from the neutron spallation source - IPNS - at Argonne National Laboratory. Spectra showing the transition from the ground Γ_5 state to the quadrupolar split Γ_3 level are shown in Fig. 1. On the other hand, in more metallic U systems, the observation of CF levels has been successful only in one compound UPd_3 . [4]

This absence of well-defined CF levels has presented major problems for theories based on localized $4f$ -like behavior, which invariably predict such states. Their absence, we believe, is caused by strong hybridization of the $5f$ electrons not only with the conduction d and s states, but also with the anion p or d states. [5]

3. Ordered magnetic systems.

In these systems, we anticipate finding magnetic excitations below the ordering temperature. Since there is considerable anisotropy in the bulk magnetization[6], we anticipate gaps in the energy spectrum even at $T = 0$. Fig. 2 shows the magnetic excitations found in USb [7], illustrating that indeed sharp well-defined modes are found. USb orders antiferromagnetically at 215K with a moment of $2.8 \mu_B$ so it is near the "localized" limit. On the other hand, systems such as UN [8] and UAs [9] also order magnetically (at 53 and 127K, respectively) but show no evidence for sharp spin waves. Instead, overdamped modes often showing the presence of strong correlations and even, in the case of UN , the ghost of an anisotropy gap are present. It should not be thought that such observations are inconsistent with a localized $5f$

picture plus strong exchange and hybridization interactions, but clearly the starting values for such an analysis are very difficult to determine experimentally. Aspects of the experimental and theoretical situation have been recently reviewed by Holden[10] and Edwards[11], respectively.

4. Paramagnetic systems - spin fluctuations and heavy fermions.

In the previous two sections, we have seen that a characteristic of $5f$ systems is that the neutron inelastic response functions tends to be either initially broad and featureless or rapidly becomes so on heating an otherwise "localized" compound such as USb. This broad and featureless characteristic is now the sole surviving feature of paramagnetic systems, whether they be so-called spin-fluctuating or heavy-fermion systems.[12] It is for that reason we wrote down in Sec. 1 the neutron cross section; see especially Eq. (8).

4.1 Experimental limitations

In most cases, the measurement of CF levels is relatively straight forward. They are sharp in energy space and have little Q dependence. Magnetic excitations are more difficult, one requires a single crystal for a start, and structure factors, which cannot be known in advance, often mean that a number of different zones must be examined before the full dispersion can be determined. However, such excitations are usually fairly sharp in $(\vec{Q}, \omega)$ space. For the diffuse relaxational spectrum the problem is much more difficult. The safest way to perform these measurements is with a time-of-flight spectrometer and polycrystalline samples. In this case, complete integrations [see Eq. (5)] can give accurate comparisons with the bulk susceptibility and one can be sure that no part of $S(Q, \omega)$ is missed. Good examples are Holland-Moritz et al.[1], and C.-K. Loong [13], the latter illustrating the importance of using high-energy neutrons from IPNS in cases where Γ

is large. However, such time-of-flight experiments are limited in their $\vec{Q}$ resolution. Of course, for a polycrystalline sample one can only define the modulus $|\vec{Q}| = Q$, and essential information may be lost. On the other hand, with a triple-axis machine the solid angle subtended is very small so that the signal observed for any one setting is very weak and careful background subtractions must be made to determine the real magnetic signal. Polarized beams can also be used but the intensities are still rather weak and the energy resolution relatively poor.[12].

Whatever the chosen experimental method careful account has to be taken that the nuclear vibrational spectrum does not contaminate the magnetic response function.[14]

4.2 Experimental results.

We show in Fig. 3 the low temperature (1.3K) spectrum of UPt_3 recently measured at Brookhaven National Laboratory.[15] The solid line is the fit with Γ [of Eq. (6)-(8)] of 20 meV. Note how the response is proportional to $M\omega$ at low energy [as Eq. (8) predicts] and at these temperatures the peak in the response function is at $\Gamma/2$. Although these data were taken on a polycrystalline sample no Q dependence was reported. More recently, Walter et al.[16] have used time-of-flight spectrometers at both IPNS and ILL to show that the response in URu_2Si_2 has no quasielastic contribution at low temperature, although $\Gamma \approx 14$ meV at high temperature, but rather a substantial "gap" of 5.5 meV in the spectrum. This gap is Q dependent, the strength being greatest at $Q \approx 0.8\text{\AA}^{-1}$. Both UPt_3 and URu_2Si_2 are heavy fermion systems, the latter orders antiferromagnetically (17K) as well as exhibiting superconductivity below 1.5K.[17]

The spin fluctuation systems such as UAl_2 have response functions very similar to the heavy fermions discussed above. In Fig. 4 we show the T-dependence of Γ [18] showing that Γ has a value of 50 meV at the lowest temperature. At all temperatures $\Gamma > kT$ so that thermally excitations clearly play only a small role. A tabulation of Γ and the electronic specific heat coefficient γ has been given by Shapiro[12] for a series of Ce and U based compounds. There is little correlation between γ and Γ . Attempts have been made to analyze the spectra in terms of spin fluctuation temperatures T_{sf} [4] and effective masses[15], but the analyses make many assumptions which can be challenged. One noticeable feature is that the U compounds have Γ values almost an order of magnitude greater than Ce compounds. This can be rationalized on the basis of there being more electrons around U than Ce, and a stronger hybridization between the U $5f$ electrons and the conduction electrons than in the Ce case. One important point to be made here is that the integrated neutron measurements are in good agreement with the bulk susceptibility [see Eq. (5)]. This means that the spectral weight of any additional $\vec{Q}$ dependent feature is probably small, unless very strong antiferromagnetic fluctuations such as in UAs[9] exist. The bulk susceptibility is at $Q = 0$; only neutrons can probe $\chi(Q,0)$ for $Q \neq 0$.

In reality, however, the neutron results for these spin fluctuating and heavy-fermion systems are disappointing. One parameter, Γ , which so far seems independent of $\vec{Q}$ and is only mildly T-dependent seems little return for so much effort! It does, of course, represent the extent of $5f$ electron hybridization and as such represents a stringent test of theories that are being proposed. Hopefully, single crystal studies will determine some $\vec{Q}$ dependence which will add to the significance of Γ . Since we are dealing with hybridization of $5f$ electrons with the

conduction electrons it may be necessary to make observations at small Q values (i.e., $Q < 0.6 \text{\AA}^{-1}$) where the conduction electron form factor $f(Q)$ is non-zero. This would require an experiment with a small-angle diffractometer equipped for inelastic magnetic scattering - by no means a trivial task!

5. Studies of transuranium systems.

The title of this short review article contains the word "actinide" and yet the discussion so far has been limited to U compounds. Clearly, to establish the systematics of the 5f series we need to extend the work beyond uranium. This presents formidable difficulties, specifically for neutron inelastic scattering, which frequently requires large quantities of materials (i.e., g rather than mg quantities) and also single crystals. To address this need a program has been started at the Transuranium Institute, Euratom, Karlsruhe to grow single crystals. A number of papers have now been published using these samples and neutron elastic scattering.[19,20]

We report here some highlights of an inelastic study of large $^{242}\text{PuSb}$ single crystals performed recently at the ILL[22] and not yet completed. PuSb is a ferromagnet with $\mu \parallel [001]$ at low temperature. The neutron study was performed on a monodomain sample (formed by field cooling) and this gives the rare possibility of studying the individual $\alpha\beta$ components of $\chi^{\alpha\beta}(\vec{Q}, \omega)$ [see Eq. (2)]. The dispersion curve is almost independent of $\vec{Q}$ with an anisotropy gap of ~ 16 meV. This corresponds to the splitting of the ground state Γ_8 quartet[22], although one would also have anticipated a second mode within the Γ_8 ground state. Of particular interest, and found only near the zone-boundary X point is the splitting of the mode into two components. Spin waves with their displacements parallel to the propagation vector have a lower energy than those with transverse displacements. This is illustrated by the experimental data in Fig. 5. Such a splitting of the modes implies

that in this region of $\vec{Q}$ space they are linearly polarized, which is certainly an unusual situation and which may be proven directly by performing an analysis of the scattered radiation. What is so unusual is that the minimum in the dispersion curve is at the zone boundary, whereas for a ferromagnet we expect such a minimum at the zone center. Attempts to understand various features of the excitation spectrum by considering anisotropic hybridization of the conduction electrons with the primarily localized $5f$ electrons are now in progress[22] and more experiments on this and other similar systems are planned.

6. Summary.

We have touched briefly in this article on some of the neutron inelastic scattering studies of actinide compounds. The task is by no means complete. We need to extend the crystal-field measurements to other U and transuranium systems (Sec. 2); we need to study more carefully the T-dependence of the excitations in localized systems (Sec. 3); we need to encourage theoretical efforts to relate Γ to either bulk measurements or to the microscopic parameters that describe the hybridization between the $5f$ and conduction electrons (Sec. 4). Studies of transuranium single crystals will continue to be very difficult, but present the rare possibility of studying the systematics of the localization that occurs as one proceeds across the actinide series.

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Figure Captions

Figure 1 Inelastic spectrum of UO_2 [3] as a function of temperature. The two peaks represent transitions from the Γ_5 ground state to the Γ_3 excited doublet, which is split by a strong quadrupole interaction. The CF splitting is essentially independent of temperature. UO_2 orders antiferromagnetically at 30.8K. The data were taken at the IPNS spallation source, Argonne.

Figure 2 The magnetic (solid and dashed) and phonon (broken lines) dispersion curves for USb [7] at low temperature. Note the minimum at the X-point, the anisotropy gap and the steep dispersion indicating strong exchange in this system which orders at 215K. The data were taken at ILL. (Note 1 THz = 4.14 meV)

Figure 3 Inelastic spectra of UPt_3 measured at Brookhaven[15]. The solid line is a fit to Eq. (8) with $\Gamma = 20$ meV. Note the peak at $\sim \Gamma/2$ which is a consequence of the detail balance factor in Eq. (8).

Figure 4 Temperature dependence of the Lorentzian half-width of UAl_2 as measured on a polycrystalline sample at ILL[17] and IPNS[18]. Note that $\Gamma/2$ is much greater than kT at low temperatures.

Figure 5 Inelastic spectra from a single crystal of $^{242}\text{PuSb}$ taken at ILL[21]. The three spectra indicate (a) that there is little dispersion across the zone since a peak appears throughout at $\sim 4\text{THz}$ (1 THz = 4.14 meV) and (b) at the zone boundary [102] two peaks are seen because a mode $S^{xx}[\text{q}00]$ is measured, whereas at [201] this mode cannot be seen and only one peak is observed.

Fig 1

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