INVESTIGATION OF THE E.P.R. OF THE Dy^{2+} ION IN CaF$_2$ IN THE SHORT-WAVELENGTH PART OF THE SUBMILLIMETER RANGE

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Abstract

Full Text

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INVESTIGATION OF THE E.P.R. OF THE Dy^{2+} ION IN CaF_2 IN THE SHORT-WAVELENGTH PART OF THE SUBMILLIMETER RANGE

The successful use of fluorite crystals (CaF_2) activated with dysprosium as the working substance of a quantum generator at a wavelength $\lambda = 2.36\,\mu$ was the reason for its comprehensive investigation in the most diverse wavelength ranges: from the optical to the centimeter range ($^{1-9}$). A number of works are devoted to the study of the system $CaF_2 : Dy^{2+}$ in a magnetic field. The E.P.R. of the three lowest Stark levels of the term $^5I_8$ was studied in ($^{7-9}$). In our works (^{10,11}), transitions between the Zeeman components of the two lower Stark levels of the ground term $^5I_8$ were investigated at a wavelength $\sim 2.06\,\mu$, and the possibility of observing transitions at a wavelength ~ 0.34 mm was reported.

Fig. 1. Experimental recording of absorption lines of $CaF_2 : Dy^{2+}$ in a magnetic field ($H_0 \parallel C_4$), corresponding to transitions $2 \to 6$ ($H_1 = 2200$ Oe) and $4 \to 6$ ($H_2 = 2560$ Oe) at $\lambda = 0.3506$ mm (time constant $\tau = 1.5$ sec).

In the present work, investigations of the E.P.R. of the Dy^{2+} ion in CaF_2 were carried out at a wavelength $\lambda \sim 0.34$ mm on a radiospectroscope developed by us (^{12}), providing a resolving power of the order of 10^4 over the entire submillimeter wavelength range. The samples had the form of a cylinder (diameter ~ 15 mm, length 20 mm) with plane-parallel end faces perpendicular to the geometric axis of the cylinder.

The orientation of the crystallographic axes was determined by us by x-ray structural analysis with an accuracy of ±5°. Samples of two types were studied:

for some, the axes C_2 and C_4 lay in the plane of the end faces; for others, the axes C_2 and C_3 did. The samples under investigation were placed vertically in a glass Dewar vessel between the poles of an electromagnet. The E.P.R. spectrum was studied at the temperature of liquid helium. We observed the dependence of the magnitude of the magnetic field corresponding to the maximum of the resonance absorption lines on wavelength in the range 0.354-0.330 mm. As can be seen from Fig. 1, the signal-to-noise ratio was ~ 100, which made it possible, in addition to measuring the position and width of the line, to carry out a comparative estimate of the intensities of the various transitions. Frequency measurements of the position of the resonance absorption lines made it possible to determine the arrangement of the lower energy levels. Measurement of the width of the lines in small fields made it possible to estimate the width of the level $T_2^{(2)}$. The intensity of the lines observed for different orientations of the crystal changed by more than a factor of 100.

We refined the value of the initial splitting between the levels $E^{(2)}$ and $T_2^{(2)}$ at 4.2° K, which proved to be equal to 29.01 ± 0.02 cm^{-1}, greater than the splitting 28.6 cm^{-1} measured at 27° K in (^{7}).

Figure 2 gives theoretically calculated curves of the dependence of the wavelengths of transitions on the magnitude of the constant magnetic field, calcu-

···obtained for these two values of the initial splitting for the orientation $H_0 \parallel C_4$. Our experimental data are marked with crosses. Using the splitting values $\Delta_1(E^{(2)} \to T_1^{(1)}) = 4.867$ cm^{-1} and $\Delta_2(E^{(2)} \to T_2^{(2)}) = 29.01$ cm^{-1}, obtained in our previous work (^{10}) and in the present work, we carried out on an electronic computer a new calculation of the splitting of the levels $E^{(2)}$, $T_1^{(1)}$, $T_2^{(2)}$ in a magnetic field (up to 10 kOe), under the same assumptions as in (^{10}), and with the same operator of interaction with the magnetic field $\mathcal{H}_{\text{mag}}$. In calculating the matrix elements of the operator $\mathcal{H}_{\text{mag}}$, the wave functions of the levels $E^{(2)}$, $T_1^{(1)}$, $T_2^{(2)}$, calculated in (^{6}), were used. The calculation showed that changing the magnitude of the initial splitting of the levels $E^{(2)}$ and $T_2^{(2)}$ does not substantially affect the character of the behavior of the Zeeman components of the levels $E^{(2)}$, $T_1^{(1)}$, and $T_2^{(2)}$, but is significant when observing transitions between the levels $E^{(2)}$ and $T_2^{(2)}$.

Figure 3 presents graphs of the dependence of the Zeeman components of the levels $E^{(2)}$, $T_1^{(1)}$, $T_2^{(2)}$ on the magnitude of the magnetic field for the three most interesting orientations $H_0 \parallel C_4, C_3, C_2$.

In Figs. 2 and 4 the experimental data are marked with crosses on the theoretically calculated graphs of the dependence of the transition wavelengths on the magnitude of the constant magnetic field. In all measurements $H_{\text{h.f.}} \perp H_0$.

For the orientations $H_0 \parallel C_4$ and $H_{\text{h.f.}} \parallel C_4' \perp H_0$, four absorption lines are observed, corresponding to transitions from levels 1, 2 to 6, 8 (the numbering of the levels is given in order of increasing energy; see Fig. 3), with the transitions

Fig. 2

Figure 2: Fig. 2

Fig. 3. Diagram of the levels $E^{(2)}$, $T_1^{(1)}$, and $T_2^{(2)}$ of the Dy^{2+} ion in CaF_2 in a magnetic field

Figure 3: Fig. 3. Diagram of the levels $E^{(2)}$, $T_1^{(1)}$, and $T_2^{(2)}$ of the Dy^{2+} ion in CaF_2 in a magnetic field

1 → 6 and 1 → 8 more intense than 2 → 6 and 2 → 8. The transitions 1 → 7 and 2 → 7 are forbidden for this orientation of H_0 and $H_{\text{h.f.}}$ relative to the crystallographic axes, and we did not observe them.

Fig. 2. Dependence of the transition wavelengths on the magnetic field for the orientation $H_0 \parallel C_4$. Solid lines: initial splitting $\Delta_2(E^{(2)} \rightarrow T_2^{(2)}) = 29.011$ cm^{-1}; dashed lines: initial splitting $\Delta_2'(E^{(2)} \rightarrow T_2^{(2)}) = 28.6$ cm^{-1}. Crosses mark the experimental data.

It is known that for the orientations $H_0 \parallel C_2$ and $H_0 \parallel C_3'$, as a result of the mixing of states in the magnetic field, all six transitions become allowed; moreover, as a rule, the intensity of the lines corresponding to transitions from level 1 is greater than from level 2. This agrees with our estimate of the probabilities of the corresponding transitions, calculated using the wave functions obtained in the calculation.

Let us note that the larger value we obtained for the initial splitting between the levels $E^{(2)}$ and $T_2^{(2)}$, in comparison with the data of work (7), may be connected with the fact that we carried out the measurements at a lower temperature (4.2° K instead of 27° K). A decrease in temperature leads to an increase in the splitting in the crystalline field, since the crystal-field potential for Dy^{2+} in CaF_2 contains terms of the 4th and 6th degrees, into which R (the distance between the Dy^{2+} ion and the nearest F^- ion) enters respectively as R^{-5} and R^{-7}. As the temperature is lowered, the crystal-field constants increase, causing an increase in the splitting between the Stark components.

Fig. 3. Diagram of the levels $E^{(2)}$, $T_1^{(1)}$, and $T_2^{(2)}$ of the Dy^{2+} ion in CaF_2 in a magnetic field (the splitting of the level $T_2^{(2)}$ for $H_0 \parallel C_3$ is not shown, since it is very close to the splitting for $H_0 \parallel C_2$).

Observation of transitions between the levels $E^{(2)}$ and $T_2^{(2)}$ in a magnetic field is of great interest from the point of view of the dynamics of laser operation on $CaF_2 : Dy^{2+}$ crystals. The level $T_2^{(2)}$ is the terminal level of the luminescent transition $T_1^{(2)}\,(^5I_7) \rightarrow T_2^{(2)}\,(^5I_8)$, on which generation with wavelength $\lambda 2.36\,\mu$ was achieved (1).

As was shown in work (5), the broadening of the luminescence line of this tran-

Fig. 4. Dependence of the wavelengths of transitions on the magnetic field for $H_0 \parallel C_2$ and $H_0 \parallel C_3$

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sition is connected mainly with the broadening of the level $T_2^{(2)}$, which is determined chiefly by one-phonon nonradiative transitions to the levels $T_1^{(1)}$ and $E^{(2)}$. The value calculated in (5) for the width of the luminescence line of the transition $T_1^{(2)}\,(^5I_7) \to T_2^{(2)}\,(^5I_8)$ is ~ 0.073 cm^{-1} at 4.2° K, while experimentally in (6) it was found that the value of the level width at the same temperature is less than 0.07 cm^{-1}.

Assuming the width of the ground state $E^{(2)}$ to be small, one can obtain from the absorption-line width of the transition $E^{(2)} \to T_2^{(2)}$ measured by us the width of the $T_2^{(2)}$ level. In magnetic fields of ~ 0.5 kOe, the line width was ~ 150 Oe, or 0.04 cm^{-1}. Thus, according to our data, the width of the $T_2^{(2)}$ level at $T = 4.2°$ K has a value of ~ 0.04 cm^{-1}, which agrees sufficiently well with the value theoretically obtained in Ref. (5).

Fig. 4. Dependence of the wavelengths of transitions on the magnetic field for $H_0 \parallel C_2$ and $H_0 \parallel C_3$

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