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ZEEMAN SPECTROSCOPY ON THE EXCITED Cr^{3+} IN Al_2O_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_2O_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.

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We report the investigation of excited Cr^{3+} magnetic impurity ions in Al_2O_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 $\Delta \simeq 29\ cm^{-1}$ are split into four Zeeman levels E_-, E_+ and $2A_-$, $2A_+$. The laser radiation of frequency $\nu_L = 891$ 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.[1]

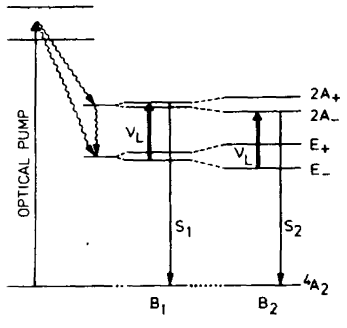

FIGURE 1 Energy levels of Cr^{3+} in Al_2O_3 (not to scale). At two magnetic fields B_1 and B_2 absorption of laser radiation leads to emission of R_2-fluorescence radiation S_1 and S_2.

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.

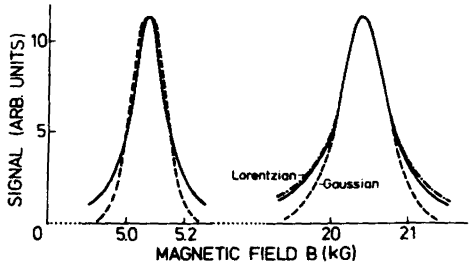

FIGURE 2
R_2-fluorescence in ruby (with about 10^{15} cm^{-3} excited Cr^{3+} ions) irradiated with the 891 GHz-radiation of an HCN-laser (note change in the magnetic field scale). The experimental curves (solid lines) are Lorentzian.

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 $g_{\parallel}$-value of the 2A-state

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

and the energy splitting at zero magnetic field

$$\Delta = h\nu_L - \frac{1}{2} \mu_B B_1 \left\{ g_{\parallel}(E) + g_{\parallel}(2A) \right\} .$$

We obtain $g_{\parallel}(2A) = 1.47 \pm 0.01$ and $\Delta = 29.25 \pm 0.01$ cm^{-1}. We have used $g_{\parallel}(E) = 2.445$ for the g-factor of the E level[2] and $\nu_L = 890.760$ GHz for the HCN-laser frequency.[3] Our experimental results are in good agreement with values obtained from fluorescence experiments.[4,5]

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 cm^{-1} phonons. From the magnetic width of the lines we obtain for both transitions a spectral width of $\Delta\nu = 360$ MHz and $T_1 = 1/2\pi\Delta\nu = 4.4 \times 10^{-10}$s. This value is in good agreement with a theoretical estimate[6] of $T_1 = 3 \times 10^{-10}$s.

The two signals shown in Fig. 2 are obtained at a fixed density N^* of excited 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 ($E_- \rightarrow 2A_+$ transition) increases quadratically with N^* while the signal height S_2 increases only linearly with N^*.

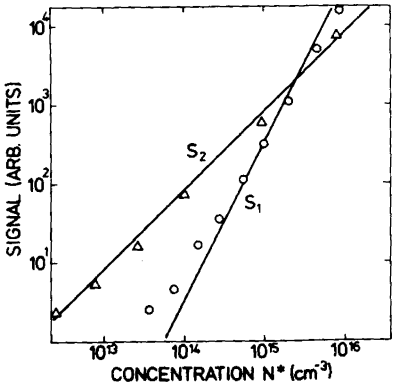

FIGURE 3
Maxima of S_1 and S_2. The signal maximum S_1 increases quadratically, the signal maximum S_2 linearly with N*.

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. [6] 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*.

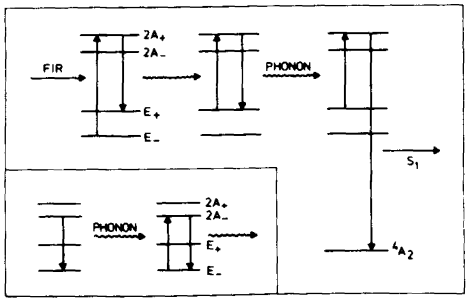

FIGURE 4
Resonant absorption and emission of phonons at the magnetic field B_1. The inset shows the phonon trapping at the magnetic field B_2.

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 $2A_- \rightarrow E_-$ 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 $E_- \rightarrow 2A_-$ 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[7]

used to describe recent bottleneck experiments[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^{-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[10] that this model describes the phonon bottleneck observed in ruby for concentrations $N^* > 10^{16}$ cm^{-3}. We suggest that spectral phonon diffusion can also explain the linear increase of the factor b found in our experiment for concentrations $N^* \approx 10^{15}$ cm^{-3} to 10^{16} cm^{-3} excited Cr^{3+}.

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

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