SPIN RELAXATION AND BREIT-RABI SPECTRA OF CUBIC Yb SALT

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We have recently shown from the Mössbauer studies of $^{170}$Yb in Cs$_2$NaYbCl$_6$ that the Yb$^{3+}$ ion experiences a cubic crystalline electric field (CEF) in this compound [1]. From the observed paramagnetic hyperfine structure it was established that the ground CEF level is a $T_6$ state, in agreement with susceptibility measurements [2]. We report in the present work the study of relaxation spectra as a function of temperature and of applied magnetic field.

The primary relaxation mechanisms we are concerned with in such an undiluted chemical compound arise through spin-lattice and spin-spin coupling. The former mechanism is strongly temperature dependent [3-4], while the latter can be dependent on magnetic field [5]. The experimentally measured relaxation time ($\tau$) is then related to the spin-lattice ($\tau_l$) and to the spin-spin ($\tau_s$) relaxation times through the relation

$$\tau^{-1} = (\tau_l)^{-1} + (\tau_s)^{-1}$$

$\tau_l$ is dependent on various phonon scattering mechanisms, viz. direct, Raman and Orbach processes. The value of $\tau_l$ is mainly governed by the distance between the paramagnetic atoms in the lattice. In the case $\tau_s$ and $\tau_l$ are of comparable magnitudes, $\tau$ itself would be temperature dependent. Such a situation is rather common in rare-earth compounds as has been demonstrated in the case of YbCl$_3$, 6 H$_2$O [6].

We have measured the temperature dependence of Mössbauer resonance spectra of Cs$_2$NaYbCl$_6$ between 1.6 and 40 K using the 84 keV transition in $^{170}$Yb. In figure 1a we show the spectrum measured at 4.2 K. The asymmetric pattern with two resonance lines is characteristic of the $T_6$ CEF level [1]. The shape of the spectrum did not change up to 20 K. A rapid decrease in the resonance fraction did not permit discernable measurements above 20 K.

In order to understand the relaxation spectra of the $T_6$ state with isotropic hyperfine coupling can be done using Hirst's theory [7]. We have however preferred to use the closed form equations provided by Gonzalez-Jimenez et al. [8]. The solid curves in figure 1a show the least-squares fit to the data using this theory. The resulting relaxation time was found to be fairly constant in the temperature range from 1.6 to 20 K, with a value of $5 \times 10^{-9}$ s.

To describe the influence of the field on the static hyperfine structure of the $T_6$ CEF level, Small fields lift the degeneracy of the two hf components shown in figure 1a, so that 20 transitions become possible. The field dependence given by the hamiltonian

$$\mathcal{H} = A_s[V\cdot S - hS_z]$$

with $h = g\mu_B H_{ext}/A_s$ and $A_s = -10.9 \pm 0.1$ mm/s, has been depicted using a Breit-Rabi diagram (Fig. 2) [9]. Figure 2 represents the energies and the intensities of the 20 transitions as a function of the external field. The details relative to such a diagram are given in [10]. The angular distribution of the various hf transitions however limits the number of observable...
ones to eight (8^-, 3^+, 4^-, 7^+, 9^-, 2^+, 6^+, 5^-) in Fig. 2) if the external field is parallel to the gamma ray absorption direction. With increasing field, four (9^-, 5^-, 2^+, 6^+) of these gradually reduce in intensity. In very high fields (~30 kG), the remaining four transitions merge into two (see Fig. 2) and the result is typical of paramagnetic hyperfine spectra with \( g_s \neq 0, \quad g_s = g_d = 0 \) observed parallel to the \( z \) axis. Increasing the field basically results in gradually decoupling the effective spin \( S = \frac{1}{2} \) of the \( \Gamma_6 \) level and the nuclear spin \( I \); finally in high fields we reach the Paschen-Back region where the electron-nuclear system is completely decoupled. It must be pointed out that the isotropic character essential to a Breit-Rabi description of the system can be perturbed by the mixing of higher CEF levels in an external field. However this is not the situation in CsNaYbCl₆ in which the first excited CEF level is a few hundred degrees above the \( \Gamma_6 \) level [11].

In an isotropic electron-nuclear coupled system the hyperfine energies depend on the applied field, which leads to a field dependent nuclear correlation time \( \tau_n \). In order to study the variation of \( \tau_n \) with field one has thus to include the dependence of hyperfine frequencies on the field in the relaxation theory. Such a theory is available from Gonzalez-Jimenez et al. [8] and we have used it to least-squares fit the data obtained in small fields (see for example, Fig. 1b). The analysis of the spectrum measured at 45 kG (Fig. 1c) is
TABLE I

Spin relaxation rate for Yb\(^{3+}\) in Cs\(_2\)NaYbCl\(_6\)
at various temperatures and external magnetic fields

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>(H_{ext}) (kG)</th>
<th>(\tau) (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.6</td>
<td>0</td>
<td>4.9 ± 0.5</td>
</tr>
<tr>
<td>4.2</td>
<td>0</td>
<td>4.7 ± 0.5</td>
</tr>
<tr>
<td>20</td>
<td>0</td>
<td>4.5 ± 0.8</td>
</tr>
<tr>
<td>1.6</td>
<td>0.30</td>
<td>6 ± 1</td>
</tr>
<tr>
<td>4.2</td>
<td>0.30</td>
<td>6 ± 1</td>
</tr>
<tr>
<td>4.2</td>
<td>0.40</td>
<td>8 ± 2</td>
</tr>
<tr>
<td>4.2</td>
<td>0.85</td>
<td>12 ± 3</td>
</tr>
<tr>
<td>4.2</td>
<td>45</td>
<td>&gt; 15</td>
</tr>
</tbody>
</table>

however done using an effective field relaxation theory [3] and including a Boltzmann distribution over the electronic levels. We find that the resulting relaxation times depend on the field and increase with increasing field. In fact the situation is such that the spectrum in 400 G may be considered nearly static i.e. \(\tau \gg \tau_0\). In figure 1b we have shown the 8 transitions with appropriate intensities as calculated from figure 2 for this particular case. The typical relaxation times with and without the field are larger than \(\tau_0\) (\(\approx 10^{-10}\) s). Hence the values of \(\tau\) deduced from the analysis (and given in Table I) are susceptible to large errors.

In Rb\(_2\)NaYbF\(_6\), \(\tau\) is of the order of \(10^{-9}\) s at 4.2 K in zero applied field so that the influence of field can be more quantitatively studied in this case [12].

In conclusion one may point out that the observed variation in \(\tau\) with field is a manifestation of the field dependence of \(\tau_0\); however as \(\tau_0\) decreases it may become comparable to \(\tau_0\) so that the nature of the \(\tau\) dependence on field cannot be solely associated with \(\tau_0\).

References


[11] Although in reference [2] the separation of excited state is given to be 85 K, a careful reanalysis of the susceptibility data yields this separation to be at least 450 K.