ZEEMAN SPECTROSCOPY ON THE EXCITED Cr$^{3+}$ IN Al$_2$O$_3$ WITH A FAR-INFRARED LASER

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Zeeman spectroscopy on the metastable E and 2A levels of excited Cr$^{3+}$ ions in Al$_2$O$_3$ has been performed with the 891-GHz radiation of an HCN-laser. The absorption by E $\rightarrow$ 2A transitions, which leads to an increase of the population of 2A levels, was detected optically by the R$_2$-fluorescence radiation. Narrow absorption lines were found at two magnetic fields B$_1$ and B$_2$, for the E $\rightarrow$ 2A$^+$ and the E $\rightarrow$ 2A$^-$ transitions, respectively. From our experiments we obtain a g-factor of 1.47 for the 2A level and an energy splitting of 29.25 cm$^{-1}$ between the E and 2A levels at zero magnetic field. We found the interesting result that the signal at B$_2$ increases linearly with the concentration of excited Cr$^{3+}$ ions while the signal at B$_1$ increases quadratically. The role of a bottleneck of 29 cm$^{-1}$ phonons generated by the far-infrared absorption is discussed.
We report the investigation of excited Cr$^{3+}$ magnetic impurity ions in Al$_2$O$_3$ by means of the 891-GHz radiation of an HCN-laser. Our method is shown in Fig. 1. A magnetic field is applied along the c-axis of an optically pumped ruby crystal immersed in liquid helium. The excited metastable E($^2E$) and 2A($^2E$) states which are separated by an energy $\Delta \approx 29 \text{ cm}^{-1}$ are split into four Zeeman levels $E_-, E_+,$ and $2A_-, 2A_+.$ The laser radiation of frequency $v_L = 891 \text{ GHz}$ can be absorbed at the magnetic field $B_1$ by the $E_- \rightarrow 2A_+$ transition and at the field $B_2$ by the $E_- \rightarrow 2A_-$ transition. The absorption of the far-infrared radiation is detected optically by observing the $R_2$-fluorescence radiation that is due to optical transitions from the 2A-levels to the $^4A_2$ ground state. The experimental arrangement is described elsewhere.

FIGURE 1 Energy levels of Cr$^{3+}$ in Al$_2$O$_3$ (not to scale). At two magnetic fields $B_1$ and $B_2$ absorption of laser radiation leads to emission of $R_2$-fluorescence radiation $S_1$ and $S_2.$
In the following we report spectroscopic data obtained with our method and furthermore, the observation of a bottleneck of the 29 cm$^{-1}$ phonons that are generated by absorption of far-infrared radiation.

**FIGURE 2**

$R_2$-fluorescence in ruby (with about $10^{15}$ cm$^{-3}$ excited Cr$^{3+}$ ions) irradiated with the 891 GHz-radiation of an HCN-laser (note change in the magnetic field scale). The experimental curves (solid lines) are Lorentzian.

Experimental results are shown in Fig. 2. We found resonant absorption at the fields $B_1 = 5.07$ kG and $B_2 = 20.46$ kG. From these values we can derive the $g$-value of the 2A-state

$$g_{||}(2A) = \frac{B_2 - B_1}{B_2 + B_1} g_{||}(E)$$

and the energy splitting at zero magnetic field

$$\Delta = h\nu_L - \frac{1}{2} \mu_B B_1 \left\{ g_{||}(E) + g_{||}(2A) \right\}.$$
We obtain \( g_{\parallel}(2A) = 1.47 \pm 0.01 \) and \( \Delta = 29.25 \pm 0.01 \text{ cm}^{-1} \). We have used \( g_{\parallel}(E) = 2.445 \) for the \( g \)-factor of the \( E \) level and \( \nu_L = 890.760 \text{ GHz} \) for the HCN-laser frequency. Our experimental results are in good agreement with values obtained from fluorescence experiments. 

In addition to the experimental results the Gaussian and Lorentzian lineshapes are also plotted in Fig. 2. For both transitions, \( E \rightarrow 2A_+ \) at the magnetic field \( B_1 \) and \( E \rightarrow 2A_- \) at the field \( B_2 \), the lineshapes are nearly Lorentzian. Therefore, we conclude that the broadening of both lines is due to the finite lifetime \( T_1 \) of the \( 2A \) levels caused by spontaneous emission of \( 29 \text{ cm}^{-1} \) phonons. From the magnetic width of the lines we obtain for both transitions a spectral width of \( \Delta \nu = 360 \text{ MHz} \) and \( T_1 = 1/2\pi \Delta \nu = 4.4 \times 10^{-10} \text{s} \). This value is in good agreement with a theoretical estimate \( T_1 = 3 \times 10^{-10} \text{s} \).

The two signals shown in Fig. 2 are obtained at a fixed density \( N^* \) of excited \( \text{Cr}^{3+} \) ions. We have varied \( N^* \) by changing the intensity of the optical pump light. We found that the halfwidths of the signals do not change appreciably with \( N^* \) while the signal heights \( S \) increase for both signals as shown in Fig. 3. We found the interesting result that the signal height \( S_1 \) at \( B_1(E \rightarrow 2A_+ \text{ transition}) \) increases quadratically with \( N^* \) while the signal height \( S_2 \) increases only linearly with \( N^* \).
FIGURE 3
Maxima of $S_1$ and $S_2$. The signal maximum $S_1$ increases quadratically, the signal maximum $S_2$ linearly with $N^*$. 

The quadratic dependence of $S_1$ on $N^*$ indicates a strong phonon bottleneck effect which is due to resonant absorption of the 29 cm$^{-1}$ phonons that are generated by the far-infrared absorption process (see Fig. 4): After a far-infrared quantum is absorbed by the $E_- \rightarrow 2A_+$ transition the $2A_+$ state decays by a $2A_+ \rightarrow E_+$ transition generating a phonon. This phonon can be re-absorbed resonantly by the $E_+ \rightarrow 2A_+$ transition because at the magnetic field $B_1$ the $E_-$ and $E_+$ levels are nearly equally populated at our crystal temperature of 2 K. The spin-flip phonon transition $2A_+ \rightarrow E_-$ is expected to be much weaker than the non-spin-flip transition. Since the far-infrared absorption coefficient for the $E_- \rightarrow 2A_+$ transition increases linearly with $N^*$ we conclude from the quadratic dependence $S_1(N^*)$ that (for $N^* > 5 \times 10^{14}$ cm$^{-3}$) the number $b$ of emission and re-absorption processes of a resonant phonon increases proportionally to $N^*$. 

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FIGURE 4
Resonant absorption and emission of phonons at the magnetic field \( B_1 \). The inset shows the phonon trapping at the magnetic field \( B_2 \).

That the signal \( S_2 \) shows a different behaviour than the signal \( S_1 \) (Fig. 3) is not understood: The resonant phonons generated in this case by \( 2A_1 \rightarrow E_1 \) transitions should also be resonantly re-absorbed and a quadratic dependence \( S_2(N^*) \) would be expected. An explanation for the observed linear dependence \( S_2(N^*) \) may be found in the absorption process: It is possible that the \( S_2 \) signal is not caused by \( E_1 \rightarrow 2A_1 \) absorption, but, by an other absorption process, f.i. a defect-induced one-phonon absorption process due to the \( Cr^{3+} \) impurity ions in the crystal. In this case the absorption coefficient for the far-infrared radiation would be independent on \( N^* \) and the factor \( b \) would, therefore, again increase linearly with \( N^* \).

We like to point out that the linear dependence of the factor \( b \) on the concentration \( N^* \) can not be explained by a bottleneck model. 

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used to describe recent bottleneck experiments\textsuperscript{7-9} because in this model $b$ is expected to increase proportionally to $(N^*)^{3/2}$. In the model it is assumed that the phonons are trapped in the crystal by resonant scattering processes in which the frequency of the 29 cm\textsuperscript{-1} phonons is not changed. We think one has, however, to regard frequency changes of the scattered phonons in the resonantly coupled electron-phonon system which leads to spectral phonon diffusion. It has been shown\textsuperscript{10} that this model describes the phonon bottleneck observed in ruby for concentrations $N^* > 10^{16}$ cm\textsuperscript{-3}. We suggest that spectral phonon diffusion can also explain the linear increase of the factor $b$ found in our experiment for concentrations $N^* = 10^{15}$ cm\textsuperscript{-3} to $10^{16}$ cm\textsuperscript{-3} excited Cr\textsuperscript{3+}.

Further experiments to generate monochromatic 29 cm\textsuperscript{-1} phonons and to study the electron-phonon interaction in ruby are in preparation.

Discussions with T. Walker are acknowledged.


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