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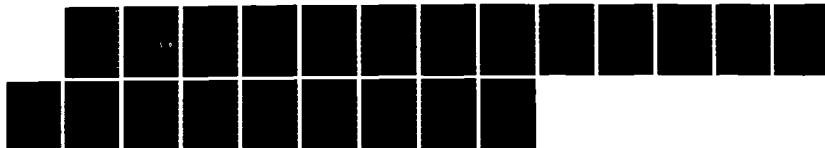
COHERENT PHONON GENERATION AND HIGH FREQUENCY PHONON
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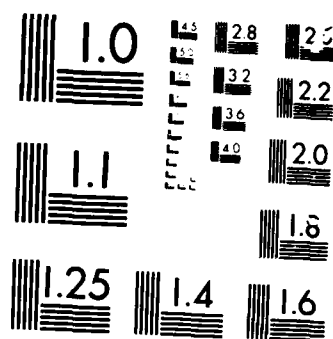
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A number of optical techniques have been developed in recent years which have been applied to study the dynamics of high frequency phonons. Monoenergetic phonons were generated by spin-lattice relaxation between two electronically excited states of an impurity ion. Resonant trapping provided the means to detect

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20. ABSTRACT CONTINUED

these phonons with good temporal resolution. VSPS was used to detect high frequency phonons over a wide frequency range with excellent temporal, spectral and spatial resolution. The life-time of a monoenergetic non-equilibrium distribution of phonons was determined by the observation of optical dephasing, using free induction decay(FID), which results from phonon-induced coherence loss(PICOLO). Non-resonant fluorescence-line-narrowing(FLN) techniques provided a means to determine the spectral distribution of non-equilibrium phonons generated by spin-lattice relaxation.

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Statement Of Problem Studied

We proposed to attempt coherent generation of high frequency phonons and to use these phonons to study high frequency phonon physics. While the results of the work described in this final report are different to a great extent from the goals of the original proposal, they are nonetheless significant and relevant to the question of stimulated phonon emission.

The reasons for these differences resulted from our discovery, shortly after receipt of funding, that our original observation of stimulated phonon emission in $\text{LaF}_3:\text{Er}^{3+}$ (Phys. Rev. B25 , 5064(1982)) was in question. When we repeated these experiments we became aware of severe problems from sample heating. This heating ($\approx 5\text{K}$) resulted in the dominance of the Orbach process in relaxing the populations among the excited state Zeeman levels, not stimulated phonon emission. While we may have observed effects of stimulated phonon emission in our original experiments, we were unable to confirm this.

As a result, we have focussed much of our attention on understanding why we were unsuccessful in observing stimulated emission in this system and to better identify the critical elements in obtaining stimulated phonon emission in this and other systems. In the process we have learned a great deal generally about phonon dynamics and have developed techniques to obtain the requisite parameters to evaluate systems for stimulated phonon emission.

We should point out however that in just this past year, stimulated phonon emission within a system, conceptually identical to the example we have been studying, has been observed by a group in the Netherlands (Miltenburg, Jongerden, Dijkhuis and de Wijn, Phonon Scattering In Condensed Matter , ed. W. Eisenmenger, K. Lassmann and S. Dottinger (Springer-Verlag, Berlin, 1984) p. 130) using the Zeeman resonance in the $E(^2E)$ state of Cr^{3+} in Al_2O_3 (ruby). The system we are studying would extend stimulated phonon emission to much higher frequencies (>200 GHz) than the ruby example which was carried out at 46 GHz. The positive results in the ruby system gives us renewed encouragement to pursue stimulated phonon emission in other systems at higher frequencies with the ultimate goal of generating coherent phonons.

In this final report we describe the main problems studied, summarize our most important results, draw conclusions from our work relevant to stimulated phonon emission, and discuss what steps need to be considered to improve our chances for the achievement of stimulated phonon emission in other systems and at higher frequencies.

In this second three year grant period we have continued to investigate high frequency phonon dynamics using a variety of optical techniques which have been specifically developed for these studies. The problems we have examined are summarized in three sections below.

1. The investigation of lifetimes of high frequency (> 200 GHz) phonons using phonon generation with excited state

spin-lattice relaxation coupled with the detection technique of hot luminescence, vibronic sideband phonon spectroscopy (VSPS), and a new technique developed with R. M. Macfarlane, phonon induced optical dephasing (PICOLO).

2. A study of the role of 2-phonon Raman processes in high frequency phonon dynamics.
3. The use of non-resonant fluorescence-line-narrowing (FLN) techniques to determine excited state resonance widths, which yield the spectral distribution of resonantly generated phonons.

These problems have been investigated in several insulating ionic solids, doped with small concentrations of impurity ions, for which the excited electronic states are accessible with visible light.

Summary Of Most Important Results

A number of optical techniques have been developed in recent years which we have applied to study the dynamics of high frequency phonons. Monoenergetic phonons were generated by spin-lattice relaxation between two electronically excited states of an impurity ion. Resonant trapping provided the means to detect these phonons with good temporal resolution. VSPS was used to detect high frequency phonons over a wide frequency range with excellent temporal, spectral and spatial resolution. The lifetime of a monoenergetic non-equilibrium distribution of phonons was determined by the observation of optical dephasing, using free induction decay(FID), which results from phonon-induced coherence loss(PICOLO). Non-resonant fluorescence-line-narrowing(FLN) techniques provided a means to determine the spectral distribution of non-equilibrium phonons generated by spin-lattice relaxation. We describe below some important details about our major results.

1. Anharmonic decay of phonons and the role of 2-phonon Raman processes in phonon dynamics.

a) Anharmonic lifetimes of low frequency optical phonons in $\text{LaF}_3:\text{Er}^{3+}, \text{Pr}^{3+}$ - (Publications 1 and 2)

Monoenergetic non-equilibrium phonons were generated at 41 and 54 cm^{-1} by single phonon relaxation between excited states of Er^{3+} . Detection was accomplished with anti-Stokes VSPS using the $^3\text{P}_0$ state of Pr^{3+} . The lifetime of these phonons

was determined by delaying the detector laser relative to the phonon generation. LaF_3 has two almost dispersionless low-lying optical phonon branches near 41 cm^{-1} , which intersect the TA mode near the zone boundary. Our analysis indicates that at 41 cm^{-1} the measured lifetime of 46 nsec is dominated by the optical mode lifetime which is determined from our experiment to be 40 nsec. At 54 cm^{-1} the LA and optical modes contribute about equally to the decay. It is very significant that these optical phonon lifetimes are two to four orders of magnitude longer than typical optical phonon lifetimes which have been measured by a number of researchers in a variety of materials. The probable explanation is that because the mode energies in this case are considerably less than the optical modes studied in most other materials, the phonon densities of states for the decay products are much lower than the typical system.

b) Detection of surface absorption of light from the resulting phonons in $\text{LaF}_3:\text{Er}^{3+}$ - (Publications 3 and 11)

Nominally transparent solids exhibit a weak surface absorption of light for reasons not completely understood. We have measured this for $1.06 \text{ }\mu\text{m}$ and $0.53 \text{ }\mu\text{m}$ laser radiation on LaF_3 and have found that it is possible to detect the resulting non-equilibrium phonons in the bulk with measurements of excited state population dynamics of Zeeman-split sublevels. Model calculations which include 2-phonon Raman processes, anharmonic decay and recombination, and elastic scattering,

confirm that for up to 15 μ sec after the heat pulse, the phonon dynamics in the detector region immediately behind the heated surface, is dominated by rapid resonant Raman processes involving the crystal field levels of the $^4S_{3/2}$ manifold of Er^{3+} . The phonons which diffuse out of the heated surface region undergo recombination which maintains the population of the very high frequency phonons much longer than their anharmonic decay times. It is these phonons which govern the population dynamics through resonant Raman scattering in the detector volume.

- c) PICOLO - A new phonon detection technique - its application to anharmonic lifetimes in $LaF_3:Pr^{3+}$ - (Publications 5, 6 and 9)

Optical dephasing by an equilibrium thermal distribution of phonons has been studied extensively in $LaF_3:Pr^{3+}$ by several researchers. In recent experiments conducted at I.B.M., San Jose, in collaboration with R. M. Macfarlane, it was demonstrated that monoenergetic non-equilibrium phonons can lead to phonon-induced coherence loss (PICOLO) when 23 cm^{-1} phonons are generated by spin-lattice relaxation between the lower two states of the 1D_2 manifold in Pr^{3+} . An analysis of the free-induction-decay (FID) in the presence of these phonons yielded a lifetime of 500 nsec for the 23 cm^{-1} phonons, in good agreement with recent results we obtained by observing the time dependence of the hot luminescence following laser excitation of the $^1D_2(II)$ state. (See d) below). This is a technique

with high sensitivity (can detect phonon occupation numbers, p , as small as 10^{-7} in some cases) which should be applicable in a number of systems.

d) Resonant Raman scattering of 23 cm^{-1} phonons in $\text{LaF}_3:\text{Pr}^{3+}, \text{Dy}^{3+}$ - (Publications 7, 10 and 13)

Resonant Raman processes lead to a dramatic reduction of the lifetime of 23 cm^{-1} phonons in doubly doped $\text{LaF}_3:\text{Pr}^{3+}, \text{Dy}^{3+}$. The lowest two crystal field states of the ground manifold of Dy^{3+} are separated by 15 cm^{-1} in zero magnetic field. A magnetic field Zeeman-splits the two levels leading to a resonance between the 23 cm^{-1} phonons and the Zeeman components of the lowest two crystal field levels of the Dy^{3+} ion at 15.9 and 23 kG. 23 cm^{-1} phonons are generated by fast spin-lattice relaxation following optical excitation of the $^1\text{D}_2(\text{II})$ state of Pr^{3+} . The lifetime of the 23 cm^{-1} phonons, which is 600 nsec at, zero field is reduced to 125 nsec for fields in the immediately vicinity of 15.9 kG, with a second reduction to 250 nsec occurring near 23 kG. Detailed calculations indicate that resonant 2-phonon Raman processes within the Dy^{3+} ground manifold are responsible for these lifetime reductions.

2. Non-Resonant Fluorescence-Line-Narrowing (FLN) - Phonon Resonance Widths

The generation of phonons by spin-lattice relaxation

between optically excited electronic states produces a distribution of phonons of spectral width equal to the resonance width between the states in question. It is important to know this spectral width in order to calculate the degree of phonon bottlenecking present, and from that, the extent to which we can deduce phonon dynamics from these studies. It is also important to know this parameter for the cases in which we are attempting to observe stimulated emission of phonons. The threshold for stimulated emission is directly related to the phonon spectral width.

With a few exceptions, resonance widths between excited electronic states have not previously been measured. The non-resonant FLN technique provides us with a very high resolution direct method for making these measurements which is of quite general applicability.

a) Zeeman resonance widths in $\text{LaF}_3:\text{Er}^{3+}$ - (Publications 8 and 12)

Non-resonant FLN was used to determine the resonance width between the Zeeman-split components of the $^4F_{9/2}$ state of Er^{3+} in LaF_3 . LaF_3 is complicated by the presence of six magnetically inequivalent sites. Because of the large anisotropy of the g-tensors of the ground and excited states, only a small misalignment splits the spectrum into three resolvable groups of sites. When the crystal was carefully aligned to minimize this effect, the resonance width was determined to be 225 MHz at 28 kG, decreasing to 150 MHz at 12 kG.

Since the fluorescence from the lower Zeeman component of the $^4F_{9/2}$ state to the two Zeeman components of the ground state ($^4I_{15/2}$) was resolved in this technique it was also possible to determine the ground state resonance width to be 90 MHz at 28 kG.

In this case the non-resonant FLN is equivalent to a high frequency (200-300 GHz) ESR measurement, but with several major advantages. The technique works at very high frequencies where standard microwave sources are not readily available. The sensitivity inherent in optical techniques make it particularly useful for excited state ESR. The ability to study a select group of ions within the inhomogeneous absorption profile provides additional selectivity not available in standard techniques.

In this particular case, the change in the resonance width and the magnitude of the excited state g-value were measured as a function of pumping frequency within the inhomogeneous profile. A program is to be developed to correlate these changes with their dependence on the crystal field parameters which are altered by the presence of impurity ions.

b) Other materials - (Publication 8)

Non-resonant FLN was used to determine excited state resonance widths between crystal field states in $Al_2O_3:Cr^{3+}$ (ruby), $BeAl_2O_4:Cr^{3+}$ (alexandrite) and $LaF_3:Pr^{3+}$. In ruby the $E \rightarrow 2A$ resonance width was found to be consistent

with the value of 570 MHz observed by Lengfellner et al (Opt. Lettr. 8 ,220(1983)) using excited state far infrared absorption.

In alexandrite our result for the $E \leftrightarrow 2A$ resonance width of 13.9 GHz compares with the values of 2.1 and 9 GHz in two different samples determined by Goossens et al (Phonon Scattering In Condensed Matter , ed. W. Eisenmenger, K. Lassmann and S. Dottinger (Springer- Verlag, Berlin, 1984), p.112) from studies of phonon dynamics in a magnetic field.

In the case of $\text{LaF}_3:\text{Pr}^{3+}$ we measured the resonance width of the ${}^1D_2(\text{II}) \leftrightarrow {}^1D_2(\text{I})$ resonance which has been used to generate 23 cm^{-1} phonons in many of our studies. When the ${}^1D_2(\text{II})$ state is excited at line center of the inhomogeneous profile ($\Delta\nu_{\text{inh}} = 3 \text{ GHz}$), a resonance width of 2 GHz is obtained, which is dominated by the homogeneous width of this state (1.4 GHz) as determined by Erickson (Opt. Commun. 15 , 246 (1975)). However when the ions are excited in the wings of the resonance the excited state resonance acquires an inhomogeneous broadening up to 10 GHz.

CONCLUSIONS RELEVANT TO STIMULATED PHONON EMISSION

1. Sample heating due to optical excitation.

Heating is a problem in optical excitation techniques. Only 0.04% of the absorbed energy, and even a smaller fraction of the energy in the laser beam (0.01%), is converted into monoenergetic phonons by spin-lattice relaxation in $\text{LaF}_3:\text{Er}^{3+}$. Surface heating due to anomalous absorption at the surface, which results in an enhanced Orbach process rather than stimulated emission, seems to be the major factor which governs the relaxation. Some bulk heating must also occur due to nonradiative relaxation which will take place on some of the excited sites due to multiphonon emission, up conversion and other pair processes which conserve energy through the generation of phonons. In the case of ruby, because of strong optical absorption of the Cr^{3+} ions, and a relatively weak coupling by the phonons to the ${}^2\text{E}(2\text{A})$ level 29 cm^{-1} above the level generating the stimulated emission, it is the stimulated emission, not the Orbach relaxation which dominates. In $\text{LaF}_3:\text{Er}^{3+}$ the reverse situation seems to hold.

2. Anharmonic phonon lifetimes

Measurements of anharmonic decay times at several frequencies in LaF_3 indicate that for phonons in the energy range $5\text{-}10\text{ cm}^{-1}$ anharmonic breakup should not be a major loss mechanism. The lifetime of LA phonons at 23 cm^{-1} is about 50

nsec. Lifetimes of 41 cm^{-1} optical phonons are about 40 nsec. Scaling the 23 cm^{-1} lifetime by ω^{-5} yields an estimated lifetime of LA phonons of about 50 μsec at 6 cm^{-1} and 4 μsec at 10 cm^{-1} . The TA phonons are expected to be much longer-lived. The lifetime of 23 cm^{-1} phonons was confirmed with a new technique, PICOLO, using optical coherent transient techniques to detect the resonant phonons.

3. Role of resonant Raman scattering.

The mechanism by which the heat generated in the optical excitation dominates over stimulated emission is identified as resonant Raman scattering (RRS), otherwise known as the Orbach process. We have identified its role based on studies of the relaxation between the Zeeman levels of excited Er^{3+} ions after generating heat at the surface of the crystal with a non-resonant infrared laser. Fits of the data to computer solutions of the rate equations demonstrate the dominant role of RRS. The importance of RRS is also demonstrated in a double resonance experiment where 23 cm^{-1} phonons obtained from relaxation of excited Pr^{3+} ions are brought into resonance with ground state Dy^{3+} ions in a double-doped sample. A reduction of the 23 cm^{-1} phonon lifetime by a factor of five occurs whenever resonance with the 23 cm^{-1} phonons occurs. The 23 cm^{-1} phonons are inelastically scattered out of the resonance channel by the RRS on the Dy^{3+} . This process has been very successfully modeled with rate equations for the system.

4. Phonon resonance widths.

We developed a technique using non-resonant FLN to obtain information on the excited state resonance widths which play an essential role in the strength of the stimulated phonon emission. The observed widths (≈ 100 MHz extrapolated to zero field) were about what we anticipated. However some very important points have resulted from this study, some of which are relevant to the subject of stimulated phonon emission.

First, we found that the magnetic field must be aligned relative to the c-axis to better than 0.1° because the spin resonance frequencies of the six magnetically inequivalent sites become sufficiently equal only for this orientation. This was undoubtedly a problem in our early experiments when we did not appreciate this critical orientation requirement.

Second, we found that the g-values for the ions depend on the optical transition frequency selected by the laser from within the inhomogeneous absorption profile. As a result there is a spread of resonance frequencies over the distribution of Er^{3+} sites. This limits the effective resonance frequency distribution of the phonons to ≈ 400 MHz. This is almost an order of magnitude greater than the ruby resonance in which de Wijn's group has demonstrated stimulated phonon emission.

Third, the linear dependence of resonance width on field indicated that inhomogeneities of the field over the $2 \times 2 \times 6$

mm^3 excited volume are non-negligible when one is considering mean free paths for stimulated phonon emission of ~ 1 mm.

We are still positive toward further efforts to establish stimulated phonon emission using Zeeman resonances of ions in solids, especially with emphasis toward higher frequencies. In selecting systems for future work, several special requirements should be considered:

- (1) Systems with only one magnetic site are to be preferred.
- (2) Larger magnetic fields should be used to reduce the mean free paths for stimulated phonon emission and to enhance the gain.
- (3) Emphasis should be placed on systems with strong optical absorption in order to enhance the creation of monochromatic phonons due to stimulated emission relative to broadband phonon production due to surface absorption.
- (4) Ions with large energy gaps to the next excited crystal field level should be selected to minimize effects of the Orbach process.
- (5) Homogeneity of the magnetic field should be a consideration in the design of an experiment, taking into account the magnitude of the g-value, the resonance width, and the mean free path for stimulated phonon emission.

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1. Laser Detection of Phonons With Anti-Stokes Sideband-Induced Absorption, R. S. Meltzer, J. E. Rives and G. S. Dixon, Proc. of the Int. Conf. on Lasers, '82, New Orleans, 1982.
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8. Excited State Electron Spin Resonance Using Non-Resonant Fluorescence Line Narrowing, D. J. Sox, S. Majetich, J. E. Rives and R. S. Meltzer, J. de Physique , Colloque C7, 46 , 493 (1985).
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Meltzer and R. M. Macfarlane, Proc. of the Second Int. Conf. on Phonon Physics, Aug. 26-30, 1985, Budapest, ed. J. Kollar, et al, In press.

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11. Role of Raman Processes In Impurity-Induced Phonon Scattering, J. E. Rives, R. S. Meltzer and G. S. Dixon, to be published.
12. Non-Resonant Fluorescence Line-Narrowing in $\text{LaF}_3:\text{Er}^{3+}$, D. J. Sox, R. S. Meltzer and J. E. Rives, to be published.
13. Reduction of 23 cm^{-1} Phonon Lifetime Due To Resonant Raman Scattering By Dy^{3+} Ions in LaF_3 , S. S. Yom, J. E. Rives and R. S. Meltzer.

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1. Stimulated Emission of Tunable High Frequency Phonons in $\text{LaF}_3:\text{Er}^{3+}$, D. J. Sox, J. E. Rives and R. S. Meltzer, Int. Conf. on Phonon Physics, Indiana University, 1981.
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