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Magnetization and spin-echo spectra of a mixing-type quadrupolar ordering in YbSb

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Abstract

Magnetization and nuclear magnetic resonance spin-echo spectra were measured to investigate a quadrupolar ordering in YbSb.

The width of the spin-echo spectra increases with applied magnetic field significantly, below a quadrupolar ordering temperature (T_Q). On the other hand, magnetization processes do not show any noticeable changes at T_Q. A model calculation using the molecular field approximation shows that the ferromagnetic components do not show a significant change, while antiferromagnetic components are induced strongly below T_Q in a magnetic field, which is qualitatively consistent with the experimental results. Furthermore, it is suggested that eqQ broadening of the spin-echo spectra is dominant at low magnetic field.

1. Introduction

A mixing type of quadrupolar ordering in YbSb has attracted much attention recently [1]. One of the important problems relating to Yb monopnictides is the unexpected splitting of the excited crystal field states (Γ_8) [2, 3]. Heavy-fermion behaviour at low temperature is also controversial, because the carrier concentration of Yb monopnictides is rather small [4]. It is plausible that quadrupolar interactions play an important role in all Yb monopnictides, even though a quadrupolar ordering occurs only in YbSb. The splitting of Γ_8 may be related to the quadrupolar interactions [5]. Low-lying excitations originating from quadrupolar interactions may cause the observed heavy-fermion behaviour. Therefore, investigation of a quadrupolar ordering in YbSb may be informative.

In this paper, we report nuclear magnetic resonance (NMR) spin-echo spectra for ^{121}Sb and magnetization measurements on YbSb. A discussion of quadrupolar ordering based on the molecular field approximation will also be given.

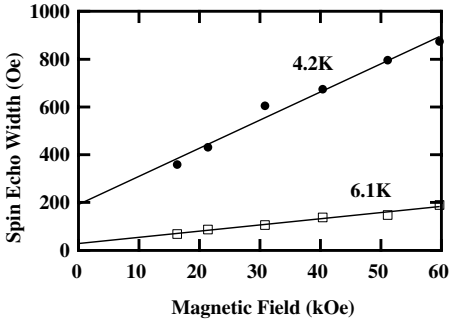

Figure 1. The field dependence of the width of the spin-echo spectra above and below T_Q.

2. Experimental results

Figure 1 shows the field dependence of the full width at half-maximum (FWHM) of nuclear spin-echo spectra. There is an abrupt increase of the width below a quadrupolar ordering temperature (T_Q). The FWHM is approximately proportional to the external magnetic field at all temperatures. The slope is enhanced strongly below T_Q. Furthermore, the value of the FWHM extrapolated to zero magnetic field is about 200 Oe, which is also strongly enhanced. The width of the spin-echo spectra above T_Q is proportional to the magnetization. The temperature dependence of the magnetization, however, does not show significant change at T_Q. This shows that the additional contribution to the width of the spin-echo spectra appears below T_Q. In the next section, we propose a quadrupolar ordering as the origin of the additional width.

3. Discussion

The Hamiltonian for the $J = 7/2$ multiplet of Yb^{3+} in cubic local symmetry examined here is the same as that used in our previous paper [1]:

$$H = H_{CF} + H_Z + H_{QQ}. \tag{1}$$

The first term represents the crystalline electric field potential, the second term the Zeeman energy, and the third term the intersite quadrupolar interactions between 4f electrons. With the fourfold axis chosen as the z-axis, the single-ion crystalline field potential is expressed as

$$H_{CF} = B_4^0(O_4^0 + 5O_4^4) + B_6^0(O_6^0 - 21O_6^4), \tag{2}$$

where the O_m^n are Stevens operators [6] and the B_m^n are the crystalline field parameters. The B_m^n have been reported from inelastic neutron scattering measurements [2]. The Zeeman energy is given as

$$H_Z = -g^J \mu_B J H. \tag{3}$$

We assume a two-sublattice model for simplicity, because the structure of the ordering is not known. Furthermore, we only investigate Γ_3-type quadrupolar interactions. Then the intersite quadrupolar interactions are given as follows:

$$H_{QQ} = -\sum_{i=1\,(i\in A)}^{N/2} \{K(u_i\langle u\rangle_B + v_i\langle v\rangle_B)\} - \sum_{j=N/2+1\,(j\in B)}^{N} \{K(u_j\langle u\rangle_A + v_j\langle v\rangle_A)\} + \frac{N}{2}\{K(\langle u\rangle_A\langle u\rangle_B + \langle v\rangle_A\langle v\rangle_B)\} \tag{4}$$

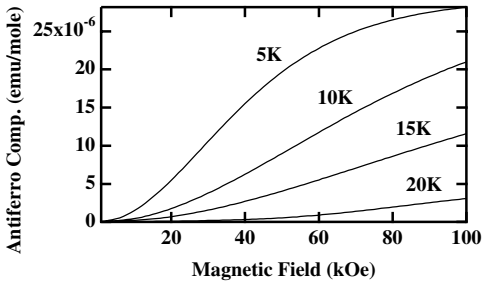

Figure 2. The magnetic field dependence of the AF component between 5 and 25 K. The magnetic field is applied along the [111] direction. The AF component at 25 K is zero throughout the magnetic field range.

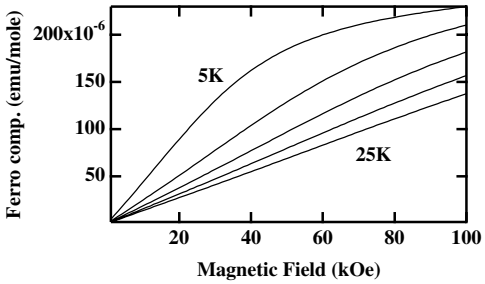

Figure 3. The magnetic field dependence of the F component between 5 and 25 K. The magnetic field is applied along the [111] direction. The F component does not show a significant change at T_Q.

where

$$u = J_z^2 - \tfrac{1}{3}J(J+1)$$
$$v = (J_x^2 - J_y^2)/\sqrt{3}. \tag{5}$$

The total Hamiltonian has been solved self-consistently, and ferromagnetic components (F components) and antiferromagnetic components (AF components) have been estimated. Here we note that the transition temperature derived from the experiment cannot be reproduced by the above molecular field calculation. Therefore, the discussion is limited to qualitative remarks. We adopt $K = 2.487$ K. The transition temperature is around 20 K in this case. Figure 2 shows the magnetic field dependence of the AF component below and above T_Q when the magnetic field is applied along the [111] direction. The AF component at 25 K is zero for the entire magnetic field range, while it increases with magnetic field below T_Q. The increase of the width of the spin-echo spectra arises from a distribution of static hyperfine fields at the Sb sites. The AF component is expected to generate static hyperfine fields, although the ordered structure is not known at present. Therefore the field dependence of the AF component is consistent with the enhancement of the spin-echo width (figure 1) below T_Q.

On the other hand, the F component does not show a significant change at T_Q. Its temperature dependence is rather gradual as shown in figure 3. This is also consistent with the temperature dependence of the magnetization. The F and AF components show almost the same behaviour when magnetic field is applied along other directions.

There is one discrepancy between the AF component and the width of the spin-echo spectra. The value of the FWHM extrapolated to zero magnetic field is rather large

(about 200 Oe), whereas the AF component is zero at zero field. This suggests that another mechanism of broadening appears below T_Q. The most probable mechanism is eqQ broadening.

4. Summary

In summary, we have measured the magnetization and ^{121}Sb spin-echo spectra of YbSb to investigate a mixing-type quadrupolar ordering. An abrupt change of the width of the spin-echo spectra at T_Q was observed, while the magnetization curve does not show any significant change. A molecular field calculation based on the crystal field interactions and Γ_3-type quadrupolar interactions shows that the AF components are strongly induced below T_Q while the F components do not show a significant change. In this way, the molecular field calculation offers a qualitative explanation for our experimental results.

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