OBSERVATION OF PHONONS GENERATED BY SPIN-LATTICE RELAXATION

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The diffusion of a "warm" band of phonons, generated by continuously saturating a spin resonance at one end of a $SrF_2:Tm^{2+}$ crystal, was measured throughout the rest of the crystal by optically monitoring the spatial distribution of the spin temperature. We interpret the experimental results in terms of a phonon transmission coefficient into the helium (~4%) and a mean free path determined by diffuse scattering (~25%) off the crystal surfaces.

Saturation of a paramagnetic spin system which relaxes by the direct one-phonon process should generate a hot band of resonant phonons in the crystal.[1] This phenomenon has been observed indirectly in strongly interacting systems as a phonon bottleneck in spin-lattice relaxation measurements.[2,3] More recently it has been observed that adiabatic fast-passage inversion of the spin resonance in Fe^{2+}-doped MgO can lead to a phonon "avalanche"[4] which if generated at one end of a rod can be detected at the other end.[5] We have generated what might be called a "warm" band of phonons by continuously saturating the spin resonance at one end of a SrF_2 crystal doped with Tm^{2+} and have detected the diffusion of these phonons throughout the rest of the crystal by optically monitoring the spatial distribution of the spin temperature. This has been attempted several times in the past using more conventional microwave methods of detection and was unsuccessful probably because either the spin system did not relax primarily by the direct one-phonon process, or the phonons were scattered by too strong an interaction with the spins, or the crystals had poor acoustical properties.[6-8] This is probably so because the values we obtain for the coupling of the phonons to the helium bath and the diffusion coefficient for the phonons along the crystals are reasonably close to the values anticipated.

Measurements were made on two crystals which contained 0.02% thulium, 90% of which was reduced to the divalent state by heating in strontium vapor. The paddle-shaped crystal *A*, whose dimensions are given in Fig. 1, was mounted with the shaded region inside a *K*-band waveguide against a side wall where the magnetic part of the rf waves are strongest. The stem was inserted through a small hole in the corner of a 3-mm thick shorting plate at the end of the waveguide. The two large surfaces on this crystal were polished with 0.5-μ diamond powder and the edges left as cut. The straight rectangular rod *B* was inserted 1.8 mm into a *K*-band waveguide through a hole in the center of a 2-mm thick shorting plate. All the surfaces on this crystal were polished.

A brass plate containing holes 1 mm in diameter centered every 2 mm was mounted next to the part of the crystal which extended out past the shorting plate. This provided a calibrated mask for the monitoring light to pass through. The monitor light came from a small tungsten lamp and passed through a filter with a 100-Å bandwidth and centered at 5800 Å, where the optical absorption bands of Tm^{2+} in SrF_2 have a large paramagnetic circular dichroism.[9] The unpolarized light, directed at 45° from the magnetic field direction for the convenience of avoiding the electromagnet's pole faces, was brought to focus on one hole in the mask. (Coming in at an angle of 45° instead of parallel to the field reduces the signal by only 30%.) The transmitted light was analyzed for the percent of circular polarization induced in the sample, which gives a measure of the spin orientation or spin temperature for a given magnetic field. Using an electro-optic po-

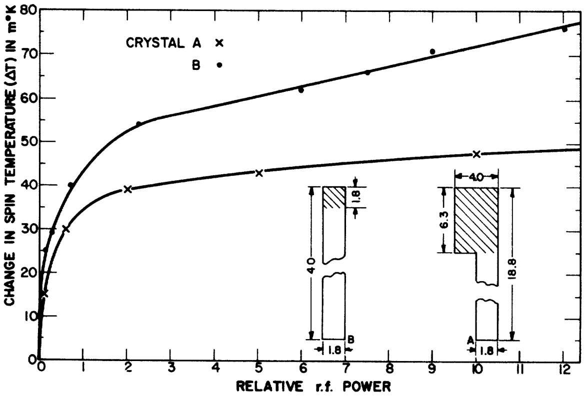

FIG. 1. Plots of the change in spin temperature as a function of applied rf power as measured 3 to 4 mm below the regions where the spins are saturated by the rf field (shaded areas). The dimensions of the crystals are in mm and both are 0.84 mm thick.

tassium dihydrogen phosphate crystal modulated at 8 kHz, followed by a linear polarizer, and a photomultiplier stabilized with feedback to keep the dc output level constant, we could detect with a 1-sec time constant changes in the spin temperature of about 1 part in 10^3.

The direct one-phonon spin-relaxation rate for Tm^{2+} in SrF_2 is independent of field orientation and given by

$$T_1^{-1} = 35H^5 \coth[g\beta H/2kT]\times 10^{-5}\ \sec^{-1}, \qquad (1)$$

where H is the field in kilogauss and T is the temperature. This rate dominates relaxation by Raman scattering of phonons at temperatures below about 5°K and cross relaxation to pairs and other impurities at fields above about 2 kG for the crystals used in these experiments. The relaxation rate at 30 GHz and 1.3°K is 6.5 $\sec^{-1}$; so the spin resonance can readily be saturated without the necessity of using a resonant microwave cavity.[10]

Data were taken by recording the fraction of induced circular polarization as measured at one hole of the mask while the magnetic field was slowly swept through one of the two Tm^{2+} spin-resonance lines. (The 100% abundant thulium isotope 169 has spin $\frac{1}{2}$.) Since the optical signal depends only on the parameter $g\beta H/kT$, the change in spin temperature can readily be obtained from

$$\delta T = -2\left[\frac{H}{T}\frac{\partial S}{\partial H}\right]^{-1}\delta S, \qquad (2)$$

where δS is the change in signal on resonance with the rf on and $\partial S/\partial H$ is the measured slope of of the signal with increasing magnetic field in the vicinity of the resonance. The factor of 2 is added because only one spin-resonance line is saturated.

Figure 1 shows that the change in spin temperature on resonance saturates at a rather low signal level in both crystals, the rf level in the waveguide being consistent with that required to saturate the spins in the waveguide. The geometry for the paddle-shaped crystal provided more isolation against rf leakage out of the hole in the waveguide, and so we could increase the rf power by 10 dB more than that shown with no measurable increase in the signal, while the slight increase with power shown after saturation for the rectangular rod is due to a small amount of rf leakage. (In this latter case we have measured ~30 dB of rf isolation.) The EPR linewidth in these crystals is 16 G, but the linewidths observed in both crystals outside the waveguide were broadened at power levels above the knee in the curves in Fig. 1. At the highest power level the linewidth observed with the paddle crystal A was 40 G. This broadening is due to complete saturation of the resonance in that part of the crystal in the waveguide by spin diffusion while on the edge of the line. The observation of saturation and line broadening at such small signal levels rules out the possibility that we are observing rf leakage out of the waveguide.

The intensity of this signal as measured along the crystal decreases exponentially as shown in Fig. 2. This can be described by the simple diffusion equation

$$K\frac{\partial^2\rho}{\partial x^2} - \tau^{-1}\rho + g = 0, \qquad (3)$$

where ρ is the excess-phonon density, K is the diffusion coefficient for phonons along the crystal, τ is the lifetime for the phonons to stay in the crystal and in the spin-resonance bandwidth, and g is the generation rate for the phonons. The excess-phonon density can be obtained directly from the measured change in the spin temperature using

$$\rho = \rho_0(\partial N/\partial T)\sqrt{2}\,\delta T, \qquad (4)$$

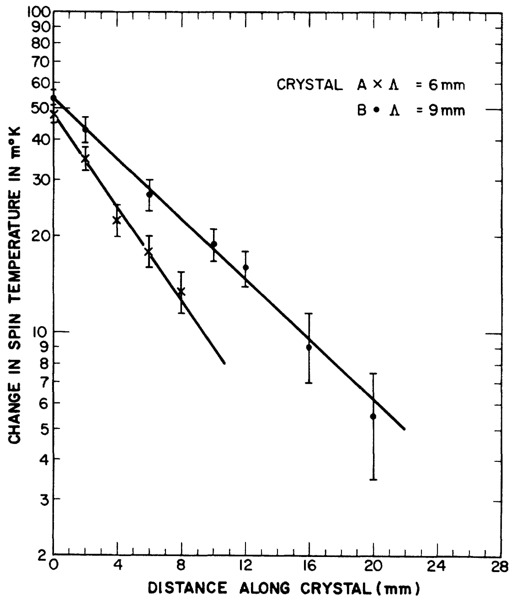

FIG. 2. Change in spin temperature along the crystals taken with the rf level set just above the point where the curves in Fig. 1 flatten out. The bath temperature is 1.3°K and the data shown for crystal A were taken at 26.6 GHz and those for crystal B at 34.4 GHz.

where $\rho_0 = 8.2\nu^2 \times 10^{-8}$ phonons/cc is the phonon density of states within the 70-MHz spin-resonance linewidth[11] and $N = [\exp(h\nu/kT)-1]^{-1}$ is the phonon occupation number. The effective change in the temperature for the phonons in the warm band is hotter than the detected change in spin temperature by the factor $\sqrt{2}$ because the phonons are distributed in frequency over an approximately Gaussian-shaped band and are detected by a group of spins with their resonances distributed over the identical line shape. This difficulty is similar to the general spectroscopy problem of trying to measure the peak intensity of a line whose width is comparable with the resolution of the spectrometer. It should be pointed out that SrF_2 is rather close to being acoustically isotropic; so the division into longitudinal and transverse phonons is a good approximation for all propagation directions. The spins interact primarily with the transverse phonons, and since the longitudinal phonon density of states at a particular frequency is less than 10% that of the transverse phonons, the longitudinal phonons can be ignored. The generation rate of the phonons can be calculated when the spins are completely saturated from

$$g = \tfrac{1}{2}T_1^{-1}N_0 \tanh^2[g\beta H/2kT], \qquad (5)$$

where N_0 is the conenctration of active ions. This is important because this is the only absolute calibrated source of phonons we know of and using it as a reference, one can measure the absolute spectral output of other sources. This generation rate also increases between the fifth and sixth power of the frequency; so the observed signal increases rapidly with frequency.

In the region where no phonons are being generated, the excess-phonon density will decrease exponentially in a characteristic length given by $\Lambda = (K\tau)^{1/2}$. If the excess-phonon density can be extrapolated from the measured region into the entire crystal using this diffusion equation, which is straightforward for the simple rectangular crystal B, then the phonon lifetime can be calculated using

$$\tau^{-1}\int \rho dV = \int g dV. \qquad (6)$$

This can be obtained by taking the volume integral of the diffusion differential equation, but physically it comes about by setting the total phonon loss rate equal to the total phonon generation rate. The data for crystal A can be reduced by extrapolating ρ as measured in the stem into the paddle and assuming that it is constant throughout the paddle region. The lifetime for the phonons in the paddle region can then be obtained by use of Eq. (4) if the loss of the phonons down the stem is taken into account.

Measurements were made with crystal B at frequencies from 29.6 to 38 GHz with the bath temperature at 1.3°K. The values obtained for the lifetime ranged from 20 to 10 μsec with a clear tendency for the higher frequencies to have shorter lifetimes. Measurements were made on crystal A at 18.9 and 26.9 GHz which gave values for τ of 24 and 12 μsec in the region of the paddle. The phonon lifetime should be determined at these frequencies and temperatures primarily by the rate phonons escape into the helium. This rate is given approximately by $Tv(4P/a)$, where T is the transmission probability, v we take as the transverse sound velocity (2.80×10^5 cm/sec), and P/a is the ratio of the perimeter of the cross section of the crystal to its cross-sectional area. For crystal B a lifetime of 12 μsec as obtained at 34.4 GHz gives $T = 3\%$. A simple acoustic impedance calculation gives $T = 4\rho_1 v_1/\rho_2 v_2 = 0.8\%$, where ρ_1 and v_1 are the density and sound velocity of helium, and ρ_2, v_2 are the density and transverse sound velocity of SrF_2. In fact, a transverse wave incident normal to the crystal surface should not couple to the longitudinal waves in the helium, but this gives what we believe to be a reasonable maximum for this transmission coefficient using a simple, classical, acoustic impedance match. The details of this coupling at the interface are intimately related to the problem of the Kapitza resistance, as encountered in thermal conductivity.

Finally, we can also obtain the diffusion coefficient K since we know Λ and τ, which in turn can give a mean free path for scattering of the phonons, $K = \frac{1}{3}v\Lambda$. Our best data for crystal B were taken at 34.4 GHz, where $\Lambda = 9$ mm and $\tau = 12.5$ μsec. This leads to a mean free path for the phonons of 6 mm. This implies a high degree of specular reflection off the surface since pure diffuse scattering would lead to a mean free path of about 1.3 mm, the "Casimir length" for this crystal. Indeed the data for crystal A, which was not polished on its edges, are consistent with a mean free path which is somewhat shorter. Electron microscope pictures of a SrF_2 surface polished the same as the crystals used for these experiments show the surface to be quite smooth on the 0.1-μ scale, which is the wavelength of 30-GHz phonons in SrF_2. The mean free path for scattering off the spins in these

0.02% crystals at 30 GHz and 1.3°K is 7 cm, and this decreases roughly as the inverse third power of the frequency.

In conclusion, we want to emphasize that besides performing a successful experiment on a long outstanding problem, this work also demonstrates most of the features of our tunable phonon detector concept.[12] Although in this problem the phonon generator is strongly dependent on the magnetic field, there are many problems where the spins can readily be tuned without affecting the phonon distribution, and so the spins can be used to measure that distribution. As examples of this we have recently measured[13] the harmonic output of piezoelectric transducers at 10 GHz and have made preliminary measurements on the spectral distribution of phonons in crystals with thermal gradients across them.[14]

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INFRARED REFLECTION SPECTRA OF $Ga_{1-x}In_xAs$: A NEW TYPE OF MIXED-CRYSTAL BEHAVIOR

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The infrared reflection spectra of $Ga_{1-x}In_xAs$ crystals reveal a new type of mixed-crystal behavior in which two bands are seen, one near the InAs Reststrahlen region and one which shifts monotonically with x between the InAs and GaAs Reststrahlen regions. A discussion is given in terms of postulated criteria using local and gap modes.

In this paper we report and offer an interpretation of measurements of the infrared reflectivity spectra of $Ga_{1-x}In_xAs$ single crystals. The spectra, which represent a new type of mixed-crystal behavior, are characterized by the occurrence of two Reststrahlen bands, a strong band which is shifted down in frequency monotonically out of the GaAs Reststrahlen region to the edge of the InAs Reststrahlen region with increasing molar fraction of InAs (i.e., increasing x) and a lower frequency weak band which occurs in the InAs Reststrahlen region. Workers had previously distinguished two types of behavior in similar mixed polar crystals.[1] Alloy crystals such as $Na_{1-x}K_xCl$[2] and $Cd_{1-x}Zn_xS$[3] have been reported to exhibit a single Reststrahlen band for all values of x. The frequencies characterizing this band are observed to be intermediate to those of the end members and shift monotonically with composition. Mixed-crystal systems exhibiting this type of behavior have been called "one-mode" systems. In contrast to this, "two-mode" behavior occurs in crystals such as $GaAs_{1-x}P_x$[4] and $CdS_{1-x}Se_x$[5]; here two reflection bands are observed for all compositions. For each band the frequencies of the optic modes lie approximately within the Reststrahlen region of the end-member constituent compounds ($x = 0$ or 1); how-