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Nuclear spin-lattice and nuclear spin-spin relaxation time measurements in EuB_6 at low temperatures using the spin-echo technique

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Abstract. Experimental results on the nuclear spin-lattice and nuclear spin-spin relaxation times in the ferromagnetic EuB_6 at temperatures below 4·2 K are presented using the external magnetic field, H_{ext}, in the range of $0 \leqslant H_{ext} \leqslant 10$ kG. Nuclear spin-spin relaxation time computed on the basis of the Suhl-Nakamura process turns out to be 3·2 μs, which compares well with the experimental value 11·1 μs obtained with the 10 kG magnetic field at 1·7 K. It is found that in the ferromagnetic EuB_6, T_1 is approximately 5×10^3 times larger than T_2 at 1·7 K with the 10 kG magnetic field. Thus the effect of T_1 on T_2 can be neglected. From the experimental value of T_2, the value of the homogeneous line broadening is found to be 14 kHz. The corresponding value obtained from the cw method is 175 kHz. This evidently shows the presence of the inhomogeneous line broadening in the cw NMR.

Keywords. Nuclear spin-lattice relaxation time; nuclear spin-spin relaxation time; spin-echo technique; EuB_6.

1. Introduction

In continuation of our earlier experimental investigations on EuB_6 using the cw NMR technique, we present here our studies on the nuclear spin-spin and nuclear spin-lattice processes using the ^{153}Eu NMR line in the spin-echo (pulsed NMR) experiments. Here, we are mainly concerned with the nuclear spin-spin relaxation time, T_2, to get information about the origin of the width of the lineshape reported by us earlier (Bajaj *et al* 1975, 1976, 1977; Isikawa *et al* 1977).

In the cw NMR, the linewidth is affected by the homogeneous and inhomogeneous broadening. On the other hand, by measuring T_2 using the pulsed NMR, one is able to get information on the homogeneous line broadening, which is very important from the physics point of view. The spin-spin relaxation time, T_2, can be obtained by measuring the echo height of a pair of pulses separated by a variable time, τ.

2. Experimental details

A standard experimental set-up locally designed and fabricated, was used for spin-echo measurements. A monostat device was used for obtaining temperatures below 4·2 K. This device provides a method for changing the pressure and keeping it constant for the duration of the actual measurement. Constancy of pressure secured by this indigenously fabricated floating cup device leads to the constancy of the temperature within 0·006 K.

The samples used in the present work were prepared by the reduction of Eu_2O_3 (purity 99·9%) by boron (purity 99·9%).

$$Eu_2O_3 + 14\,B = 2EuB_6 + B_2O_3.$$

In the first step, Eu_2O_3 and boron were mixed in the desired proportion and pressed in the shape of cylindrical pellets. In the second step, the system was sealed in an evacuated silica tube and then heated to 1100°C for 20 hours in an electric furnace. In this step, the reaction was complete. In the third step, the sample was washed with a 1 : 1 solution of HCl and distilled water and was melted by using a plasma jet. This step eliminated the traces of B_2O_3 remaining in the sample.

The sample thus obtained was checked by x-ray diffraction and magnetization measurements. X-ray diffraction studies clearly showed that our sample has only EuB_6 and does not contain any other phase. The lattice parameter obtained is 4·179Å, which is in agreement with that reported in the literature (4·178Å).

Table 1. Summary of experimental results for EuB_6 in the ferromagnetic region using spin-echo technique.

Temperature (K)	Magnetic field H_{ext} (kG)	Nuclear spin-spin relaxation time (transverse relaxation time) T_2 (μs)
1·7	0	$4·5 \pm 0·4$
	1·5	$4·5 \pm 0·4$
	3·0	$5·7 \pm 0·6$
	6·0	$9·6 \pm 1·0$
	7·5	$11·0 \pm 1·0$
	10·0	$11·0 \pm 1·0$
4·2	0	$4·5 \pm 0·4$
	10	$8·9 \pm 0·9$
		Nuclear spin-lattice relaxation time (T_1) or longitudinal relaxation time (ms)
1·7	10	54 ± 5

3. Results

Experiments for the nuclear spin-spin relaxation time were performed at 1·7 and 4·2 K, respectively. Results are presented in figures 1 and 2. It is clear from figure 1 that the external field up to 10 kG is sufficient to remove the effect of the domain walls. The echo amplitude varies with the pulse interval exponentially, suggesting that the effect of the self-diffusion is rather small. In figure 3, we show the experimental results for T_1 obtained by the saturation recovery method. Although the saturation is not complete, the value of T_1 can be estimated from this figure. Table 1 presents a summary of the values obtained for the nuclear spin-spin and nuclear spin-lattice relaxation times.

4. Discussion

It appears worthwhile here to examine the possibility of having the Suhl-Nakamura interaction mechanism (Suhl 1958, 1959; Nakamura 1959) in EuB_6. In this interaction process, each nuclear spin sees the electronic spin on its own ion, through the effective hyperfine coupling A **I**·**S**. The electronic spins of all the ions are coupled by exchange interaction. An interaction of the nuclei therefore arises via the low-

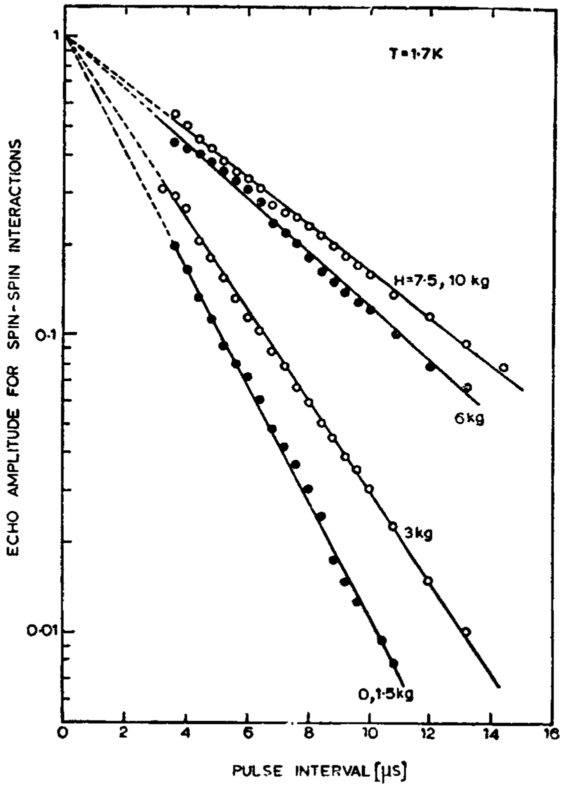

Figure 1. Nuclear spin-spin interaction process in EuB_6 at 1·7 K with different values of the external magnetic field.

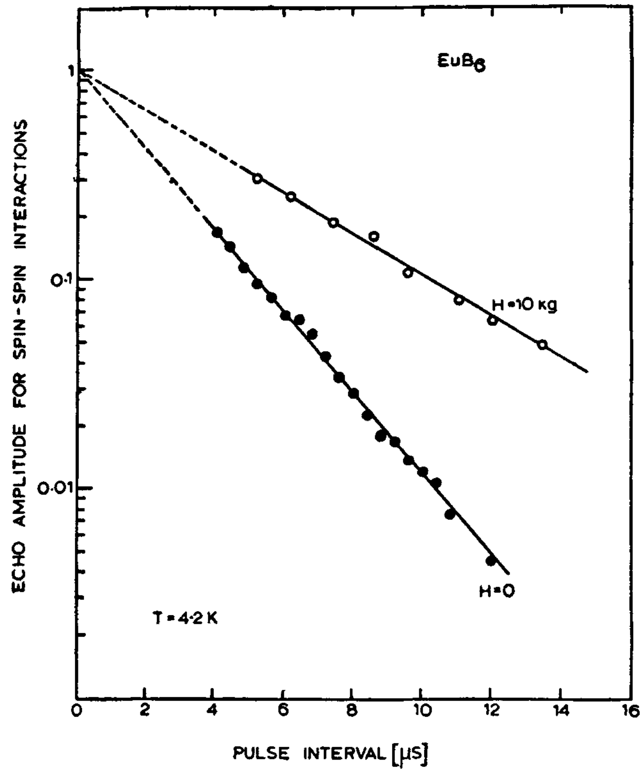

Figure 2. Nuclear spin-spin interaction process in EuB_6 at 4·2 K with and without external magnetic field.

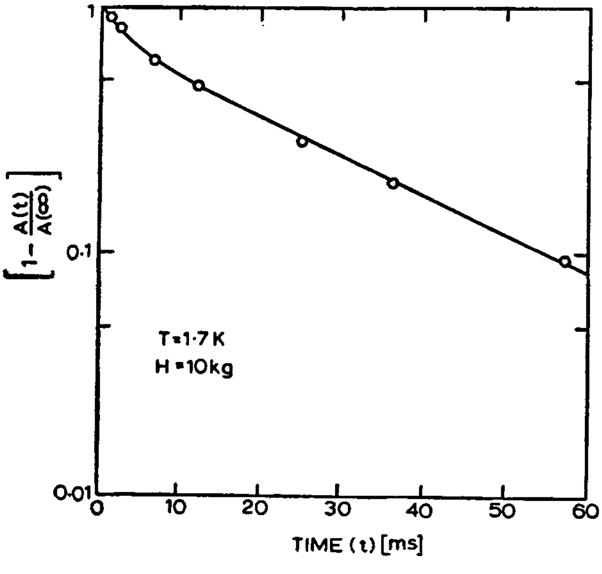

Figure 3. Analysis of the data recorded for the nuclear spin-lattice relaxation time (T_1) at 1·7 K with an external magnetic field of 10 kG using the saturation recovery method.

lying excited states (spin waves) of the electronic system as intermediate states. That is to say, a nuclear spin excites a spin wave through the hyperfine coupling and another nuclear spin causes it to be reabsorbed through its hyperfine coupling. For a cubic ferromagnet, summation of this process over all the possible virtual spin-wave states leads to nuclear interaction of the form

$$H_{\text{eff}} = -\frac{A^2 S}{8\pi g \mu_B H_{\text{exch}}} \sum_{i \neq j} \left\{ \frac{a}{r_{ij}} \exp\left[-(H_{\text{int}}/H_{\text{exch}})^{1/2} \; r_{ij}/a \right] I_i^{-} I_j^{+} \right\} \quad (1)$$

where r_{ij} is the distance between the sites i and j, a the lattice spacing, A the hyperfine coupling constant, S the ionic spin, H_{exch} the effective exchange field, H_{int} the effective dc field (i.e., the applied steady field minus the demagnetization field). I_i stands for $I_i^x \pm iI_i^y$, where I_i^x, I_i^y are the nuclear spin components in a plane normal to the quantization direction, which is taken to be the direction of H_{int}. Demagnetization effects other than in H_{int}, have been neglected, the energy of a spin wave quantum wave number k being taken in the form

$$\hbar\omega_k = g\mu_B(H_{\text{int}} + H_{\text{exch}} \; a^2k^2). \quad (2)$$

The Suhl-Nakamura interaction leads to a nearly Gaussian line profile, with root-mean-square width given by the Van Vleck formula. The range of this interaction depends on the relative magnitude of the exchange energy and the anisotropy energy. The contribution of the transverse relaxation rate due to the Suhl-Nakamura interaction process is given by

$$T_{SN}^{-1} = a[I(I+1)/(24\pi)]^{1/2} \, (\omega_{\text{exch}}/\omega_{\text{int}})^{1/4} \, [A^2S/\hbar^2\omega_{\text{exch}}]. \quad (3)$$

In our case, exchange frequency, $\omega_{\text{exch}} = 2JS/\hbar = 1{\cdot}61 \times 10^{11}\ s^{-1}$, $A = h\nu/S = 2.84 \times 10^{-19}$ erg, $\omega_{\text{int}} = 2\pi\gamma M_s = 10^{11}\ s^{-1}$, $I = 5/2$, $S = 7/2$, $a =$ natural abundance of ^{153}Eu $= 0.52$. Using these values, it is found that the nuclear spin-spin relaxation time due to the Suhl-Nakamura interaction mechanism is 3·2 μs. In spite of the rough estimation, this value is rather near to our experimental value of 11.1 μs. It is therefore regarded, that the nuclear spin-spin relaxation process in this sample may originate due to the interaction mechanisms of the Suhl-Nakamura type. However, it is difficult to say anything about the exact contribution of the Suhl-Nakamura process in the ferromagnetic EuB_6. To be more precise, a good explanation is provided by this type of process qualitatively while the quantitative agreement is not so encouraging in the present case. Experiments and theoretical studies on the antiferromagnetic state of EuB_6 have been presented elsewhere (Bajaj 1977).

So far we have considered only the transverse relaxation time, T_2. Actually the transverse relaxation time, T_2, also contains the effect of the longitudinal relaxation time, T_1 (Bloch 1946). But in EuB_6, T_1 is approximately 5×10^3 times larger than T_2 at 1·7 K with 10 kG magnetic field. Therefore we can ignore the effect of T_1 on T_2.

From the value of T_2 obtained from the experiment, it is found that the homogeneous line broadening in EuB_6 is about 14 kHz. On the other hand, the linewidth obtained from the cw method is 175 kHz. This evidently shows the presence of the inhomogeneous line broadening in the cw NMR.

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