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SPIN RELAXATION AND BREIT-RABI SPECTRA OF CUBIC Yb SALT (*)

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Résumé. — Le spectre Mössbauer de ^{170}Yb dans le composé cubique $\text{Cs}_2\text{NaYbCl}_6$ a été mesuré en fonction de la température et d'un champ magnétique externe. Le spectre hyperfin du niveau fondamental Γ_6 est fortement influencé par de faibles champs magnétiques. Les détails sont décrits par un diagramme de Breit-Rabi. La fréquence de relaxation de spin de Yb^{3+} dépend du champ externe mais est indépendante de la température jusqu'à 20 K.

Abstract. — The Mössbauer spectrum of ^{170}Yb in cubic $\text{Cs}_2\text{NaYbCl}_6$ has been measured as a function of temperature and external field. The hyperfine spectrum due to Γ_6 CEF level, which is the ground state in this compound, is strongly influenced by small magnetic fields. The details are described using a Breit-Rabi diagram. The spin relaxation time of Yb^{3+} is strongly dependent on the external field but is independent of temperature up to 20 K.

We have recently shown from the Mössbauer studies of ^{170}Yb in $\text{Cs}_2\text{NaYbCl}_6$ 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 Γ_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 (τ) is then related to the spin-lattice (τ_1) and to the spin-spin (τ_s) relaxation times through the relation

$$(\tau^{-1}) = (\tau_1)^{-1} + (\tau_s)^{-1} \quad (1)$$

τ_1 is dependent on various phonon scattering mechanisms, viz. direct, Raman and Orbach processes. The value of τ_s is mainly governed by the distance between the paramagnetic atoms in the lattice. In the case τ_1 and τ_s are of comparable magnitudes, τ itself would be temperature dependent. Such a situation is rather common in rare-earth compounds as has been demonstrated in the case of $\text{YbCl}_3 \cdot 6\text{H}_2\text{O}$ [6].

We have measured the temperature dependence of Mössbauer resonance spectra of $\text{Cs}_2\text{NaYbCl}_6$ 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 Γ_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.

The analysis of the paramagnetic relaxation spectra of the Γ_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 5×10^{-9} s.

In order to understand the relaxation spectra of this compound in applied magnetic fields one has to describe the influence of the field on the static hyperfine structure of the Γ_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

$$\mathcal{H} = A_s[\bar{I} \cdot \bar{S} - hS_z] \quad (2)$$

with $h = g\mu_B H_{\text{ext}}/A_s$, 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

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FIG. 1. — Mössbauer spectra of $\text{Cs}_2\text{NaYbCl}_6$ at 4.2 K for different external fields: a) 0 kG, b) 0.4 kG, c) 45 kG.

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 $g_z \neq 0$, $g_x = g_y = 0$ observed parallel to the z axis. Increasing the field basically results in gradually decoupling the effective spin $S = \frac{1}{2}$ of the Γ_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 situa-

FIG. 2. — Breit-Rabi diagram: a) energies and b) intensities of the Mössbauer transitions as a function of external magnetic field.

tion in $\text{Cs}_2\text{NaYbCl}_6$ in which the first excited CEF level is a few hundred degrees above the Γ_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 (τ_c). In order to study the variation of τ 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 Yb³⁺ in Cs₂NaYbCl₆ at various temperatures and external magnetic fields

Temperature K	H_{ext} kG	τ ns
1.6	0	4.9 ± 0.5
4.2	0	4.7 ± 0.5
20	0	4.5 ± 0.8
1.6	0.30	6 ± 1
4.2	0.30	6 ± 1
4.2	0.40	8 ± 2
4.2	0.85	12 ± 3
4.2	45	> 15

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. $\tau \gg \tau_c$. 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 τ_c ($\approx 10^{-10}$ s). Hence the values of τ deduced from the analysis (and given in Table I) are susceptible to large errors.

In Rb₂NaYbF₆, τ 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 τ with field is a manifestation of the field dependence of τ_s ; however as τ_s decreases it may become comparable to τ_1 so that the nature of the τ dependence on field cannot be solely associated with τ_s .

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