[13]M. D. Sturge (unpublished).

[14]The Zeeman effect is quite insensitive to the inclusion of second-order terms, or to reasonable variations in ζ and k.

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[16]We could reduce the size of the matrix by a factor of 3 by choosing linear combinations of vibronic states which transform as Γ_6, Γ_7, and Γ_8. However, the probability of error in this process is not negligible, and in a real basis the degeneracies of the computed energy levels provide a valuable check on the accuracy of the calculation.

[17]Our calculation differs from that of Ham and Schwarz and O'Brien (Ref. 18) in that our basis states are those of the distorted rather than the undistorted complex.

[18]F. S. Ham, W. M. Schwarz, and M. C. M. O'Brien, Phys. Rev. 185, 548 (1969); F. S. Ham and W. M. Schwarz (unpublished).

[19]J. S. Griffith, *Theory of Transition Metal Ions* (Cambridge U. P., Cambridge, England, 1961), Table A20.

[20]In I we obtained $x=2.6$ for the $^4T_2$ term of V^{2+} in $KMgF_3$. If V_τ is the same as for Co^{2+}, we find $y=0.1$ for $\hbar\omega_\tau=100$ cm^{-1}. The shifts (relative to the Γ_7 ground state) are -8 cm^{-1} for Γ_6, -5 cm^{-1} for Γ_8^a, and -2 cm^{-1} for Γ_8^b. These are quite appreciable relative to the spin-orbit splittings (39, 28, and 15 cm^{-1}, respectively). However, they are maximum shifts, and $\hbar\omega_\tau$ would not need to be much larger for them to be negligible.

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Spin and Phonon Relaxation of V^{4+} in TiO_2 †

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The spin-lattice relaxation of V^{4+} in TiO_2 is measured at liquid-helium temperatures using electron-spin-echo techniques. The T_1 relaxation decay function is represented by an expression which is the sum of two exponential functions. The decay constants for the two exponentials are 0.11 and 0.70 sec and they do not depend on temperature or sample size. The sample is annealed in air and the relaxation decay constants become 0.16 and 1.10 sec. This is interpreted as one constant being related to the equilibration of a "hot" spin system and a set of resonant phonon modes, and the other decay constant being related to the relaxation of the phonon mode by defect scattering.

I. INTRODUCTION

Spin-lattice relaxation[1] is of continuing interest because the process of energy flow from defect ion sites to the modes of a crystal lattice has important consequences on laser efficiency in solid-state laser crystals. The pulse techniques used for many years in nuclear-spin-relaxation experiments[2] are finding increased application in electron-spin-relaxation studies. When the coupling between the spin system and the phonon system is strong enough and the phonon relaxation slow enough, the "phonon bottleneck" can be observed.[3] In this paper we present the results of a spin-lattice-relaxation study of V^{4+} paramagnetic impurities in TiO_2 (rutile) using an electron-spin-echo[4] pulse-sequence excitation of the spin system and the subsequent relaxation of the spin and phonon systems back to thermal equilibrium.

In Sec. II of this paper, we present a standard spin-echo vector-model discussion of the spin excitation and relaxation. We then discuss the results we would expect from this simple theory. In Sec. III, we present our experimental procedure and results and show how our results differ from the simple vector model. In Sec IV, we discuss a more general theory of spin-lattice relaxation to take into account such processes as the stimulated emission of phonons and nonequilibrium phonon distributions which arise from the strong spin-phonon coupling. We use this more general picture to explain our experimental results. In Sec. V, we point out some conclusions and speculations that come out of the more general picture.

II. SIMPLE VECTOR MODEL OF RELAXATION

The dynamics of a spin system can be represented by a magnetization vector whose motion is similar to the motion of a classical gyroscope. The dynamics of our four-pulse–spin-echo–relaxation experiment can be described by this vector model. In our relaxation measurements, we apply a pulse pair followed by another pulse pair. The separation between the pulse pairs is much longer than the transverse relaxation time T_2. The two pulse pairs are identical and the separation between the individual pulses of a pair is shorter than T_2 and will produce an echo.

Initially, the magnetization vector is aligned along the positive z axis. An intense microwave pulse is applied to the spin system and, in a frame of ref-

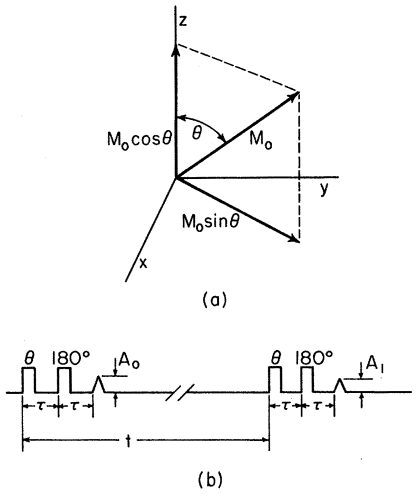

FIG. 1. (a) Spin-system magnetization after a θ pulse where the magnetization vector M_0 has a z component of $M_0\cos\theta$ and a transverse component in the x-y plane of $M_0\sin\theta$. (b) Pulse sequence used to measure the T_1 relaxation time. θ and 180° are the excitation angles of the two pulses of the echo sequence which are separated by the time τ. The second pulse pair is applied at the time t. (The pulse lengths are small compared to τ and t.)

erence rotating about the z axis with a frequency equal to the applied microwave field frequency, will rotate or excite the magnetization vector $\vec{M}$ to an angle θ from the z axis, as shown in Fig. 1. After the first pulse, the magnetization component in the x-y plane decays or dephases with a characteristic time constant $1/\Delta\omega$, where $\Delta\omega$ is the inhomogeneous linewidth. A second pulse is applied which rotates each individual magnetization spin vector by 180° and produces the spin echo.[4] The amplitude of the first echo, A_0, is dependent upon the relaxation time T_2 and the transverse magnetization that is excited by the first pulse. If the time between pulses τ is held fixed, we have the following relationship:

$$A_0 = kM_0\sin\theta,$$

where k is some constant which depends on T_2 and τ, M_0 is the magnetization along the z axis before the first pulse is applied, θ is the angle of excitation of the first pulse, and A_0 is the amplitude of the first echo.

After a time t, which is much longer than T_2 but comparable to T_1, we apply a second pulse pair similar to the first. The second echo amplitude A_1 is proportional to the transverse magnetization that is excited at time t by the first pulse of the second pulse pair and the amount of magnetization that is along the z axis at the time t. Thus we have

$$A_1 = kM_z(t)\sin\theta,$$

where k and θ are the same as for the first pulse pair and $M_z(t)$ is the magnetization along the z axis at time t.

If we assume a simple exponential relaxation of the z component of magnetization, we obtain

$$M_z(t) = M_0 + M_0(\cos\theta - 1)e^{-t/T_1},$$

where t is the time between the pulse pairs and T_1 is the longitudinal relaxation time. The ratio of A_1/A_0 is given by

$$1 - A_1/A_0 = (1-\cos\theta)e^{-t/T_1}.$$

In the liquid-helium temperature range, standard electron-paramagnetic-relaxation theory[1] gives a direct relaxation process with a rate proportional to the temperature T, a Raman-process rate proportional to T^7 or T^9, and, if the crystal-field energy Δ is smaller than the highest phonon energy, an Orbach process proportional to $e^{\Delta/kT}$. In the case of V^{4+} in TiO_2 we would expect a T_1 given by

$$1/T_1 = AT + BT^7,$$

where A and B are constants of proportionality.

If we assume that the phonon system is heated by the spin system, then the above equation must be modified to include the phonon bottleneck. In a pulse saturation experiment like that of Scott and Jeffries,[5] one would obtain a T^2 temperature dependence in place of the direct process.

In our experiments, our pulse lengths are shorter and our amplitudes are much greater than those used in pulse saturation measurements. We should not be too surprised to find different experimental results, but the nature of the difference is not clear from the simple vector model. We have to develop a more realistic model to handle all the experimental conditions that may be imposed on a spin system. Before we discuss another model, we shall present our experimental results for spin-lattice relaxation of V^{4+} in TiO_2.

III. EXPERIMENTAL PROCEDURE AND RESULTS

The pulse spectrometer is capable of producing microwave pulses at 9.3 GHz with a power of up to 800 W for pulse lengths that can be continuously varied from 20 to 100 nsec. The separation between pulses of a pulse pair can be varied from 0.1 μsec to 10 sec and the time delay between pairs can be varied from 50 msec to 5 sec. The sample was a disk of rutile with a diameter of about 1 cm and a thickness of about 2 mm, and doped with 0.05-at.% V^{4+} paramagnetic impurities. The sample was placed in a waveguide (0.900×0.100 in.) and cooled by liquid helium, whose temperature was varied from 4.2 to about 1.8 °K. After the sample came to thermal equilibrium, two pulse pairs were ap-

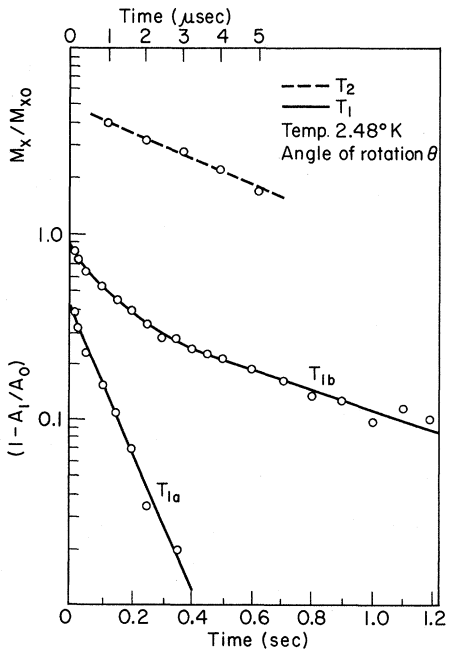

FIG. 2. Semilog plot of the ratio $1-A_1/A_0$ for one of the relaxation measurements. The straight-line T_{1a} curve is obtained from subtracting off the straight-line T_{1b} part of the nonlinear T_1 curve.

plied and the two echoes were superimposed on the same photograph of the oscilloscope trace. The ratio of the echo amplitudes A_1/A_0 was then measured in the photograph.

The T_1 relaxation of all the hyperfine lines of the vanadium spectrum behaved in the same way. A typical plot of $1-A_1/A_0$ is shown in Fig. 2, which also shows the T_2 decay curve. The excitation angle or angle of rotation of the first pulse is expressed by some factor times θ which is an unknown but high power level. The T_1 decay curve can be represented by the sum of two exponential decay curves. At very low power levels, the T_1 decay curve becomes a simple exponential. Table I gives a summary of our experimental results. Our T_1 decay can be represented by the expression

$$1-A_1/A_0=(1-\cos\theta)\,(ae^{-t/T_{1a}}+be^{-t/T_{1b}})\,,$$

where a and b are constants and T_{1a} and T_{1b} are the two relaxation decay constants.[6]

The most striking result is that the two T_1 decay constants do not depend on temperature from 1.8 to 4.2 °K. We changed the thickness of the disk by about 10% and saw no change in the decay constants. We changed the condition of the surface from a smooth surface to a rough surface and saw no change in the decay constants. We annealed the rutile sample for 2 days in air at 850 °C and observed some change in the decay constants as noted in the table. During the measurements the pulse widths were fixed at 50 nsec and the pulse separation in the pairs was fixed at 1.5 μsec. We noted also that the point where the T_1 decay curve would intercept the vertical axis at time zero is fixed once a certain power is reached. The applied power was increased by a factor of 16 above the plateau power level and the curve intercept remained fixed at about $1-A_1/A_0\cong 0.7\pm 0.1$.

In summary, our experimental results show (i) a spin-lattice-relaxation curve that can be represented by the sum of two exponentials; (ii) the relaxation time constants are *independent of temperature up to* 4.2 °K, independent of thickness and surface conditions; (iii) one of the relaxation time constants depends on annealing history; (iv) the relaxation shows some nonlinear behavior in the 0–50-msec time domain.

IV. MORE GENERAL THEORY

In most electron-paramagnetic-relaxation experiments, the paramagnetic spins are contained in a dielectric crystal as impurities and the crystal is placed in a microwave cavity. To consider the interaction between the spin system and a field, we first specify the modes of the field. The effect of the field on one individual spin is then obtained as dependent on the excitation of all the modes of the field which are usually assumed to be in thermal equilibrium.

When more than one spin is placed in the field, there then arises between the individuals an interaction or coupling that is mediated by the field. One then finds that processes can occur where the field energy can be exchanged between the individual spins. These processes modify the interaction between the spin system and the field.

Dicke[7] and others[8] have discussed this problem in the form of N two-level systems coupled to a field system. In these discussions, the first step in the analysis is to define states for the whole spin system. These states take into consideration the degeneracy that is inherent in a system with many identical energy levels. The states thus defined may have phase factors between them. These phase factors arise from the field in which the spin is imbedded.

The electron-paramagnetic-spin system can couple to two fields, the electromagnetic (EM) field of the microwave cavity which usually excites the spins and the phonon field of the crystal which usually relaxes the spins. Since the velocity of light is so much greater than the velocity of sound in a crystal, the wavelength of the EM field is much longer than the wavelength of the phonon field at the same frequency. Thus if the spin states are defined with phase factors for the EM field, then the

TABLE I. Summary of experimental results.

Angle of rotation[a]	Temp (°K)	a	T_{1a} (sec)	b	T_{1b} (sec)	T_2 (μsec)
1.0θ	1.30	0.23	0.10	0.35	0.65	7.0
1.4θ	1.30	0.28	0.10	0.42	0.69	6.0
2.0θ	1.30	0.24	0.11	0.56	0.62	5.0
2.8θ	1.30	0.33	0.11	0.54	0.82	4.0
4.0θ	1.30	0.27	0.07	0.58	0.61	4.0
1.0θ	1.87	0.23	0.10	0.28	0.70	6.5
1.4θ	1.87	0.26	0.10	0.44	0.60	5.0
2.0θ	1.87	0.37	0.10	0.46	0.70	4.5
2.8θ	1.87	0.26	0.11	0.56	0.70	4.0
4.0θ	1.87	0.25	0.08	0.57	0.66	4.0
1.0θ	2.48	0.22	0.08	0.39	0.62	7.0
1.4θ	2.48	0.39	0.11	0.42	0.75	5.0
2.0θ	2.48	0.36	0.15	0.46	0.71	4.5
2.8θ	2.48	0.37	0.12	0.48	0.65	4.0
4.0θ	2.48	0.38	0.09	0.48	0.72	4.0
1.0θ	4.20	0.20	0.10	0.35	0.51	6.0
1.4θ	4.20	0.40	0.12	0.34	0.75	5.0
2.0θ	4.20	0.40	0.12	0.44	0.70	4.0
2.8θ	4.20	0.36	0.12	0.47	0.65	4.0
4.0θ	4.20	0.37	0.11	0.46	0.60	4.0
After the sample was annealed the following data were obtained:						
θ_1	4.20	0.20	0.16	0.60	1.10	5.0

[a] θ is an arbitrary excitation angle which cannot be calculated because of uncertainties in the reflection coefficient.

phase factors will not be right for the phonon field. Thus if the spin system is excited by the EM field, then the spin system will have high "cooperation," as Dicke[7] called it, for a long wavelength or a small quasi-$\vec{k}$-vector. But since the spin system cannot have high cooperation for two widely differing wavelengths, the spin system must have low cooperation for the phonon $\vec{k}$ vector. The spin system must alter the phase relations so that energy may flow to the phonon system. In general, this is accomplished by spin-spin interactions and by inhomogeneous broadening. This is noted by Buley and Cummings.[8]

Let us briefly review Dicke's presentation by assuming that the individual particles have two energy levels and thus have an effective spin of $\frac{1}{2}$. Each particle of spin has S_x^i, S_y^i, and S_z^i as observables, and S_z^i is taken as diagonal in the energy representation. For our system, the energy is the Zeeman energy $h\Omega$, where Ω is the Larmor frequency. All the spins are assumed to have the same frequency. (A static distribution of frequencies gives rise to a decay of the free induction signal and is related to the shape of the echo.) To consider the system of N spin-$\frac{1}{2}$ particles, we form the product states of the individual states. These product states are specified by a total spin S which is analogous to Dicke's "cooperation number," the energy m which is the total energy of the spin system and is the eigenvalue of the total energy operator S_z, and some quasi-$\vec{k}$-vector which specifies the phases between the individual spins. Each of the operators and eigenvalues should be subscripted with a quasi-$\vec{k}$-vector. But for our work we want to focus upon the state of affairs when the spin system has acquired the proper phases so that high cooperation occurs for a $\vec{k}$ vector of the phonon system. We assume that there are enough spins such that a large number of spins have frequencies equal to a phonon mode frequency. So, there is large degeneracy with respect to each phonon mode. The amount of degeneracy is approximately $N_{\text{tot}}/\rho\,\Delta\omega$, where N_{tot} is the total number of spins, ρ is the density of phonon modes, and $\Delta\omega$ is the static linewidth of the spin system.

Thus we assume that our S and m refer to the cooperation and energy of those spins on resonance with a phonon mode $\vec{k}$. Also defined are S_+ and S_- which are the raising and lowering operators, respectively, for the total energy of the spin system. We will not need to carry along the $\vec{k}$-vector subscript. Thus the state of the spin system is given by

$$|S, m\rangle ,$$

where $-S \leq m \leq S$ and $0 \leq S \leq \frac{1}{2}N$, and N here refers to the number of degenerate spins on resonance with the mode $\vec{k}$. The raising and lowering opera-

tors act on the state

$$S_+|S,m\rangle = [S(S+1)-m(m+1)]^{1/2}|S,m+1\rangle,$$

$$S_-|S,m\rangle = [S(S+1)-m(m-1)]^{1/2}|S,m-1\rangle.$$

It will be convenient for our discussion to change the notation slightly and define a new energy number $n=m+S$ which is a measure of the energy above the lowest energy state. In this new representation we have

$$S_+|S,n\rangle = [(2S-n)(n+1)]^{1/2}|S,n+1\rangle,$$

$$S_-|S,n\rangle = [(2S-n+1)(n)]^{1/2}|S,n-1\rangle,$$

where now $0 \le n \le 2S$ and $0 \le S \le \frac{1}{2}N$.

As mentioned by Dicke, "resonance trapping" and "superradiance" appear naturally in the formalism. The value of S is determined by the amount of phase coherence or cooperation there is between the individual spins for this particular wave vector.

The modes of the phonon field are specified by a wave vector $\vec{k}$ and each mode can be excited to some energy level. The energy levels for each mode are the energy levels of a simple harmonic oscillator. The phonon number p, which is the eigenvalue of the operator $b^\dagger b$, is a measure of the energy in the phonon mode. There is also a $b^\dagger$ and a b operator for each of the phonon modes.

We will focus our attention upon the interaction between a degenerate set of spins on resonance and with the appropriate $\vec{k}$ vector of a phonon mode. The coupling between the spin system and the phonon system will be given by a term

$$\mathcal{H}_{int} = hw(S_+ b + S_- b^\dagger),$$

where w is the interaction frequency. The constant w is a result of considering the details of how the phonon mode causes fluctuations in the crystal field which is coupled through spin-orbit coupling to the spin to cause its relaxation. We will apply standard perturbation theory to the coupled spin-phonon system. But we must keep in mind that this approach is valid after T_2, that is, after all the quantum-mechanical superposition has disappeared. Only then can we be sure that our systems can be specified by the quantum numbers n and p.

The time rate of change of the energy of the spin system is then given by

$$\frac{dn}{dt} = T_P(n \to n+1) - T_P(n \to n-1),$$

where T_P is the transition probability. We then have

$$T_P(n \to n+1) = (2\pi/h^2)|\langle n,p|\mathcal{H}_{int}|n+1,\ p-1\rangle|^2 \rho(\Omega)$$

$$= 2\pi w^2 \rho(\Omega)(2S-n)(n+1)p,$$

where $\rho(\Omega)$ is the density of phonon states at the frequency Ω. Similarly, we have

$$T_P(n \to n-1) = (2\pi/h^2)|\langle n,p|\mathcal{H}_{int}|n-1,p+1\rangle|^2 \rho(\Omega)$$

$$= 2\pi w^2 \rho(\Omega)(2S-n+1)(n)(p+1).$$

Thus we obtain for the time rate of change of the spin-system energy n

$$\frac{dn}{dt} = 2\pi w^2 \rho(\Omega)[2S(p-n) - n(2p+1-n)].$$

The above equation is nonlinear and contains stimulated-emission phenomena. When n is near to its maximum value of $2S$, which corresponds to complete population inversion, the equation is approximately

$$\left.\frac{dn}{dt}\right|_{n=2S} \cong -2\pi w^2 \rho(\Omega) 2S(p+1),$$

and hence n decays with a rate that depends on S and on the number of phonons. However, when n is near S, which corresponds to spin-system saturation, the above equation is approximately

$$\left.\frac{dn}{dt}\right|_{n=S} \cong -2\pi w^2 \rho(\Omega) S(S+1),$$

where now n decays with a rate that depends on $S(S+1)$ and not on the phonons. This is the superradiant region and corresponds to stimulated emission dominating the relaxation process.

In the region where n is near zero, which is the region of interest for this work, we have

$$\frac{dn}{dt} = 2\pi w^2 \rho(\Omega) 2S(p-n). \tag{1}$$

Also, we have for the rate of change of the phonon number p, where we assume an exponential relaxation of the phonon mode to thermal equilibrium,

$$\frac{dp}{dt} = -2\pi w^2 \rho(\Omega) 2S(p-n) + \frac{1}{\tau}(p^0 - p), \tag{2}$$

where τ is the relaxation time for the phonon mode, and p^0 is the thermal equilibrium phonon number. Equations (1) and (2) are two coupled linear equations which can be solved to obtain two time constants, as shown in the Appendix,

$$\frac{1}{T_{1a}} = W + \frac{1}{2\tau} + \left(W^2 + \frac{1}{4\tau^2}\right)^{1/2},$$

$$\frac{1}{T_{1b}} = W + \frac{1}{2\tau} - \left(W^2 + \frac{1}{4\tau^2}\right)^{1/2},$$

where $W = 4\pi w^2 S \rho(\Omega)$.

The solution for the spin number $n(t)$ is given by

$$n(t) = \tfrac{1}{2}\{n(0) - \cot\tfrac{1}{2}\alpha[p(0) - T_{1a}p^0/\tau]\}e^{-t/T_{1a}}$$

$$+\tfrac{1}{2}\{n(0)+\cot\tfrac{1}{2}\alpha[p(0)-T_{1b}p^0/\tau]\}e^{-t/T_{1b}},$$

where $n(0)$ and $p(0)$ are the spin-system excitation number and the phonon-system number, respectively, at a time equal to approximately zero, but after a suitable time for coherence effects to be gone, and $\tan\alpha = 2W\tau$.

In order to estimate the spin and phonon number at approximately zero, let us assume that the phonon-relaxation time is infinite and that the following equations for n and p apply:

$$\frac{dn}{dt} = W(2S(p-n) - n(2p+1-n)),$$

$$\frac{dp}{dt} = -W(2S(p-n) + n(2p+1-n)).$$

One solution to the above system of equations is $n(t)+p(t) = \text{const}$. So we have $n+p = n_{exc}+n^0+p^0$, where n_{exc} is the spin number that is excited by the first microwave pulse, n^0 is the thermal equilibrium number of spins, and p^0 is the thermal equilibrium number of phonons.

Also, the above equations have an equilibrium solution for when $\dot{n}$ and $\dot{p}$ are zero. If we take $n+p = c$, where c is a constant, we have

$$\bar{n} = \tfrac{1}{3}(c+2S) - \tfrac{1}{3}(c^2+4S^2)^{1/2},$$

where $\bar{n}$ is the equilibrium value of spin excitation after the spin and phonons have come to equilibrium, but before the phonon mode can relax. To make an estimate of $\bar{n}$, we assume that n^0 and p^0 are each of the order of $kT/\hbar\Omega$. The value of n_{exc} depends on the first pulse and has a maximum value of $2S$. As noted in Dicke,[7] the value of S in the high-temperature approximation is $N\hbar/4kT$, which in our case is much larger than n^0 and p^0. Therefore, we have that $\bar{n}$ is approximately $2S/3 + kT/6\hbar\Omega$.

Since $S_z = -S\cos\theta$ and $S_z = n - S$, we have that $1-\cos\theta = n/S$. Therefore the value of $1-\cos\theta$ at approximately zero is given by n/S or $\frac{2}{3}+kT/6\hbar\Omega S$, which for our experimental conditions is about 0.67. This is to be compared with our experimental results of 0.7 ± 0.1 with an uncertain temperature dependance.

To compare the theory of this part with our experimental results, we see that the theory depicts a decay curve with two exponentials, and a saturation point or plateau value for $1-\cos\theta$ at approximately zero. Any excess excitation of the spin system above $2S$ would quickly be radiated through stimulated emission of photons or phonons. If we take values of $W = 2.01\ \text{sec}^{-1}$ and $1/\tau = 6.51\ \text{sec}^{-1}$, we obtain the following values:

$$T_{1a} = 0.11 \text{ sec}, \quad T_{1b} = 0.70 \text{ sec},$$
$$a = 0.30, \qquad b = 0.40,$$

which are consistent with our experimental results before annealing the sample. After annealing, we obtain the following values:

$$T_{1a} = 0.16 \text{ sec}, \quad T_{1b} = 1.10 \text{ sec},$$
$$a = 0.20, \qquad b = 0.60,$$

which give us $W = 1.19\ \text{sec}^{-1}$ and $1/\tau = 4.77\ \text{sec}^{-1}$.

V. CONCLUSION

The more general theory that we are presenting seems to explain most of the experimental results. It is just the application of the rate equations to a system of N two-level systems coupled to a phonon system. However, care must be exercised not to apply the equations too near the time that the pulses are applied as coherent effects modify the motion of the system and hence cannot be described by rate equations. More general coherent effects lead to phenomena such as self-induced transparency.[9] The problem of coherent phonon generation via photon stimulation leads to a difficulty as seen in this more general description. The change of the cooperation number S from a value appropriate for the photons to a value appropriate for phonons has to be accomplished for the stimulated emission of phonons to occur. This is somewhat like a change in the $\vec{k}$ vector for the photon field to the $\vec{k}$ vector of the sound field at the same frequency. This then requires a T_2-type process which is fast enough not to limit the pumping of energy into the phonon system.

The value of S for a given quasi-$\vec{k}$-vector is limited by $\frac{1}{2}N$, where N is the number of spin in resonance with the phonon mode $\vec{k}$. This depends on the linewidth $\Delta\omega$ of the spin system. An estimate of the value of N per mode is $N_{tot}/\Delta\omega\rho(\omega)$, where N_{tot} is the total number of spin, $\Delta\omega$ is the linewidth, and $\rho(\omega)$ is the density of phonon state. For our experiment, this is about 10^8.

Also note that the T_2 relaxation time depends on the power or the angle of excitation. We are continuing our study of this effect.

APPENDIX

We want to solve the following system of linear differential equations:

$$\frac{dn}{dt} = W(p-n), \quad \frac{dp}{dt} = -W(p-n) + \frac{1}{\tau}(p^0-p).$$

We define the vector (n,p) such that

$$\frac{d}{dt}\begin{pmatrix} n \\ p \end{pmatrix} = \begin{pmatrix} -W & W \\ W & -W-R \end{pmatrix}\begin{pmatrix} n \\ p \end{pmatrix} + \begin{pmatrix} 0 & 0 \\ 0 & R \end{pmatrix}\begin{pmatrix} n^0 \\ p^0 \end{pmatrix},$$

where $R = 1/\tau$. The solution to the homogeneous part has eigenvalues

$$\lambda = -W - \tfrac{1}{2}R \pm (W^2 + \tfrac{1}{4}R^2)^{1/2},$$

which are two two time constants

$$1/T_{1a} = W + \tfrac{1}{2}R + (W^2 + \tfrac{1}{4}R^2)^{1/2},$$

$$1/T_{1b} = W + \tfrac{1}{2}R - (W^2 + \tfrac{1}{4}R^2)^{1/2}.$$

The eigenvalue $1/T_{1a}$ has as an eigenvector

$$\begin{pmatrix} \cos\frac{1}{2}\alpha \\ -\sin\frac{1}{2}\alpha \end{pmatrix},$$

and $1/T_{1b}$, the eigenvector

$$\begin{pmatrix} \cos\frac{1}{2}\alpha \\ \sin\frac{1}{2}\alpha \end{pmatrix},$$

where $\tan\alpha = 2W/R$.

Thus the most general solution to the homogeneous equation is written as

$$\begin{pmatrix} n(t) \\ p(t) \end{pmatrix} = A\, e^{-t/T_{1a}} \begin{pmatrix} \cos\frac{1}{2}\alpha \\ -\sin\frac{1}{2}\alpha \end{pmatrix} + B\, e^{-t/T_{1b}} \begin{pmatrix} \cos\frac{1}{2}\alpha \\ \sin\frac{1}{2}\alpha \end{pmatrix},$$

where A and B are arbitrary constants.

The solution to the inhomogeneous part is obtained in the standard way to obtain the complete solution, where A and B become

$$A = (e^{t/T_{1a}} - 1)\, T_{1a}\, p^0 R/2 \sin\tfrac{1}{2}\alpha + A_0,$$

$$B = (e^{t/T_{1b}} - 1)\, T_{1b}\, p^0 R/2 \sin\tfrac{1}{2}\alpha + B_0,$$

where A_0 and B_0 are determined by the initial conditions and given by

$$A_0 = \tfrac{1}{2}[n(0)/\cos\tfrac{1}{2}\alpha - p(0)/\sin\tfrac{1}{2}\alpha],$$

$$B_0 = \tfrac{1}{2}[n(0)/\cos\tfrac{1}{2}\alpha + p(0)/\sin\tfrac{1}{2}\alpha],$$

where $n(0)$ and $p(0)$ are the excitation number for the spin system and phonon system, respectively, at time zero.

Therefore the first component of this vector solution gives us for the time dependence of the spin-system excitation number

$$n(t) = \tfrac{1}{2}\{n(0) - \cot\tfrac{1}{2}\alpha[p(0) - p^0 T_{1a} R]\}\, e^{-t/T_{1a}} + \tfrac{1}{2}\{n(0) + \cot\tfrac{1}{2}\alpha[p(0) - p^0 T_{1b} R]\}\, e^{-t/T_{1b}}.$$

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Multiple Scattering of Heavy Ions of keV Energies Transmitted through Thin Carbon Films

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This paper describes experimental studies of multiple scattering of 13 different heavy ions with $3 \leq Z_1 \leq 30$ in the energy range of 200–1000 keV, transmitted through thin (8–25 $\mu g/cm^2$) carbon foils. The agreement between our experimental data, recent theoretical calculations, and published experimental data is found to be satisfactory, although a small systematic deviation exists for thicker foils.

I. INTRODUCTION

For several years, multiple scattering of light ions of high energies has been extensively studied.[1] Recently, there has been a growing interest in the investigation of multiple scattering in very thin foils at energies low enough for a classical theoretical treatment to be applicable.[2] This interest is motivated both experimentally and theoretically.

A knowledge of multiple-scattering angular distributions is necessary in several cases. For example, in experiments with heavy ions, multiple scattering very often appears as an undesirable process decreasing the resolution of the experi-