1. 8. PAC Spectroscopy on Tb at 77K and RT

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Introduction

In a previous report in this volume\(^1\), we have shown calculated results of the nuclear energy levels and the transition probability among them when both the magnetic and electric terms are present. There, the cases where the magnetic and electronic interactions have a comparable magnitude were studied and hence an exact calculation was necessary.

Here in this paper, we will report a result of PAC spectroscopy on Tb at RT and 77K. Since Tb has a hcp structure, an EFG with \(\eta=0\) should be present at the probe site at RT where it is paramagnetic. Also the \(V_{xx}\) axis in a hcp structure has been known to be parallel to the crystal c-axis. On the other hand, it is ferromagnetic at 77K with keeping the hcp structure. So we should have both the magnetic and electric interactions at 77K in Tb. By measuring the PAC spectrum at both temperatures, therefore, we can study how these two interactions will be reflected at PAC spectrum. Our main concern is to determine the angle between the \(V_{xx}\) axis and the magnetization direction, \(\beta\) (see Fig. 1(a) in Ref.(1)).

As will be shown later, the magnitude of the magnetic interaction at 77K was found much larger than the electric for Tb. So we are in the range where \(\gamma(=\omega_E/\omega_0)>>20\), which is a limiting case of the calculated ones in the previous paper where the cases \(\gamma<20\) were studied. As will be shown later, the exact calculation results are found to converge to the present limiting case quite nicely, of which details will be described in the present.

Experimental

PAC source of \(^{111}\text{In}+\text{Tb}\) was prepared either by the melting or a diffusion method. Since almost the same results were obtained for the specimens prepared by the two methods, we will show that by the melting method.

PAC spectrum of Tb at RT and 77K

Fig. 1(a) shows the PAC spectrum for Tb measured at RT in the paramagnetic region and Fig. 1(b) at 77K in ferromagnetic region. The RT spectrum is characterized by the low frequency component of which period is about 240ns. On the other hand the 77K spectrum is characterized by a high frequency component of which period is 16.5ns, though
the amplitude of the spectrum shows a trend of a beating. The beating indicates that the spectrum consists of the several signals of which frequencies are different by a small amount from each other.

Fig. 2(a) and (b) shows the corresponding Fourier spectra to show that the RT spectrum consists of 3 different frequencies of $27(\omega_i)$, $54(2\omega_i)$ and $81 \text{Mrad/s}(3\omega_i)$. This is an expected result for the electric interaction for $I=5/2$ spin in the EFG with $\eta=0$. On the other hand, the 77K Fourier spectrum shows two components at 356 and 408 Mrad/s. The average of them, $382 \text{Mrad/s}$, gives the absolute magnitude of the hyperfine field for Tb at 77K as $27T$.

Most importantly in the present, the separation between the two components at 77K, $52 \text{Mrad/s}$, is found very close to the value of $2\omega_i = 54 \text{Mrad/s}$ of the spectrum at RT. This observation will be discussed in terms of a simplified model of the magnetic sublevels splitting due to the quadrupole interactions and the transition probabilities among them in the next section.

No difference was observed in the spectrum with the application of an external magnetic field (0.27 T), Fig. 1(e) and Fig. 2(c), or after the removal of a thin oxide layer from the specimen surface, Fig. 1(d) and Fig. 2(d). The former indicates that the field strength is too weak to align the magnetization direction. The latter indicates that the spectrum is due to the $^{111}$In in the bulk metallic phase.

**Discussion**

**Range of $y$**

The ratio of the observed $\omega_H$ to $\omega_i$ is about 15. Since $\omega_i=6\omega_e$, the ratio $(\omega_H/\omega_e)=y$ is as high as 90 far above the range calculated in the previous paper, namely $y<20$. So we may use an approximation where it is valid when $\omega_H >> \omega_e$.

**Analysis of the results on a simplified model**

In the limit $\omega_H >> \omega_e$, the eigenvalues for spin I in both a magnetic field and an EFG with $\eta=0$ is given by eq.(1); 

$$E_m = -\omega_H m + \omega_e (1/2)(3 \cos^2 \beta - 1) (3m^2 - I(I+1))$$  \hspace{1cm} (1)

$\omega_H$ and $\omega_e$ are magnetic and electric interaction frequency, respectively. Explicitly, $\omega_H=g_eH/h$ and $\omega_e=V_{zz}/h$ 4I(2I-1). $\beta$ is the angle between EFG principal axis and the magnetization direction and all other symbols have conventional meanings. This approximation is equivalent to neglect the off diagonal terms in the hamiltonian matrix elements. (Eq.(1) in Ref.(1)). Then the transition frequencies $(E_m-E_n/h)$ for $m-m'=1$ transitions are given by Eq.(2)
(m=5/2⇒3/2) \quad \omega_{H} + 6\omega_{q}(3 \cos^{2}\beta-1) \\
(m=3/2⇒1/2) \quad \omega_{H} + 3\omega_{q}(3 \cos^{2}\beta-1) \\
(m=1/2⇒-1/2) \quad \omega_{H} \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \quad \q
Comparison with the exact calculation

Finally we will compare the present results of a high magnetic field limit with those of the exact calculation. The six magnetic sublevels was found to increase almost linearly for $y>20$ with the same quadrupole splitting as given by Eq.(2). (see Fig. 1(a) in Ref.(1)) in the exact calculation. So the Eq.(2) in the present should be a good approximation for the magnetic sublevels when $y>20$.

Also the transition probability for $m-m'=1$ (see Fig. 2(c) in ref.(1) ) converges to the same values as in eq.(4) in the present for $(5/2\leftrightarrow 3/2), (-5/2\leftrightarrow -3/2)$ as well as $(3/2\leftrightarrow 1/2), (-3/2\leftrightarrow -1/2)$ transitions with increasing $y$. Also the probability for $(1/2\leftrightarrow -1/2)$ transition tends to be null with the increasing $y$, which also shows a good agreement with the present approximation. So one can safely use the high magnetic field approximation if $y>20$.

For $y<20$, however, one can not use the approximations and must use the results of the exact calculation, since the magnetic sublevels as well as the transition probabilities are considerably different from those in the high field limit. (see Fig. 2-4 in Ref.1).

References

1) Hanada R., see an article in this volume.
3) Legvold S., "Rare Earth Metals and Alloys" in Ferromagnetic Materials vol.1 ed. by Wohlfarth EP., pub. by North-Holland pub.co.(1980).

Fig. 1. PAC spectrum for Tb.
(a) measured at RT. (b) measured at 77K. (c) 77K with vertical magnetic field with no change in the spectrum. (d) surface oxide removed with no change in the spectrum.

Fig. 2. Fourier spectrum for Fig.1 Note that the $2\omega_i$ at RT is almost equal to the separation at 77K. For details, see text.