

RESONANCE TRAPPING OF MONOCHROMATICALLY GENERATED 29 cm^{-1} PHONONS IN STRONGLY EXCITED RUBY CRYSTALS

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We report on a study of the resonance trapping of monochromatically generated 29 cm^{-1} phonons in strongly excited ruby crystals. By use of a pulsed far infrared laser we generated, in homogeneously excited crystals, monochromatic phonon pulses and studied the trapping and decay of these phonons. We found that the trapping time increased with the square root of the concentration of excited Cr^{3+} ions and that it depended on the crystal dimension. We show, furthermore, that the trapping time depends on crystal quality and that it can be strongly influenced by inhomogeneous stress and by X-ray irradiation. We used a model of spectral-spatial phonon diffusion for a description of the experimental results.

1. Introduction

Resonance trapping of 29 cm^{-1} phonons in ruby crystals due to elastic resonance scattering at excited Cr^{3+} ions, with electronic transitions between the \bar{E} and $2\bar{A}$ levels, has been studied intensively. Due to the resonance scattering phonons can be spatially trapped. For crystals containing large concentrations of excited Cr^{3+} ions it was found that the trapping time was much larger than the ballistic time of flight through the crystals. The trapping time was, however, orders of magnitude smaller than expected from simple diffusive propagation due to elastic resonance scattering. The question which processes shorten the trapping time and by which processes trapped phonons finally escape from a resonance volume has attracted much interest. This led to a discussion of various inelastic scattering processes for high frequency phonons, namely scattering at exchange-coupled Cr^{3+} ions [1,2], at exchange-coupled excited Cr^{3+} ions [3] and at impurities other than Cr^{3+} ions [4]; for recent summaries see refs. [5,6]. Because the origins of inelastic scattering processes for 29 cm^{-1} phonons in ruby are still not yet well understood, we have performed a new

study using an experimental technique that was modified in comparison to earlier techniques. In our experiment we generated *monochromatic* phonons in optically pumped ruby and tried to reduce additional broadband phonon generation. Broadband phonons – in addition to monochromatic phonons – occur if phonons are generated by optical pumping into the broad absorption bands of ruby. Then, by nonradiative transitions to the \bar{E} and $2\bar{A}$ levels broadband phonons and by $2\bar{A} \rightarrow \bar{E}$ relaxation monochromatic phonons are generated. Direct pumping of the $2\bar{A}$ level leads, by $2\bar{A} \rightarrow \bar{E}$ relaxation, to monochromatic phonons. At strong excitation, i.e. for large concentrations of excited Cr^{3+} ions, however, excited state absorption of Cr^{3+} ions and subsequent relaxation to the \bar{E} and $2\bar{A}$ levels may again lead to broadband phonon generation; a quantitative discussion to these points is given elsewhere [6]. In our experiment presented in this paper, we used a far infrared laser for monochromatic phonon generation and avoided broadband phonon generation by the strong optical pumping by delaying the far infrared laser pulse with respect to the optical pump pulse. Thus, it was possible to study the dynamics of the monochromatically generated phonons

without additional effects from broadband phonons.

2. Experimental method

Phonons at an energy of 29 cm^{-1} were generated monochromatically with a far infrared laser. In the experiment we used a pulsed, TEA CO_2 laser pumped, D_2O zigzag laser, with a resonator and Michelson output coupling design. Laser emission was obtained either at a frequency of 836 GHz or at 779 GHz. The spectral width of the radiation was about 0.1 GHz, the pulse duration 100 ns and the pulse energy $2 \times 10^{-5} \text{ J}$.

By applying a magnetic field, the energy separation between \bar{E} and $2\bar{A}$ sublevels was tuned to the frequency ν_L of the far infrared laser radiation. Then, far infrared absorption occurred by $\bar{E}_+ \rightarrow 2\bar{A}_-$ transitions (fig. 1(a)) and, by relaxation processes $2\bar{A}_- \rightarrow \bar{E}_+$ and $2\bar{A}_- \rightarrow \bar{E}_-$, respectively, two monochromatic phonon packets were generated at frequencies ν_L and $\nu_L + (\mu_B/h)g_E B$, where μ_B is Bohr's magneton, h Planck's constant, $g_E = 2.445$ the effective g value of the \bar{E} level and B the magnetic field strength for B applied parallel to the crystal c -axis. The corresponding relaxation rates for spinflip and non-spinflip transitions are $T_{1-+}^{-1} = 2 \times 10^9 \text{ s}^{-1}$ and $T_{1--}^{-1} = 7 \times 10^7 \text{ s}^{-1}$. Both phonon packets are re-absorbed and determine the population of the $2\bar{A}$ level that is probed by R_2 fluorescence. The population of the \bar{E} level is probed by R_1 fluorescence. In order to study frequency-shifted phonons we

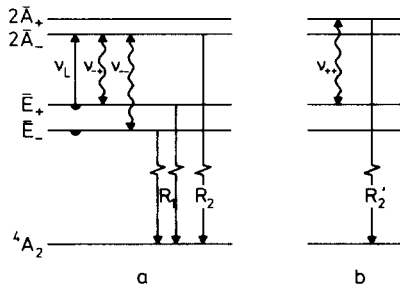


Fig. 1. Monochromatic generation of 29 cm^{-1} phonons in ruby due to electronic transitions in excited Cr^{3+} ions and detection by R_2 fluorescence (a) and detection of spectrally shifted phonons by observation of R_2' fluorescence (b).

have also observed radiation from the $2\bar{A}_+$ level; the corresponding phonon-induced R_2' fluorescence (fig. 1(b)) was much weaker than the phonon-induced R_2 radiation.

We excited the ruby samples with frequency-doubled radiation (wavelength 530 nm) of a pulsed Nd:YAG laser via the $4T_2$ absorption band. By radiationless transitions into the \bar{E} and $2\bar{A}$ levels a high-frequency broadband phonon spectrum and, by the $2\bar{A} \rightarrow \bar{E}$ relaxation, additional (resonant) 29 cm^{-1} phonons are created. All these phonons escape into the surrounding liquid helium bath in a time of about $100 \mu\text{s}$. After this time the $2\bar{A}$ level is almost unpopulated while the \bar{E} level (decay time $\approx 5 \text{ ms}$) is still nearly fully populated. By delaying the far-infrared pulse relative to the optical excitation pulse an interaction of phonons generated by far-infrared absorption with phonons created by the optical pumping process has been avoided.

The high pulse energy (maximum 0.15 J) of the Nd:YAG laser system enabled us to reach *homogeneous* strong excitation in relatively large samples. Thus, the resonance volume was only limited by the sample surfaces. We determined the concentration N^* of excited Cr^{3+} ions from the saturation of the R_1 intensity with Nd:YAG laser pulse energy occurring when nearly half of all Cr^{3+} ions were in the excited state. By irradiating an optically excited crystal with a far-infrared laser beam that had a diameter larger than the sample size we obtained almost homogeneous phonon excitation in the whole crystal.

Our samples had dimensions of $2 \times 2 \times 4 \text{ mm}^3$ and $1 \times 1 \times 4 \text{ mm}^3$ and were doped with 0.05 wt.% Cr_2O_3 . The samples were mounted in a superconducting magnet with the c -axis parallel to the magnetic field. The optical excitation beam was parallel to the c -axis and fluorescence radiation was observed perpendicular to the excitation beam. For separation of R_2 from R_1 fluorescence radiation and of the Zeeman components of the R_2 line we used a double grating monochromator. Phonon-induced R_2 radiation was detected with a photomultiplier in combination with a photon counting technique using a transient recorder as time resolving element. The R_2 signal counts were integrated over typically 2×10^3 laser shots.

3. Experimental results

An experimental R_2 signal curve, for the maximum possible concentration N^* of excited Cr^{3+} ions ($N^* \approx 8 \times 10^{18} \text{ cm}^{-3}$), is shown in fig. 2. The signal increases very fast, corresponding to the short far-infrared laser pulse, and then decreases slowly. Before the arrival of the far-infrared pulse there are almost no phonons remaining from the optical excitation; the delay between excitation pulse and far-infrared pulse was about 10^{-3} s. The signal curve (points) shows a first fast decay followed by an exponential decay over several time constants. The population ratio for the $2\bar{A}$ and \bar{E} levels obtained from the ratio of the fluorescence intensities was about 5×10^{-4} ; this ratio is also a measure of the phonon occupation number for the phonons generated by the far-infrared excitation. The first fast decay gives evidence for fast spectral redistribution of the monochromatically generated phonons. We suggest that during the nonexponential decay the original narrow phonon distribution broadens and that a fixed frequency distribution has been established later as indicated by the exponential decay for later times; this will be discussed in detail in the next chapter. In other words, our results seem to indicate a fast phonon redistribution from a narrow into a broad spectral distribution within a time of less than 100 ns and a slow exponential decay of the broad distribution with a time constant larger than 100 ns.

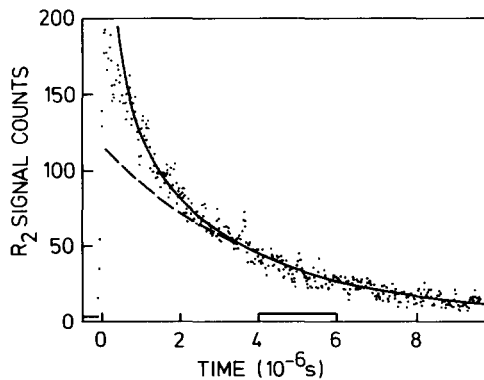


Fig. 2. Phonon-induced R_2 signal (points) and theoretical curve (solid line); dashed, exponential slope. The time $t = 0$ corresponds to a time immediately after phonon injection.

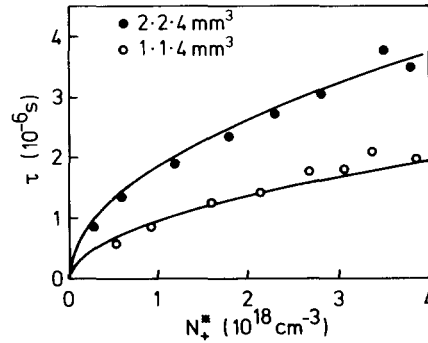


Fig. 3. Trapping time of 29 cm^{-1} phonons versus concentration of excited Cr^{3+} ions in crystals with different dimensions, and theoretical curves.

We have measured the decay of the R_2 signal at different concentrations N^* of excited Cr^{3+} ions in the \bar{E} state and determined from the exponential part of the decay curve the trapping time τ . The N^* dependence of τ , shown in fig. 3, can be described by a square root function; we used as a measure of excited Cr^{3+} ions the concentration N_+^* ($\approx N^*/2$) of ions in the \bar{E}_+ state. We observed a trapping time $\tau \approx 4 \mu\text{s}$ at our maximum concentration ($8 \times 10^{18} \text{ cm}^{-3}$) of excited Cr^{3+} ions that seems to be the largest observed trapping time in a magnetic field in comparison to other trapping experiments with high frequency phonons in ruby. A saturation of the trapping time in strongly excited samples as reported for other experiments [6] was not found in the present experiment.

The comparison of results for two homogeneously excited samples with different volumes (fig. 3) suggests a linear increase of the trapping time with increasing size of the resonance volume; the samples were cut from the same large crystal. The observation of a dependence of the trapping time τ on the size of the resonance volume indicates that spatial diffusion is involved in the phonon loss mechanism. However, since the trapping times do not show a quadratic dependence on crystal dimension, the phonon escape cannot be described by a simple diffusion behavior.

We have magnetically tuned the $\bar{E}_+ \rightarrow 2\bar{A}_-$ resonance at constant far infrared frequency, thus generating phonons in the center and in the wing of the resonance line respectively. We found (fig.

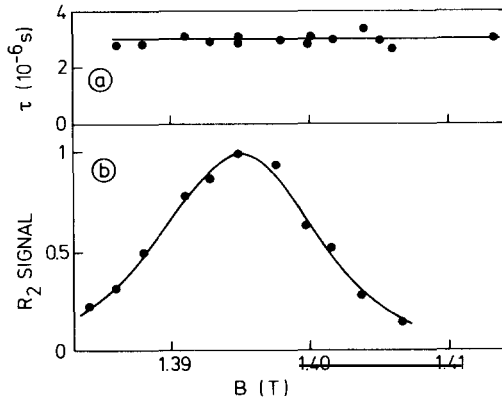


Fig. 4. Magnetic tuning of the $\bar{E}_+ \rightarrow 2\bar{A}_-$ resonance at constant far-infrared frequency (836 GHz) for $N^* \approx 3 \times 10^{18} \text{ cm}^{-3}$; (a) trapping time and (b) R_2 signal (at time $t = 0$).

4(a)) that the trapping time was a constant, i.e. it was independent of the phonon generation frequency. This result gives strong evidence that fast spectral diffusion occurred. We note that the width of the R_2 signal curve (fig. 4(b)) corresponds to the $\bar{E} \rightarrow 2\bar{A}$ resonance linewidth of 370 MHz*. Thus, our experiment indicates that fast spectral redistribution occurs at least over a frequency range of the order of the resonance linewidth.

We have also observed phonons with large frequency shift by detecting inelastically scattered phonons at a frequency ν_{++} (fig. 1(b)); in this case we monitored the occupation of the $2\bar{A}_+$ level by observing R'_2 radiation. The phonons have been generated at a frequency (≈ 896 Hz) by the transition $2\bar{A}_- \rightarrow \bar{E}_+$ (fig. 1(a)). A $2\bar{A}_+$ population is only possible if phonons increase their frequency by about 45 GHz, i.e. by 5% relative to the generation frequency. The R'_2 signal curve due to optical transitions from the $2\bar{A}_+$ state to the ground state (fig. 5(a)) is shown in comparison with the R_2 signal curve due to transitions from the $2\bar{A}_-$ state

* In an earlier experiment [6] performed with cw excitation of ruby and by the use of a cw far infrared laser it was found that the width of the R_2 signal curve was narrower than the $\bar{E} \rightarrow 2\bar{A}$ resonance linewidth. We suggest that the maximum concentration of excited Cr^{3+} ions was much less than in the experiment described in this paper and that the excited state concentration reported in ref. [6] may have been overestimated by an order of magnitude.

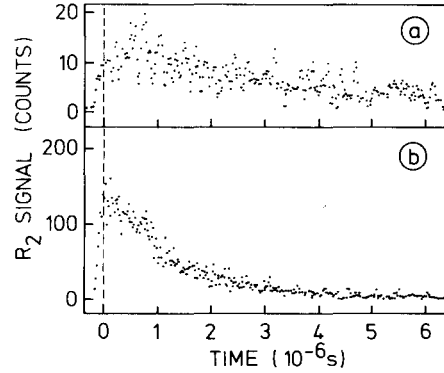


Fig. 5. Observation of phonons with large frequency shift; (a) R'_2 signal (after 7×10^4 sweeps) and (b) R_2 signal (10^3 sweeps).

(fig. 5(b)). The ratio of the maxima of the R'_2 and R_2 intensities and therefore of the occupation numbers of the two $2\bar{A}$ levels was $N_{2\bar{A}_+}/N_{2\bar{A}_-} = 1/1600$, i.e. only a small part of the phonons was shifted to give a population in the $2\bar{A}_+$ level. This experiment indicates that the fast spectral redistribution – if it occurs as we assume – does lead to a phonon distribution with a width much smaller than 45 GHz.

In fig. 6 the maximum R_2 signal for different concentrations of excited Cr^{3+} ions is shown. The R_2 signal increases linearly with N^* . Since the absorption coefficient $\sigma_0 \times N^*$ for far-infrared radiation, where $\sigma_0 \approx 3 \times 10^{-19} \text{ cm}^2$ is the cross section for absorption of far-infrared radiation by the $\bar{E}_- \rightarrow 2\bar{A}_-$ transitions at resonance, increases

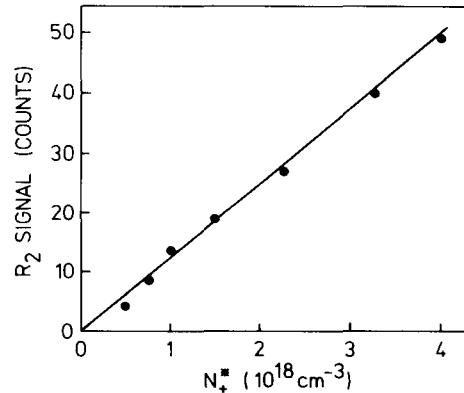


Fig. 6. R_2 signal (for $t = 0$) versus concentration of excited Cr^{3+} ions.

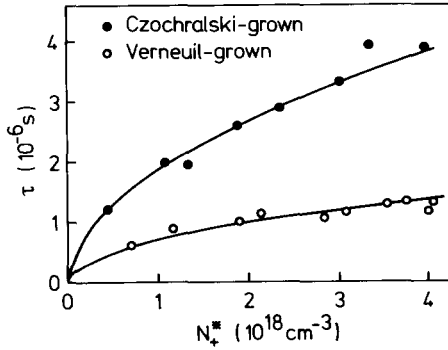


Fig. 7. Trapping time for different crystals.

linearly with N^* , our result indicates that the initial $2\bar{A}$ population per generated phonon is independent on N^* . This is expected for strong trapping and in case that the phonons are on “speaking terms” with the $\bar{E} \rightarrow 2\bar{A}$ resonance line.

We have studied the influence of crystal quality on the trapping time. We investigated the trapping time in a Verneuil-grown and a Czochralski-grown ruby crystal that had equal Cr^{3+} concentrations (0.05%) and equal dimensions ($2 \times 2 \times 4 \text{ mm}^3$). We found that the trapping time at a fixed N^* in a Czochralski crystal was three times the trapping time in a Verneuil crystal (fig. 7). Verneuil crystals are known to have stronger inhomogeneous strain field within the sample and higher dislocation densities than Czochralski crystals. Evidence for different strain fields follows directly from a comparison of the $\bar{E} \rightarrow 2\bar{A}$ far-infrared resonance lines: we found that for Verneuil crystals the width is almost twice as large as for Czochralski crystals [7]. The observed shorter trapping time in a Verneuil crystal may therefore be due to inelastic phonon scattering caused by strain fields or dislocations.

Finally we studied the influence of X-ray irradiation on the trapping time (fig. 8). We irradiated a Czochralski-grown crystal for 100 min with X-rays. After X-ray irradiation the R_1 fluorescence was reduced by about five percent. We suggest that about five per cent of chromium ions were transformed into Cr^{2+} ions and Cr^{4+} ions. After X-ray irradiation the trapping times were reduced to one fourth of the values determined before irradiation. UV radiation (120 min) from

an argon ion laser (wavelength 350.7 nm and 356.9 nm) transformed Cr^{2+} and Cr^{4+} ions back into Cr^{3+} ions. After UV irradiation we observed the same trapping time as before X-ray irradiation. This experiment is consistent with an earlier experiment [4] that showed that the trapping can be reduced by X-ray irradiation. In these earlier studies reduction of the trapping was attributed to elastic phonon scattering at Cr^{2+} ions. These ions are to a certain degree also present in unirradiated crystals and may play an essential role for inelastic scattering and phonon loss processes.

4. Theoretical analysis

Summarizing our experimental results, the trapping time can be described by the expression

$$\tau = Ad(N^*)^{1/2}, \quad (1)$$

where N^* is the concentration of excited Cr^{3+} ions, d a characteristic crystal dimension and A a constant that depends on crystal quality and crystal treatment.

We describe the $\bar{E} \rightarrow 2\bar{A}$ resonance line of excited Cr^{3+} ions by the Lorentzian lineshape function

$$g(\nu) = \frac{\Gamma^2/4}{(\nu - \nu_0)^2 + \Gamma^2/4}, \quad (2)$$

where ν_0 is the resonance frequency for the $\bar{E} \rightarrow 2\bar{A}$ transition at zero magnetic field and Γ (≈ 0.4 GHz) the halfwidth of the resonance line.

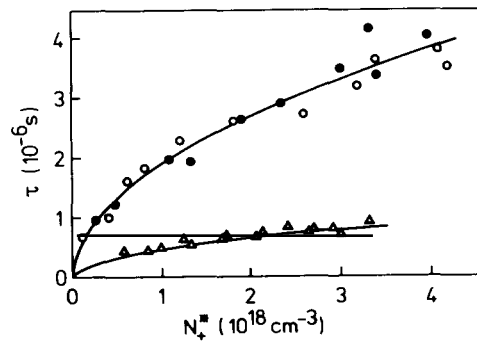


Fig. 8. Trapping time for different concentrations of excited Cr^{3+} ions in an X-ray irradiated sample; circles, before X-ray irradiation; triangles, after X-ray irradiation; points, after UV irradiation.

Assuming, first, that only elastic resonance scattering occurs, we have a diffusion coefficient for phonons in the line center of [6]

$$D_0 = \frac{1}{6} v^2 Z(\nu) \Gamma T_{-+} (N_+^*)^{-1}, \quad (3)$$

where v ($\approx 6.7 \times 10^5$ cm/s) is an average velocity of sound, $Z(\nu)$ ($\approx 10^8$ s/cm³) the density of states for the resonant phonons, Γ (≈ 0.4 GHz) the halfwidth of the $\bar{E} \rightarrow 2\bar{A}$ resonance line, T_{-+} the $2\bar{A}_- \rightarrow \bar{E}_+$ relaxation time and N_+^* ($\approx N^*/2$) the concentration of excited Cr³⁺ ions in the \bar{E}_+ state. In eq. (3) it is taken into account that we have in the magnetic field two phonon packets, which are coupled via the $2\bar{A}$ level and therefore have equal population and that phonons at frequency ν_{+-} diffuse much faster than phonons at frequency ν_{--} . For the largest concentration N^* we find $D_0 \approx 10$ cm² s⁻¹. We introduce a characteristic crystal dimension d by the expression

$$\frac{\pi}{d} = \left[\left(\frac{\pi}{d_1} \right)^2 + \left(\frac{\pi}{d_2} \right)^2 + \left(\frac{\pi}{d_3} \right)^2 \right]^{1/2}, \quad (4)$$

where d_1 , d_2 and d_3 are sample dimensions, and find a diffusive escape time $\tau_d = \pi^{-2} d^2 D_0^{-1}$, which shows dependencies on d and N^* that disagree with experiment. Furthermore, τ_d is much larger than the observed phonon trapping times. Thus, pure spatial diffusion cannot describe the phonon escape as is well known from earlier experiments.

For a theoretical analysis, we will now use a model of spectral-spatial diffusion [8,9]. In addition to resonance scattering we introduce inelastic scattering and assume that the inelastic scattering occurs in steps so that an inelastically scattered phonon can still be resonantly scattered at excited Cr³⁺ ions. We define a spectral diffusion coefficient

$$D_\nu = \nu_R^2 \tau_{in}^{-1}, \quad (5)$$

where ν_R is an average frequency shift in an inelastic scattering process and τ_{in}^{-1} an average inelastic scattering rate. We assume for simplification that D_ν is independent of frequency. A spectral-spatial diffusion equation is then given by

$$\frac{\partial f}{\partial t} = D_0 g^{-1} \nabla^2 f + D_\nu \frac{\partial^2 f}{\partial \nu^2}, \quad (6)$$

where $f(x, y, z, \nu, t)$ is the distribution function of the phonons. For the conditions of our experiment, namely monochromatic excitation in the line center at time $t = 0$, an approximate solution of eq. (1) is given by [8–10]

$$f = (2\pi \sinh(2t/\tau))^{-1/2}, \quad (7)$$

where

$$\tau = \frac{1}{2\pi} \Gamma (D_0 D_\nu)^{-1/2} d \quad (8)$$

is the trapping time. For large times the distribution function has a gaussian profile of halfwidth

$$\gamma = 2(D_\nu \tau)^{1/2}, \quad (9)$$

and decreases exponentially with time constant τ .

A comparison of eq. (7) with experiment is shown in fig. 2. The theoretical curve describes reasonably well the observed R_2 signal curve. A discrepancy at small times may be due to limitations of our spectral-spatial diffusion model for small times.

Since D_0 decreases as $(N^*)^{-1}$ and D_ν is assumed to be independent of N^* , the theoretical trapping time of eq. (8) shows a square-root dependence of N^* and it increases linearly with d . The theory (solid line in fig. 3) can well describe the N^* dependence of the trapping time as well as its dependence on crystal dimension; the analysis delivers $D_\nu \approx 5 \times 10^{23}$ s⁻³; taking account of phonon reflection at the crystal surfaces, the effective crystal dimensions may be larger than our characteristic dimension d , and D_ν may therefore be slightly smaller. It follows from eq. (9) that the spectral width of a phonon packet reaches a value of $\gamma \approx 2 \times 10^9$ Hz that is four times the halfwidth of the $\bar{E} \rightarrow 2\bar{A}$ resonance line.

We note that we assumed in our analysis that the scattering of a phonon at an excited Cr³⁺ ion does not take time, i.e. that the collision time is negligible compared with the mean time of flight τ_{res} between two successive collisions. However, this condition may not be fulfilled, especially for phonons in the line center. Since in our experiment the concentration of excited Cr³⁺ ions was much larger than the number of phonon states within the halfwidth Γ , $N^* \gg Z(\nu_0)\Gamma$ ($\approx 3 \times 10^{16}$ cm³), it was expected that phonons at resonance

have a collision time, which corresponds to the lifetime T_1 of the $2\bar{A}$ level, that is much larger than τ_{res} , namely $T_1/\tau_{\text{res}} = N^*/[Z(\nu_0)\Gamma]$. We have performed an analysis of our experiment taking into account this finite collision time by introducing reduced elastic and inelastic diffusion coefficients in eq. (6) and by taking into account the splitting of the energy levels by a magnetic field. We were, however, not able to obtain a reasonable agreement between such a modified spectral-spatial diffusion model and our experimental results. We suggest that phonons in the resonance line center undergo strong fast spectral diffusion and that immediately after phonon generation already a broadened phonon distribution is reached; such a distribution may be the initial distribution for which eq. (6) can be applied. A strong inelastic scattering with small frequency steps may be due to scattering at exchange-coupled Cr^{3+} pairs [1]. A fast initial spectral redistribution process is consistent with the experimental signal curve at small times (fig. 4).

Our analysis therefore gives evidence that monochromatically generated phonons undergo a first fast spectral diffusion and that a broadened phonon distribution is established. The further behavior of the phonon distribution can be described by the model of spectral-spatial diffusion of eq. (6). As we have seen, this model describes well the N^* dependence and the volume dependence of the trapping time τ . According to the model, influences of crystal defects that lead to inelastic phonon scattering can be described by modifying the spectral diffusion constant D_ν . Irradiation of the crystal with X-rays, i.e. most likely creation of Cr^{2+} ions, increases D_ν and leads, according to eq. (9), to a reduction of τ (fig. 8). The detailed processes which influence the inelastic phonon lifetime are not known.

We finally point out that the observation of phonons far away from the resonance frequency (fig. 5) cannot be explained by our simple spectral-diffusion model. According to our analysis, there should be no signal of the strength we observed (though it was a weak signal). It is most likely that phonons undergo inelastic scattering with a variety of frequency shifts ν_R and that therefore phonons are observable still at large

frequency distances compared to the originally generated phonons.

5. Conclusions

We have presented a method for monochromatic phonon generation in ruby crystals that contained large concentrations N^* of excited Cr^{3+} ions. We found that the phonon trapping time increased as $(N^*)^{1/2}$ and increased also with increasing sample size. The trapping time was different for Czochralski- and Verneuil-grown crystals and could be strongly influenced by X-ray irradiation. The experimental results are analyzed by use of a model of spectral-spatial phonon diffusion. The origin of inelastic scattering processes that give rise to spectral diffusion is not yet known.

Acknowledgements

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