EFFECT OF OPTICAL RADIATION ON THE ELECTRIC QUADRUPOLE INTERACTION OF A PARAMAGNETIC-ION NUCLEUS

Sh. Sh. Bashkirov and E. K. Sadykov
Kazan' State University
Submitted 30 January 1966
ZhETF Pis'ma 3, No. 6, 240-243, 15 March 1966

Bersuker and Kovarskii [1] advanced the idea that a Mossbauer line can be shifted with the aid of optical radiation. We have examined the effect of optical radiation on the electric quadrupole interaction between a paramagnetic-ion nucleus and its surroundings.

The electric field gradient (EFG) on the nucleus of a paramagnetic ion with non-zero orbital angular momentum is determined essentially by the contribution of the electron shell of the ion. The temperature averaging over the Stark levels of the ion in the crystal explains well the observed temperature dependence of the quadrupole splitting of the Mossbauer line. This averaging is in accord with the fact that the lifetimes of the ions at the Stark sublevels are short compared with the time of quadrupole precession [2].

The optical radiation can produce an appreciable population of some band of upper levels of the ion in the crystal, and this in turn can change the average value of the EFG on the nucleus and consequently its quadrupole-interaction energy.

It must be noted that the ion lifetime in the upper band may turn out to be appreciably longer than the nuclear moment quadrupole precession time, and then the static averaging of the EFG will be effected separately for the ions in the upper and lower states respectively. We can then expect the Mossbauer spectrum to have, besides the main minima, additional minima corresponding to the ions in the upper energy states.

The contribution of the m-th band of levels to the EFG tensor is

$$\langle q_{ij}^{(m)}\rangle_T = \frac{\sum_n \langle m,n|\hat{q}_{ij}|m,n\rangle \exp(-E_{mn}/kT)}{\sum_n \exp(-E_{mn}/kT)}, \qquad (1)$$

where the summation is over the Stark sublevels of the band and $\hat{q}_{ij}$ is the EFG operator on the nucleus. If the times of transitions between bands are shorter than the nuclear moment quadrupole precession time, then the average EFG acting on the nucleus is

$$\langle q_{ij}\rangle_T = \frac{\sum_m N_m \langle q_{ij}^{(m)}\rangle_T}{\sum_m N_m}, \qquad (2)$$

where N_m is the population of the m-th band.

By way of examples we have studied the ions Tm^{3+}, Tm^{2+}, and Yb^{3+} placed in a crystalline field of cubic symmetry with superposition of a relatively weak component of stronger symmetry [2-4]. Calculations carried out for the Tm^{3+} ion (using the parameters of the field in thulium ethyl sulfate [2]) lead in practice to a negative result, and the expected change in EFG under the influence of the radiation does not exceed several per cent at best. On the other hand, as shown by calculations, an appreciable effect can be expected in the case of Tm^{2+} and Yb^{3+}.

For the Yb^{3+} and Tm^{2+} ions, the ground state is $4f^{13}\ {}^2F$. Under the influence of a cubic-symmetry field, the lower level ${}^2F_{7/2}$ splits into a quadruplet Γ_8 and two doublets, while the level ${}^2F_{5/2}$ splits into a quadruplet Γ_8 and a doublet (Fig. 1a). In a field of lower symmetry, we have 4 and 3 Kramers doublets, respectively (Fig. 1b). The intensity of the lower-symmetry field, which can be characterized by the ratio δ_1/Δ_1, was varied over a wide range.

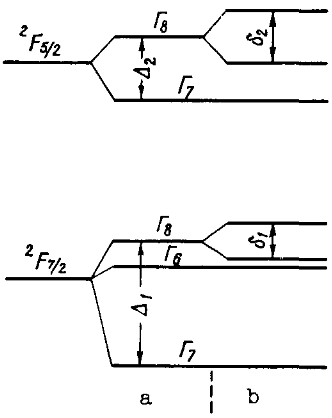

Fig. 1

	m \ Δ_1/kT	4.12	2.06	1.37	1.03	0.82	0.69
$\delta_1/\Delta_1 = 6.5$	7/2	0.003	0.020	0.028	0.026	0.024	0.022
	5/2	0.068	0.227	0.245	0.227	0.209	0.187
$\delta_1/\Delta_1 = 13$	7/2	0.006	0.040	0.055	0.053	0.051	0.050
	5/2	0.160	0.450	0.502	0.462	0.409	0.374

Cases of tetragonal, trigonal, and rhombic symmetry were considered. The calculations were made by the method of equivalent operators. The table lists the values of the EFG ($\langle q_{zz}^{(m)}\rangle_T$ as calculated by formula (1), in units of 10^{18} V/cm^2) for the levels ${}^2F_{7/2}$ and ${}^2F_{5/2}$ in a field of tetragonal symmetry for two values of δ_1/Δ_1 as a function of the temperature. We see that the values of $\langle q_{zz}^{7/2}\rangle$ and $\langle q_{zz}^{5/2}\rangle$ differ so much, that appearance of the effect can be expected even at relatively low population of the upper levels (not higher than 10%). As $T \to 0$, and also if the temperature changes appreciably, $\langle q_{zz}^{(m)}\rangle$ tends to zero, since the doublets of the cubic representation Γ_7 make no contribution to the EFG. In many Yb^{3+} compounds the field components of lower symmetry are so intense that the EFG on the nucleus is determined principally by the contribution of the lower Kramers doublet [5,6]. In the latter case the effect will be observed at low temperatures ($T \approx 4.2°K$).

The authors sincerely thank S. A. Al'tshuler for discussion of the question.

[1] I. B. Bersuker and V. A. Kovarskii, JETP Letters 2, 286 (1965), transl. p. 182

[2] R. G. Barnes, R. L. Mossbauer, E. Kankeleit, and J. M. Poindexter, Phys. Rev. 136A, 175 (1964).

[3] Z. Kiss, ibid. 127, 718 (1962).

[4] R. Pappalardo and D. L. Wood, J. Chem. Phys. 33, 1734 (1960).

[5] S. Hufner, Z. Physik 169, 417 (1962).

[6] F. E. Wagner, F. W. Stanek, P. Kienle, and H. Eicher, ibid. 166, 1 (1962).