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Neutron inelastic scattering experiments on uranium antimonide

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Résumé. — Nous présentons la première observation par diffusion inélastique de neutrons d'une excitation collective élémentaire dans un composé d'actinide métallique. Cette excitation est polarisée longitudinalment par rapport à la direction du moment magnétique. Les seuls autres modes magnétiques observés se situent à des énergies supérieures à 6 THz et ne présentent pratiquement pas de dispersion.

Abstract. — We report the first observation by neutron inelastic scattering of a collective elementary excitation in a metallic actinide compound. The excitation is longitudinally polarized with respect to the direction of the magnetic moment. The only other magnetic modes observed are at energies above 6 THz, and are essentially dispersionless.

1. Introduction. — At the present time models for the behaviour of metallic actinide (5f) systems are based on the (4f) systems. Thus, USb with the NaCl crystal structure ($a_0 = 6.197 \, \text{Å}$) and the type-I magnetic structure ($T_N = 241 \, \text{K}$) might be expected to have a similar electronic structure as NdSb, which has the same crystal ($a_0 = 6.235 \, \text{Å}$) and magnetic ($T_N = 13.6 \, \text{K}$) structure [1]. Both compounds would then be basically f\(^2\) configurations, developing almost the full free-ion moment of 3.27 \(\mu_B\) at low temperature [2] (2.98 \(\mu_B\) in NdSb and 2.82 \(\mu_B\) in USb). The aim of the present experiment is to measure the microscopic exchange and crystal-field interactions.

2. Experimental and results. — The experiments were performed on a crystal assembly, consisting of six oriented single crystals (total volume of 0.2 cc), with the triple axis spectrometers IN8 and IN3 at the High Flux Reactor, Institut Laue Langevin, Grenoble.

The type-I antiferromagnetic structure [2] of USb consists of ferromagnetic (001) sheets stacked in the simple alternating sequence + - + -. The moments are parallel to [001] and the crystal contains three domains corresponding to the moments being aligned along each cube axis. In the present experiments, since most of the measurements have been done at or near the X points [110] and [001], where the structure factors for two out of three domains are very small, the excitations may be identified with a single (001) domain [1]. In an exact analogy with the critical scattering experiments [3] on USb, the excitations at wavevectors [0, 0, 1 + \(\xi\)] are transversal only, whereas those at [1 + \(\xi\), 1 + \(\xi\), 0] consist of both transverse and longitudinal components. The first important observation is shown in figure 1, in which the excitation at the X point is clearly seen at [110] (Fig. 1a), but not at, or nor, [001] (Fig. 1b). We conclude that the collective excitation is almost entirely longitudinal in nature.

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The dispersion relations as measured to date are summarized in figure 2. The following points are important:

1) Two types of magnetic excitations are found. At low energies we see a longitudinal mode with considerable dispersion. At higher energies we see at least two broad, and essentially dispersionless, magnetic modes. The polarization of these modes is not simply longitudinal, since they are also seen at wave vectors [0, 0, 1 + ξ]. These high-energy modes have very little temperature dependence.

2) The collective excitation is completely degenerate with the zone boundary phonon at [110] with an energy of 1.53 ± 0.05 THz. In figure 1c we show a constant energy scan at 1 THz through the [110] position. This confirms that no lower branch of the magnetic excitation exists.

3) At wave vectors not too close to the point X the magnetic excitation has a dispersion of (6.7 ± 0.2) THz – Å in both the [ξξ0] and [00ξ] directions.

3. Discussion. — The most important point to emerge from the present studies, which are still in progress, is that the dynamic behaviour of the collective excitation in USb cannot be understood with simple crystal-field theory. This can be demonstrated by recalling that the polarization of the wave functions necessary to obtain the full ordered moment tends to produce eigenvectors composed mainly of single | M_i > components, as for example in the heavy rare-earth metals. Since a longitudinal excitation corresponds to a J_z matrix element between similar components, this situation occurs when the exchange is small, as for example [4] in Pr. Usually the longitudinal modes are very difficult to observe [1, 5].

Our inability to understand the dynamics does not mean that the crystal-field interactions are negligible. The dispersionless high-energy modes at ~ 6 THz may well be related to single-ion excitations. The magnitude of the energy splitting, ~ 6 THz, is almost identical to that between the J_z^I states predicted by our form-factor measurements [2], so that both the static and dynamic measurements show that an excited state exists at ~ 6 THz above the ground state.

In discussing the spin correlations [3] observed in the critical regime in USb we introduced the idea of a molecular orbital approach involving the hybridization between the magnetic 5f electrons and the anion p orbitals. The excitation observed in USb at low temperatures would seem further evidence for the need for such a model. The excitation is truly Ising in nature corresponding to correlations in the J_z components.

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