atoms are present in the alloy to give the needed accuracy. We may recall that the moments are obtained from intensities of peaks, not from line position as is the case for the hyperfine fields. The study of the ordered alloy of 25% Rh can probably provide the needed information on the FeII moment. The task becomes considerably easier if one can obtain Fe$_3$Al-type ordering. As already mentioned, our effort to obtain this type of ordering was not successful. At present, we are attempting to obtain a single crystal of 25% Rh ordered in CsCl type.

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Field Dependence of the Direct-Process Spin-Lattice Relaxation Time in Rare-Earth Salts

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The field dependence of the direct-process spin-lattice relaxation time for rare-earth salts having $g_1=0$ is computed taking into account dipolar coupling between the spins. Comparison is made with the corresponding relaxation time calculated in a modified effective-field approximation, and the presence of terms not found in the modified effective-field formula is noted. Conditions that must obtain for the effective-field approximation to be valid are discussed, and a numerical evaluation of the relaxation time for dysprosium ethyl sulfate is given. Extension of the calculation to salts not having $g_1=0$ is also considered.

I. INTRODUCTION

THE purpose of this note is to extend to one-phonon processes the calculation of the field dependence of the spin-lattice relaxation time. Because of the complexity of the equations we will present explicit expressions for the relaxation time only for those salts having $g_1=0$. The formalism used will be that of Van Vleck,[1] Gorter,[2] and Hebel and Slichter,[3] while the notation will be similar to that of Orbach,[4] who computed the field dependence of the two-phonon process. Although we restrict our treatment to spin-lattice relaxation in rare earth salts, the theory is also applicable to the iron group salts, with certain provisos that are discussed in OI. Mention must also be made of the work of Bolger[5] who has considered the field dependence of the spin-lattice relaxation time from a somewhat different point of view.

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[4] R. Orbach, Proc. Roy. Soc. (London) A264, 458 (1961); A264, 485 (1961). Hereafter referred to as OI and OII.

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II. DERIVATION OF THE GENERAL FORMULA FOR THE RELAXATION TIME

We begin with the general expression for the relaxation time,[3] T_1,

$$\frac{1}{T_1} = \frac{1}{2}\frac{\sum_{\alpha,\beta}(E_\alpha-E_\beta)^2 W_{\alpha\beta}}{\sum_\alpha E_\alpha{}^2},\qquad(1)$$

where $W_{\alpha\beta}$ is the net relaxation rate between the levels α and β, which have energies E_α and E_β. The spin-lattice relaxation process takes place via the orbit-lattice interaction as discussed in detail in OI. We restrict our attention to Kramers salts, the extension to non-Kramers salts being straightforward. In the case of Kramers salts the states $|\alpha\rangle$ and $|\beta\rangle$ form a time-conjugate doublet in the absence of perturbations. A perturbation is, therefore, introduced to remove the degeneracy of the ground state and to couple in the excited states so that the relaxation process might take place. In our problem the perturbation V' is the sum of the dipolar and Zeeman interactions. That is,

$$V'=V_{\rm dip}+V_z,\qquad(2)$$

where

$$V_z=\sum_j \beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(j)},\qquad(3)$$

and

$$V_{\rm dip}=\beta^2\Lambda^2\sum_{(j,k)}\frac{1}{r_{jk}{}^3}(A+B+C+D+E+F),\qquad(4)$$

with[6]

$$A = (J_z^{(i)}J_z^{(k)})(1 - 3\cos^2\theta_{jk})$$

$$B = -\tfrac{1}{4}(J_-^{(i)}J_+^{(k)} + J_+^{(i)}J_-^{(k)})(1 - 3\cos^2\theta_{jk})$$

$$C = -\tfrac{3}{2}(J_+^{(i)}J_z^{(k)} + J_z^{(i)}J_+^{(k)})\sin\theta_{jk}\cos\theta_{jk}e^{-i\varphi_{jk}}$$

$$D = -\tfrac{3}{2}(J_-^{(i)}J_z^{(k)} + J_z^{(i)}J_-^{(k)})\sin\theta_{jk}\cos\theta_{jk}e^{i\varphi_{jk}} \quad (5)$$

$$E = -\tfrac{3}{4}(J_+^{(i)}J_+^{(k)})\sin^2\theta_{jk}e^{-2i\varphi_{jk}}$$

$$F = -\tfrac{3}{4}(J_-^{(i)}J_-^{(k)})\sin^2\theta_{jk}e^{2i\varphi_{jk}}.$$

After rearranging terms we can write V_{dip} as follows:

$$V_{\text{dip}} = \sum_{(j,k),p,q} a_{pq}{}^{jk}O_1{}^p(j)O_1{}^q(k)Y_2{}^{p+q}(jk)^*. \quad (4')$$

The $a_{pq}{}^{jk}$ are the appropriate coefficients in the expansion (5); $p, q = 0, \pm 1$; and $O_1{}^\pm(j) = \mp J_\pm^{(i)}$. In these expressions the symbol β denotes the Bohr magneton, Λ is the Landé g factor, and $\mathbf{H}$ denotes the external magnetic field. The bracketed indices (j,k) indicate that each pair of ions is to be counted once.

In the presence of V' the state vectors for the jth spin become

$$|\alpha_j\rangle' = |\alpha_j\rangle + \left\{ \frac{\langle \pm\tfrac{1}{2}q_j | V' | \alpha_j\rangle}{-\Delta_q} | \pm\tfrac{1}{2}q_j\rangle + \frac{\langle \pm\tfrac{1}{2}r_j | V' | \alpha_j\rangle}{-\Delta_r} | \pm\tfrac{1}{2}r_j\rangle + \cdots \right\}, \quad (6)$$

$$|\beta_j\rangle' = |\beta_j\rangle + \left\{ \frac{\langle \pm\tfrac{1}{2}q_j | V' | \beta_j\rangle}{-\Delta_q} | \pm\tfrac{1}{2}q_j\rangle + \frac{\langle \pm\tfrac{1}{2}r_j | V' | \beta_j\rangle}{-\Delta_r} | \pm\tfrac{1}{2}r_j\rangle + \cdots \right\}, \quad (7)$$

where $|\pm\tfrac{1}{2}q_j\rangle$, $|\pm\tfrac{1}{2}r_j\rangle$, $\cdots$, refer to the excited doublets of the jth spin, separated from the ground doublet by energies Δ_q, Δ_r, $\cdots$, respectively. With these perturbed state vectors one proceeds directly to compute the net transition rate by methods outlined in OI.[7] Making the assumption $kT \gg \langle V'\rangle$ and including only the first excited doublet, we find

$$W_{\alpha\beta} = \frac{3\delta_{\alpha\beta}{}^2 kT}{2\pi\hbar^4\rho V^5} \sum_{i,j} \frac{\sin k_0 r_{ji}}{k_0 r_{ij}} \langle \alpha | V_{\text{OL}}^{(i)} | \beta\rangle' \langle \beta | V_{\text{OL}}^{(i)} | \alpha\rangle'$$

$$= \frac{6\delta_{\alpha\beta}{}^2 kT}{\pi\rho V^5\hbar^4\Delta_q{}^2} \left\{ \sum_{i,j} \frac{\sin k_0 r_{ij}}{k_0 r_{ij}} \langle \alpha | (\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)} + \sum_{k,p,q} a_{pq}{}^{jk}Y_2{}^{p+q}(jk)^*O_1{}^p(j)O_1{}^q(k)) | \tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j | V_{\text{OL}}^{(i)} | \beta\rangle\langle\beta | V_{\text{OL}}^{(i)} | \tfrac{1}{2}q_i\rangle \right.$$

$$\left. \times \langle\tfrac{1}{2}q_i | (\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)} + \sum_{m,r,s} a_{rs}{}^{im}Y_2{}^{r+s}(im)^*O_1{}^r(i)O_1{}^s(m)) | \alpha\rangle \right\}. \quad (8)$$

In (8) we have introduced the following symbols: $\delta_{\alpha\beta} = E_\alpha - E_\beta = $ energy difference between perturbed doublet states, $\rho = $ density, $V = $ velocity of sound, $k_0 = \delta_{\alpha\beta}/\hbar V$, $T = $ temperature in °K, $V_{\text{OL}}^{(i)} = $ orbit-lattice interaction of ith spin, and $k = $ Boltzmann's constant.

The state vectors $|\alpha\rangle$ are state vectors in the space of the entire system of N spins, and $|\tfrac{1}{2}q_j\rangle$ is a state vector in the space of the jth spin.

Letting

$$\Gamma_j = (\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)} + \sum_{m,r,s} a_{rs}{}^{jm}Y_2{}^{r+s}(jm)^*O_1{}^r(j)O_1{}^s(m)) | \tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j | V_{\text{OL}}^{(i)}, \quad (9)$$

and

$$M = \frac{3kT}{\pi\rho V^5\hbar^4\Delta_q{}^2}, \quad (10)$$

we obtain for the relaxation time

$$\frac{1}{T_1} = \frac{M \displaystyle\sum_{\alpha,\beta,i,j} \frac{\sin k_0 r_{ij}}{k_0 r_{ij}}(E_\alpha - E_\beta)^4\langle\alpha | \Gamma_i | \beta\rangle\langle\beta | \Gamma_j{}^\dagger | \alpha\rangle}{\displaystyle\sum_\alpha E_\alpha{}^2}, \quad (11)$$

where, as before, α and β refer to states of the N spin system. Noting that

$$\langle\alpha | V' | \alpha\rangle = E_\alpha, \quad (12)$$

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[6] N. Bloembergen, E. M. Purcell, and R. V. Pound, Phys. Rev. **73**, 679 (1948).

[7] See, in particular, Sec. 4.

(11) can be written as

$$\frac{1}{T_1} = M\left(\sum_\alpha E_\alpha{}^2\right)^{-1}\left(\sum_{\alpha,\beta,i,j}\frac{\sin k_0 r_{ij}}{k_0 r_{ij}}\langle\alpha|\,V'V'\Gamma_i - 2V'\Gamma_i V' + \Gamma_i V'V'\,|\beta\rangle\langle\beta|\,V'V'\Gamma_j{}^\dagger - 2V'\Gamma_j{}^\dagger V' + \Gamma_j{}^\dagger V'V'\,|\alpha\rangle\right)$$

$$=\frac{M\displaystyle\sum_{i,j}\frac{\sin k_0 r_{ij}}{k_0 r_{ij}}\,\mathrm{tr}[V',[V',\Gamma_i]][V',[V',\Gamma_j{}^\dagger]]}{\mathrm{tr}\,V'^2}. \tag{13}$$

III. SPECIALIZATION $g_\perp = 0$ SALTS

Since the evaluation of (13) becomes prohibitively complex for the general rare earth salt, we will restrict our attention to salts having $g_\perp = 0$. As all matrix elements of J_x and J_y within the ground doublet vanish for these salts, we can write

$$V_z = \sum_j \beta\Lambda H J_z{}^{(j)}\cos\theta, \tag{14}$$

$$V_{\mathrm{dip}} = \sum_{(i,j)} a_{00}{}^{ij}Y_2{}^0(ij)J_z{}^{(i)}J_z{}^{(j)}, \tag{15}$$

$$\Gamma_j = \left(\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(j)} + \sum_{r,m} a_{r0}{}^{jm}Y_2{}^r(jm)^* O_1{}^r(j)J_z{}^{(m)}\right)|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}. \tag{16}$$

We have noted that because Γ_j operates only between ground doublets of spins other than the jth we need keep only terms containing $J_z{}^{(m)}$ in its expansion $(m\neq j)$. Since the trace is independent of representation we can work in the representation in which the state vectors are products of the state vectors of the individual spins. For $g_\perp = 0$ salts,

$$\sum_{\alpha,\beta}\langle\alpha|\,V'V'\Gamma_i - 2V'\Gamma_i V' + \Gamma_i V'V'\,|\beta\rangle\langle\beta|\,V'V'\Gamma_j{}^\dagger - 2V'\Gamma_j{}^\dagger V' + \Gamma_j{}^\dagger V'V'\,|\alpha\rangle$$

$$\propto \langle\alpha_i|\Gamma_i|\beta_i\rangle\langle\alpha_j|\beta_j\rangle\langle\beta_i|\alpha_i\rangle\langle\beta_j|\Gamma_j{}^\dagger|\alpha_j\rangle = 0 \quad (i\neq j), \tag{17}$$

the last step following from the observation that V' has only terms in $J_z{}^{(i)}$ and $J_z{}^{(j)}$, while Γ_j has only terms in $J_z{}^{(k)}$ for $k\neq j$.

Remembering that Γ_j connects only states $|\alpha_j\rangle$ and $|\beta_j\rangle$ for which $\langle\alpha_j|J_z{}^{(j)}|\alpha_j\rangle = -\langle\beta_j|J_z{}^{(j)}|\beta_j\rangle$, we can write

$$\sum_{\alpha,\beta}\langle\alpha|\,V'V'\Gamma_j - 2V'\Gamma_j V' + \Gamma_j V'V'\,|\beta\rangle\langle\beta|\,V'V'\Gamma_j{}^\dagger - 2V'\Gamma_j{}^\dagger V' + \Gamma_j{}^\dagger V'V'\,|\alpha\rangle = 16\,\mathrm{tr}\,V'^4\Gamma_j\Gamma_j{}^\dagger. \tag{18}$$

The equation for the direct process spin-lattice relaxation time of $g_\perp = 0$ salts is thus written

$$1/T_1 = 16M\sum_j \mathrm{tr}\,V'^4\Gamma_j\Gamma_j{}^\dagger/\mathrm{tr}\,V'^2. \tag{19}$$

The evaluation of $\mathrm{tr}\,V'^4\Gamma_j\Gamma_j{}^\dagger$ is left to the Appendix. We give only the result.

$$\mathrm{tr}\,V'^4\Gamma_j\Gamma_j{}^\dagger = |\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}|\beta_j\rangle|^2\{\alpha^4\cos^4\theta(\beta\Lambda H)^6|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}|\tfrac{1}{2}q_j\rangle|^2 + \alpha^6\cos^4\theta(\beta\Lambda H)^4\sum_m|H_m{}^{(j)}|^2 + 6\alpha^8(\beta\Lambda H)^2$$

$$\times\cos^2\theta\sideset{}{'}\sum_{k,m} Y_2{}^0(jk)^2(a_{00}{}^{jk})^2|H_m{}^{(j)}|^2 + 6\alpha^6\cos^2\theta(\beta\Lambda H)^4|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}|\tfrac{1}{2}q_j\rangle|^2\sum_m Y_2{}^0(jm)^2(a_{00}{}^{jm})^2$$

$$+ 6\alpha^8\cos^2\theta(\beta\Lambda H)^2\sum_m Y_2{}^0(jm)^2(a_{00}{}^{jm})^2|H_m{}^{(j)}|^2 + 12\alpha^8(\beta\Lambda H)^2\cos^2\theta\sideset{}{'}\sum_{k,m} Y_2{}^0(jk)$$

$$\times Y_2{}^0(jm)a_{00}{}^{jk}a_{00}{}^{jm}H_k{}^{(j)}H_m{}^{(j)*} + \alpha^8(\beta\Lambda H)^2|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}|\tfrac{1}{2}q_j\rangle|^2\sum_m Y_2{}^0(jm)^4(a_{00}{}^{jm})^4$$

$$+ 3\alpha^8(\beta\Lambda H)^2|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}|\tfrac{1}{2}q_j\rangle|^2\sideset{}{'}\sum_{k,m} Y_2{}^0(jk)^2 Y_2{}^0(jm)^2(a_{00}{}^{jk})^2(a_{00}{}^{jm})^2 + \alpha^{10}\sum_k Y_2{}^0(jk)^4(a_{00}{}^{jk})^4|H_k{}^{(j)}|^2$$

$$+ \alpha^{10}\sideset{}{'}\sum_{k,m} Y_2{}^0(jk)^4(a_{00}{}^{jk})^4|H_m{}^{(j)}|^2 + 3\alpha^{10}\sideset{}{'}\sum_{k,m,n} Y_2{}^0(jk)^2 Y_2{}^0(jm)^2(a_{00}{}^{jk})^2(a_{00}{}^{jm})^2|H_n{}^{(j)}|^2$$

$$+ 6\alpha^{10}\sideset{}{'}\sum_{k,m} Y_2{}^0(jm)^2 Y_2{}^0(jk)^2(a_{00}{}^{jm})^2(a_{00}{}^{jk})^2|H_m{}^{(j)}|^2 + 4\alpha^{10}\sideset{}{'}\sum_{k,m} Y_2{}^0(jm)^3 Y_2{}^0(jk)(a_{00}{}^{jm})^3(a_{00}{}^{jk})$$

$$\times[H_m^{(i)}H_k^{(i)*}+H_m^{(i)*}H_k^{(i)}]+12\alpha^{10}\sum_{k,m,n}{}' Y_2^0(jk)^2Y_2^0(jm)Y_2^0(jn)(a_{00}^{jk})^2(a_{00}^{jm})(a_{00}^{jn})H_m^{(i)}H_n^{(i)*}$$

$$+4\alpha^6(\beta\Lambda H)^4\cos^3\theta\sum_k a_{00}^{jk}Y_2^0(jk)[\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle H_k^{(i)*}+H_k^{(i)}\langle\tfrac{1}{2}q_j|\hat{h}\cdot\mathbf{J}^{(i)}|\alpha_j\rangle]$$

$$+4\alpha^8(\beta\Lambda H)^2\cos\theta\sum_k (a_{00}^{jk})^3Y_2^0(jk)^3[\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle H_k^{(i)*}+H_k^{(i)}\langle\tfrac{1}{2}q_j|\hat{h}\cdot\mathbf{J}^{(i)}|\alpha_j\rangle]+12\alpha^8(\beta\Lambda H)^2\cos\theta\sum_{k,m}{}' Y_2^0(jk)^2$$

$$\times Y_2^0(jm)(a_{00}^{jk})^2(a_{00}^{jm})[\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle H_m^{(i)*}+H_m^{(i)}\langle\tfrac{1}{2}q_j|\hat{h}\cdot\mathbf{J}^{(i)}|\alpha_j\rangle]\}, \quad (20)$$

where

$$\alpha=|\langle\alpha_j|J_z^{(i)}|\alpha_j\rangle|=g_{11}/2\Lambda, \quad (21)$$

$$\hat{h}=\mathbf{H}/|\mathbf{H}|, \quad (22)$$

$$H_m^{(i)}=\langle\alpha_j|\sum_r O_1^r(j)a_{r0}^{jm}Y_2^r(jm)^*|\tfrac{1}{2}q_j\rangle. \quad (23)$$

The prime on the summation symbol means that none of the indices that are summed over are equal. One obtains as well[8]

$$\mathrm{tr}V'^2=\sum_j \alpha^2(\beta\Lambda H)^2\cos^2\theta+\tfrac{1}{2}\alpha^4\sum_{j,k} (a_{00}^{jk})^2Y_2^0(jk)^2. \quad (24)$$

If we define

$$(1/T_{10})=16\alpha^2(\beta\Lambda H)^2\cos^2\theta M|\langle\tfrac{1}{2}q_j|V_{\mathrm{OL}}^{(i)}|\beta\rangle|^2|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2$$

$$=\frac{12g_{11}^2H^4\beta^4\Lambda^2\cos^2\theta kT}{\pi\rho V^5\hbar^4\Delta_q^2}|\langle\tfrac{1}{2}q_j|V_{\mathrm{OL}}^{(i)}|\beta_j\rangle|^2|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2, \quad (25)$$

the inverse direct-process spin-lattice relaxation time in the absence of dipolar coupling,[9] we find, after substituting (20) and (24) into (19) and rearranging terms,

$$\frac{1}{T_1}=\frac{1}{T_{10}}(H^2\cos^2\theta+\tfrac{1}{2}K^2)^{-1}\Bigg(H^2\cos^2\theta+\frac{2K'^2\cos^2\theta}{\sin^2\theta}+6K^2+\frac{12K^2K'^2}{H^2\sin^2\theta}-\frac{12\alpha^4\sum_m Y_2^0(jm)^2|H_m^{(i)}|^2(a_{00}^{jm})^2}{(\beta\Lambda)^4H^2|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}$$

$$+\frac{12\alpha^4\sum_{k,m} Y_2^0(jk)Y_2^0(jm)a_{00}^{jk}a_{00}^{jm}H_k^{(i)}H_m^{(i)*}}{(\beta\Lambda)^4H^2|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}-\frac{2\alpha^4\sum_m Y_2^0(jm)^4(a_{00}^{jm})^4}{(\beta\Lambda)^4H^2\cos^2\theta}-\frac{4K'^2\alpha^4\sum_m Y_2^0(jm)^4(a_{00}^{jm})^4}{(\beta\Lambda H)^4\cos^2\theta\sin^2\theta}$$

$$+\frac{3K^4}{H^2\cos^2\theta}+\frac{6K^4K'^2}{H^4\cos^2\theta\sin^2\theta}+\frac{16\alpha^6\sum_m Y_2^0(jm)^4(a_{00}^{jm})^4|H_m^{(i)}|^2}{(\beta\Lambda)^6H^4\cos^2\theta|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}-\frac{12\alpha^4K^2\sum_{k,m} Y_2^0(jk)^2(a_{00}^{jk})^2|H_m^{(i)}|^2}{(\beta\Lambda)^4H^4\cos^2\theta|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}$$

$$-\frac{8\alpha^6\sum_{k,m} Y_2^0(jk)^3Y_2^0(jm)(a_{00}^{jk})^3a_{00}^{jm}[H_k^{(i)}H_m^{(i)*}+H_k^{(i)*}H_m^{(i)}]}{(\beta\Lambda)^6H^4\cos^2\theta|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}$$

$$+\frac{12\alpha^6\sum_{k,m,n} Y_2^0(jk)^2Y_2^0(jm)Y_2^0(jn)(a_{00}^{jk})^2a_{00}^{jm}a_{00}^{jn}H_m^{(i)}H_n^{(i)*}}{(\beta\Lambda)^6H^4\cos^2\theta|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}$$

$$+\frac{4\alpha^2\cos\theta\sum_k Y_2^0(jk)a_{00}^{jk}[\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle H_k^{(i)*}+H_k^{(i)}\langle\tfrac{1}{2}q_j|\hat{h}\cdot\mathbf{J}^{(i)}|\alpha_j\rangle]}{(\beta\Lambda)^2|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}$$

$$-\frac{8\alpha^4\sum_k Y_2^0(jk)^3(a_{00}^{jk})^3[\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle H_k^{(i)*}+H_k^{(i)}\langle\tfrac{1}{2}q_j|\hat{h}\cdot\mathbf{J}^{(i)}|\alpha_j\rangle]}{(\beta\Lambda)^4H^2\cos\theta|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}$$

$$+\frac{2\alpha^4\sum_{k,m} Y_2^0(jk)^2Y_2^0(jm)(a_{00}^{jk})^2(a_{00}^{jm})[\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle H_k^{(i)*}+H_k^{(i)}\langle\tfrac{1}{2}q_j|\hat{h}\cdot\mathbf{J}^{(i)}|\alpha_j\rangle]}{|\langle\alpha_j|\hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2}\Bigg). \quad (26)$$

In anticipation of the discussion given below, we have introduced the symbols

$$K^2=\left(\frac{\alpha}{\beta\Lambda}\right)^2\sum_k (a_{00}^{jk})^2Y_2^0(jk)^2, \quad (27)$$

[8] OII, Eqs. (16) and (21).
[9] OI, Eq. (25).

and

$$K'^2 = \frac{\alpha^2 \sin^2\theta \sum_m |H_m^{(j)}|^2}{2(\beta\Lambda)^2 |\langle \alpha_j | \hat{h} \cdot \mathbf{J}^{(j)} | \tfrac{1}{2} q_j \rangle|^2}. \tag{28}$$

An important simplification in the expression for the relaxation time results when we limit consideration to those salts for which all matrix elements of J_z between the ground doublet and the first excited doublet vanish.[10] Because only J_+ and J_- connect the first excited doublet with the ground doublet,

$$H_m^{(j)} = -\frac{3\beta^2\Lambda^2 \sin\theta_{jm} \cos\theta_{jm}}{2r_{jm}^3} [e^{-i\varphi_{jm}} \langle \alpha_j | J_+^{(j)} | \tfrac{1}{2} q_j \rangle + e^{i\varphi_{jm}} \langle \alpha_j | J_-^{(j)} | \tfrac{1}{2} q_j \rangle]. \tag{29}$$

If there is an absence of a single preferred direction in the plane perpendicular to the crystal axis, as is the case for cubic and hexagonal symmetries, we observe the following properties of the $H_m^{(j)}$:

$$\begin{aligned}
\sum_m H_m^{(j)} Y_2^0(jm) a_{00}^{jm} &= \sum_m H_m^{(j)} Y_2^0(jm)^2 (a_{00}^{jm})^2, \\
&= \sum_m H_m^{(j)} Y_2^0(jm)^3 (a_{00}^{jm})^3, \\
&= 0.
\end{aligned} \tag{30}$$

The expression for the relaxation time is, thus, considerably simplified. We obtain

$$\begin{aligned}
\frac{1}{T_1} = \frac{(H^2\cos^2\theta + \tfrac{1}{2}K^2)^{-1}}{T_{10}H^4 \cos^2\theta \sin^2\theta} &\bigg[H^4 \cos^4\theta (H^2\sin^2\theta + 2K'^2) + 6H^2K^2\cos^2\theta (H^2\sin^2\theta + 2K'^2) + 3K^4(H^2\sin^2\theta + 2K'^2) \\
&- \frac{12\alpha^4(H^2\cos^2\theta + K^2)\sum_m Y_2^0(jm)^2(a_{00}^{jm})^2 |H_m^{(j)}|^2}{(\beta\Lambda)^4 |\langle \alpha_j | J_\perp^{(j)} | \tfrac{1}{2} q_j \rangle|^2} - \frac{2\alpha^4(H^2\sin^2\theta + 2K'^2)\sum_m Y_2^0(jm)^4 (a_{00}^{jm})^4}{(\beta\Lambda)^4} \\
&+ \frac{16\alpha^6 \sum_m Y_2^0(jm)^4 (a_{00}^{jm})^4 |H_m^{(j)}|^2}{(\beta\Lambda)^6 |\langle \alpha_j | J_\perp^{(j)} | \tfrac{1}{2} q_j \rangle|^2} \bigg].
\end{aligned} \tag{31}$$

Furthermore, K'^2 is independent of θ and is given by

$$K'^2 = \frac{\alpha^2 \sum_m |H_m^{(j)}|^2}{2(\beta\Lambda)^2 |\langle \alpha_j | J_\perp^{(j)} | \tfrac{1}{2} q_j \rangle|^2}. \tag{28'}$$

Since $1/T_{10} \propto H^4$, $1/T_1$ approaches a constant nonzero value in the limit as H approaches zero. This limit is given by

$$\begin{aligned}
\frac{1}{T_1} \sim \frac{2}{T_{10}H^4\cos^2\theta \sin^2\theta K^2} &\bigg[6K^4K'^2 - \frac{4\alpha^4 K'^2 \sum_m Y_2^0(jm)^4 (a_{00}^{jm})^4}{(\beta\Lambda)^4} + \frac{16\alpha^6 \sum_m Y_2^0(jm)^4 (a_{00}^{jm})^4 |H_m^{(j)}|^2}{(\beta\Lambda)^6 |\langle \alpha_j | J_\perp^{(j)} | \tfrac{1}{2} q_j \rangle|^2} \\
&- \frac{12\alpha^4 K^2 \sum_m Y_2^0(jm)^2 (a_{00}^{jm})^2 |H_m^{(j)}|^2}{(\beta\Lambda)^4 |\langle \alpha_j | J_\perp^{(j)} | \tfrac{1}{2} q_j \rangle|^2} \bigg].
\end{aligned} \tag{32}$$

The fact that $1/T_1$ is not equal to zero in the absence of an external magnetic field is a direct consequence of the dipolar coupling of the excited doublet to the ground doublet. In the opposite limit, when the external magnetic field is much greater than the dipolar field, $1/T_1 \sim 1/T_{10}$.

IV. DISCUSSION OF RESULTS

We wish to compare (31) with a corresponding expression for the relaxation time calculated in an approximation that is a slight modification of the effective-field approximation of Brons[11] and Van Vleck.[1] In the effective-field approximation the factors of H arising in the expression for the relaxation time calculated in the absence of dipolar coupling are averaged over a mean square dipolar field K^2, with the distribution $\mathcal{P}(H')$ given by

$$\mathcal{P}(H')dH' = (2\pi K^2)^{-1/2} \exp\left(-\frac{(H-H')^2}{2K^2} \right)dH'. \tag{33}$$

[10] This is the case for nearly all $g_\perp = 0$ salts.
[11] F. Brons, thesis, Gronigen, 1938 (unpublished).

The inverse relaxation time in the absence of dipolar coupling must be interpreted as having a factor of H^6 in the numerator (arising from $\mathrm{tr}V'^4\Gamma_j\Gamma_j^\dagger$) and a factor of H^2 in the denominator (arising from $\mathrm{tr}V'^2$). It must also be noted that the factor of H^6 in the numerator is the product of a factor of H^4 arising from the matrix elements of V_z taken within the ground doublet and a factor of H^2 arising from matrix elements of V_z between the ground doublet and the excited doublet. Since in $g_\perp=0$ salts only the term $\beta^2\Lambda^2 J_z^{(j)}J_z^{(k)}(1-3\cos^2\theta_{jk})$ contributes to the matrix elements of V_{dip} within the ground doublet whereas the full set of terms, (5), contributes to the matrix elements of V_{dip} between the ground doublet and the first excited doublet, we argue that the factors of H^4 and H^2 are to be averaged over different mean square dipolar fields, K^2 and K'^2.[12] We obtain for the relaxation time in our modified effective-field approximation,

$$\frac{1}{T_1}=\frac{1}{T_{10}}\frac{(H^4\cos^4\theta+6H^2K^2\cos^2\theta+3K^4)(H^2+3K'^2)}{H^4\sin^2\theta\cos^2\theta(H^2\cos^2\theta+\tfrac{1}{2}K^2)}. \tag{34}$$

In particular, for those salts having no nonzero elements of J_z between the ground doublet and the first excited doublet the modified effective-field approximation yields

$$\frac{1}{T_1}=\frac{1}{T_{10}}\frac{(H^4\cos^4\theta+6H^2K^2\cos^2\theta+3K^4)(H^2\sin^2\theta+2K'^2)}{H^4\sin^2\theta\cos^2\theta(H^2\cos^2\theta+\tfrac{1}{2}K^2)}. \tag{35}$$

The difference between (35) and (34) arises from the fact that in (34) both H_z^2 and $H_\perp^2$ are averaged, whereas in (35) only $H_\perp^2$ is averaged.

Our use of the effective-field approximation differs somewhat from that of Van Vleck. We have averaged the factors H^2 and H^4 over different dipolar fields, whereas Van Vleck has averaged them over the same dipolar field. Our expression for the effective field approximation to the relaxation time is thus not the same as the expression found in his paper.

Comparison of (35) with (31) indicates that the modified effective-field approximation omits the terms

$$-\frac{2\alpha^4(H^2\sin^2\theta+2K'^2)\sum_m Y_2^0(jm)^4(a_{00}{}^{jm})^4}{(\beta\Lambda)^4}, \tag{36}$$

$$-\frac{12\alpha^4(H^2\cos^2\theta+K^2)\sum_m Y_2^0(jm)^2(a_{00}{}^{jm})^2|H_m^{(j)}|^2}{(\beta\Lambda)^4|\langle\alpha_j|J_\perp^{(j)}|\tfrac{1}{2}q_j\rangle|^2}, \tag{37}$$

and

$$\frac{16\alpha^6\sum_m Y_2^0(jm)^4(a_{00}{}^{jm})^4|H_m^{(j)}|^2}{(\beta\Lambda)^6|\langle a_j|J_\perp^{(j)}|\tfrac{1}{2}q_j\rangle|^2}, \tag{38}$$

from the exact relaxation time formula. Since each of these terms falls off at least as rapidly as $1/r_{jm}^{12}$, it is to be expected that their contribution to the relaxation time will be small. We presently show that for at least one rare salt this is the case.

Ignoring the contributions of (36), (37), and (38) we see that (35) and (31) are the same, permitting us to identify the mean square dipolar fields, K^2 and K'^2, with the right-hand sides of (27) and (28).

Our results are also to be compared with the results of OII in which it was shown that for the two-phonon process the exact field dependence of the relaxation time for $g_\perp=0$ salts was identical to that predicted by Van Vleck on the basis of the effective-field approximation. This is to be expected since in the two-phonon process relaxation time, a factor of H^2, is averaged over a mean square field as contrasted with the factors of H^4 and H^2 that are averaged in the direct-process relaxation time. As Van Vleck[1] pointed out, the mean square dipolar field can be adjusted to yield the correct second moment, but then it does not necessarily yield the correct fourth (or higher) moments.

In view of the complexity of (26) it is evident that there is little to be gained from an evaluation of the direct-process relaxation time for rare earth salts not having $g_\perp=0$. We can say, however, that we would obtain in the most general expression for the inverse relaxation time terms of the form $(\sin k_0 r_{ij}/k_0 r_{ij})(E_\alpha-E_\beta)^4\langle\alpha|\Gamma_i|\beta\rangle\langle\beta|\Gamma_j^\dagger|\alpha\rangle$ $(i\ne j)$ which take into account correlated relaxation processes and whose importance has been emphasized in OII.

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[12] We will presently identify the mean square dipolar fields, K^2 and K'^2, introduced here, with the right-hand sides of (27) and (28).

V. NUMERICAL EVALUATION OF THE RELAXATION TIME FORMULAS

We now wish to consider the numerical evaluation of (31) and (33) having in mind a particular salt, dysprosium ethyl sulfate. For this salt the ground doublet and first excited doublet are given by[13]

$$0.964|\pm 9/2\rangle - 0.261|\mp 3/2\rangle - 0.053|\mp 15/2\rangle, \tag{39}$$

and

$$0.872|\pm 7/2\rangle - 0.489|\mp 5/2\rangle. \tag{40}$$

It is seen that only J_+ and J_- connect the first excited doublet with the ground doublet. Hence,

$$H_m{}^{(j)} = -10.2\beta^2\Lambda^2 \sin\theta_{jm} \cos\theta_{jm} e^{-i\varphi_{jm}}/r_{jm}{}^3. \tag{41}$$

In the evaluation of K^2 and K'^2 we use the results of Daniels[14] and Ketelaar[15] and find

$$K^2 = 2.02 \times 10^5 \text{ Oe}^2, \tag{42}$$

and

$$K'^2 = 1.86 \times 10^4 \text{ Oe}^2; \tag{43}$$

so that, in the effective field approximation for dysprosium ethyl sulfate,

$$\frac{1}{T_1} \simeq \frac{1}{T_{10}} \frac{(H^4 \cos^4\theta + 1.21 \times 10^6 H^2 \cos^2\theta + 1.23 \times 10^{11})(H^2 \sin^2\theta + 3.72 \times 10^4)}{H^4 \cos^2\theta \sin^2\theta (H^2 \cos^2\theta + 1.01 \times 10^5)}, \tag{44}$$

where H is measured in Oe.

Consideration of the crystal structure of the ethyl sulfates shows that, of the terms (36), and (37), and (38) that have been neglected in making the effective field approximation, only the first gives rise to a noticeable correction to the expression for the relaxation time. Making this correction, we find

$$\frac{1}{T_1} \simeq \frac{1}{T_{10}} \frac{(H^4 \cos^4\theta + 1.21 \times 10^6 H^2 \cos^2\theta + 1.11 \times 10^{11})(H^2 \sin^2\theta + 4.77 \times 10^4)}{H^4 \cos^2\theta \sin^2\theta (H^2 \cos^2\theta + 1.01 \times 10^5)}. \tag{45}$$

The size of the correction, 1.11×10^{11} Oe^4 instead of 1.23×10^{11} Oe^4, is an indication of the accuracy of the modified effective-field approximation in this case.

ACKNOWLEDGMENTS

APPENDIX

We now consider the evaluation of $\mathrm{tr} V'^4 \Gamma_j \Gamma_j{}^\dagger$. Since the trace over a single spin operator vanishes, we can separate the trace terms into two distinct groups,

$$\mathrm{tr}(V_z{}^4 + 6V_{\mathrm{dip}}{}^2 V_z{}^2 + V_{\mathrm{dip}}{}^4)[(\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j| V_{\mathrm{OL}}{}^{(i)}|\beta_j\rangle\langle\beta_j| V_{\mathrm{OL}}{}^{(i)}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)})$$

$$+ \sum_{r,s,k,m} J_z{}^{(k)} J_z{}^{(m)} Y_2{}^r(jk)^* Y_2{}^s(jm)^* a_{r0}{}^{ik} a_{s0}{}^{im} O_1{}^r(j)|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j| V_{\mathrm{OL}}{}^{(i)}|\beta_j\rangle\langle\beta_j| V_{\mathrm{OL}}{}^{(i)}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|O_1{}^s(j)], \tag{A1}$$

and

$$\mathrm{tr}(4V_z{}^3 V_{\mathrm{dip}} + 4V_z V_{\mathrm{dip}}{}^3)\{\sum_{r,m} a_{r0}{}^{im} Y_2{}^r(jm)^* J_z{}^{(m)}[(\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j| V_{\mathrm{OL}}{}^{(i)}|\beta_j\rangle\langle\beta_j| V_{\mathrm{OL}}{}^{(i)}|\tfrac{1}{2}q_j\rangle$$

$$\times\langle\tfrac{1}{2}q_j|O_1{}^r(j) + O_1{}^r(j)|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j| V_{\mathrm{OL}}{}^{(i)}|\beta_j\rangle\langle\beta_j| V_{\mathrm{OL}}{}^{(i)}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|\beta\Lambda\mathbf{H}\cdot\mathbf{J}^{(i)}]\}. \tag{A2}$$

We first evaluate the $V_z{}^4$ term in (A1):

$$\mathrm{tr} V_z{}^4 \Gamma_j \Gamma_j{}^\dagger = \alpha^4 \cos^4\theta (\beta\Lambda H)^6 |\langle\alpha_j| \hat{h}\cdot\mathbf{J}^{(i)}|\tfrac{1}{2}q_j\rangle|^2 |\langle\tfrac{1}{2}q_j| V_{\mathrm{OL}}{}^{(i)}|\beta_j\rangle|^2$$

$$+ \alpha^6 \cos^4\theta (\beta\Lambda H)^4 (\sum_m |H_m{}^{(i)}|^2)|\langle\tfrac{1}{2}q_j| V_{\mathrm{OL}}{}^{(i)}|\beta_j\rangle|^2 \tag{A3}$$

[13] M. J. D. Powell and R. Orbach, Proc. Phys. Soc. (London) 78, 753 (1961).
[14] J. M. Daniels, Proc. Phys. Soc. (London) 66, 673 (1953).
[15] J. A. A. Ketelaar, Physica 4, 619 (1937).

where θ is the angle between the crystal axis and the direction of the external magnetic field. The symbol $H_m{}^{(j)}$ is defined by (23).

Similarly, we observe

$$\mathrm{tr}6V_{\mathrm{dip}}{}^2V_z{}^2\Gamma_j\Gamma_j{}^\dagger = 6\alpha^6(\beta\Lambda H)^4\cos^2\theta\,|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle|^2|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_k Y_2{}^0(jk)^2(a_{00}{}^{jk})^2$$

$$+6\alpha^4(\beta\Lambda H)^2\cos^2\theta\sum_{k,m,n,p}\mathrm{tr}J_z{}^{(k)}J_z{}^{(m)}J_z{}^{(n)}J_z{}^{(p)}Y_2{}^0(jm)Y_2{}^0(jk)a_{00}{}^{jm}a_{00}{}^{jk}$$

$$\times\langle\alpha_j|\sum_r O_1{}^r(j)Y_2{}^r(jn)^*a_{r0}{}^{jn}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|\sum_s O_1{}^s(j)Y_2{}^s(jp)^*a_{s0}{}^{jp}|\alpha_j\rangle$$

$$=6\alpha^6(\beta\Lambda H)^4\cos^2\theta\,|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle|^2|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_k Y_2{}^0(jk)^2(a_{00}{}^{jk})^2$$

$$+6\alpha^8(\beta\Lambda H)^2\cos^2\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_k Y_2{}^0(jk)^2(a_{00}{}^{jk})^2|H_k{}^{(j)}|^2$$

$$+12\alpha^8(\beta\Lambda H)^2\cos^2\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)Y_2{}^0(jm)a_{00}{}^{jk}a_{00}{}^{jm}H_k{}^{(j)}H_m{}^{(j)*}$$

$$+6\alpha^8(\beta\Lambda H)^2\cos^2\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)^2(a_{00}{}^{jk})^2|H_m{}^{(j)}|^2. \quad (A4)$$

The prime on the summation symbol means that none of the indices that are summed over are equal.

Finally, we obtain

$$\mathrm{tr}V_{\mathrm{dip}}{}^4\Gamma_j\Gamma_j{}^\dagger = \alpha^4(\beta\Lambda H)^2|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle|^2|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m,n,p}\mathrm{tr}J_z{}^{(k)}J_z{}^{(m)}J_z{}^{(n)}J_z{}^{(p)}Y_2{}^0(jk)Y_2{}^0(jm)$$

$$\times Y_2{}^0(jn)Y_2{}^0(jp)a_{00}{}^{jk}a_{00}{}^{jm}a_{00}{}^{jn}a_{00}{}^{jp}+\alpha^4\sum_{k,m,n,p,q,t}\mathrm{tr}J_z{}^{(k)}J_z{}^{(m)}J_z{}^{(n)}J_z{}^{(p)}J_z{}^{(q)}J_z{}^{(t)}$$

$$\times|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2a_{00}{}^{jk}a_{00}{}^{jm}a_{00}{}^{jn}a_{00}{}^{jp}\langle\alpha_j|\sum_r O_1{}^r(j)Y_2{}^r(jq)^*a_{r0}{}^{jq}|\tfrac{1}{2}q_j\rangle$$

$$\times\langle\tfrac{1}{2}q_j|\sum_s O_1{}^s(j)Y_2{}^s(jt)^*a_{s0}{}^{jt}|\alpha_j\rangle$$

$$=\alpha^8(\beta\Lambda H)^2|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2|\langle\tfrac{1}{2}q_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\alpha_j\rangle|^2\sum_k Y_2{}^0(jk)^4(a_{00}{}^{jk})^4$$

$$+3\alpha^8(\beta\Lambda H)^2|\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle|^2|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)^2Y_2{}^0(jm)^2(a_{00}{}^{jk})^2(a_{00}{}^{jm})^2$$

$$+\alpha^{10}|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_k Y_2{}^0(jk)^4(a_{00}{}^{jk})^4|H_k{}^{(j)}|^2+\alpha^{10}|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)^4$$

$$\times(a_{00}{}^{jk})^4|H_m{}^{(j)}|^2+3\alpha^{10}|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m,n}{}' Y_2{}^0(jk)^2Y_2{}^0(jm)^2(a_{00}{}^{jk})^2(a_{00}{}^{jm})^2|H_n{}^{(j)}|^2$$

$$+6\alpha^{10}|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)^2Y_2{}^0(jm)^2(a_{00}{}^{jk})^2(a_{00}{}^{jm})^2|H_m{}^{(j)}|^2$$

$$+4\alpha^{10}|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)^3Y_2{}^0(jm)(a_{00}{}^{jk})^3a_{00}{}^{jm}[H_k{}^{(j)}H_m{}^{(j)*}+H_k{}^{(j)*}H_m{}^{(j)}]$$

$$+12\alpha^{10}|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m,n}{}' Y_2{}^0(jk)^2Y_2{}^0(jm)Y_2{}^0(jn)(a_{00}{}^{jk})^2a_{00}{}^{jm}a_{00}{}^{jn}H_m{}^{(j)}H_n{}^{(j)*}. \quad (A5)$$

We now evaluate (A2). We obtain for the first term

$$\mathrm{tr}4V_z{}^3V_{\mathrm{dip}}\Gamma_j\Gamma_j{}^\dagger = 4\alpha^6(\beta\Lambda H)^4\cos^3\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_k Y_2{}^0(jk)a_{00}{}^{jk}[\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle H_k{}^{(j)*}$$

$$+H_k{}^{(j)}\langle\tfrac{1}{2}q_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\alpha_j\rangle]. \quad (A6)$$

And we find for the second term

$$\mathrm{tr}4V_zV_{\mathrm{dip}}{}^3\Gamma_j\Gamma_j{}^\dagger = 4\alpha^4(\beta\Lambda H)^2\cos\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m,n,p}\mathrm{tr}J_z{}^{(k)}J_z{}^{(m)}J_z{}^{(n)}J_z{}^{(p)}Y_2{}^0(jk)Y_2{}^0(jm)Y_2{}^0(jn)$$

$$\times a_{00}{}^{jk}a_{00}{}^{jm}a_{00}{}^{jn}[\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|\sum_r O_1{}^r(j)Y_2{}^r(jp)^*a_{r0}{}^{jp}|\alpha_j\rangle$$

$$+\langle\alpha_j|\sum_r O_1{}^r(j)Y_2{}^r(jp)^*a_{r0}{}^{jp}|\tfrac{1}{2}q_j\rangle\langle\tfrac{1}{2}q_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\alpha_j\rangle]$$

$$=4\alpha^8(\beta\Lambda H)^2\cos\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_k Y_2{}^0(jk)^3(a_{00}{}^{jk})^3[\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle H_k{}^{(j)*}$$

$$+H_k{}^{(j)}\langle\tfrac{1}{2}q_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\alpha_j\rangle]+12\alpha^8(\beta\Lambda H)^2\cos\theta\,|\langle\tfrac{1}{2}q_j|\,V_{\mathrm{OL}}{}^{(j)}\,|\beta_j\rangle|^2\sum_{k,m}{}' Y_2{}^0(jk)^2Y_2{}^0(jm)$$

$$\times(a_{00}{}^{jk})^2a_{00}{}^{jm}[\langle\alpha_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\tfrac{1}{2}q_j\rangle H_m{}^{(j)*}+H_m{}^{(j)}\langle\tfrac{1}{2}q_j|\,\hat{h}\cdot\mathbf{J}^{(j)}\,|\alpha_j\rangle]. \quad (A7)$$

Summing (A3), (A4), (A5), (A6), and (A7) we immediately obtain (20).