

Effect of lattice optical vibrations in the spin-lattice relaxation of Ce^{3+} and Yb^{3+} in BaY_2F_8 single crystals

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The temperature, orientation, and field dependences of the spin-lattice relaxation times of Ce^{3+} and Yb^{3+} in BaY_2F_8 single crystals are investigated. The luminescence and Raman scattering of light are also studied. It is concluded that optical phonons of energy $\sim 60 \text{ cm}^{-1}$ make a substantial contribution to the rate of the spin-lattice relaxation of the systems on top of the contribution of the acoustic phonons. A field dependence, determined by the optical phonons, of the spin-lattice relaxation rate is observed for the Yb^{3+} ion for the first time.

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In investigations of spin-lattice relaxation (SLR) of paramagnetic impurities in crystals, exponential temperature dependences of the SLR rate are frequently observed, of the form

$$T_1^{-1} = B \exp(-\Delta/kT). \quad (1)$$

As a rule, this dependence is attributed to the substantial role played in SLR by Urbach-Aminov processes through the excited Stark level with energy $\Delta < \Theta$, where Θ is the Debye temperature of the crystal.¹ However, similar exponential T_1^{-1} dependences are possible also for Raman two-phonon processes if the phonon spectrum of the crystal contains either local or quasilocal impurity vibrations of noticeably pronounced intensity,²⁻⁴ or else optical oscillations of the lattice at the frequency $\omega = \Delta/\hbar$ (Ref. 5). Several papers have already been published⁵⁻⁸ in which the observation of the dependence of the type (1) is attributed to optical oscillations of the crystal. It must be borne in mind, however, that only a combination of measurements of the spin-lattice relaxation and the investigation by other methods can lead to sufficiently reliable conclusions concerning the nature of the temperature dependence (1).

The purpose of the present work was an all-inclusive study of the Raman processes of SLR in BaY_2F_8 single crystals. The crystals were activated with Ce^{3+} and Yb^{3+} ions, in which the distance to the nearest excited Stark levels is usually sufficiently large, so that the Urbach-Aminov relaxation processes are not very effective. As a result of the investigation we have succeeded in showing that the observed dependences of the type (1) with $\Delta \approx 60 \text{ cm}^{-1}$ are due for both ions to Raman processes through optical vibrations of the lattice.

Data on the study of EPR of rare-earth ions in BaY_2F_8 are contained in our preceding paper.⁹ We present here only the principal values of the g -factors: $g_x = 2.702$, $g_y = 2.143$, $g_z \leq 0.74$ for Ce^{3+} and $g_x = 1.994$, $g_y = 2.063$, $g_z = 5.049$ for Yb^{3+} . Measurements of the SLR time T_1 were carried out for $BaY_2F_8 + 0.5\% Ce^{3+}$ and $BaY_2F_8 + 0.1\% Yb^{3+}$ samples in the temperature range 1.5–40 K at frequencies 9.4 and 24.2 GHz and at different orientations of the external magnetic field H relative to the crystal axes. Times T_1 longer than 10^{-5} sec were measured by the pulsed saturation meth-

od, while those shorter than 10^{-7} sec were determined from the spin-phonon broadening of the EPR lines.

For the Ce^{3+} ion, the measurements of T_1 were carried out in the orientation $H \parallel x$ (see Fig. 1), and the results agree well with expressions of the type

$$T_1^{-1} = AT + CT^9 + Be^{-\Delta/T}. \quad (2)$$

In this case the parameters are $C = 7.4 \cdot 10^{-6} \text{ sec}^{-1} \cdot \text{K}^{-9}$, $B = 2.8 \cdot 10^9 \text{ sec}^{-1}$, $\Delta = 92 \text{ K} \equiv 64 \text{ cm}^{-1}$ and do not depend on the frequency, while $A = 0.4 \text{ sec}^{-1} \cdot \text{K}^{-1}$ for 9.4 GHz and $A = 4 \text{ sec}^{-1} \cdot \text{K}^{-1}$ for 24.2 GHz. For the Yb^{3+} ions, T_1 was measured in the orientations $H \parallel z$ and $H \parallel y$ (see Fig. 2). The results are well described by an expression of the type

$$T_1^{-1} = A_1 T + C_1 T^9 + B_1 e^{-\Delta_1/T} + B_2 e^{-\Delta_2/T}. \quad (3)$$

It was found that the parameters are $C_1 = 2.9 \cdot 10^{-6} \text{ sec}^{-1} \cdot \text{K}^{-9}$, $B_2 = 6 \cdot 10^{12} \text{ sec}^{-1}$, $\Delta_1 = 89 \text{ K} \equiv 62 \text{ cm}^{-1}$, $\Delta_2 = 294 \text{ K} \equiv 204 \text{ cm}^{-1}$ on either the orientation or the frequency, while the parameters A_1 and B_1 depend both on the orientation and on the frequency and their values are $A_1 = 0.14 \text{ sec}^{-1} \cdot \text{K}^{-1}$, $B_1 = 5 \cdot 10^7 \text{ sec}^{-1}$ for 9.4 GHz and

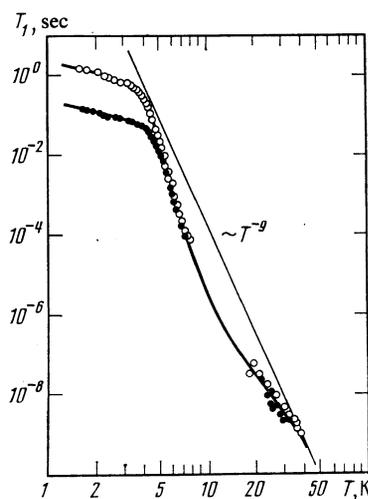


FIG. 1. Temperature dependence of the time T_1 of the Ce^{3+} ions in BaY_2F_8 at $H \parallel x$: $\circ - \nu = 9.4 \text{ GHz}$, $\bullet - \nu = 24.2 \text{ GHz}$. Solid lines—calculation by formula (2).

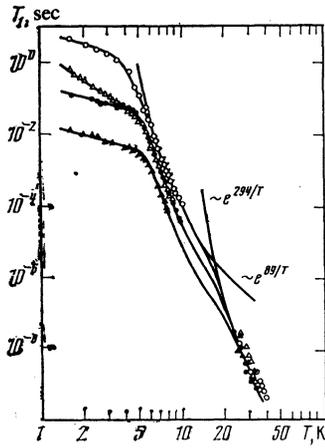


FIG. 2. Temperature dependence of the time T_1 of the ions Yb^{3+} in BaY_2F_8 : \circ — $\text{H}\parallel z$, $\nu = 9.4$ GHz; \bullet — $\text{H}\parallel z$, $\nu = 24.2$ GHz; \triangle — $\text{H}\parallel y$, $\nu = 9.4$ GHz; \blacktriangle — $\text{H}\parallel y$, $\nu = 24.2$ GHz. Solid lines—calculation by formula (3).

$A_1 = 4.3 \text{ sec}^{-1} \cdot \text{K}^{-1}$, $B_1 = 2 \cdot 10^8 \text{ sec}^{-1}$ for 24.2 GHz at $\text{H}\parallel z$, and $B_1 = 2 \cdot 10^8 \text{ sec}^{-1}$ for 9.4 GHz and $A_1 = 47 \text{ sec}^{-1} \cdot \text{K}^{-1}$, $B_1 = 8.7 \cdot 10^8 \text{ sec}^{-1}$ for 24.2 GHz at $\text{H}\parallel y$. It is seen from Figs. 1 and 2 that below 4 K direct processes predominate in the SLR of the Ce^{3+} and Yb^{3+} ions. For the Yb^{3+} ion in the y orientation at the frequency $\nu = 9.4$ GHz an anomalous dependence $T_1^{-1} = 0.48 T^{2.5}$ is observed and is apparently due to SLR under cross-relaxation conditions.¹⁰ In the temperature region 4–5 K, for a better description of the experimental temperature dependences of T_1^{-1} , we introduced in expressions (2) and (3) terms of the type T^9 , which are obviously due to the usual Raman second-order SLR via acoustic phonons. At temperatures above 5 K, processes that depend exponentially on the temperature (1) begin to predominate in the SLR of Ce^{3+} and Yb^{3+} , with $\Delta = 92$ K for Ce^{3+} and $\Delta_1 = 89$ K for Yb^{3+} . It was verified that there are no angular and field dependences of B for Ce^{3+} . For Yb^{3+} , however, both angular and field dependences of B_1 were observed, and the angular dependence is apparently the derivative of the field dependence, inasmuch as in all the measurements the coefficient B_1 is determined by the value of H . To describe the temperature dependence of T_1 of Yb^{3+} ions in the region $20 < T < 40$ K it was necessary to introduce in (3) a second exponential term. In this case no field dependence was observed here.

As already mentioned, exponential temperature dependences of T_1^{-1} are possible both for Urbach-Aminov processes, and for Raman processes. Without additional experimental data, it is impossible to draw any reliable conclusions concerning the nature of these dependences. Our case is made more complicated also by the fact that for Yb^{3+} the coefficient B_1 is field-dependent. A dependence of this type for exponential relaxation processes was not observed earlier. In addition, attention is called to the equality of the value of Δ for Ce^{3+} to that of Δ_1 for Yb^{3+} (the difference is within the limits of the measurement errors), which suggests that they are of the same origin. Taking all this into account, additional investigations were performed.

To verify the presence or absence of Stark levels with $\Delta_1 \approx 89$ K and $\Delta_2 \approx 294$ K in the case of Yb^{3+} , through which Urbach-Aminov processes can be realized, we plotted for the $\text{BaY}_2\text{F}_8 + 0.1\% \text{Yb}^{3+}$ sample at the temperatures 77 and 4.2 K the luminescence spectra corresponding to the transitions ${}^2F_{5/2} \rightarrow {}^2F_{7/2}$. The spectrometer was an MDR-2 monochromator with a diffraction grating having 600 lines/mm. The investigation has shown that there are no excited levels with energy lower than 80 cm^{-1} in the Stark structure. Consequently the exponential term in (3) with $\Delta_1 = 89 \text{ K} \approx 62 \text{ cm}^{-1}$ cannot be due to the Urbach-Aminov process. In addition, it was observed that four most intense lines stand out in the luminescence spectrum. Assuming that these lines correspond to transitions from the lower level ${}^2F_{5/2}$ of the term to the Stark levels ${}^2F_{7/2}$ of the term, we determined the positions of the ${}^2F_{7/2}$ term, namely 0, 205, 213, and 445 cm^{-1} . The position of the first excited Stark levels ($205 \text{ cm}^{-1} \approx 295$ K) agreed well with the value $\Delta_2 = 294$ K in (3). Since it is necessary also to satisfy the condition $\Delta_2 < \Theta$ to be able to realize the Urbach-Aminov process through this level, we have estimated the value of Θ for BaY_2F_8 using the formula¹¹

$$\Theta = \frac{\hbar}{k} \left[\frac{9q}{4\pi} \left(\frac{N\rho}{M} \right) / \left(\frac{1}{v_l^3} + \frac{2}{v_t^3} \right) \right]^{1/3}, \quad (4)$$

where q is the number of atoms in the molecule, M is the molecular weight, ρ is the crystal density, and v_l and v_t are the longitudinal and transverse sound velocities in the crystal. The structural data¹² yield for BaY_2F_8 the value $\rho = 5.04 \text{ g/cm}^3$. The speeds of sound ($v_l = 5.56 \text{ km/sec}$, $v_t = 2.84 \text{ km/sec}$) were measured by us in a certain arbitrary direction of the crystal by the method of reflected ultrasound pulse at a frequency 13.4 MHz at liquid-nitrogen temperature. The value $\Theta = 394$ K calculated from formula (4) turned out to be larger than the energy 295 K of the first excited Stark level of Yb^{3+} , so that it can be concluded that the second exponential term in (3) is due to the Urbach-Aminov process via a Stark level with $\Delta = 295$ K.

Next, to observe the possible vibrational energy level with $\Delta \approx 90$ K, we investigated the spectra of the Raman scattering of light. These spectra were obtained at room temperature for the following samples: 1) $\text{BaY}_2\text{F}_8 + 0.1\% \text{Yb}^{3+} + 0.1\% \text{Nd}^{3+}$; 2) $\text{BaY}_2\text{F}_8 + 0.05\% \text{Er}^{3+}$; 3) $\text{BaY}_2\text{F}_8 + 0.5\% \text{Er}^{3+}$, using a Raman-scattering spectrometer of the "Coderg" Company (France), in which the exciting light source is an He-Ne laser ($\lambda_0 = 632.8 \text{ nm}$). The scattered light was registered at an angle 90° to the incident light. The paramagnetic impurities in the samples were monitored by an EPR method. For all the samples we observed reliably in the Raman-scattering spectra a line with $\Delta = 60 \text{ cm}^{-1} \approx 86.4$ K, and no influence of impurities were noted on its position and intensity. The results offer evidence of the presence in the vibrational spectrum of BaY_2F_8 crystal of singled-out oscillations with energy 60 cm^{-1} , belonging to the optical branch.

Thus, the good agreement between the values Δ and Δ_1 obtained from the SLR of Ce^{3+} and Yb^{3+} , on the one hand, and the position of the vibrational level that manifests itself

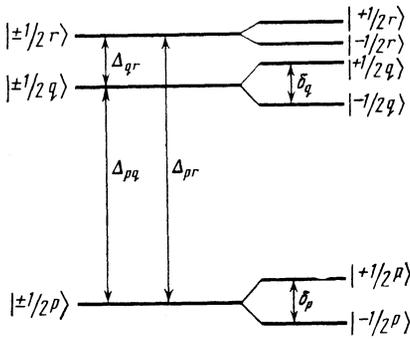


FIG. 3. Energy level scheme of a Kramers ion in the crystalline and magnetic fields.

in the Raman spectra, on the other, as well as the absence of a Stark level for Yb^{3+} , in this region, enable us to conclude that the exponential terms in (2) and (3), with $\Delta \approx \Delta_1 \approx 90 \text{ K} \approx 63 \text{ cm}^{-1}$, are due to Raman two-phonon processes with predominant participation of optical phonons of energy $\sim 60 \text{ cm}^{-1}$ in the relaxation.

Special notice should be taken of the observation, for the Yb^{3+} ion, of a field dependence of the contribution made to the SLR rate by optical vibrations of the lattice, since no such dependence was observed earlier. In Ref. 5 is given a theoretical analysis of the rate of the SLR due to optical lattice vibrations, but the field dependence in this case was not predicted. Orbach¹³ has predicted theoretically that an $H^2 T^7$ dependence will predominate in the rate of the SLR of Kramers ions over the ordinary T^9 dependence, if the Stark structure of the ion has closely lying levels as was indeed observed experimentally in Refs. 14 and 15. Using the procedure of Refs. 5 and 13, we obtained an expression for the rate of the Raman SLR due to optical vibrations of the lattice under condition of closely lying Stark levels of a Kramers ion. The energy levels scheme of the ion is shown in Fig. 3.

For simplicity, just as in Ref. 13, we take into account the contribution made to the SRL only by the first excited level $|\pm \frac{1}{2} q\rangle$ and ignore the contribution from the following levels. Since in our case the second excited state $|\pm \frac{1}{2} r\rangle$ is located close to the first $|\pm \frac{1}{2} q\rangle$, we shall take into account the addition made by the magnetic field to the state $|\pm \frac{1}{2} q\rangle$ from the state $|\pm \frac{1}{2} r\rangle$ (Ref. 13):

$$\begin{aligned} \left| -\frac{1}{2} q' \right\rangle &= \left| -\frac{1}{2} q \right\rangle - \frac{\beta \Delta H \langle -\frac{1}{2} r | \mathbf{J} | -\frac{1}{2} q \rangle}{\Delta_{rq}} \left| -\frac{1}{2} r \right\rangle, \\ \left| +\frac{1}{2} q' \right\rangle &= \left| +\frac{1}{2} q \right\rangle - \frac{\beta \Delta H \langle +\frac{1}{2} r | \mathbf{J} | +\frac{1}{2} q \rangle}{\Delta_{rq}} \left| +\frac{1}{2} r \right\rangle, \end{aligned} \quad (5)$$

where Δ_{rq} is the energy distance between the states $|\pm \frac{1}{2} r\rangle$ and $|\pm \frac{1}{2} q\rangle$. The probability of the transition $|+\frac{1}{2} p\rangle \rightarrow |-\frac{1}{2} p\rangle$ due to the spin-lattice interaction per unit time can be written in the form¹³

$$W_{|+\frac{1}{2} p\rangle \rightarrow |-\frac{1}{2} p\rangle} = \left(\frac{2\pi}{\hbar} \right) \frac{V}{4\pi^4} \iint \left| \left\langle -\frac{1}{2} p \middle| V_{eff} \middle| \frac{1}{2} p \right\rangle \right|^2 \times \delta(\hbar\omega_{nm} - \hbar\omega_{n'm'} - \delta_p) k_{nm}^2 k_{n'm'}^2 dk_{nm} dk_{n'm'}, \quad (6)$$

where

$$\begin{aligned} & \left| \left\langle -\frac{1}{2} p \middle| V_{eff} \middle| \frac{1}{2} p \right\rangle \right| \\ &= \left| \sum_{\substack{n, m, \\ n', m'}} \left\{ \frac{\langle -\frac{1}{2} p | V_n^m \epsilon_{nm} | \frac{1}{2} q' \rangle \langle \frac{1}{2} q' | V_n^{m'} \epsilon_{n'm'} | \frac{1}{2} p \rangle}{-\Delta_{pq} + \hbar\omega_{n'm'}} \right. \right. \\ & \left. \left. + \frac{\langle -\frac{1}{2} p | V_n^{m'} \epsilon_{n'm'} | -\frac{1}{2} q' \rangle \langle -\frac{1}{2} q' | V_n^m \epsilon_{nm} | \frac{1}{2} p \rangle}{-\Delta_{pq} - \hbar\omega_{nm}} \right\} \right|. \quad (7) \end{aligned}$$

The last expression is written under the assumption that the quantity $(\delta_p + \delta_q)/2$ is negligibly small compared with Δ_{pq} and with the energies of the created ($\hbar\omega_{nm}$) and of the annihilated ($\hbar\omega_{n'm'}$) phonons. By ϵ_{nm} we mean here the same as in Ref. 13, namely the average deformation which is independent of the direction. Substituting (5) in (7) and taking terms of first order only into account, relative to $(\beta \Delta H / \Delta_{rq})$, we obtain

$$\begin{aligned} & \left| \left\langle -\frac{1}{2} p \middle| V_{eff} \middle| \frac{1}{2} p \right\rangle \right| \\ &= \left| \sum_{\substack{n, m, \\ n', m'}} \left[P_{nn'}^{mm'} \left(\frac{1}{-\Delta_{pq} + \hbar\omega_{n'm'}} - \frac{1}{-\Delta_{pq} - \hbar\omega_{nm}} \right) \right. \right. \\ & \left. \left. - \frac{\beta \Delta H \langle \frac{1}{2} r | \mathbf{J} | \frac{1}{2} q \rangle}{\Delta_{rq}} Q_{nn'}^{mm'} \left(\frac{1}{-\Delta_{pq} + \hbar\omega_{n'm'}} + \frac{1}{-\Delta_{pq} - \hbar\omega_{nm}} \right) \right] \right| \\ & \times \left| \frac{\hbar k_{nm} k_{n'm'} N_{n'm'}^{1/2} (N_{nm} + 1)^{1/2}}{2M (\omega_{nm} \omega_{n'm'})^{1/2}} \right|, \quad (8) \end{aligned}$$

where

$$\begin{aligned} P_{nn'}^{mm'} &= \langle -\frac{1}{2} p | V_n^m | \frac{1}{2} q \rangle \langle \frac{1}{2} q | V_n^{m'} | \frac{1}{2} p \rangle, \\ Q_{nn'}^{mm'} &= \langle -\frac{1}{2} p | V_n^m | \frac{1}{2} q \rangle \langle \frac{1}{2} r | V_n^{m'} | \frac{1}{2} p \rangle \\ & + \langle -\frac{1}{2} p | V_n^m | \frac{1}{2} r \rangle \langle \frac{1}{2} q | V_n^{m'} | \frac{1}{2} p \rangle. \end{aligned} \quad (9)$$

In (8) we took into account the fact that

$$\begin{aligned} | \langle N+1 | \epsilon_{nm} | N \rangle | &= k_{nm} [\hbar(N_{nm} + 1) / 2M\omega_{nm}]^{1/2}, \\ | \langle N-1 | \epsilon_{n'm'} | N \rangle | &= k_{n'm'} [\hbar N_{n'm'} / 2M\omega_{n'm'}]^{1/2}, \\ N_{nm} &= [\exp(-\hbar\omega_{nm}/kT) - 1]^{-1}, \end{aligned} \quad (10)$$

where M is the mass of the crystal. For optical phonons we shall use for the wave vector a simplified dispersion relation of the type⁵

$$k = \pi(\omega_0 - \omega) / 2a\Delta\omega, \quad (11)$$

where ω_0 is the angular frequency of the optical phonons with $k = 0$, a is the interatomic distance, and $\Delta\omega$ is the width of the band of the optical lattice vibrations. Then, substituting (8) and (11) in (6) and performing slight transformations under the assumption that the Zeeman splitting of the level $|\pm \frac{1}{2} p\rangle$ is $\delta_p \ll \Delta\omega$, and that $\Delta\omega \ll \omega_0$, we obtain

$$\begin{aligned}
& W_{|1/2p\rangle \rightarrow |-1/2p\rangle} \\
&= \left(\frac{\pi}{2a\Delta\omega} \right)^{10} \frac{1}{8\pi^3\rho^2} \left| \sum_{\substack{n,m \\ n',m'}} [P_{nn'}^{mm'} \left(\frac{1}{-\Delta_{pq} + \hbar\omega_0} - \frac{1}{-\Delta - \hbar\omega_0} \right) \right. \right. \\
&\quad \left. \left. - \frac{\beta\Delta\mathbf{H}\langle 1/2r|\mathbf{J}|1/2q\rangle Q_{nn'}^{mm'}}{\Delta_{rq}} \left(\frac{1}{-\Delta_{pq} + \hbar\omega_0} + \frac{1}{-\Delta_{pq} - \hbar\omega_0} \right) \right] \right|^2 \\
&\quad \times \frac{e^{\hbar\omega_0/kT}}{\omega_0^2 (e^{\hbar\omega_0/kT} - 1)^2} \int_{\omega_0 - \Delta\omega}^{\omega_0} (\omega_0 - \omega)^8 d\omega, \quad (12)
\end{aligned}$$

where $\rho = M/V$ is the density of the crystal. We introduce next the notation

$$\sum_{\substack{n,m \\ n',m'}} P_{nn'}^{mm'} \equiv P, \quad \sum_{\substack{n,m \\ n',m'}} Q_{nn'}^{mm'} \equiv Q, \quad \hbar\omega_0 \equiv \Delta_0,$$

where \mathbf{h} is a unit vector of the magnetic field \mathbf{H} . Assuming that $\Delta_0^2 \ll \Delta_{pq}^2$ and $kT \ll \Delta_0$, we obtain ultimately an expression for the rate of the SLR:

$$\begin{aligned}
T_1^{-1} &= 2W_{|1/2p\rangle \rightarrow |-1/2p\rangle} = \frac{\pi^7 \hbar^2 |P|^2}{9 \cdot 2^{10} a^{10} \rho^2 \Delta\omega \Delta_{pq}^4} \\
&\times \left| 1 - \frac{\beta\Delta\langle 1/2r|\mathbf{hJ}|1/2q\rangle \Delta_{pq} Q}{\Delta_0 \Delta_{rq} P} H \right|^2 \exp\left(-\frac{\Delta_0}{kT}\right). \quad (13)
\end{aligned}$$

At

$$\left| \frac{\beta\Delta\langle 1/2r|\mathbf{hJ}|1/2q\rangle \Delta_{pq} Q}{\Delta_0 \Delta_{rq} P} H \right| \ll 1$$

we obtain the expression

$$T_1^{-1} = \frac{\pi^7 \hbar^2 |P|^2}{9 \cdot 2^{10} a^{10} \rho^2 \Delta\omega \Delta_{pq}^4} \exp\left(-\frac{\Delta_0}{kT}\right), \quad (14)$$

which is similar to (15) in Ref. 5, differing only by the coefficient in front of the exponential, due to the error incurred in Ref. 5. If

$$\left| \frac{\beta\Delta\langle 1/2r|\mathbf{hJ}|1/2q\rangle \Delta_{pq} Q}{\Delta_0 \Delta_{rq} P} H \right| \gg 1,$$

then

$$\begin{aligned}
T_1^{-1} &= \frac{\pi^7 \hbar^2 |\beta\Delta\langle 1/2r|\mathbf{hJ}|1/2q\rangle Q|^2}{9 \cdot 2^{10} a^{10} \rho^2 \Delta\omega \Delta_{pq}^2 \Delta_0^2 \Delta_{rq}^2} \\
&\times H^2 \exp\left(-\frac{\Delta_0}{kT}\right). \quad (15)
\end{aligned}$$

Putting

$$D = \frac{\pi^7 \hbar^2 |P|^2}{9 \cdot 2^{10} a^{10} \rho^2 \Delta\omega \Delta_{pq}^4}, \quad (16)$$

$$d = -\frac{\beta\Delta\langle 1/2r|\mathbf{hJ}|1/2q\rangle \Delta_{pq} Q}{\Delta_0 \Delta_{rq} P},$$

we write down expression (13) in the form

$$T_1^{-1} = D(1 + Hd)^2 \exp(-\Delta_0/kT), \quad (17)$$

and, comparing (17) with (1), we have

$$B = D(1 + Hd)^2. \quad (18)$$

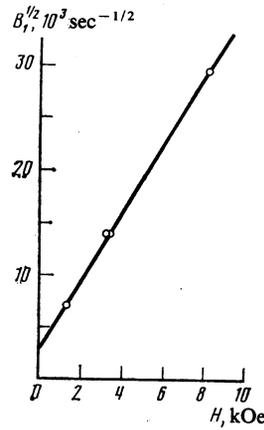


FIG. 4. Field dependence of the coefficient B_1 for Yb^{3+} ions in BaY_2F_8 . The solid line is drawn in accordance with formula (18) with the parameters given in the text.

Figure 4 shows the field dependence of the coefficient B_1 for Yb^{3+} . The circles correspond to the experimental values of B_1 . The solid line is a plot of expression (18) with parameters $D = 8 \times 10^6 \text{ sec}^{-1}$ and $d = 1.1 \times 10^{-3} \text{ Oe}^{-1}$. It is seen that expression (18) describes the experimental data very well. For the Ce^{3+} ion it follows from experiment that $D \equiv B = 2.8 \cdot 10^9 \text{ sec}^{-1}$ and $d \leq 10^{-5} \text{ Oe}^{-1}$.

We now estimate theoretically the values of D and d . For Yb^{3+} we have $\Delta_{pq} = 205 \text{ cm}^{-1}$, $\Delta_{rq} = 8 \text{ cm}^{-1}$, $\Delta_0 = 60 \text{ cm}^{-1}$, and, assuming that $Q \approx 2P$ and $\langle 1/2r|\mathbf{hJ}|1/2q\rangle \approx 1$, we obtain $d \approx 10^{-4} \text{ Oe}^{-1}$. Since $\rho = 5.04 \text{ g/cm}^3$ and $a \approx 2.3 \times 10^{-8} \text{ cm}$ for BaY_2F_8 , it follows that assuming $\Delta\omega = 5 \text{ cm}^{-1} \equiv 10^{12} \text{ sec}^{-1}$ and $|P| = \Delta_{pq}^2$ we obtain $D \approx 3 \times 10^8 \text{ sec}^{-1}$. For Ce^{3+} , assuming $\Delta_{pq} \sim \Delta_{rq}$, we obtain $d \approx 10^{-5} \text{ Oe}^{-1}$, and an estimate of D was given above. Recognizing the assumptions made above, we can assume that the estimates of D and d are close enough to their experimental values.

Thus, our comprehensive investigations have shown that an important contribution is made to the spin-lattice relaxation of the ions Ce^{3+} and Yb^{3+} in BaY_2F_8 by the Raman processes with participation of optical lattice vibrations with energy $\sim 60 \text{ cm}^{-1}$. The observed field dependence of the rate of the SLR of the Yb^{3+} ions is well explained by taking into account the proximity of the first and second excited Stark levels.

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