Suppression of stimulated phonon emission in ruby by a magnetic-field gradient

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(Received 6 May 1996)

It is demonstrated that a magnetic-field gradient can suppress amplified stimulated emission of phonons taking place within the Zeeman-split $\bar{E}(^2E)$ doublet of Cr^{3+} in 700 ppm ruby. The doublet is initially inverted by selective pulsed optical pumping. The field gradient produces a gradient in the transition frequency, thereby reducing the gain of the amplifying medium. The experiments are described with rate equations in which the gain is accounted for by the interaction length over which the phonons remain at resonance. [S0163-1829(97)05505-7]

I. INTRODUCTION

In ruby it has been shown that phonons resonant with the Zeeman-split optically inverted $\bar{E}(^2E)$ doublet can participate in amplified stimulated emission, leading to strong phonon avalanches.[1,2] In these experiments it has also been demonstrated[2] that the end faces of the crystal may form a phonon ''cavity,'' in which the phonons travel back and forth. The phonon beam appears to be highly directional for a pencil-shaped optically excited zone in conjunction with specular reflection by the crystalline end faces.

The present paper shows that the phonon amplification can be reduced, and even completely suppressed, by a gradient in the magnetic field providing the $\bar{E}(^2E)$ splitting. Such a gradient introduces a gradual spatial variation of the resonance frequency,[3] and, as a result, shortens the length over which the propagating phonons remain at resonance with the excited centers. The phonon amplification then is controlled in a way which resembles optical Q switching. A prerequisite for this phenomenon to occur is of course a significant directionality of the phonon amplification along the field gradient. To describe the results, use is made of the model based on rate equations that has been worked out in detail in Ref. 2. A natural ingredient of the model is the distance over which phonon amplification takes place.

II. EXPERIMENTS

The experiments were performed on a Czochralski-grown single crystal of ruby with a Cr^{3+} concentration of 700 at ppm and with dimensions of $1.9\times1.9\times6$ mm^3. The crystalline c axis was at an angle $\theta=69°$ with respect to the longest dimension. The crystal faces were polished to a roughness of 0.2 μm. The sample was mounted in a bath cryostat, and cooled with superfluid He to suppress thermally induced relaxation processes, notably Orbach processes via $2\bar{A}(^2E)$ and Raman processes. An electromagnet provided a uniform magnetic field of 3.0 T parallel to the longest dimension of the crystal, i.e., at an angle of 69° with respect to the c axis. In this configuration, the Zeeman splitting of the $\bar{E}(^2E)$ excited state ($g=2.445$) amounts to $g\mu_B B\cos\theta/hc=1.23$ cm^{-1}, while the direct relaxation time between the $\bar{E}(^2E)$ levels equals $T_1=2.2$ ms.[4]

To generate a magnetic-field gradient in the direction of the external field, two oppositely wound 20-turn coils were positioned around the sample. These coils had an inner diameter of 2.6 mm, and their centers were 4.0 mm apart. Field gradients dB/dx of up to 0.10 T/mm could thus be produced in the center of the crystal with a rise time of about 2 μs. These gradients require excitation currents of up to 10 A, as calculated from the extended Helmholtz geometry.

A dye laser, pumped by a Q-switched Nd:YAG laser, achieved selective population of the upper $\bar{E}(^2E)$ level, $\bar{E}(^2E)_+$, with light pulses of 10-ns duration. For the present experiments, pulse energies of 1 mJ or less were adequate. The laser beam, focused to a diameter of about 0.2 mm, produced a cylindrical excited zone parallel to the magnetic field. To measure the populations of the $\bar{E}(^2E)$ excited levels as a function of time, the luminescence emanating from the crystal at right angles to the laser beam was analyzed by means of a 1.8-m single monochromator followed by standard photon-counting and averaging equipment. The temporal resolution was limited to 50 ns.

III. RESULTS

Figure 1 demonstrates the central point of this paper: A magnetic-field gradient of sufficient magnitude inhibits the development of an avalanche of phonons resonant with $\bar{E}(^2E)$. The population N_- of the lower $\bar{E}(^2E)$ level, $\bar{E}(^2E)_-$, is plotted as a function of time for various field

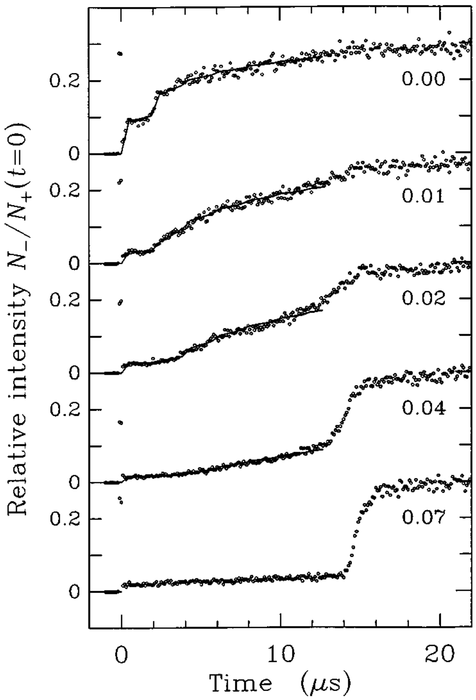

FIG. 1. Suppression of stimulated phonon emission after inversion of $\overline{E}(^2E)$ in ruby by a magnetic-field gradient. Field gradients of up to 0.07 T/mm, as indicated by the labels, are switched on 1 μs before the laser pulse, and switched off 14 μs later. All other conditions are held the same. The initial inversion amounts to $N_+/\rho\Delta\nu = 3.0\times10^5$. Gradients of 0.07 T/mm and larger completely quench the phonon avalanche at this inversion.

gradients superimposed on a constant homogeneous field of 3.0 T under otherwise the same conditions. The field gradient is switched on well before the laser pulse excites $\overline{E}(^2E)_+$, and is switched off about 14 μs later, which is much longer than it takes for the avalanche to develop at the given pumping power. Comparison of the traces shows that a field gradient can indeed be used to control the process of stimulated emission. For field gradients larger than 0.007 T/mm, the avalanche is completely suppressed at this laser power. In these extreme cases, the amplification is frustrated until the field gradient is removed, and the medium recovers its original gain.

The repeated amplification observed in Fig. 1 in the presence of smaller field gradients is a consequence of the cavity formed by the boundaries of the crystal. As is discussed in Ref. 2, phonons that have left the active zone reflect from the end faces of the crystal acting as partial mirrors, and so a fraction of them actually returns to the active zone. This leads to a recurrence of the phonon amplification, which shows up as a further increase of N_-. Detailed inspection of the phonon dynamics has shown that the growth of N_- is particularly enhanced after the phonons have completed a full round trip through the crystal. Whereas the avalanche developing immediately after the laser pulse exhibits a recurrence (upper traces in Fig. 1), this phenomenon is not observed for the avalanches starting up after removal of the field gradient. The first reason for this is that at the time the field gradient is switched off phonons originating from heating by the laser have long since disappeared, so that the avalanche must be initiated by thermal phonons and phonons produced by spontaneous emission. The second reason lies in the time constant of the coils, which compares with the round-trip time of the phonons.

To describe the experiments, we rely on the model developed in Ref. 2, which allows us to accommodate the presence of the gradient field via a shortening of the length over which the propagating phonons stay at resonance of the medium. In this model, the phonon avalanche scales with the integral

$$N_0(x_0,x_1) = \frac{1}{vT_1}\int_{x_0}^{x_1} n_0(x)\,dx, \tag{1}$$

in which the integration is restricted to that part of the excited zone in which the phonons are resonant with the $\overline{E}(^2E)$ two-level system. In Eq. (1), v is the velocity of the transverse acoustic phonons, and T_1 is the direct spin-phonon relaxation time ($T_1 = 2.2$ ms in the present configuration). The quantity $n_0(x) = (N_+ - N_-)/\rho\Delta\nu$, in which $\rho\Delta\nu$ is the density of phonon modes resonant with the $\overline{E}(^2E)$ transition, represents the initial inversion created by the laser pulse. In view of the relatively weak optical absorption, $n_0(x)$ may be assumed not to vary significantly with x. Introducing the resonance length

$$\Lambda = x_1 - x_0, \tag{2}$$

we thus have $N_0(x_0,x_1) = n_0\Lambda/vT_1$. The linewidth $\Delta\nu$ is known from independent measurements[5] to be $\Delta\nu = 95\pm10$ MHz. Because the phonon amplification is primarily due to transverse acoustic phonons, we adopt the Debye approximation for ρ. For the present phonon energy, we then have $\rho = 1.29\times10^5\ \mathrm{cm}^{-3}$ s, so that $\rho\Delta\nu = 1.23\times10^{13}\ \mathrm{cm}^{-3}$.

Quantitative results for Λ as a function of the field gradient may be arrived at by applying the model inclusive of the recurrence of the stimulated emission in the way described in Ref. 2. Apart from Λ, the parameters to be adjusted are the number of phonons starting up the first stage of the amplification (more precisely the combination εP_0 in Ref. 2), the coefficient R for specular reflection by the end faces, the ratio ε'/ε between the fraction of phonon modes participating in the later stages of the amplification and the fraction participating in the initial stage, and finally the combination $\varepsilon' l \tau_a$, where the time constant τ_a is a measure of the anharmonic coupling among the resonant phonons and l is the spatial extent of the phonon pulse.

Calculations show that anharmonic coupling is responsible for a loss of the resonant phonons predominantly by upconversion to phonons of double the energy. This loss mechanism is quadratic in the phonon occupation number, and becomes sizable in avalanches achieving high occupations. Note that anharmonic coupling is also effective during the free flight of the phonons from one passage through the resonant zone to the next. The proportionality constant N_+/P between the initial inversion ($N_- \approx 0$) and the laser pulse power follows from the absorption of the laser pulse and the size of the focus, but is left to vary to the extent permitted by the quite substantial uncertainties in these quantities. Furthermore, the amplitude P_0 of the initial phonon pulse cannot correspond to temperatures higher than, say, 10

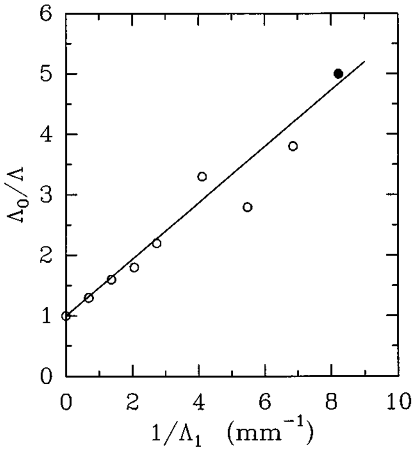

FIG. 2. Fitted results for $1/\Lambda$, normalized to a zero field gradient, as a function of $1/\Lambda_1$. The open circles refer to results from Fig. 1 and five similar traces. The solid circle is derived from Fig. 3 in comparison with Fig. 4. The straight line represents Eq. (4).

K in consideration of the fact that no Orbach relaxation via $2\bar{A}(^2E)$ is observed when stimulated emission is eliminated by pumping into $\bar{E}(^2E)_-$ instead of $\bar{E}(^2E)_+$.

The drawn curves in Fig. 1 are the results of fits of the model to the experimental traces obtained by adjustment of the above fitting parameters. Because all conditions except the field gradient were the same from trace to trace, a single comprehensive fit has been made to the top four traces in Fig. 1 together with five similar traces not shown in the figure. Except for Λ, which was allowed to vary from trace to trace, all fitting parameters were adjusted simultaneously.

The fitted results for the resonance length Λ versus the field gradient allow us to derive a value for Λ in the absence of a field gradient. Indeed, in the latter case Λ does not diverge, but reaches a limit that is determined by macroscopic inhomogeneities either in the splitting parameters or in the constant field supplied by the electromagnet. We denote this zero-inhomogeneity limit by Λ_0. We further define Λ_1 as the contribution to Λ that is induced by the applied field gradient. For each trace such as the ones in Fig. 1, Λ_1 is directly known from the strength of the gradient field in relation to the linewidth of the transition. More specifically, upon noting that the resonance frequency of the $\bar{E}(^2E)$ centers varies in space according to $d\nu/dx=(\nu/B)(dB/dx)$, we have

$$\Lambda_1=B\left(\frac{\Delta\nu}{\nu}\bigg/\frac{dB}{dx}\right). \tag{3}$$

For a gradient of 0.10 T/mm, corresponding to the maximum current used, we thus find $\Lambda_1=0.077\pm0.008$ mm. On heuristic grounds we adopt the plausible assumption that the effects of Λ_0 and Λ_1 add inversely, and thus write for the fitted Λ

$$\frac{1}{\Lambda}=\frac{1}{\Lambda_0}+\frac{1}{\Lambda_1}. \tag{4}$$

In Fig. 2, the fitted results for $1/\Lambda$ are plotted as a function of $1/\Lambda_1$. Also included in Fig. 2 is a more precise point derived

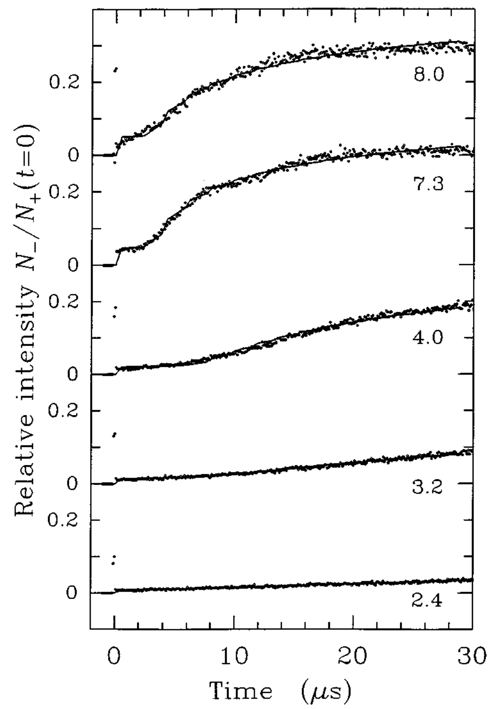

FIG. 3. The growth of N_- as a function of the time after the inverting laser pulse for a series of pumping powers in a field gradient of 0.06 T/mm. The labels denote the initial inversion $N_+/\rho\Delta\nu$ in units 10^5. For the smaller inversions, stimulated emission does not start until after 30 μs, when the field gradient is switched off.

from a comparison discussed below of a set of traces at a field gradient of 0.06 T/mm (Fig. 3) with a similar set in a zero field gradient (Fig. 4). Within errors, a linear dependence of $1/\Lambda$ versus $1/\Lambda_1$ is indeed found to hold, which confirms the assumption made in Eq. (4). The straight line in Fig. 2 directly yields $\Lambda_0=0.55\pm0.1$ mm, which compares with the results from other experiments.[2,5] It also is in conformity with the findings by Jessop and Szabo[6] and Muramoto[7] on macroscopic inhomogeneities in ruby.

With regard to the other fitting parameters derived from Fig. 1, the coefficient for specular reflection turned out to be $R=0.3\pm0.1$, while $\varepsilon P_0\sim4\times10^{-4}$ and $\varepsilon'/\varepsilon\sim10^{-4}$. It is noted that ε'/ε can be varied by an order of magnitude to either side without substantial deterioration of the quality of the fits, provided τ_a is simultaneously adjusted such as to leave the product $\varepsilon' l\tau_a$ approximately invariant. The smallness of ε'/ε is a manifestation of the strong directionality of the phonon beam reflecting back and forth between the crystal end faces. From the initial increase of the experimental traces, we estimate that the initiating phonon pulse has a duration of approximately 0.5 μs, corresponding to $l\approx0.3$ cm. Assuming further, somewhat arbitrarily, that the initial temperature of the phonon pulse corresponds to approximately 8 K, we arrive at $\varepsilon\sim0.5$ and $\varepsilon'\sim3\times10^{-5}$. With the fitted value $\varepsilon' l\tau_a\approx3\times10^{-6}$ cm s, we then find $\tau_a\sim0.3$ s for the anharmonic decay.

We finally briefly discuss a series of time traces at various optical pumping powers and a fixed field gradient of 0.06 T/mm (Fig. 3). These traces first show that in a field gradient the initial inversion need be sufficiently high for a phonon

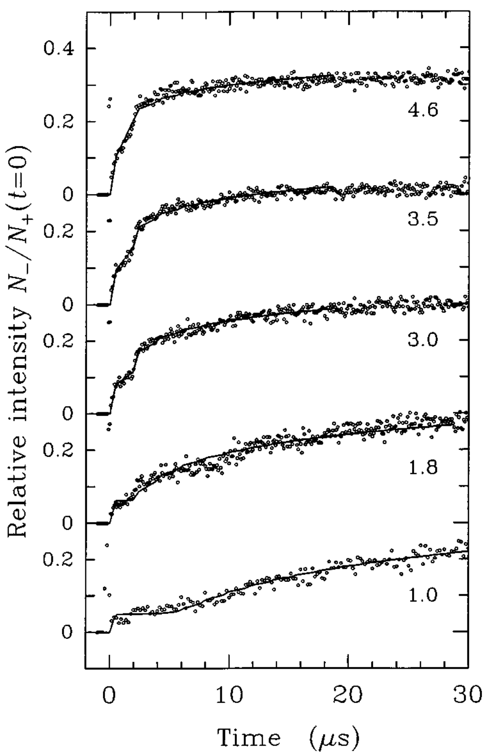

FIG. 4. The growth of N_- as a function of the time after the inverting laser pulse for a series of pumping powers in the absence of a field gradient. The labels denote the initial inversion $N_+/\rho\Delta\nu$ in units of 10^5.

avalanche to develop. Comparison with Fig. 4, which presents a series of traces at similar pumping powers in a zero field gradient, furthermore shows that a sizable field gradient is extremely detrimental to the development of the avalanche. The drawn curves in Figs. 3 and 4 are again fits to the model of Ref. 2 with inclusion of recurrence, accomplished in the way described above. In both figures, the model was fitted simultaneously to all traces by adjustment of a single set of parameters, except, of course, the number of phonons participating in the first stage of the amplification. The ratio of the Λ's pertaining to the two figures, i.e., Λ/Λ_0, was found to be 5.0 at the relevant gradient field. This result is inserted in Fig. 2 as the solid circle. Since it is based on a complete series of traces rather than a single trace, it is substantially more precise than the entries derived from Fig. 1. As for the remaining parameters, their output values compare with the results given in connection with Fig. 1 above.

IV. CONCLUSIONS

In conclusion, we have demonstrated that the stimulated emission of resonant phonons by an inverted medium can be controlled by a magnetic-field gradient to the extent of complete suppression. In the present case, the inverted medium consisted of the $\overline{E}(^2E)$ Kramers doublet in dilute ruby, the upper level of which was populated by optical pumping. Phonons that have traveled beyond a resonance distance determined by the linewidth and the field gradient on the one hand and intrinsic inhomogeneities on the other no longer take part in the process of stimulated emission. The reduction of the resonance length by the field gradient is direct experimental evidence of the directionality of the phonon amplification, and shows that the resonance distance determines the gain of the amplifying medium. In the case of complete frustration of the avalanche, removal of the field gradient leads to a delayed startup of stimulated emission. In this respect, there is a similarity with optical Q switching. An analysis of the experimental data yielded a value of 0.55 ± 0.1 mm for the resonance distance in the absence of a field gradient.

ACKNOWLEDGMENTS

The work was supported by the Netherlands Foundation Fundamenteel Onderzoek der Materie (FOM), the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO), the U.S. National Science Foundation (Grant No. DMR 9015468), and NATO (Collaborative Research Grant No. 890931).

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