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# Quadrupolar Nuclear Spin-Lattice Relaxation in Crystals with Body-Centered Cubic Lattice Structure

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In a recent paper, Van Kranendonk has estimated the effect of lattice vibrations in contributing to the spin-lattice relaxation time due to quadrupole interaction in crystals with a simple cubic structure. In the present paper, his method is extended to the case of crystals with a body-centered cubic structure.

## INTRODUCTION

Pound[1] first showed that in crystals, the quadrupole interaction of nuclei with surrounding charges contributes effectively to the spin-lattice relaxation time in nuclear magnetic resonance experiments. Van Kranendonk[2] has developed a method for calculating the relaxation time due to the perturbation of this quadrupole interaction by the lattice vibrations. The idea is essentially similar to Waller's[3] mechanism for paramagnetic relaxation due to lattice vibrations, but we have now to deal with electric quadrupole interaction and not magnetic dipole interaction as discussed by Waller and later adapted to the nuclear case of Bloembergen *et al.*[4] Besides, Van Kranendonk uses a system of quantized oscillators to represent the lattice vibrations instead of the classical Fourier expansion employed by Waller. Van Kranendonk mentions the effect of polarization of the atomic orbitals by the nuclear quadrupole moment, discussed in detail by Sternheimer[5] and his collaborators, and also the effect of the lattice vibrations in distorting the little covalent binding that may exist between neighboring atoms. This too would lead to incipient departures of the electron distribution round the nucleus from spherical symmetry.† Both these effects lead to an effective departure of the $eqQ$ at the position of the nucleus from that expected by a simple interaction between the quadrupole moment of the nucleus and the field-gradient due to surrounding point charges in the ionic model. Van Kranendonk takes this into account by his simple "one-parameter" model, introducing a parameter $\gamma$ so that each neighboring charge is taken as $\gamma e$ instead of $e$, where $\gamma$ is to be obtained from the measured relaxation times. It was expected that for ions like $\mathrm{Cs}^+$, where the Sternheimer effect leads to a total induced moment of $\gamma_\infty Q$,[5] with $\gamma_\infty$ calculated to be $-143.5$, this effect could be much more substantial in contributing to $\gamma$ than the distortion in the covalency. As a matter of fact, we expect $\gamma^2$ to be approximately equal to $(1+|\gamma_\infty|)^2$ in this case. We therefore felt it necessary to extend Van Kranendonk's ionic model calculations to the CsCl lattice which has a body-centered cubic structure. We have presented the results of this calculation in the next section.

We have closely followed the notations in Van Kranendonk's paper (which we shall refer to henceforth as I) as far as the general expression for the transition probability is concerned, but of course we need a larger number of $L$ and $N$ functions because of the larger number of nearest neighbors, *viz.*, 8 in a body-centered lattice as compared to 6 in a simple cubic lattice. As our calculations are based on Van Kranendonk's paper, they naturally involve the same assumptions, *viz*:

(a) The nuclei whose resonance is under study are situated at equivalent lattice sites and are therefore subject to similar quadrupolar interaction with the surroundings.

(b) The nuclear magnetic dipolar interactions between the species are assumed to have negligible effect on the quadrupolar relaxation.

(c) The crystal is ionic, so that the charges may be regarded as roughly concentrated around the respective lattice sites.

(d) The frequency spectrum of the lattice vibrations is approximated by an isotropic Debye expression.

## RESULTS

Van Kranendonk's expression for the probability of a transition between states characterized by $m$ and $(m+\mu)$ of the central nuclear spin, ($m\hbar$ and $(m+\mu)\hbar$ representing eigenvalues of $I_z$ the $Z$ direction being that of the external magnetic field), due to the quadrupole interaction, is

$$P(m,m+\mu)=\frac{|Q_{\mu m}|^2}{2\pi^3 d^2 v^2}\int_0^{\omega_m}\frac{k^4|f(\omega)|^2}{\omega^2}\frac{e^x}{(e^x-1)^2}M_\mu(ka)d\omega, \quad (1)$$

where $Q_{\mu m}=\langle m+\mu|Q_\mu|m\rangle$, the quadrupole moment tensor of the nucleus $Q$ being defined as in Eq. (8) of I in the conventional manner after Casimir.[6] Also,

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[1] R. V. Pound, Phys. Rev. **79**, 685 (1950).

[2] J. Van Kranendonk, Physica **20**, 781 (1954).

[3] I. Waller, Phys. **79**, 370 (1932).

[4] Bloembergen, Purcell, and Pound, Phys. Rev. **73**, 679 (1948).

[5] Foley, Sternheimer, and Tycko, Phys. Rev. **93**, 734 (1954); R. Sternheimer and H. M. Foley, Phys. Rev. **102**, 731 (1956).

† Refer to recent paper by K. Yoshida and T. Moriya, J. Phys. Soc. (Japan) **11**, 33 (1956) and the note at the end of the present paper.

[6] H. B. G. Casimir, *Interaction between Atomic Nuclei and Electrons* (Teylers Tweede Genootschap, Haarlem, 1936).

$d$ = mass density of the crystal; $v$ = velocity of propagation of long-wavelength sound waves independent of direction of polarization; and $k=1/\omega$, the wave number of the lattice waves. $\rho(\omega)$ is defined so that the number of lattice oscillators with frequency between $\omega$ and $\omega+d\omega$ is given by

$$\rho(\omega)d\omega=\frac{V}{2\pi^2 v}f(\omega)d\omega, \tag{2}$$

$V$ representing the volume of the crystal.

$$M_\mu(ka)=\sum_{s,s'}\sum_{ijlm}N_{\mu ijlm}L_{sil;s'jm}(ka), \tag{3}$$

with

$$L_{sil;s'jm}(ka)=\{B_i(\mathbf{k},s)B_l(\mathbf{k},s)\}\{B_j(\mathbf{k}',s')B_m(k',s')\},\quad N_{\mu ijlm}=A_{\mu ij}:A_{\mu lm}, \tag{4}$$

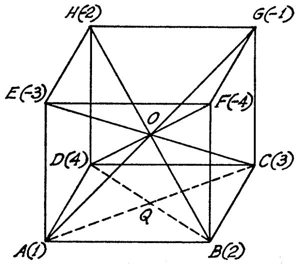

FIG. 1. Picture of a b.c.c. lattice.

the colon referring to the tensor scalar product. Here the $B_i(\mathbf{k},s)$ are defined by:

$$B_i(\mathbf{k},s)=\delta(s,1)(\cos\mathbf{k}\cdot\mathbf{a}_i-1)+\delta(s,2)\sin(\mathbf{k}\cdot\mathbf{a}_i), \tag{5}$$

where $s$ is a number which is equal to 1 or 2, and

$$\mathbf{a}_i=\mathbf{R}_i-\mathbf{R}_0, \tag{6}$$

$\mathbf{R}_i$ and $\mathbf{R}_0$ representing the equilibrium positions of the central and $i$th nuclei, respectively. $\mathbf{k}$ and $\mathbf{k}'$ represent the directions of wave propagation and the curly brackets in (4) indicate averaging over all directions of $\mathbf{k}$. The averaging occurs in expanding the effective potential $V(\mathbf{r})$ at the position of the $i$th nucleus in terms of increasing order in the relative displacements $\mathbf{r}_i$ of the surrounding ions, with respect to the displacement of the central nucleus; i.e.,

$$V(\mathbf{r})=\sum_{\mu=-2}^{+2}W_\mu Y_\mu(\mathbf{r}), \tag{7}$$

with

$$W_\mu=A_\mu+\sum_i \mathbf{A}_{\mu i}\cdot\mathbf{r}_i+\sum_{ij}A_{\mu ij}:\mathbf{r}_i\mathbf{r}_j+\cdots. \tag{8}$$

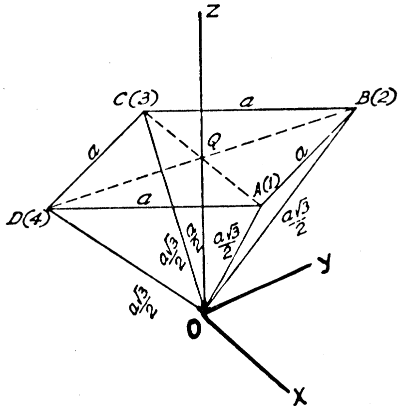

FIG. 2. Portion of a b.c.c. lattice indicating significance of $\{S_pS_q\}$ terms; $p, q=1, 2, 3$.

We shall neglect first-order vibrational processes represented by the second term on the right hand side of Eq. (8). Also the results derived in this paper do not apply at very low temperatures. Following Van Kranendonk, we choose the unit cell so that it contains only one central nucleus, and we have to carry out the sum over all the lattice points within the unit cell. Our nucleus under study being now at the center of the body-centered lattice, as in Fig. 1, it suffices to carry out summations over $i,j=\pm1, \pm2, \pm3, \pm4$. Also, since the central lattice site is one of inversion symmetry, we have the further condition $s=s'$, as in I. Referring to Figs. 1 and 2, we find that out of $2\times8^4$ functions $L_{sil;sjm}$ for a body-centered cubic lattice, only the following different ones occur:

$s=2$:

$L_{2il;2jm}=\pm\{s_1{}^2\}^2=\pm L_1,\quad i=\pm l,\quad j=\pm m.$

$L_{2il;2jm}=\pm\{s_1{}^2\}\{s_1s_2\}=\pm L_2,\quad i=\pm l;\quad j=1,\ m=2, 4, -3,$
$\qquad j=2,\ m=1, 3, -4,$
$\qquad$ etc.;

i.e., point $m$ lies at points like $B$ and $D$ if $j$ is at $A$.

$L_{2il;2jm}=\pm\{s_1{}^2\}\{s_1s_3\}=\pm L_3,\quad i=\pm l;\quad j=1,\ m=3, -2, -4;$
$\qquad j=2,\ m=1, 3, -4;$
$\qquad$ etc.;

i.e., point $m$ lies at points like $C$ when $j$ lies at $A$.

$L_{2il;2jm}=\{s_1s_2\}^2=L_4,\quad i=1,\ l=2, 4, -3;\quad j=1,\ m=2, 4, -3;$
$\qquad i=2,\ l=1, 3, -4;\quad j=2,\ m=1, 3, -4;$
$\qquad$ etc.;

i.e., $l$ lies at points like $B$ and $D$ when $i$ is at $A$; $m$ lies at points like $B$ and $D$ when $j$ is at $A$.

$$L_{2il;2jm}=\{s_1s_3\}^2=L_5 \qquad \begin{array}{l} i=1,\quad l=3,\,-2,\,-4;\quad j=1,\quad m=3,\,-2,\,-4; \\ i=2,\quad l=4,\,-1,\,-3;\quad j=2,\quad m=4,\,-3,\,-1; \\ \text{etc.}; \end{array}$$

i.e., $l$ lies at points like $C$ when $i$ is at $A$; $m$ lies at points like $C$ when $i$ is at $A$.

$$L_{2il;2jm}=\{s_1s_2\}\{s_1s_3\}=L_6, \qquad \begin{array}{l} i=1,\quad l=2,\,4,\,-3;\quad j=1,\quad m=2,\,3,\,-4; \\ i=2,\quad l=1,\,3,\,-4;\quad j=2,\quad m=4,\,-3,\,-1; \\ \text{etc.}; \end{array}$$

i.e., $l$ lies at points like $B$ and $D$ when $i$ is at $A$; $m$ lies at points like $C$ when $j$ is at $A$. (9)

$s=1$:

$$\begin{array}{ll} L_{1il;1jm}=\{c_1^2\}^2=L_7, & ijlm \text{ same as for } L_1, \\ \quad =\{c_1^2\}\{c_1c_2\}=L_8, & ijlm \text{ same as for } L_2, \\ \quad =\{c_1^2\}\{c_1c_3\}=L_9, & ijlm \text{ same as for } L_3, \\ \quad =\{c_1c_2\}^2=L_{10}, & ijlm \text{ same as for } L_4, \\ \quad =\{c_1c_3\}^2=L_{11}, & ijlm \text{ same as for } L_5, \\ \quad =\{c_1c_2\}\{c_1c_3\}=L_{12}, & ijlm \text{ same as for } L_6. \end{array} \tag{10}$$

$$\begin{array}{ll} c_r=\cos(\mathbf{k}\cdot\mathbf{a}_r)-1, & \\ s_r=\sin(\mathbf{k}\cdot\mathbf{a}_r); & r=1,\,2,\,3. \end{array} \tag{11}$$

In the case of $L_1$, the positive or negative sign is taken according as whether positive and negative signs occur in both the equalities of $i$ with respect to $l$ and $j$ with respect to $m$ or in one of them only. In $L_2$ and $L_3$, the positive or negative sign is taken according as $i=+1$ or $i=-1$, respectively.

The averages over all the directions of $k$ are given by:

$$\begin{aligned} \{s_1^2\}&=\tfrac{1}{2}-\tfrac{1}{2}f(\sqrt{3}y), \\ \{s_1s_2\}&=\tfrac{1}{2}f(y)-\tfrac{1}{2}f(\sqrt{2}y), \\ \{s_1s_3\}&=\tfrac{1}{2}f(\sqrt{2}y)-\tfrac{1}{2}f(y), \\ \{c_1^2\}&=\tfrac{3}{2}-2f(\tfrac{1}{2}\sqrt{3}y)+\tfrac{1}{2}f(\sqrt{3}y), \\ \{c_1c_2\}&=1-2f(\tfrac{1}{2}\sqrt{3}y)+\tfrac{1}{2}f(\sqrt{2}y)+\tfrac{1}{2}f(y), \\ \{c_1c_3\}&=1-2f(\tfrac{1}{2}\sqrt{3}y)+\tfrac{1}{2}f(\sqrt{2}y)+\tfrac{1}{2}f(y). \end{aligned} \tag{12}$$

where $f(y)=\sin y/y$, $y=ka$; $a$ being the side of the b.c.c. lattice.

Just as in I, we can now write (3) as:

$$\begin{aligned} M_\mu(y)&=\sum_{n=1}^{12}N_{\mu n}L_n(y), \\ N_{\mu 1,\,2,\,3}&=\sum_{ijlm}\pm N_{\mu ijlm}, \\ N_{\mu n}&=\sum_{ijlm}N_{\mu ijlm}, \quad (n\neq 1,\,2,\,3) \end{aligned} \tag{13}$$

so that from (1),

$$P(m,\,m+\mu)=c_{\mu m}T^{*2}\sum_{n=1}^{12}N_{\mu n}D_n(T^*), \tag{14}$$

$$D_n(T^*)=T^*\int_0^{1/T^*}\frac{x^2e^x}{(e^x-1)^2}L_n(cT^*x)dx, \tag{15}$$

$$c_{\mu m}=\left(\frac{3|Q_{\mu m}|^2}{\pi d^2v^3a^3}\right), \quad c=k_ma=(6\pi^2)^{\frac{1}{3}}, \tag{16}$$

where we have assumed a Debye distribution so that $f(\omega)=1$. $\Theta$ is the Debye temperature defined by

$$\hbar\omega_m=\hbar vk_m=K\Theta, \tag{17}$$

$K$ being the Boltzmann constant. $T^*$ is the reduced temperature $T/\Theta$. The $D_n(T^*)$ functions are related to the $L_1$ functions discussed and shall be discussed further subsequently.

We now proceed to calculate the functions $N_{\mu n}$: Using a one-parameter method as in I, i.e., regarding the charge on each adjacent lattice site as $q=\gamma e$, we have the potential at a point distant $r$ from the position of a central nucleus given by:

$$V(\mathbf{r})=\textstyle\sum_i V_i(\mathbf{r}-\mathbf{r}_i), \tag{18}$$

$V_i(\mathbf{r})$ representing the potential at $\mathbf{r}$ arising from the charge on nucleus $i$ when its displacement $\mathbf{r}_i=\mathbf{s}_i-\mathbf{s}_0$ relative to the displacement $s$ of the central nucleus is zero. ($i=1,\,2,\,3,\,4,\,-1,\,-2,\,-3,\,-4$, as indicated in

Fig. 1.) We get easily,

$$
\begin{aligned}
V_1(\mathbf{r}) &= \frac{2q}{81\sqrt{3}a^5}[54r^4-360r^2x^2-180r^2z^2-360\sqrt{2}xzr^2+280x^4+560\sqrt{2}x^3z+840x^2z^2+280\sqrt{2}x^3z+70z^4],\\
V_2(\mathbf{r}) &= A[54r^4-360r^2y^2-180r^2z^2-360\sqrt{2}r^2yz+280y^4+560\sqrt{2}y^3z+840y^2z^2+280\sqrt{2}yz^3+70z^4],\\
V_3(\mathbf{r}) &= A[54r^4-360r^2x^2-180r^2z^2+360\sqrt{2}r^2xz+280x^4-560\sqrt{2}x^3z+840x^2z^2-280\sqrt{2}xz^3+70z^4],\\
V_4(\mathbf{r}) &= A[54r^4-360r^2y^2-180r^2z^2+360\sqrt{2}r^2yz+280y^4-506\sqrt{2}y^3z+840y^2z^2-280\sqrt{2}yz^3+70z^4],
\end{aligned}
\tag{19}
$$

where $A=2q/81\sqrt{3}a^5$ and $V_i(\mathbf{r})=V_{-i}(\mathbf{r})$.

From these relations, we get the tensors $A_{\mu ij}$:

$$
\begin{aligned}
A_{011}&=A\begin{bmatrix} 204 & 0 & -60\sqrt{2} \\ 0 & -36 & 0 \\ -60\sqrt{2} & 0 & -168 \end{bmatrix}, &
A_{022}&=A\begin{bmatrix} -36 & 0 & 0 \\ 0 & 204 & -60\sqrt{2} \\ 0 & 60\sqrt{2} & -168 \end{bmatrix},\\
A_{033}&=A\begin{bmatrix} 204 & 0 & 60\sqrt{2} \\ 0 & -36 & 0 \\ 60\sqrt{2} & 0 & -168 \end{bmatrix}, &
A_{044}&=A\begin{bmatrix} -36 & 0 & 0 \\ 0 & 204 & 60\sqrt{2} \\ 0 & 60\sqrt{2} & -168 \end{bmatrix},\\
A_{111}&=A\begin{bmatrix} -300\sqrt{2} & -360\sqrt{2}i & 816 \\ -360\sqrt{2}i & 180\sqrt{2}i & -144i \\ 816 & -144i & 70\sqrt{2} \end{bmatrix}, &
A_{144}&=A\begin{bmatrix} -360\sqrt{2}i & 360\sqrt{2} & -144 \\ 360\sqrt{2}i & 60\sqrt{2}i & -816i \\ -144 & -816i & -240\sqrt{2}i \end{bmatrix},\\
A_{211}&=A\begin{bmatrix} 24 & 126i & 120\sqrt{2} \\ 126i & -144 & 90\sqrt{2}i \\ 120\sqrt{2} & 90\sqrt{2}i & 120 \end{bmatrix}, &
A_{222}&=A\begin{bmatrix} 144 & 126i & 90\sqrt{2}i \\ 126i & -24 & -120\sqrt{2} \\ 90\sqrt{2}i & -120\sqrt{2} & -120 \end{bmatrix},\\
A_{233}&=A\begin{bmatrix} 24 & 126i & -120\sqrt{2} \\ 126i & -144 & -90\sqrt{2}i \\ -120\sqrt{2} & -90\sqrt{2}i & 120 \end{bmatrix}, &
A_{244}&=A\begin{bmatrix} 144 & 126i & -90\sqrt{2}i \\ 126i & -24 & 120\sqrt{2} \\ -90\sqrt{2}i & 120\sqrt{2} & -120 \end{bmatrix},
\end{aligned}
\tag{20}
$$

where $A_{\mu ii}=A_{\mu,-i,-i}$ and $A_{\mu ii}=A_{-\mu ii}{}^*$.

From these values of $A_{\mu ij}$, we get finally:

$$
\begin{aligned}
&N_{01}=16\times 85536A^2=N_{07},\\
&N_{02}=0=N_{08},\\
&N_{03}=0=N_{09},\\
&N_{04}=N_{05}=16\times 41904A^2=N_{0,10}=N_{0,11},\\
&N_{06}=0=N_{0,12},\\
&N_{1,1}=16\times 25666084A^2=N_{1,7},\\
&N_{1,2}=N_{1,8}=0,\\
&N_{1,3}=N_{1,9}=0,\\
&N_{1,4}=N_{1,5}=16\times 90142A^2=N_{1,10}=N_{1,11},\\
&N_{1,6}=N_{1,12}=0,\\
&N_{2,1}=16\times 157464A^2=N_{2,7},\\
&N_{2,2}=N_{2,8}=0,\\
&N_{2,3}=N_{2,9}=0,\\
&N_{2,4}=N_{2,5}=4\times 51984A^2=N_{2,10}=N_{2,11},\\
&N_{2,6}=N_{2,12}=0.
\end{aligned}
\tag{21}
$$

From these values of $N_{\mu n}$, we get:

$$
P(m,m+\mu)=\gamma^2|Q_{\mu m}|^2CT^{*2}E_\mu(T^*),
\tag{22}
$$

where

$$
C=64e^2/(6561\pi d^2v^3a^{13})
$$

and

$$
\begin{aligned}
E_0(T^*)&=85536(D_1+D_7)+41904(D_4+D_5+D_{10}+D_{11}),\\
E_{\pm 1}(T^*)&=25666084(D_1+D_7)\\
&\quad+90142(D_4+D_5+D_{10}+D_{11}),\\
E_{\pm 2}(T^*)&=157464(D_1+D_7)\\
&\quad+12996(D_4+D_5+D_{10}+D_{11}).
\end{aligned}
\tag{23}
$$

The relevant curves for $L_1(y)$, $L_4(y)$, $L_5(y)$ and $L_7(y)$, $L_{10}(y)$, $L_{11}(y)$, which are the only ones necessary from a consideration of the equations (21) for $N_{\mu n}$, are plotted in Fig. 3, ranging from 0 to a value greater than $C$. Numerical values of the $D$ functions are given in Table I. Besides, asymptotic expressions for the relevant $D$

TABLE I. Numerical values of the integrals $D_n(T^*)$.

| $T^*$ | $D_1(T^*)$ | $D_4(T^*)$ | $D_7(T^*)$ | $D_{10}(T^*)$ |
|---|---|---|---|---|
| 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 0.1 | 0.1901 | 0.0200 | 0.1236 | 0.0300 |
| 0.2 | 0.4343 | 0.0354 | 0.7478 | 0.2087 |
| 0.3 | 0.4105 | 0.0316 | 0.9941 | 0.2774 |
| 0.4 | 0.3890 | 0.0272 | 0.9477 | 0.2777 |
| 0.5 | 0.3324 | 0.0230 | 0.8518 | 0.2513 |
| 0.6 | 0.2930 | 0.0205 | 0.7763 | 0.3130 |
| 0.7 | 0.2536 | 0.0172 | 0.6885 | 0.2030 |
| 0.8 | 0.2347 | 0.0158 | 0.6255 | 0.1819 |
| 0.9 | 0.1964 | 0.0154 | 0.5782 | 0.1723 |
| 1.0 | 0.1888 | 0.0124 | 0.5271 | 0.1511 |
| ∞ | 0.1912 | 0.0128 | 0.2703 | 0.0778 |

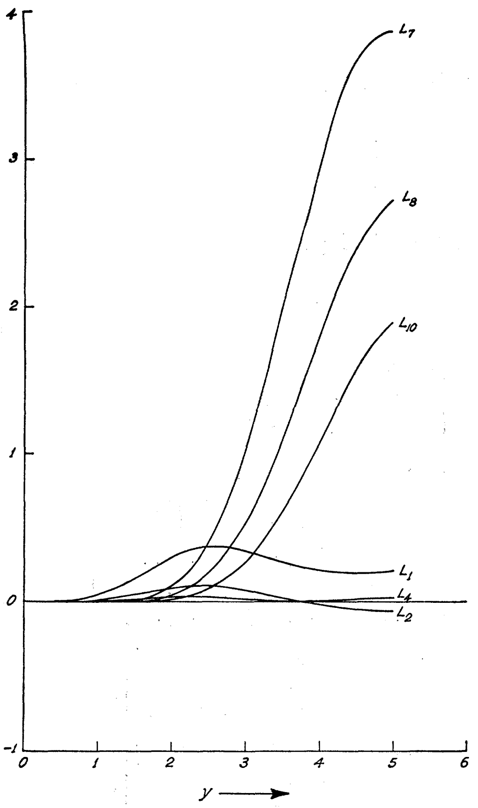

FIG. 3. Plot of the relevant $L_n(y)$ functions.

functions for low- and high-temperature ranges can be obtained as in I. Thus, for low temperatures, replacing the upper limit of the $D_n$ integrals by $\infty$, we can easily obtain, using Eqs. (12) and (15):

$$\begin{aligned} D_1' &\approx 1.05\times 10^4 T^{*5}, \\ D_4' = D_5' &\approx 1.17\times 10^3 T^{*5}, \\ D_7' &\approx 4.48\times 10^8 T^{*9}, \\ D_{10}' = D_{11}' &\approx 1.49\times 10^7 T^{*9}. \end{aligned} \tag{24}$$

Thus, at low temperatures, remembering the $T^{*2}$ factor in the expression for $P(m,m+\mu)$ in (14), the transition probability and hence the inverse relaxation time is proportional to $T^{*7}$. At high temperatures, expanding $D_n(T^*)$ in descending powers of $T^*$, we have as in I:

$$D_n(T^*) = L_{n0} - 1/12 L_{n2}(cT^*)^{-2} + \cdots, \tag{25}$$

where

$$L_{np} = \frac{1}{c}\int_0^c x^p L_n(x)dx.$$

We get numerical values for $L_{n0}$ and $L_{n2}$ and hence $D_n(T^*)$, using (12).

$$\begin{aligned} D_1(T^*) &= 0.1912 - 0.09560/T^{*2} + \cdots, \\ D_4(T^*) &= 0.0128 - 0.00463/T^{*2} + \cdots, \\ D_7(T^*) &= 0.2703 - 0.4032/T^{*2} + \cdots, \\ D_{10}(T^*) &= 0.0778 - 0.1262/T^{*2} + \cdots. \end{aligned} \tag{26}$$

To get the relaxation time(s),[6] a suitable combination of the transition probabilities $P_{m,m+\mu}$ should be taken, depending upon the experimental conditions. At high temperatures including room temperature the observed relaxation times should be proportional to $\gamma^{-2}T^{*-2}$. The proportionality constant depends on the nucleus and crystal structure under consideration.

## CONCLUSION

The calculations for the body-centered cubic lattice is essentially similar to that for a simple cubic lattice structure, and indeed Van Kranendonk's method may be used to calculate relaxation times for any type of lattice, using a suitable coordinate system. Unfortunately, the relaxation time for $Cs^{133}$ in cesium halide crystals has not been measured so that we cannot compare $\gamma^2$ with Sternheimer's[5] $(1+|\gamma_\infty|)^2$ which, from the value of $\gamma_\infty$ mentioned in the introduction, is of the order of $2\times 10^4$ in this case.

## ACKNOWLEDGMENTS

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*Note added in proof.*—After this paper was sent for publication, the paper by Yoshida and Moriya, referred to earlier, appeared. These authors calculated the contribution to the relaxation process from the distortion of the covalent bonding between the atoms and rejected the direct ionic contribution to the relaxation process, which they estimated as being an order of magnitude $10^{-4}$ smaller, without the antishielding Sternheimer factor. However, for $Br^-$ and $I^-$ ions, the cases in which they were interested, values of $\gamma_\infty$ of about $-100$ and $-200$, respectively, would not be unreasonable, considering Sternheimer and Foley's recent values of $-70.7$ and $-143.5$, respectively, in the neighboring ions $Rb^+$ and $Cs^+$. $(1+|\gamma_\infty|)^2$ would then be about $10^4$ and $10^4\times 4$ for $Br^-$ and $I^{-1}$, respectively, so that both the mechanisms would be of comparable importance.

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[6] F. Reif, Phys. Rev. **100**, 1597 (1956).