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Localized excitations in UPdSn

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We have measured the inelastic neutron-scattering response of UPdSn at various temperatures using the HET and PHAROS spectrometers at the ISIS and MLNSC facilities, respectively. UPdSn shows some quasielastic scattering, which may be attributed to the hybridization of the 5f electrons with the conduction electrons. Furthermore, we find a clear excitation around 40meV above 40K in addition to the phonon contribution. While this excitation may be indicative of crystal fields in UPdSn, its strong temperature dependence seems to contradict a simple crystal-field picture. Below T_N, the unusual temperature dependence may be attributed to magnetically-driven distortions (and subsequent changes in the local surrounding of the U ions), but there is some evidence that other additional mechanism(s) may contribute above T_N. Some possible mechanisms will be discussed.

KEYWORDS: inelastic neutron scattering, crystal fields, UPdSn

Among the uranium intermetallics, UPdSn has attracted recent attention as it is believed to be one of the rare examples where the 5f electrons are believed to be fairly localized.[1] Therefore, UPdSn is a prime candidate for crystal-field effects, and its observation in uranium intermetallics is extremely rare. UPdSn has two antiferromagnetic transition at about 40 and 25K.[2]

Here, we report on inelastic-neutron-scattering data on UPdSn using the HET and PHAROS spectrometers at the ISIS and MLNSC facilities, respectively. The experiments were performed on 25g of polycrystalline UPdSn, and data were at various temperatures using an incident energy of 100meV.

The HET data are displayed in Fig. 1, where the scattering response of UPdSn at 20 and 100K is shown. The symbols represent the response as seen in HET's low-angle detector bank (sensitive to scattering with small momentum transfer, Q), while the solid line represents the response seen in the high-angle detectors (large Q). Scaled appropriately the high-angle response provides an estimate of the scattering contribution due to lattice vibrations (phonons), while any additional scattering seen in the low-angle detectors must be magnetic in origin. Clearly, there are additional 'magnetic' features. First, at 100K, there is some quasielastic scattering (below 10meV), which is barely present at 20K. Secondly, there is a pronounced excitation around 40-50meV.

The quasielastic term seen at 100K may originate in the hybridization of 5f electrons with the conduction electrons.[3] In the magnetically-ordered state (at 20 K), on the other hand, some kind of a 'hybridization gap' may have formed. Recent band-structure calculations on UPdSn by Sandratskii and Kübler[4] find indeed a local minimum in the density of states at E_F. Furthermore, as the 5f electrons carry a magnetic moment, one would expect subsequent gapping in the magnon-excitation spectrum, and bulk results provide evidence for its presence.[5]

The excitation around 40meV (at 100K) may signal crystal fields in UPdSn. The strong energy shift (~10meV) on going to 20K could be a consequence of magnetically-driven distortions at the magnetic transitions,[6] because of subsequent changes in the local symmetry of the U ions. For better understanding of the nature of this excitation, we performed additional neutron-scattering studies at various temperatures using PHAROS (Fig. 2). Although the statistics is relatively poor (because of limited beam time), the excitation is clearly visible at all temperatures. After subtraction of a phonon background (estimated from HET data), we fitted the excitation to a single Gaussian (solid lines in Fig. 2). There is only little temperature dependence in the location of the excitation above T_N, which is followed by a strong shift below T_N (where also the magnetically-driven distortion occur). While this can be expected within crystal-field picture, we also find an unusual temperature dependence of the integrated intensities in the paramagnetic range. Only for temperatures above 100K, we find a meaningful value (~3) for the ratio of the degeneracies of the first excited and the ground state. Any extension toward lower temperatures yield unphysical large values. On the basis of the present results, however, it is not unambiguous whether the observed feature consists out of a single or possibly more excitations. Nevertheless, the above observation may indicate that there is some other physics which contributes to this excitation.

It may be tempting to ascribe the excitation to an anisotropy gap in a similar manner as done recently for URhAl.[7] However, the fact that the excitation almost vanishes at 300K contradicts bulk magnetic results, where huge magnetic anisotropy even at room temperature is found.[1]

Another alternative explanation is that there is some interaction in terms of spin-waves or magnon-phonon hybridization.[8] However, one expects such correlations to be largely absent above T_N.

Therefore, it seems likely that the observed excitation is of crystal-field origin, but some other physics interfere with the crystal fields. Some insight may be gained by comparing the inelastic-neutron-scattering data with the bulk observations. Such analysis is presently underway.

In conclusion, we believe that the 5f electrons in UPdSn are somewhat itinerant (yielding a quasielastic response), but that the hybridization of 5f electrons with the conduction electrons is too weak to completely smear out localized crystal-field excitations.

figure captions:

Fig. 1: Inelastic-scattering response of UPdSn using 100meV incident energy at 20 and 100K, taken on the HET spectrometer. The solid line represents the 20K phonon estimate obtained from the high-angle detector bank (see text).

Fig. 2: Inelastic-scattering response of UPdSn using 100meV incident energy at various temperatures, as taken on the PHAROS spectrometer. The solid lines represent the Gaussian fits to the excitation after subtraction of an estimated phonon background.

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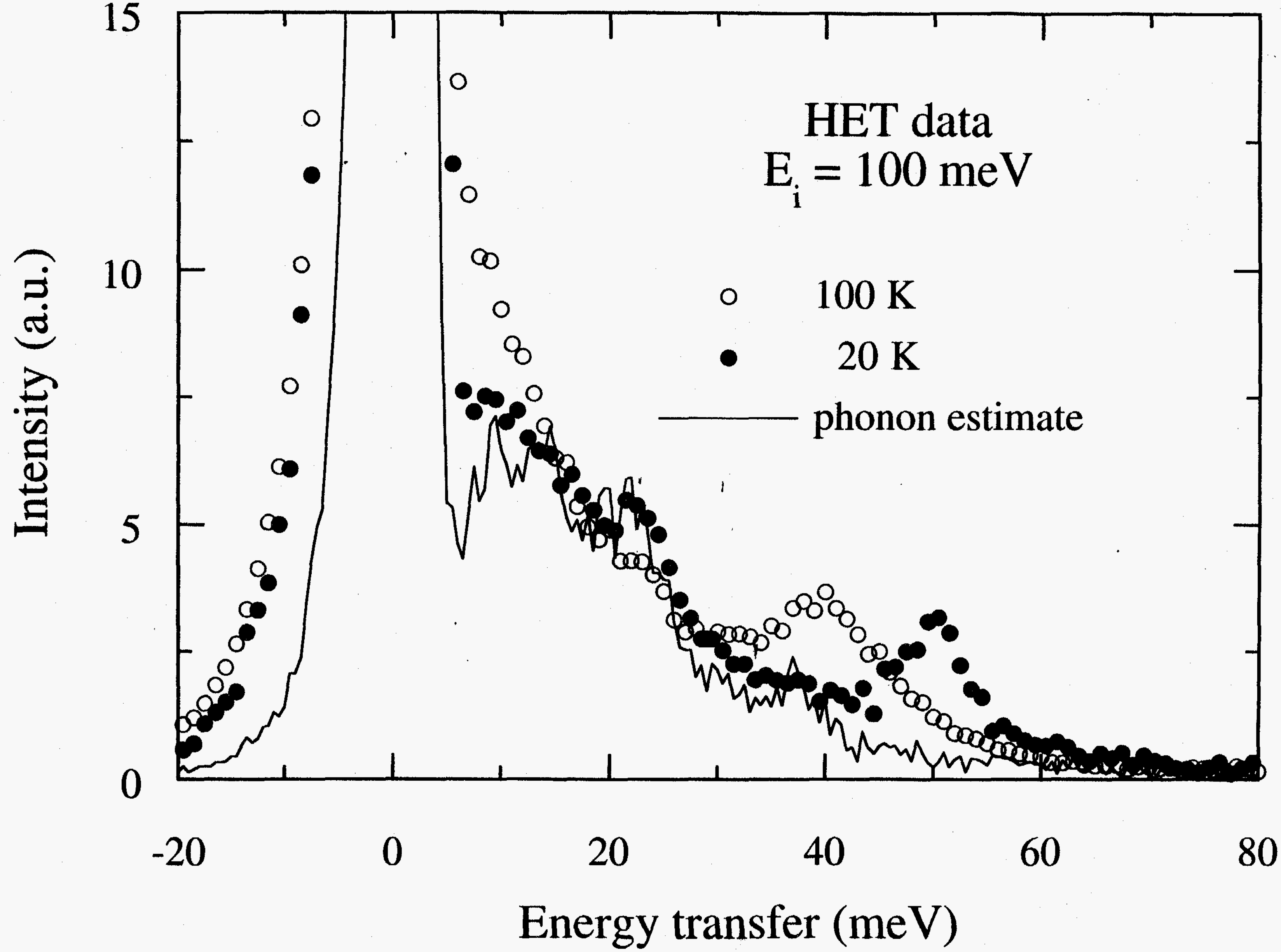
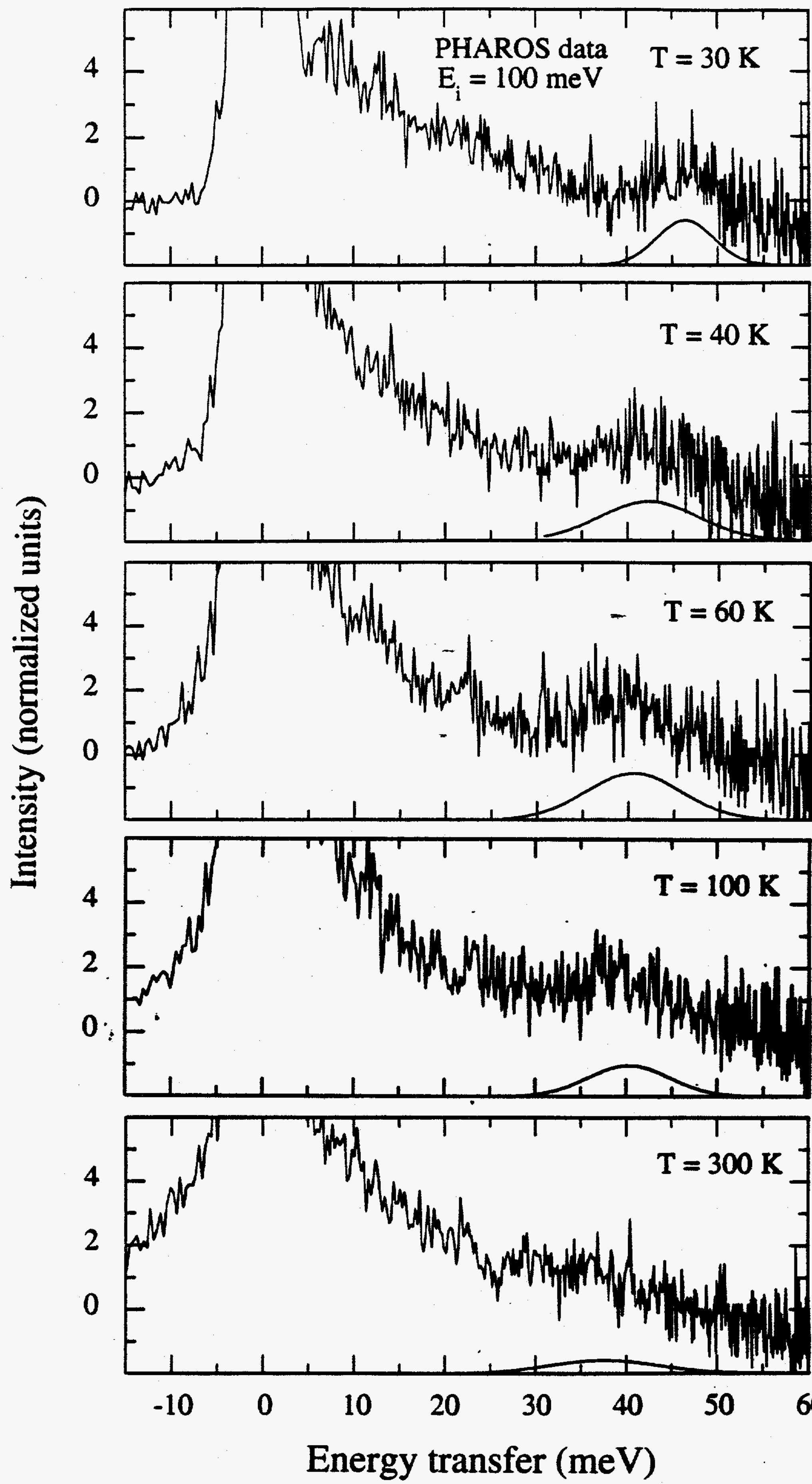

HET data
E_i = 100 meV
100 K
20 K
phonon estimate
Intensity (a.u.)
Energy transfer (meV)
15
10
5
0
-20
0
20
40
60
80



PHAROS data
E_i = 100 meV
T = 30 K
T = 40 K
T = 60 K
T = 100 K
T = 300 K
Intensity (normalized units)
Energy transfer (meV)
-10
0
10
20
30
40
50
60
0
2
4