

Magnetic dynamics in correlated electron metals poised between localization and itinerancy

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Abstract

Despite of the impressive range of experimental information on the physical properties of plutonium almost nothing is known on the magnetic dynamics in all plutonium phases. The plausible magnetic spectral response in fcc Pu has been calculated on the basis of the single-ion Kondo model and comparison with a model system CeNi, which is a good analogue of fcc Pu regarding the characteristic Kondo energy scale and partial delocalization of the f-electrons. A broad peak centered at about 40 meV dominates in the spectrum at low temperatures. The most interesting energy transfer range for the inelastic neutron scattering experiments measurements on fcc Pu is 20–150 meV. Magnetic excitations in itinerant systems like Cr and α -Pu are discussed.

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1. Introduction

Excitation spectra are indispensable experimental data to the understanding of the ground state and the major interactions in plutonium. In particular inelastic neutron scattering (INS) experiments can yield valuable answers with respect to the puzzling question raised over the absence of any evidence of magnetic moments in plutonium [1]. Despite of the impressive range of information which has been attained by measurements of physical properties and theoretical studies the investigation of the dynamic properties of plutonium is by no means complete [2]. INS is very limited in the case of plutonium to say the least and almost nothing is known on the magnetic spectral response in this metal. However, a lot of INS data has been collected for other strongly correlated metals and intermetallics with electrons balanced between itinerant and localized states. Pu is not unique among metallic systems regarding such phenomena as

the unit cell volume collapse and the partial delocalization of the f-electrons. Cerium and Ce-based compounds demonstrate many peculiarities of the 4f electron shell similar to the 5f shell in Pu. Cerium-based systems with a wide range of the characteristic Kondo energies have been studied by INS intensively (see [3,4] and references therein). The concept of shifted homology of Ce and Pu is known for a long time [5]. It is based on the electron hole symmetry between the wave functions of the f^n and f^{6-n} ions. Ce and Pu ions have a similar degree of delocalization of the f-electrons. Thus, Ce compounds could serve as good model systems to judge about the plausible magnetic spectral response in Pu and its compounds. The magnetic spectra of Ce-based compounds contain various contributions. It is generally assumed that, in the state with large hybridization and relatively high Kondo energies, one broad inelastic peak centered at an energy of several tens to some hundreds of millielectronvolts dominates at low temperatures. The broad peak is a single-ion spectral feature and almost Q -independent (except the dependence of the magnetic form factor on the absolute value of Q). However, this physical picture is oversimplified, since extra peaks show up in the INS spectra of some Ce-based Kondo systems. These peaks reflect the coherent origin of the ground state in the Kondo lattices. In addition to coherent peaks reflecting dynamic magnetic

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correlations in the Ce sublattice, other magnetic spectral features have been observed in cerium-based Kondo compounds: intermultiplet transitions, modified crystal field excitations, magnetic excitations coupled to phonons.

Nothing is known on the magnetic spectral response in α -Pu which is considered as a system where 5f electrons are believed to participate in bonding. Magnetic excitations in itinerant systems are usually very broad extending beyond hundreds millielectronvolts since the characteristic energy scales are huge and the excitations velocities are provided by the Fermi energy. Elemental Cr is an archetypical itinerant system extensively studied by the INS technique. Investigation of the magnetic excitations in Cr could be useful for the understanding of the magnetic dynamics in α -Pu.

The main goal of the present study was to calculate the plausible magnetic excitation spectrum of fcc Pu on the basis of the INS data collected in Ce-based systems and the analysis of the thermodynamic properties of Pu. We discuss the most interesting neutron energy transfer range to study magnetic excitations in fcc Pu and the feasibility of the INS experiments on ^{239}Pu -based samples.

2. Results and discussion

In order to choose the best model cerium system to understand magnetic dynamics in fcc Pu one should take into account the characteristics Kondo energy scale. Plutonium demonstrates many typical features of Kondo systems in its macroscopic physical properties (specific heat, magnetic susceptibility, resistivity, etc.), so the idea to analyze the magnetic spectral response in Pu on the basis of the Kondo approach looks reasonable at first glance. The value of the Sommerfeld coefficient in fcc Pu $\gamma = 65 \text{ mJ}/(\text{mol K}^2)$ [6] is indicative of the moderately heavy fermion or even intermediate valence behaviour. Among Ce-base compounds, CeNi has the same value of the linear specific heat coefficient γ [7]. The ground state in this system is a many-particle non-magnetic singlet. It is worthwhile to note that, very similar to the instabilities of the 5f shell in plutonium, CeNi demonstrates a first order phase transition with a 12% volume collapse [8].

The magnetic excitation spectrum of the 4f analogue of plutonium CeNi have been measured by INS on both single crystal and polycrystalline samples. ^{60}Ni isotope was used to suppress the phonon contribution to the scattering. Details of the experiments are provided in [9]. Figs. 1 and 2 show the magnetic contributions measured on a single crystal and polycrystalline sample, respectively. The spectra can be described by three peaks: one broad with $E_0 = 45 \text{ meV}$ and half-width at half-maximum about 40 meV and two relatively narrow at $E_0 = 18$ and 34 meV. The broad spectral feature is a single-ion peak typical for Ce-based Kondo compounds. Single-ion response is by far the major spectral feature contributing more than 90% to the total magnetic scattering. Two narrow peaks correspond to almost dispersionless collective excitations with intensity strongly dependent on the wavevector \mathbf{Q} . These peaks somehow reflect the coherent ground state in the system but their exact origin remains unclear.

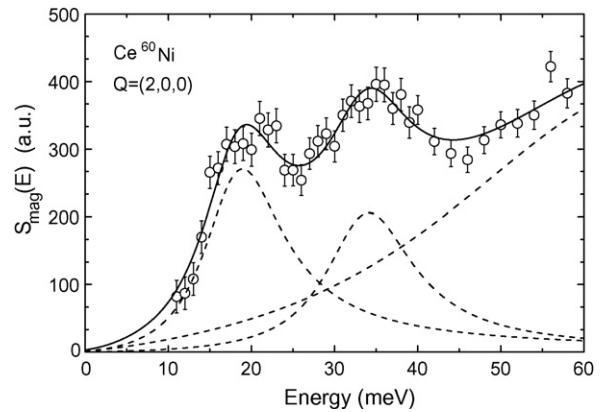


Fig. 1. Magnetic spectral response of the single crystal of Ce^{60}Ni measured at $T = 11 \text{ K}$ at the zone center along \mathbf{a} -direction $\mathbf{Q} = (2, 0, 0)$. Lines: fit of the magnetic response by three inelastic Lorentzians. The magnetic contributions have been normalised by the Ce^{3+} magnetic formfactor to $\mathbf{Q} = 2.9 \text{ \AA}^{-1}$.

Now we are coming to the main problem—calculation of the magnetic spectrum in fcc Pu. Coherent spectral features are likely to show up in fcc Pu. Nevertheless, we assume that the broad single-ion peak is the major spectral feature in fcc Pu. The characteristic energy scale of this peak is provided by the value of the Sommerfeld coefficient within the Fermi-liquid approach [10,11]:

$$\gamma = N_A \pi^2 k_B < n_f > \frac{1}{3E_0} \frac{N-1}{N} \quad (1)$$

A reasonable estimate from the specific heat data is $E_0 = 30 \text{ meV}$ assuming the occupancy of the 5f shell $\langle n_f \rangle = 0.83$ and the effective degeneracy of the ground state N ($2 < N < 6$ since $J = 5/2$ multiplet dominates in fcc Pu). This Kondo energy scale yields the value of the static magnetic susceptibility in fcc Pu at low temperatures $\chi \sim 540 \mu\text{emu}/\text{mol}$ which is very close to the experimental value obtained for stabilized δ -Pu [12]. Thus, the characteristic energy scale of about 30 meV is consistent with both specific heat and magnetic susceptibility within the Fermi liquid physical picture applied to fcc Pu at low temperatures.

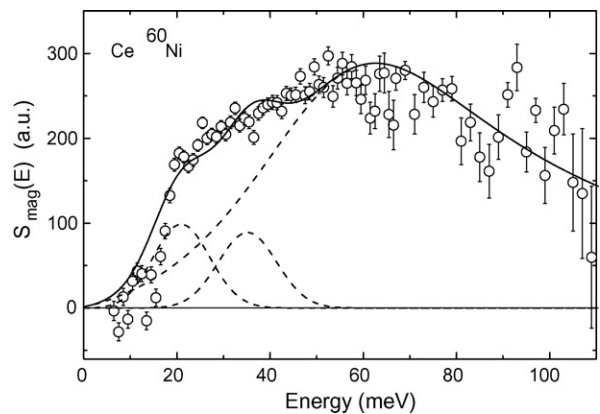


Fig. 2. Magnetic component of the INS spectrum of Ce^{60}Ni polycrystal at $T = 12 \text{ K}$. Lines: fit by three inelastic Lorentzians, one single-ion peak centered at about 60 meV and two coherent peaks with the energy positions fixed at $E = 18$ and 34 meV.

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