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PARAMAGNETIC RELAXATION TO A BOTTLENECKED LATTICE: DEVELOPMENT OF THE PHONON AVALANCHE*

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The problem of imprisoned or "bottlenecked" phonons, first introduced by Van Vleck,[1] has recently received considerable attention.[2-4] The term refers to a situation in which lattice vibrations interact more strongly with a paramagnetic transition of the same frequency than with their thermal environment. To date, the strongest reported evidence for a bottleneck appears to be the observation of size- and concentration-dependent characteristic times in the decay of a paramagnetic resonance signal after excitation by a saturating microwave pulse.[2-4] In this Letter we report what would appear to be much more compelling evidence for a bottleneck.

If one *inverts*, rather than saturates, a spin-resonance line and *no* bottleneck is present, the intensity of the line subsequently decays to its thermal equilibrium value in routine exponential fashion, characterized at low temperatures by the direct paramagnetic relaxation time T_{1d}. On the other hand, when a strong bottleneck exists, the inverted line starts to decay exponentially at the direct rate, but because of the bottleneck, there should be an attendant buildup of excitation among the resonant lattice modes which should cause a catastrophic decrease in T_{1d} itself and a sudden decay of the line. The process should cease when the paramagnetic ions and resonant phonons reach a common temperature—corresponding closely to saturation of the spin-resonance line—and afterward the ions and phonons should relax jointly, over a far longer time that is limited by the rate of de-excitation of the phonons. Thus one expects the spin-resonance line to exhibit a brief delay, a sudden decay with a knee at saturation, and a subsequent slow decay to thermal equilibrium. This "phonon avalanche" should depend most critically on the initial degree of inversion: The decay should become less abrupt as the initial value of inversion is reduced.

In Fig. 1 we show paramagnetic-relaxation curves for the cerium resonance in $(\mathrm{La}_{0.998}\mathrm{Ce}_{0.002})_2\mathrm{Mg}_3(\mathrm{NO}_3)_{12}\cdot 24\mathrm{H}_2\mathrm{O}$, a material in which other evidence has suggested a bottleneck.[2] Inversion was accomplished by adiabatic fast passage. A bimodal cavity was used, one mode for fast passage (which required about 50 watts of microwave power) and the other for monitoring the resonance signal (about 0.1 microwatt). Except for the bimodal cavity the spectrometer has been described elsewhere.[5] The frequency was 11.1 Gc/sec, and the Zeeman field was directed perpendicular to the c axis of the crystal. The sample measured 7 mm×5 mm×2.5 mm, and was surrounded by liquid helium. To obtain a relaxation plot, the Zeeman field was first swept up through the resonant value for the fast passage frequency, was then held at a constant "soaking" value for an adjustable time, and was finally swept further upward through the resonant value for the monitoring mode. The entire procedure was repeated at a different soaking time to obtain another data point. The earliest point in the Figure corresponds to no soaking interval and represents

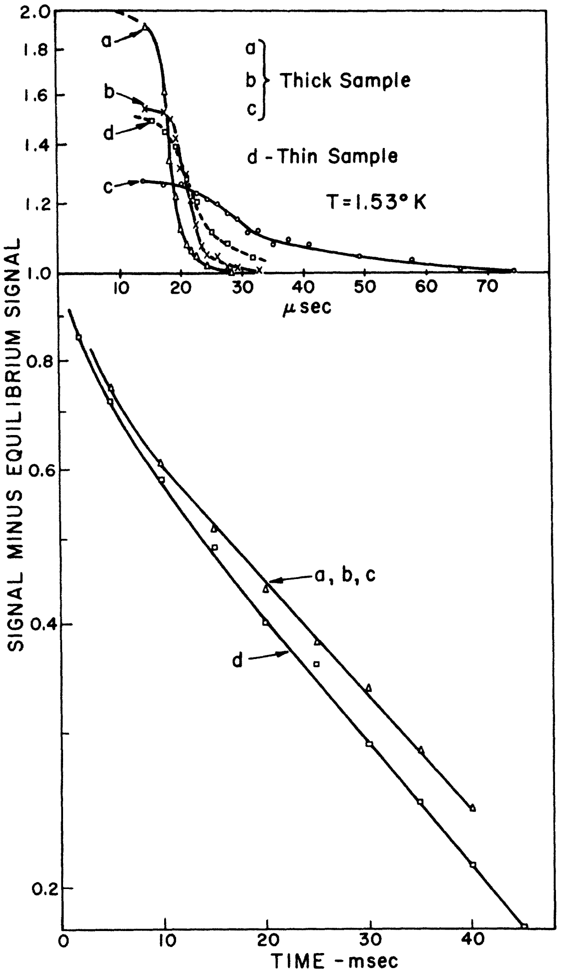

FIG. 1. Normalized plot of recovery after inversion, for different values of initial inversion and for two sample sizes. Complete inversion is 2.0; saturation is 1.0. Fast passage occurs at $t = 0$. Lower curves are continuations of upper curves.

the time required to sweep the Zeeman frequency from one mode to the other. Time resolution was, in fact, limited by the rate at which the Zeeman field could be changed; the above sequence was adopted in order to minimize the number of changes in the time derivative of the field. The initial degree of inversion could be altered by lowering the power level of the microwave pulse; however, it was found difficult to reproduce a given initial condition with precision (except the one corresponding to full power).

The most germane features of Fig. 1 are the delayed onset of the sudden decrease in inversion (visible only at reduced initial values of inversion), the flattening out at saturation, the enormous difference in time scale between inverted and "normal" decays, and the blunting of the fast decay when the initial degree of inversion is reduced. No particular attention was devoted to the long-time behavior, except to insure that it was, indeed, much longer than the decay before saturation and had a rate consistent with results obtained elsewhere.[2]

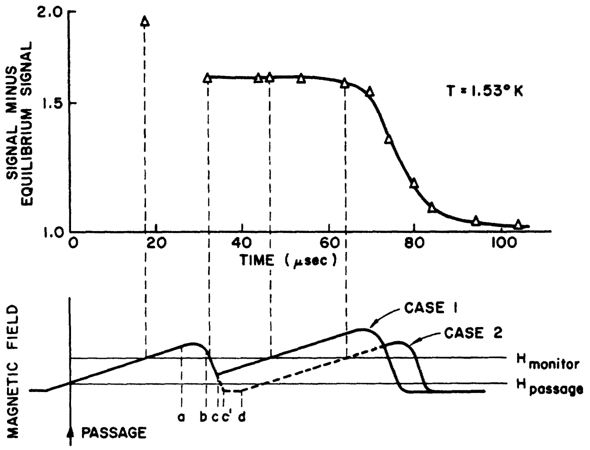

FIG. 2. Normalized plot of inverted portion of recovery curve, using field sweep illustrated. Fast passage is at $t = 0$.

Some measurements were made on the same sample after thinning it to 1.2 mm. At about the same degree of initial inversion, the logarithmic slope during the avalanche was always less (compare curves *b* and *d* of Fig. 1).

A remarkable change occurred when the timing sequence was altered. If the field was swept in the manner of Fig. 2, the avalanche seemed to occur in two distinct steps, the second being consistently delayed by about 70 microseconds. Although it is the end product of a complicated sequence of field variations, the curve can be fully explained by one assumption: that an avalanche is inhibited if the Zeeman frequency of the resonance line is not allowed to dwell at the frequency of its emitted phonons.[6] Thus for case (1), the avalanche starts at (*a*) but is quenched at (*b*). The dwell time at (*c*) is too short, and the remaining inversion too small, to allow a significant reduction of the signal by another avalanche. However, if the second sweep is delayed, as in case (2), enough to allow a dwell time (*c*′-*d*), another avalanche does start and is not quenched until (*d*).

One can evidently manipulate the Zeeman field in order to inhibit an avalanche; that is, one can prevent the generated phonons from coming to speaking terms[1] with the paramagnetic transition. This fact may have interesting practical implications.

Finally, we mention some peculiarities in line shape observed during the avalanche. Just after fast passage, the (inverted) line shape was ordinary; however, during the avalanche the center of the line reached saturation ahead of the wings; that is, a hole appeared in the inverted line. The wings then caught up and the shape appeared undistorted between saturation and thermal equilibrium. Linewidth, incidentally, was roughly 2-3 gauss, about right for inhomogeneous broadening.[7] This behavior is reminiscent of the onset of oscillations in a two-level maser[8]; indeed, the mechanism we propose is quite analogous, with resonant phonons playing the role of electromagnetic radiation.[9]

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