SPATIAL EFFECTS IN THE THEORY OF THE PHONON AVALANCHE*

R. I. Joseph, David H. K. Liu, and Peter E. Wagner
Department of Electrical Engineering, The Johns Hopkins University, Baltimore, Maryland 21218
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A set of coupled, nonlinear integro-differential equations is proposed to describe the space-time variation of the spin and phonon excitations in a paramagnetic crystal under conditions appropriate to a phonon avalanche. An effective phonon lifetime is derived and found to exceed the simple time-of-flight lifetime by orders of magnitude. For the case of an infinite slab, the theory is shown to be capable of remedying the inability of earlier models to describe the time evolution of the average magnetization.

The phonon avalanche in paramagnetic crystals is well documented experimentally,[1,2] but the existing theoretical model is too crude to provide more than semiquantitative agreement with experimental data. One of the major deficiencies in this theory is its failure to allow for a spatial variation in the magnetization and the phonon excitation. This omission is especially serious in light of the fact that direct measurements of the phonon excitation as a function of position now appear to be feasible by the technique of Brillouin scattering; and measurements of the position dependence of the magnetization, by the use of Faraday rotation, are also possible (at least in principle).

In the present Letter we propose a theory which accounts explicitly for a spatial variation in magnetization and phonon density. Calculations of the spin populations and phonon density as functions of position and time are made for a simple geometry. In the case of the magnetization, it is shown that the present theory is capable of providing a greatly improved fit to experimental data.

The theory is based on a pair of equations which are closely related to Holstein's[3] theory of the imprisonment of resonance radiation in gases. The first of these is the rate equation

$$\partial[n_+(\vec{r},t)-n_-(\vec{r},t)]/\partial t = -2A\{[p(\vec{r},t)+1]n_+(\vec{r},t)-p(\vec{r},t)n_-(\vec{r},t)\}, \tag{1}$$

where n_+ and n_- are the population densities of the paramagnetic levels, p is the excitation number of the phonons which interact with the spins, A is the spontaneous-emission rate for the resonant spin-phonon interaction, $\vec{r}$ is position, and t time. The second equation is the conservation law

$$\partial p(\vec{r},t)/\partial t = -\tfrac{1}{2}B\partial[n_+(\vec{r},t)-n_-(\vec{r},t)]/\partial t$$
$$-\int d\vec{r}'\, G(\vec{r}',\vec{r})\{\partial p(\vec{r}',t')/\partial t'\}_{t'=t-|\vec{r}-\vec{r}'|/v} - [p(\vec{r},t)-p_0]T_p^{-1}(\vec{r}). \tag{2}$$

In the first term on the right, B^{-1} is the number of lattice states per unit volume in interaction with the ions. In the second term the integration is over the volume of the sample and expresses the fact that phonons originating anywhere within the sample can interact with spins in a volume element $d\vec{r}$ at $\vec{r}$. The quantity $G(\vec{r}',\vec{r})d\vec{r}$ is the probability that a phonon originating at $\vec{r}'$ survives over the path $\vec{r}-\vec{r}'$ without any interaction and then, within $d\vec{r}$, interacts with a spin. The acoustical waves are assigned a single, isotropic velocity v. We assume G to be isotropic; so

$$G(\vec{r}',\vec{r})d\vec{r}' = G(\rho)\rho^2 d\rho d\Omega = -(1/4\pi)[\partial T(\rho)/\partial\rho]d\rho d\Omega, \tag{3}$$

where $\rho = |\vec{r}-\vec{r}'|$, $d\Omega$ is an element of solid angle measured about $\rho=0$, and $T(\rho)$ is the probability that a phonon traverses a distance ρ, in any direction, without incident, and is given by

$$T(\rho) = \int d\nu\, P(\nu)\exp[-\rho k(\nu)], \tag{4}$$

where $P(\nu)$ is the normalized emission spectrum, and $k(\nu)$ is the sum of the coefficients for absorption and stimulated emission via the spin-phonon interaction. Both coefficients are assumed to have the same spectrum; hence $k(\nu)$ is proportional to the total density of spins.

The environment external to the crystal is not included in the volume integral; so we introduce the last term in Eq. (2) to express the net loss of phonons which escape from $d\vec{r}$ and do not interact at all until they reach the surface of the sample. There they are assumed to be destroyed in such a way as to maintain the excitation number at its thermal value p_0. The phonon lifetime $T_p(\vec{r})$ can be written

$$T_p^{-1}(\vec{r}) = \int \frac{d\Omega_s}{4\pi} \frac{v}{\rho_s}\left[1-4\pi\int_{\rho=0}^{\rho_s} d\rho\, G(\rho)\right] = \frac{v}{4\pi}\int \frac{d\Omega_s}{\rho_s} T(\rho_s), \tag{5}$$

where $\rho_s = |\vec{r}-\vec{r}_s|$, $\vec{r}_s$ lies on the surface, and $d\Omega_s$ is subtended by a surface element centered at $\vec{r}_s$.

The initial conditions appropriate to experiment are $p(\vec{r},0)=p_0$ and $n_+(\vec{r},0)-n_-(\vec{r},0)=u_i(n_{-0}-n_{+0}) = u_i N(2p_0+1)^{-1}$, where N is the spin density, u_i the initial degree of inversion, and the subscript zero denotes thermal equilibrium.

We now apply the theory to an infinite slab of thickness L. The distance measured from the center in a direction normal to the walls is z. For definiteness, we take the spectrum to be Gaussian and homogeneous, that is, $k(\nu)\propto\rho(\nu)$ and

$$P(\nu) = (\pi^{1/2}\Delta\nu)^{-1}\exp[-(\nu-\nu_0)^2/(\Delta\nu)^2], \tag{6}$$

where ν_0 is the Larmor frequency. Then

$$T(\rho) = 2\pi^{-1/2}\int_0^\infty du\, \exp\{-u^2-k(0)\rho\exp(-u^2)\} \tag{7}$$

with $k(0) = NAv^2/8\pi^{3/2}\nu_0^2\Delta\nu$. Typically, $k(0)\sim 50\ \mathrm{cm}^{-1}$.

In one dimension, Eqs. (1) and (2) become

$$\partial u(z,t)/\partial t = -T_{1D}^{-1}[1+u(z,t)+u(z,t)y(z,t)], \tag{8}$$

and

$$\frac{\partial y(z,t)}{\partial t} = -S_0\frac{\partial u(z,t)}{\partial t} - \int_{-L/2}^{L/2} dz'\, H(|z-z'|)\left\{\frac{\partial y(z',t')}{\partial t'}\right\}_{t'=t-|z-z'|/v} - y(z,t)T_p^{-1}(z), \tag{9}$$

where $u=(n_+-n_-)/(n_{-0}-n_{+0})$, $y=(p-p_0)/(p_0+\frac{1}{2})$, $T_{1D}^{-1}=A(2p_0+1)$, $S_0=B(n_{-0}-n_{+0})/(2p_0+1)$, and

$$H(\xi) = \frac{k(0)}{\pi^{1/2}}\int_0^\infty du\, e^{-2u^2}\int_1^\infty \frac{dw}{w}\exp[-k(0)\xi e^{-u^2}w]. \tag{10}$$

The phonon lifetime becomes

$$v^{-1}T_p^{-1}(z) = C(\tfrac{1}{2}L+z)(\tfrac{1}{2}L+z)^{-1} + C(\tfrac{1}{2}L-z)(\tfrac{1}{2}L-z)^{-1}, \tag{11}$$

where

$$C(\xi) = \pi^{-\frac{1}{2}}\int_0^\infty du\, e^{-u^2}\int_1^\infty \frac{dw}{w^2}\exp[-k(0)\xi e^{-u^2}w]. \tag{12}$$

There is no straightforward way to solve Eqs. (8) and (9). It can, however, be shown that $H(|z-z'|)$ has a logarithmic singularity at $z=z'$ and dies off rapidly with the argument when $k(0)L$ is large. Thus we have approximated the integral of Eq. (9) by $F(z)\partial y(z,t)/\partial t$, where

$$F(z) = \int_{-L/2}^{L/2} dz'\, H(|z-z'|) = 1-[C(\tfrac{1}{2}L+z)+C(\tfrac{1}{2}L-z)]. \tag{13}$$

This step simplifies the problem enormously because it decouples the equations for different z. Equation (9) can then be written

$$\partial y(z,t)/\partial t = -S(z)\partial u(z,t)/\partial t - y(z,t)[T_p^{\mathrm{eff}}(z)]^{-1} \tag{14}$$

with $[S(z)/S_0]^{-1} = T_p^{\mathrm{eff}}(z)/T_p(z) = 1+F(z)$.

We have numerically evaluated the functions $S(z)$ and $T_p^{\mathrm{eff}}(z)$ and in Fig. 1 we plot them for several choices of $k(0)L$. The reduction of S below S_0 is modest; it occurs because spins in a given dz are allowed to interact with phonons not only within dz, but outside this region as well. The spatial varia-

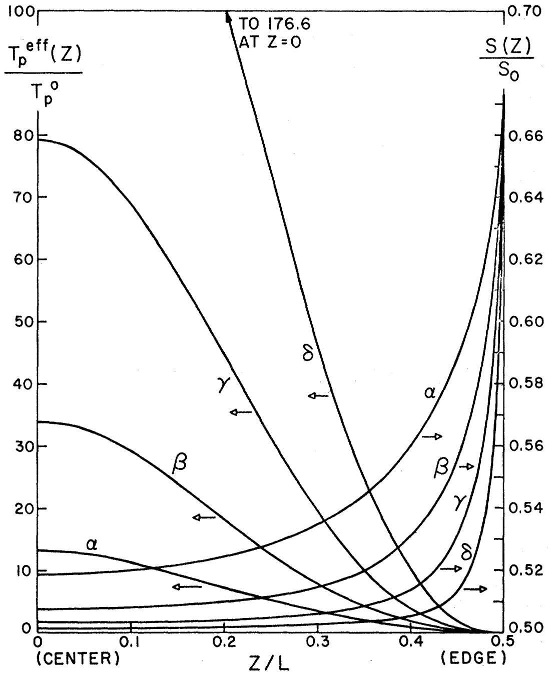

FIG. 1. Variation with position z/L of coefficients $S(z)/S_0$ (right-hand scale) and $T_p^{\rm eff}(z)/T_p^0$ ($T_p^0 = L/v$) (left-hand scale) appearing in Eq. (14) for various choices of $k(0)L$: α, 6.25; β, 12.5; γ, 25; δ, 50. Note that $S(z) = S(-z)$ and $T_p^{\rm eff}(z) = T_p^{\rm eff}(-z)$.

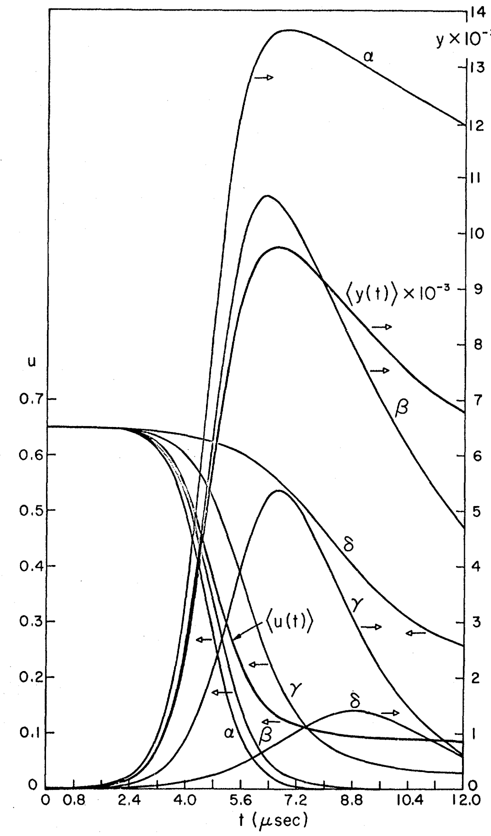

FIG. 2. Reduced magnetization $u = (n_+ - n_-)/(n_{-0} - n_{+0})$ (left-hand scale) and reduced phonon excitation $y = (p - p_0)/(p_0 + \frac{1}{2})$ (right-hand scale) as functions of time for various positions in sample z/L: α, 0.0; β, 0.32; γ, 0.4; δ, 0.432. Also shown are spatial averages $\langle u(t)\rangle$ $[= (\frac{1}{2}L)^{-1}\int_0^{L/2} dz\, u(z,t)]$ and $\langle y(t)\rangle$. Figure corresponds to $S_0 = 4.5 \times 10^4$, $T_p^0 = 1\ \mu$sec, $k(0)L = 12.5$, and $T_{1D} = 10$ msec. Note that $u(z,t) = u(-z,t)$ and $y(z,t) = y(-z,t)$.

tion of $T_p^{\rm eff}$, on the other hand, is enormous. Its enhancement near the center over the simple time-of-flight lifetime L/v (by a factor of ~100) shows dramatically how few phonons in this region are able to escape without interacting with ions. Preliminary experimental results in our laboratory on dilute $Ce_2Mg_3(NO_3)_{12} \cdot 24H_2O$ are consistent with a phonon lifetime of this order.

Equations (8) and (14) can be solved for u and y only by numerical methods. In Fig. 2 we show a representative calculation of the time dependence at several different depths in the slab. The spatial averages $\langle u(t)\rangle$ and $\langle y(t)\rangle$ are also shown. The curves of Fig. 2 give quantitative expression to the intuitive idea that the avalanche ought to be more severe in the center of the slab, simply because of the enhancement of $T_p^{\rm eff}$ by trapping. This conclusion differs considerably from earlier interpretations.[1,4]

The only quantity for which reliable data presently exist[1] is $\langle u(t)\rangle$. In previous attempts[1] at fitting experimental decay curves, we always found that the decay at early times was too abrupt to be compatible with the rather long observed decay tails. Some improvement could be obtained by the application of a large, arbitrary shift of the theoretical curves to earlier times; however, the discrepancy was never very satisfactorily resolved in this way. It is clear from Fig. 2, and on physical grounds as well, that these difficulties are remedied by the present model. The decay at early times is dominated by spins far from the surface where $T_p^{\rm eff}$ is large and the avalanche is fast. After these spins have decayed to saturation, the remaining inversion is contributed by spins nearer the surface, where $T_p^{\rm eff}$ is small and the avalanche is therefore relatively slow. The present theory can also explain the detailed size dependence of the experimental results.

At present we are attempting a quantitative fit to existing experimental data. Part of the reason for presenting the theory before this timeconsuming analysis has been completed is to stimulate experimental work on the measurement of $p(\vec{r}, t)$ by Brillouin scattering. These experiments are extremely difficult, and, hopefully, it will be encouraging to have available a tractable theory for analysis of the experimental results.

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MAGNETOREFLECTION DICHROISM OF THE LOW-ENERGY EXCITON OF RbI

R. K. Ahrenkiel
Research Laboratories, Eastman Kodak Company, Rochester, New York 14650

and

K. J. Teegarden
University of Rochester, Rochester, New York 14624
(Received 22 November 1968)

The differential magnetoreflectance of the 5.5-eV exciton in RbI is observed for right- and left-hand circularly polarized light. An analysis of the Zeeman effect results in an effective g factor of about 0.012. Explanations are offered for such an anomalously small g factor.

Recent work has utilized the technique of circular dichroism or Faraday rotation to probe the Zeeman effect in broad absorption bands arising from color centers in solids.[1,2] In the case of exciton transitions, the large absorption coefficient necessitates the use of thin films in absorption measurements. We have developed a method for studying the Zeeman effect in insulators with broad exciton lines by observing changes in reflectivity induced by a magnetic field. A similar technique has been used to study plasma effects in metals.[3]

In this method the difference in specular reflection for right- and left-hand circularly polarized light was detected by a photomultiplier and fed to a lock-in amplifier. The polarization sense of the incident light was driven from plus to minus (right to left) at 400 Hz by a Baird-Atomic Pockels cell.[4] The magnetic field of a 50-kG superconducting solenoid was parallel to the k vector of the incident light.

The differential reflectance data obtained in this way for RbI are shown in Fig. 1. The quantity measured was $\Delta R/R$, where $\Delta R = (R_+ - R_-)$ is the difference between the reflectivity for right- and left-hand circularly polarized light. One would like to relate ΔR to the differential absorption or circular dichroism of the exciton line $\Delta\alpha = (\alpha_+ - \alpha_-)$.

An attempt was made to calculate the change in the reflective phase angle $\Delta\theta$ by means of a Kramers-Kronig-type integration over the differential reflectivity spectrum.

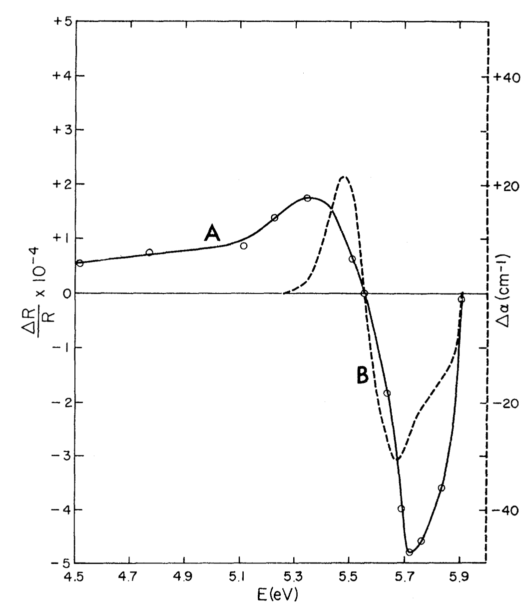

FIG. 1. Curve A is a smoothed plot of the $\Delta R/R$ data for RbI. The data are given by the open circles. Curve B is the deduced $\Delta\alpha$ using the scheme outlined in the text.