plane.

In order to estimate the effect of the diffraction of the primary beam on the secondary emission we assumed that the diffracted beams would contribute to the yield in the same way as the "normal" monotonic increase in yield with increasing angle of incidence predicts. The proposed theory explains qualitatively the experimentally observed features; calculations are in progress and have given satisfactory results so far.

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[3] R. W. Soshea (private communication).

[4] See G. P. Thomson and W. Cochrane, *Theory and Practice of Electron Diffraction* (MacMillan and Company, London, 1939), p. 80.

NUCLEAR RELAXATION VIA QUADRUPOLE COUPLING[*†]

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Several papers[1-4] have reported calculations of the transition probabilities for quadrupolar nuclear spin-lattice relaxation and considerable data are becoming available.[5-10] Nevertheless, three important problems concerning quadrupolar relaxation have not been solved. The first problem is to express the relaxation time, T_1, in terms of the predicted transition probabilities in order to obtain a prediction that may be compared with experiment. The second problem involves the angular dependence of T_1 as the orientation of the crystal is varied with respect to the magnetic field. Experimental T_1's are isotropic, but the theoretical transition probabilities have a strong angular dependence. The third problem concerns the temperature dependence of the quadrupolar T_1. Khutsishvili[11] has proposed a high-temperature "four phonon" process as opposed to the Raman "two phonon" process considered by Van Kranendonk.[1] This Letter is concerned with these three problems.

Pound[12] first demonstrated and discussed the importance of the nuclear quadrupole moment in determining the spin-lattice relaxation time. The first detailed theory was developed by Van Kranendonk.[1] Using the approximations that the lattice vibrations could be described by a Debye spectrum and that the lattice could be described by an array of point charges located at the lattice sites, he showed that the dominant quadrupolar relaxation mechanism was a Raman process in which one quantum of lattice vibration is absorbed, another quantum emitted, and a nucleus makes a $\Delta m = \pm 1$ or ± 2 transition. He obtained explicit expressions for the $\Delta m = 1$ and 2 transition probabilities, W_1 and W_2, for the NaCl lattice.

The spin systems we consider are those in crystals of cubic symmetry with no static quadrupole interactions. The nuclear energy levels are equally spaced and are given by the Zeeman interaction, $E(m) = -\gamma\hbar Hm$. Abragam and Proctor[13] have shown that the concept of spin temperature is valid for such systems when the interactions between nuclei are stronger than the interactions between the lattice and the nuclei (i.e., $T_2 \ll T_1$). Using the spin temperature concept, an explicit relation between relaxation times and transition probabilities has recently been given by Hebel and Slichter.[14] This relation is

$$\frac{1}{T_1} = \frac{1}{2}\,\frac{\sum_{m,\mu}(E_m - E_{m+\mu})^2 W(m, m+\mu)}{\sum_m E_m{}^2}, \qquad (1)$$

where the energy values, E_m, are chosen such that the trace of the energy matrix is zero and $W(m, m+\mu)$ is the probability of a nucleus making a transition from state m to $m+\mu$.

Starting with the quadrupole Hamiltonian of Cohen and Reif,[15] we have carried out calculations for the transition probabilities for a point-charge zincblende lattice. The predicted value of $W(m, m+\mu)$ is

$$W(m, m+\mu) = C\,|Q_{\mu m}|^2 T^{*2} E(T^*) E_\mu(\infty), \qquad (2)$$

where $C = \sqrt{3}/8\pi d^2 v^3 r^3$, r = interatomic distance, d = density, v = velocity of sound, $T^* = T/\theta$ (θ is the Debye temperature), $Q_{\mu m}$ is the matrix element for the quadrupole operator between states m and $m+\mu$, $E(T^*)$ is a numerical function given by Van Kranendonk, and the $E_\mu(\infty)$ are

$$E_{\pm 1}(\infty) = \frac{(\gamma e)^2}{r^{10}}[723.4 - 312\alpha^2],$$

$$E_{\pm 2}(\infty) = \frac{(\gamma e)^2}{r^{10}}[645.4 + 78\alpha^2], \quad (3)$$

where $\alpha^2 = \alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2$ and $\alpha_1, \alpha_2, \alpha_3$ are the direction cosines between the magnetic field and the [100], [010], [001] cyrstalline directions. The notation is the same as that used by Van Kranendonk.

When the expressions for the transition probabilities are substituted into Eq. (1), the result is

$$\frac{1}{T_1} = \frac{1}{40}\frac{(2I+3)}{I^2(2I-1)}(eQ)^2 C T^{*2} E(T^*)[E_1(\infty) + 4E_2(\infty)], \quad (4)$$

where the term $f(I) = (2I+3)/I^2(2I-1)$ contains the entire dependence of T_1 on the nuclear spin. This spin function is generally valid for the NaCl, CsCl, and ZnS lattices because the lattice details are contained in the $E_\mu(\infty)$ terms. While it has been recognized that T_1 would depend on the nuclear spin,[5] the explicit dependence has not previously been derived.

When the values of $E_\mu(\infty)$ are inserted in Eq. (4), it becomes

$$\frac{1}{T_1} = \frac{1}{40}\frac{(2I+3)}{I^2(2I-1)}\frac{(e^2\gamma Q)^2}{r^{10}} C T^{*2} E(T^*)(3305). \quad (5)$$

T_1 is isotropic because the angular dependences of the transition probabilities exactly cancel. This isotropy holds also if one uses Van Kranendonk's[1] transition probabilities for the NaCl lattice or Yosida and Moriya's[3] transition probabilities for the NaCl and CsCl lattices. [Because of a different choice of quadrupole Hamiltonian their coefficients of α^2 in $E_2(\infty)$ are 1/16 those of α^2 in $E_1(\infty)$. This is balanced by a factor of two difference in the $Q_{\mu m}$'s so that the predicted relaxation is still isotropic.] Our T_1 measurements on In^{115} in a single crystal of InSb at 77°K give an isotropic T_1 of 0.80 ± 0.05 second. Other investigations[5,9] have reported isotropic T_1's in the alkali halides.

The spin function, $f(I)$, has been experimentally verified with measurements on AlSb and RbCl. These results are

$$\left[\frac{Q(\mathrm{Sb}^{123})}{Q(\mathrm{Sb}^{121})}\right]^2 = 1.63, \quad \frac{f(7/2)}{f(5/2)} = 0.425,$$

$$\frac{T_1(\mathrm{Sb}^{121})}{T_1(\mathrm{Sb}^{123})} = \left(\frac{Q}{Q}\right)^2 \frac{f}{f} = 0.69 \text{ (theory)},$$

$$\frac{T_1(\mathrm{AlSb}^{121})}{T_1(\mathrm{AlSb}^{123})} = 0.75 \pm 0.10 \text{ (exp.)};$$

$$\left[\frac{Q(\mathrm{Rb}^{85})}{Q(\mathrm{Rb}^{87})}\right]^2 = 4.28, \quad \frac{f(5/2)}{f(3/2)} = 0.24,$$

$$\frac{T_1(\mathrm{Rb}^{87})}{T_1(\mathrm{Rb}^{85})} = \left(\frac{Q}{Q}\right)^2 \frac{f}{f} = 1.027 \text{ (theory)},$$

$$\frac{T_1(\mathrm{Rb}^{87}\mathrm{Cl})}{T_1(\mathrm{Rb}^{85}\mathrm{Cl})} = 1.23 \pm 0.40 \text{ (exp.)}.$$

Figure 1 shows the results of temperature dependence measurements of I^{127} in KI between 77°K and 800°K. The data lie close to the $T_1 \propto T^{-2}$ line, the predicted high-temperature dependence for the Raman "two phonon" process. The $T_1 \propto T^{-4}$ line is arbitrarily drawn through

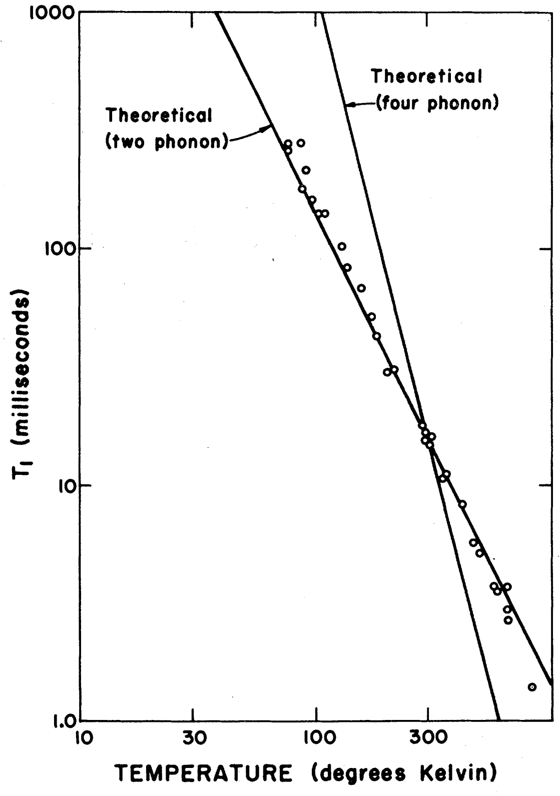

FIG. 1. Relaxation times of I^{127} in'KI. The "two phonon" process predicts $T_1 \propto T^{-2}$ and the "four phonon" process predicts $T_1 \propto T^{-4}$.

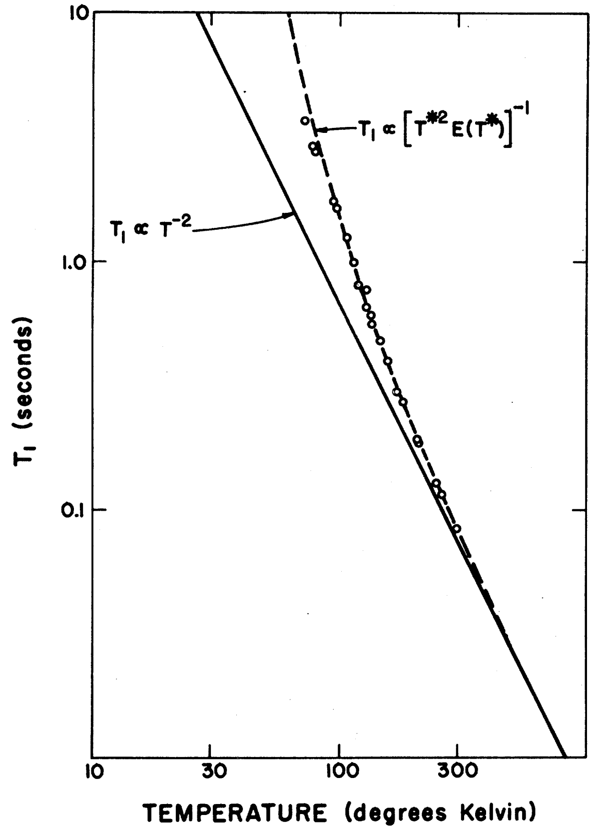

FIG. 2. Relaxation times of In^{115} in InP. Because $T < \theta$, the data deviate from the straight line, $T_1 \propto T^{-2}$; the dashed line is calculated using Van Kranendonk's function, $E(T^*)$.

300°K. It is apparent that the T_1 dependence of the "four phonon" process proposed by Khutsishvili[11] does not fit the data.

The temperature dependence for the Raman process deviates from T^{-2} for $T < \theta$ and is given by Van Kranendonk's function $E(T^*)$. Figure 2 shows the results of T_1 measurements on In^{115} in InP. The straight line is $T_1 \propto T^{-2}$ and the dashed line is the predicted temperature dependence using $E(T^*)$. The line is calculated using the room temperature T_1 and a Debye temperature of 400°K which gives the best fit to the data. The scatter in the data gives a ±25°K uncertainty for the Debye temperature (which is not available from other sources).

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