not fit here since even pure $SrTiO_3$ crystals exhibit remanent polarization up to about 70°K, a temperature slightly below the cubic-tetragonal transition point (~80°K).

The dielectric properties of $SrTiO_3$ reported by other authors[16,19,24] have the same trend as ours, except for those of Smolenskii,[25] who observed a dielectric constant peak between 20° and 30°K on ceramic $SrTiO_3$. One possible explanation for this conflict seems to be that his sample was not very pure. Figures 5 and 12 show that a

[24] A. Linz, Jr., Phys. Rev. **91**, 753 (1953).
[25] G. A. Smolenskii, Izvest. Akad. Nauk S.S.S.R., Ser. Fiz. **20**, 149 (1956).

small addition of Ca^{2+} (for example) can change the dielectric properties of $SrTiO_3$ drastically.

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Spin Absorption of Solids*

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A study of the spin absorption lines of all orders was made. Rules for obtaining the absorption operator of any line were found. With magnetic dipole-dipole and exchange interactions taken into account, and with the assumption that the Zeeman energy is dominant, the zeroth and the second moments of the following four lines were evaluated for powders: (1) first Larmor line, parallel field, (2) second Larmor line, parallel field, (3) second Larmor line, perpendicular field, (4) low-frequency line, perpendicular field. The contribution of exchange to the second moment was found to be the same for the four cases treated, while the total intensity has the ratio 1:1:1:3/2. Agreement with existing experimental data is good.

I. INTRODUCTION

THE first Larmor line of a solid in a strong, constant, and perpendicular magnetic field, with the magnetic dipole-dipole and exchange interactions taken into account, was first studied by Van Vleck,[1] who calculated its shape function up to the fourth moment. Later, Wright[2] extended the moment method and applied it especially to low frequency lines. In recent years other satellite lines have also become of experimental interest.[3] Furthermore, in general, the Hamiltonian of a single molecule is not necessarily the Zeeman energy, and the method Wright developed cannot be applied directly. It is the purpose of this paper to cope with this situation. Rules will be given for obtaining moments of all lines, thus avoiding the elaborate derivation necessary in the previous method. The rules are

applicable to Hamiltonians of all kinds, provided that the perturbing part of the Hamiltonian is small enough for the perturbation method to be valid.

II. ABSORPTION OPERATOR

Let us assume that the Hamiltonian of a system is described by

$$\mathcal{3C} = \mathcal{3C}_0 + \mathcal{3C}', \tag{1}$$

where $\mathcal{3C}'$ is the perturbing potential small compared with $\mathcal{3C}_0$.

Let us represent $\mathcal{3C}'$ as an integral over its spectrum,

$$\mathcal{3C}' = \int_{-\infty}^{\infty} \mathcal{3C}'(\omega)d\omega, \tag{2}$$

where

$$[\mathcal{3C}_0, \mathcal{3C}'(\omega)] = \hbar\omega\mathcal{3C}'(\omega). \tag{3}$$

Under an extremely small perturbing potential $\mathfrak{M}(t)$, $\mathfrak{M}(t) \ll \mathcal{3C}'$, the probability of transition from one state to another, both eigenstates of $\mathcal{3C}$ when $t=0$, is propor-

[1] J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).
[2] A. Wright, Phys. Rev. **76**, 1826 (1949).
[3] A. G. Anderson, Phys. Rev. **115**, 863 (1959).

tional to the square of the matrix element

$$\int_0^T \langle \exp(-i\mathfrak{IC}T/\hbar)\psi_n | \exp[-i\mathfrak{IC}(T-t)/\hbar]\mathfrak{M}(t) \exp(-i\mathfrak{IC}t/\hbar) | \psi_m \rangle dt$$

$$= \left\langle \psi_n \left| \int_0^T \exp(i\mathfrak{IC}t/\hbar)\mathfrak{M}(t) \exp(-i\mathfrak{IC}t/\hbar)dt \right| \psi_m \right\rangle. \quad (4)$$

Physically, expression (4) says that the wave function, initially in state ψ_m, propagates to time t with the propagation factor $\exp(-i\mathfrak{IC}t/\hbar)$; there it is scattered by the potential $\mathfrak{M}(t)$, and then propagates to time T with the propagation factor $\exp[-i\mathfrak{IC}(T-t)/\hbar]$. The matrix element is then taken between this wave function and the wave function initially in state ψ_n, which is $\exp(-i\mathfrak{IC}T/\hbar)\psi_n$ at time T; and, since the system may be scattered at any time t in the time interval $(0,T)$, a time integral is taken from zero to T.

Now, we know that

$$\exp(i\mathfrak{IC}t/\hbar) = \exp[i(\mathfrak{IC}_0 + \mathfrak{IC}')t/\hbar]$$

$$= \left\{ 1 + \frac{i}{\hbar}\int_0^t \mathfrak{IC}'(t_1)dt_1 + \left(\frac{i}{\hbar}\right)^2 \int_0^t dt_1 \int_0^{t_1} \mathfrak{IC}(t_2)\mathfrak{IC}'(t_1)dt_2 + \cdots \right.$$

$$\left. + \left(\frac{i}{\hbar}\right)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} \mathfrak{IC}(t_n)\cdots\mathfrak{IC}'(t_2)\mathfrak{IC}'(t_1)dt_n + \cdots \right\} \exp(i\mathfrak{IC}_0 t/\hbar), \quad (5)$$

where

$$\mathfrak{IC}'(t) \equiv \exp(i\mathfrak{IC}_0 t/\hbar)\mathfrak{IC}' \exp(-i\mathfrak{IC}_0 t/\hbar), \quad (6)$$

and

$$\exp(-i\mathfrak{IC}t/\hbar) = \exp[-i(\mathfrak{IC}_0 + \mathfrak{IC}')t/\hbar]$$

$$= \exp(-i\mathfrak{IC}_0 t/\hbar)\left\{ 1 - \frac{i}{\hbar}\int_0^t \mathfrak{IC}'(t_1)dt_1 + \left(-\frac{i}{\hbar}\right)^2 \int_0^t dt_1 \int_0^{t_1} \mathfrak{IC}'(t_1)\mathfrak{IC}'(t_2)dt_2 + \cdots \right.$$

$$\left. + \left(-\frac{i}{\hbar}\right)^n \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} \mathfrak{IC}'(t_1)\mathfrak{IC}'(t_2)\cdots\mathfrak{IC}'(t_n)dt_n. \quad (7)$$

We have, from (5) and (7)

$$\int_0^T \exp(i\mathfrak{IC}t/\hbar)\mathfrak{M}(t) \exp(-i\mathfrak{IC}t/\hbar)dt$$

$$= \int_0^T \mathfrak{M}'(t)dt + \left(\frac{i}{\hbar}\right)\int_0^T dt \int_0^t [\mathfrak{IC}'(t_1),\mathfrak{M}'(t)]dt_1 + \left(\frac{i}{\hbar}\right)^2 \int_0^T dt \int_0^t dt_1 \int_0^{t_1} [\mathfrak{IC}'(t_2),[\mathfrak{IC}'(t_1),\mathfrak{M}'(t)]]dt_2 + \cdots$$

$$+ \left(\frac{i}{\hbar}\right)^n \int_0^T dt \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{n-1}} [\mathfrak{IC}'(t_n),\cdots[\mathfrak{IC}'(t_1),\mathfrak{M}'(t)]\cdots]dt_n + \cdots, \quad (8)$$

where

$$\mathfrak{M}'(t) = \exp(i\mathfrak{IC}_0 t/\hbar)\mathfrak{M}(t) \exp(-i\mathfrak{IC}_0 t/\hbar). \quad (9)$$

Let us decompose $\mathfrak{M}(t)$ into its Fourier components:

$$\mathfrak{M}(t) = \int_{-\infty}^{\infty} \mathfrak{M}_{\omega_0} \exp(-i\omega_0 t)d\omega_0, \quad (10)$$

and then further decompose $\mathfrak{M}_{\omega_0}$ into its spectrum:

$$\mathfrak{M}_{\omega_0} = \int_{-\infty}^{\infty} \mathfrak{M}_{\omega_0}(\omega)d\omega, \quad (11)$$

where

$$[\mathfrak{IC}_0,\mathfrak{M}_{\omega_0}(\omega)] = \hbar\omega\mathfrak{M}_{\omega_0}(\omega). \quad (12)$$

Then

$$\exp(i\mathcal{3C}_0 t/\hbar)\mathfrak{M}_{\omega 0}(\omega)\exp(-i\mathcal{3C}_0 t/\hbar)=e^{i\omega t}\mathfrak{M}_{\omega 0}(\omega).$$

We then have

$$\mathfrak{M}'(t)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathfrak{M}_{\omega 0}(\omega)e^{i(\omega-\omega 0)t}d\omega d\omega_0. \tag{13}$$

Similarly, from (2), (3), and (6), we have

$$\mathcal{3C}'(t)=\int_{-\infty}^{\infty}\mathcal{3C}'(\omega)e^{i\omega t}d\omega. \tag{14}$$

The nth term at the right side of (8) is therefore equal to

$$\left(\frac{i}{\hbar}\right)^{n}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\big[\mathcal{3C}'(\omega_n),\big[\mathcal{3C}'(\omega_{n-1}),\cdots\big[\mathcal{3C}'(\omega_1),\mathfrak{M}_{\omega 0}(\omega)\big]\cdots\big]\big]d\omega_n\cdots d\omega_1 d\omega_0 d\omega$$
$$\cdot\left\{\int_0^T dt\int_0^t dt_1\cdots\int_0^{t_{n-1}}\exp[i\omega_n t_n+\cdots+i\omega_1 t_1+i(\omega-\omega_0)t]dt_n\right\}. \tag{15}$$

The dominant term of (15) is equal to, as $T\rightarrow\infty$,

$$\left(\frac{i}{\hbar}\right)^{n}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\frac{\big[\mathcal{3C}'(\omega_n),\big[\mathcal{3C}'(\omega_{n-1}),\cdots\big[\mathcal{3C}'(\omega_1),\mathfrak{M}_{\omega 0}(\omega)\big]\cdots\big]\big]\pi\delta(\omega+\omega_1+\cdots+\omega_n-\omega_0)}{i^n(\omega_n)(\omega_n+\omega_{n-1})\cdots(\omega_n+\omega_{n-1}+\cdots+\omega_2+\omega_1)}d\omega d\omega_0\cdots d\omega_n$$

$$=\pi\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\frac{\big[\mathcal{3C}'(\omega_n),\big[\mathcal{3C}'(\omega_{n-1}),\cdots\big[\mathcal{3C}'(\omega_1),\mathfrak{M}_{\omega 0}(\omega)\big]\cdots\big]\big]\delta(\omega+\omega_1+\cdots+\omega_n-\omega_0)}{(\hbar\omega_n)(\hbar\omega_n+\hbar\omega_{n-1})\cdots(\hbar\omega_n+\hbar\omega_{n-1}+\cdots+\hbar\omega_2+\hbar\omega_1)}d\omega d\omega_0\cdots d\omega_n. \tag{16}$$

The physical meaning of (16) is clear: $\mathcal{3C}'(\omega_j)$ changes the system to a state the energy of which is higher by an amount of $\hbar\omega_j$ than that of the initial state; therefore, the successive applications of $\mathcal{3C}'(\omega_n)$, $\mathcal{3C}'(\omega_{n-1})\cdots\mathcal{3C}'(\omega_2)$, $\mathcal{3C}'(\omega_1)$, $\mathfrak{M}_{\omega 0}(\omega)$ change the system to a state with an energy higher by an amount of $\hbar(\omega_n+\omega_{n-1}+\cdots+\omega_2+\omega_1+\omega)$ than that of the initial state, and the law of the conservation of energy demands that this should be equal to the energy absorbed from $\mathfrak{M}(t)$ which is $\hbar\omega_0$. There are many alternative ways to apply a given group of $\mathcal{3C}'(\omega_j)$'s and $\mathfrak{M}_{\omega 0}(\omega)$, and (16) indicates that all should be taken into account, with the weighting factor

$$[(\hbar\omega_n)(\hbar\omega_n+\hbar\omega_{n-1})\cdots(\hbar\omega_n+\hbar\omega_{n-1}+\cdots+\hbar\omega_1)]^{-1},$$

for each.

In most cases, the spectrum of $\mathcal{3C}'$ is discrete; i.e.,

$$\mathcal{3C}'=\sum_{\alpha}\int_{-\infty}^{\infty}\mathcal{3C}'(\omega_\alpha)\delta(\omega-\omega_\alpha)d\omega=\sum_{\alpha}\mathcal{3C}'(\omega_\alpha). \tag{17}$$

Let us assume that the external field also has a discrete spectrum and further has a definite frequency; i.e.,

$$\mathfrak{M}(t)=e^{-i\omega_0 t}\sum_{\beta}\mathfrak{M}(\omega_\beta)=e^{-i\omega_0 t}\sum_{\beta}\int_{-\infty}^{\infty}\mathfrak{M}(\omega_\beta)\delta(\omega-\omega_\beta)d\omega. \tag{18}$$

With the omission of the factor π, expression (16) is then reduced to the form

$$\sum_{\alpha_n}\cdots\sum_{\alpha_1}\sum_{\beta}\frac{\big[\mathcal{3C}'(\omega_{\alpha_n}),\big[\mathcal{3C}'(\omega_{\alpha_{n-1}}),\cdots\big[\mathcal{3C}'(\omega_{\alpha_1}),\mathfrak{M}(\omega_\beta)\big]\cdots\big]\big]\delta(\omega_{\alpha_n}+\cdots+\omega_{\alpha_1}+\omega_\beta-\omega_0)}{(\hbar\omega_{\alpha_n})(\hbar\omega_{\alpha_n}+\hbar\omega_{\alpha_{n-1}})\cdots(\hbar\omega_{\alpha_n}+\hbar\omega_{\alpha_{n-1}}+\cdots+\hbar\omega_{\alpha_1})}. \tag{19}$$

With proper choice of the α_j's, expression (19) will account for the absorption of the line we are interested in, and henceforth expression (19), with the delta function omitted and with the correct α_j's chosen, will be called the

absorption operator of the line. Expression (8) is now reduced to, aside from a factor of π,

$$\sum_{\beta} \mathfrak{M}(\omega_\beta)\delta(\omega_\beta-\omega_0)+\sum_{\alpha 1}\sum_{\beta}\frac{[\mathfrak{K}'(\omega_{\alpha 1}),\mathfrak{M}(\omega_\beta)]}{\hbar\omega_{\alpha 1}}\delta(\omega_{\alpha 1}+\omega_\beta-\omega_0)$$

$$+\sum_{\alpha 1}\sum_{\alpha 2}\sum_{\beta}\frac{[\mathfrak{K}'(\omega_{\alpha 2}),[\mathfrak{K}'(\omega_{\alpha 1}),\mathfrak{M}(\omega_\beta)]]}{(\hbar\omega_{\alpha 2})(\hbar\omega_{\alpha 2}+\hbar\omega_{\alpha 1})}\delta(\omega_{\alpha 1}+\omega_{\alpha 2}+\omega_\beta-\omega_0)$$

$$+\cdots+\sum_{\alpha 1}\sum_{\alpha 2}\cdots\sum_{\alpha n}\sum_{\beta}\frac{[\mathfrak{K}'(\omega_{\alpha n}),[\mathfrak{K}'(\omega_{\alpha n-1}),\cdots[\mathfrak{K}'(\omega_{\alpha 1}),\mathfrak{M}(\omega_\beta)]\cdots]]}{(\hbar\omega_{\alpha n})(\hbar\omega_{\alpha n}+\hbar\omega_{\alpha n-1})\cdots(\hbar\omega_{\alpha n}+\cdots+\hbar\omega_{\alpha 1})}\delta(\omega_{\alpha n}+\cdots+\omega_{\alpha 1}+\omega_\beta-\omega_0)+\cdots. \quad (20)$$

The terms neglected in (15) give higher order corrections to the absorption operator, line shifts, and broadening of the absorption lines. We shall neglect the higher order corrections to the absorption operator, and take care of the broadening by the moment method of Van Vleck.[1]

As an illustration of the use of (19), let us consider a spin system in a strong, constant magnetic field, with magnetic dipole-dipole and exchange interactions between the spins. This is the case investigated by Van Vleck[1] and Wright.[2]

The Hamiltonian of this spin system is given as

$$\mathfrak{K}=\mathfrak{K}_0+\mathfrak{K}', \quad (21)$$

where

$$\mathfrak{K}_0=-Hg\beta\sum_i S_{zi}, \quad (22)$$

and

$$\mathfrak{K}'=\mathfrak{K}'(0)+\mathfrak{K}'(-2g\beta H\hbar^{-1})$$

$$+\mathfrak{K}'(2g\beta H\hbar^{-1})+\mathfrak{K}'(-g\beta H\hbar^{-1})+\mathfrak{K}'(g\beta H\hbar^{-1})$$

$$\equiv\mathfrak{K}_0'+\mathfrak{K}_{-2}'+\mathfrak{K}_2'+\mathfrak{K}_{-1}'+\mathfrak{K}_1',$$

$$\mathfrak{K}'(-2g\beta H\hbar^{-1})\equiv\mathfrak{K}_{-2}'=\sum_{i,j\neq}D_{ij}S_{+i}S_{+j},$$

$$\mathfrak{K}'(2g\beta H\hbar^{-1})\equiv\mathfrak{K}_2'=\sum_{i,j\neq}D_{ij}{}^*S_{-i}S_{-j}, \quad (23)$$

$$\mathfrak{K}'(-g\beta H\hbar^{-1})\equiv\mathfrak{K}_{-1}'=\sum_{i,j\neq}E_{ij}S_{+i}S_{zj},$$

$$\mathfrak{K}'(g\beta H\hbar^{-1})\equiv\mathfrak{K}_1'=\sum_{i,j\neq}E_{ij}{}^*S_{-i}S_{zj},$$

$$\mathfrak{K}'(0)\equiv\mathfrak{K}_0'=\sum_{i,j\neq}C_{ij}S_{zi}S_{zj}+\sum_{i,j\neq}A_{ij}S_{+i}S_{-j}.$$

One readily may prove that $[\mathfrak{K}_0,\mathfrak{K}'(\omega)]=\hbar\omega\mathfrak{K}'(\omega)$, and the definition here is therefore in agreement with (3). A physical way to see this is to notice that S_z, S_+, and S_- change the z component of the spin by the amount of $0, +1, -1$, respectively. Therefore, $\mathfrak{K}_0'$, $\mathfrak{K}_{-2}'$, $\mathfrak{K}_2'$, $\mathfrak{K}_{-1}'$, $\mathfrak{K}_1'$ change the z component of the spin of the system by the amount of $0, +2, -2, +1, -1$, respectively; hence the energy changes are $0, -2g\beta H$, $2g\beta H, -g\beta H, g\beta H$, respectively.

The coefficients $A, B, C, D,$ and E are defined as

$$A_{ij}=\tfrac{1}{2}[\tilde{A}_{ij}+\tfrac{1}{2}g^2\beta^2 r_{ij}{}^{-3}(3\gamma_{ij}{}^2-1)]=\tfrac{1}{2}(\tilde{A}_{ij}+\tfrac{1}{2}B_{ij}),$$

$$B_{ij}=g^2\beta^2 r_{ij}{}^{-3}(3\gamma_{ij}{}^2-1),$$

$$C_{ij}=\tfrac{1}{2}[\tilde{A}_{ij}-g^2\beta^2 r_{ij}{}^{-3}(3\gamma_{ij}{}^2-1)]=\tfrac{1}{2}(\tilde{A}_{ij}-B_{ij}), \quad (24)$$

$$D_{ij}=-3g^2\beta^2 r_{ij}{}^{-3}(\alpha_{ij}{}^2-\beta_{ij}{}^2-2i\alpha_{ij}\beta_{ij})/8,$$

$$E_{ij}=-\tfrac{3}{2}g^2\beta^2 r_{ij}{}^{-3}\gamma_{ij}(\alpha_{ij}-i\beta_{ij}),$$

where α_{ij}, β_{ij}, and γ_{ij} are the direction cosines of $\mathbf{r}_{ij}$ (see Fig. 1) and $\tilde{A}_{ij}$ is the exchange constant defined by Van Vleck.[1] The reader should notice that the Hamiltonian used here is the same as that used by Van Vleck and Wright, with a slightly different notation. The truncated Hamiltonian defined by Van Vleck is the part of the complete Hamiltonian which commutes with $\mathfrak{K}_0$, and is therefore the sum of $\mathfrak{K}_0$ and $\mathfrak{K}_0'$, so we have

$$\mathfrak{K}_{00}=\mathfrak{K}_0+\mathfrak{K}_0'. \quad (25)$$

The absorption lines centering at $g\beta H\hbar^{-1}$, $2g\beta H\hbar^{-2}$, and $3g\beta H\hbar^{-1}$ will henceforth be called the first, the second, and the third Larmor line, respectively. We will find the absorption operators for these lines when $\mathfrak{M}(t)$ is an oscillating magnetic field both in the x direction (perpendicular field) and in the z direction (parallel field).

For perpendicular oscillating field,

$$\mathfrak{M}(t)=e^{-i\omega_0 t}g\beta S_x=\tfrac{1}{2}e^{-i\omega_0 t}g\beta S_+ +\tfrac{1}{2}e^{-i\omega_0 t}g\beta S_-. \quad (26)$$

It is easy to identify $\tfrac{1}{2}g\beta S_+$ and $\tfrac{1}{2}g\beta S_-$ as $\mathfrak{M}(-g\beta H\hbar^{-1})$ and $\mathfrak{M}(g\beta H\hbar^{-1})$, respectively.

For the first Larmor line in a perpendicular field, $\omega_0\approx g\beta H\hbar^{-1}$, and the lowest nonvanishing term in (20),
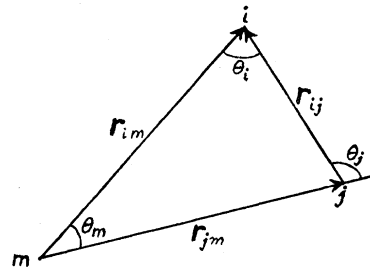

FIG. 1. Coordinate geometry used.

which is called the absorption operator O, is

$$O = \mathfrak{M}(g\beta H) = \tfrac{1}{2}g\beta S_-. \tag{27}$$

For the second Larmor line in a perpendicular field, the lowest nonvanishing term in (20) is

$$O = \frac{[\mathfrak{IC}_1', \mathfrak{M}(g\beta H)]}{g\beta H} = \frac{[\mathfrak{IC}_1', \tfrac{1}{2}g\beta S_-]}{g\beta H}. \tag{28}$$

For the third Larmor line in a perpendicular field, the lowest nonvanishing term in (20) is

$$O = \frac{[\mathfrak{IC}_1', [\mathfrak{IC}_1', \tfrac{1}{2}g\beta S_-]]}{(g\beta H)(2g\beta H)} + \frac{[\mathfrak{IC}_2', [\mathfrak{IC}_2', \tfrac{1}{2}g\beta S_+]]}{(2g\beta H)(4g\beta H)}$$
$$+ \frac{[\mathfrak{IC}_2', [\mathfrak{IC}_0', \tfrac{1}{2}g\beta S_-]]}{(2g\beta H)(2g\beta H)}. \tag{29}$$

For the low-frequency line in a perpendicular field, the lowest nonvanishing term in (20) is

$$O = \frac{[\mathfrak{IC}_1', \tfrac{1}{2}g\beta S_+]}{g\beta H} + \frac{[\mathfrak{IC}_{-1}', \tfrac{1}{2}g\beta S_-]}{-g\beta H}. \tag{30}$$

Next, let us consider the parallel field case, i.e.,

$$\mathfrak{M}(t) = e^{-i\omega_0 t} g\beta S_z. \tag{31}$$

It is easy to see that

$$g\beta S_z = \mathfrak{M}(0). \tag{32}$$

For the first Larmor line in a parallel field,

$$O = \frac{[\mathfrak{IC}_1', g\beta S_z]}{g\beta H} = \frac{\mathfrak{IC}_1'}{H}. \tag{33}$$

For the second Larmor line in a parallel field,

$$O = \frac{[\mathfrak{IC}_2', g\beta S_z]}{2g\beta H} = \frac{\mathfrak{IC}_2'}{H}. \tag{34}$$

For the third Larmor line in a parallel field,

$$O = \frac{[\mathfrak{IC}_1', [\mathfrak{IC}_2', g\beta S_z]]}{(g\beta H)(3g\beta H)} + \frac{[\mathfrak{IC}_2', [\mathfrak{IC}_1', g\beta S_z]]}{(2g\beta H)(3g\beta H)}$$
$$= \frac{g\beta[\mathfrak{IC}_1', \mathfrak{IC}_2']}{2(g\beta H)^2}. \tag{35}$$

For the low-frequency line in a parallel field,

$$O = \frac{[\mathfrak{IC}_1', [\mathfrak{IC}_{-1}', g\beta S_z]]}{(\hbar\omega_1)(\hbar\omega_1 + \hbar\omega_{-1})} + \frac{[\mathfrak{IC}_{-1}', [\mathfrak{IC}_1', g\beta S_z]]}{(\hbar\omega_1)(\hbar\omega_1 + \hbar\omega_{-1})}$$
$$+ \frac{[\mathfrak{IC}_2', [\mathfrak{IC}_{-2}', g\beta S_z]]}{(\hbar\omega_2)(\hbar\omega_2 + \hbar\omega_{-2})} + \frac{[\mathfrak{IC}_{-2}', [\mathfrak{IC}_2', g\beta S_z]]}{(\hbar\omega_{-2})(\hbar\omega_{-2} + \hbar\omega_2)}$$
$$= \frac{-g\beta[\mathfrak{IC}_1', \mathfrak{IC}_{-1}']}{(\hbar\omega_1)(\hbar\omega_{-1})} - \frac{2g\beta[\mathfrak{IC}_2', \mathfrak{IC}_{-2}']}{(\hbar\omega_2)(\hbar\omega_{-2})}$$
$$= \frac{g\beta[\mathfrak{IC}_1', \mathfrak{IC}_{-1}']}{(g\beta H)^2} + \frac{g\beta[\mathfrak{IC}_2', \mathfrak{IC}_{-2}']}{2(g\beta H)^2}. \tag{36}$$

As is well known, and included here only for the sake of completeness, the zeroth moment of a line is

$$\int_0^\infty f(\nu)d\nu = \sum_{i,j} |O_{ij}|^2 = \sum_{i,j} O_{ij}O_{ji}^+$$
$$= \mathrm{Tr}(OO^+). \tag{37}$$

where O^+ is the Hermitian conjugate of O.

In general, the $2n$th moment of the absorption line is

$$\langle \nu^{2n} \rangle = \frac{(-1)^n \mathrm{Tr}([\mathfrak{IC}_{00},[\mathfrak{IC}_{00},\cdots[\mathfrak{IC}_{00},O]\cdots]][\mathfrak{IC}_{00},[\mathfrak{IC}_{00},\cdots[\mathfrak{IC}_{00},O^+]\cdots]])}{\hbar^{2n}\,\mathrm{Tr}(OO^+)}, \tag{38}$$

where there are n $\mathfrak{IC}_{00}$'s in the above multiple commutation bracket.

Let us summarize the foregoing by formulating the rules for obtaining the absorption operator as follows:

(1) Choose the fewest number possible of $\mathfrak{IC}'(\omega_\alpha)$'s, the successive applications of which together with the chosen $\mathfrak{M}(\omega_\beta)$ will give an energy change equal to the energy of absorption of the line of interest. (If there are several ways of achieving this, all of them have to be taken into account.)

(2) The absorption operator is the sum of all the multiple commutators of the $\mathfrak{IC}(\omega_\alpha)$'s with $\mathfrak{M}(\omega_\beta)$, divided by the weighting factor $(\hbar\omega_{\alpha n})(\hbar\omega_{\alpha n} + \hbar\omega_{\alpha n-1})\cdots(\hbar\omega_{\alpha n} + \hbar\omega_{\alpha n-1} + \cdots + \hbar\omega_{\alpha 1})$:

$$O = \sum_{\alpha n}\cdots\sum_{\alpha 1}\sum_\beta \frac{[\mathfrak{IC}(\omega_{\alpha n}), [\mathfrak{IC}(\omega_{\alpha n-1}), \cdots[\mathfrak{IC}(\omega_{\alpha 1}), \mathfrak{M}(\omega_\beta)]\cdots]]}{(\hbar\omega_{\alpha n})(\hbar\omega_{\alpha n} + \hbar\omega_{\alpha n-1})\cdots(\hbar\omega_{\alpha n} + \cdots + \hbar\omega_{\alpha 1})}. \tag{39}$$

$$\omega_{\alpha n} + \cdots + \omega_{\alpha 1} + \omega_\beta = \omega_0$$

(3) The moments of a line can then be obtained by employing (38).

In the following four sections, moments of four absorption lines will be computed for a powder.

III. SECOND LARMOR LINE, PERPENDICULAR FIELD

The zeroth and the second moments of this line are given as

$$\int_0^\infty f(\nu)d\nu = \mathrm{Tr}([\mathfrak{IC}_1{}', g\beta S_-][g\beta S_+, \mathfrak{IC}_{-1}{}'])/4g^2\beta^2 H^2, \tag{40}$$

$$\langle \nu^2 \rangle = \frac{\mathrm{Tr}([\mathfrak{IC}_{00},[\mathfrak{IC}_1{}', g\beta S_-]][\mathfrak{IC}_{00},[H_{-1}{}', g\beta S_+]])}{h^2\,\mathrm{Tr}([\mathfrak{IC}_1{}', g\beta S_-][g\beta S_+, \mathfrak{IC}_{-1}{}'])}. \tag{41}$$

Now,

$$[\mathfrak{IC}_1{}', S_-] = -\sum_{i,j\neq} E_{ij}{}^* S_{-i}S_{-j}, \tag{42}$$

$$[\mathfrak{IC}_{00},[\mathfrak{IC}_1{}', S_-]] = 4\sum_{i,j,m\neq}(C_{ij}E_{jm}{}^* - A_{ij}E_{im}{}^*)S_{zi}S_{-j}S_{-m} + 4\sum_{i,j\neq}C_{ij}E_{ij}{}^*S_{zi}S_{-i}S_{-j} - 4\sum_{i,j\neq}A_{ij}E_{ij}{}^*S_{zi}S_{-j}{}^2$$

$$+2\sum_{i,j\neq}C_{ij}E_{ij}{}^*S_{-i}S_{-j} - 2Hg\beta\sum_{i,j\neq}E_{ij}{}^*S_{-i}S_{-j}, \tag{43}$$

and

$$\mathrm{Tr}([\mathfrak{IC}_{00},[\mathfrak{IC}_1{}', S_-]][\mathfrak{IC}_{00},[\mathfrak{IC}_{-1}{}', S_+]])$$

$$= 32H^2 g^2\beta^2 S^2(S+1)^2\sum_{i,j\neq}|E_{ij}|^2/9 + 32S^2(S+1)^2[2S(S+1)/3-\tfrac{1}{2}]$$

$$\times(\sum_{i,j\neq}(C_{ij}{}^2+2A_{ij}{}^2)|E_{ij}|^2)/15 + 64S^3(S+1)^3\sum_{i,j,m\neq}|C_{ij}E_{jm}{}^* - A_{ij}E_{im}{}^*|^2/27$$

$$+\frac{64S^3(S+1)^3}{27}\sum_{i,j,m\neq}(C_{ij}E_{jm}{}^* - A_{ij}E_{im}{}^*)(C_{im}E_{jm}-A_{im}E_{ij}). \tag{44}$$

The details of the calculation are omitted and only the result is given here[4]:

$$\langle \Delta\nu^2 \rangle = \left\{ \frac{[12S(S+1)-9]}{35}\sum_{i,j\neq}r_{ij}{}^{-12} + S(S+1)\left[\sum_{i,jm\neq,}r_{ij}{}^{-6}r_{im}{}^{-6}\left(\frac{1}{2}+\frac{8\cos^2\theta_i}{7}-\frac{15\cos^4\theta_i}{14}\right)\right.\right.$$

$$\left.\left. +\sum_{i,j,m\neq}r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\left(-\frac{8}{7}+2\cos^2\theta_m+\frac{8\cos^2\theta_i}{7}-\frac{6\cos^2\theta_i\cos^2\theta_j}{7}\right)\right]\right\}\bigg/(g^{-4}\beta^{-4}h^2\sum_{i,j\neq}r_{ij}{}^{-6})$$

$$+\{[1.2(S+1)-0.9]\sum_{i,j\neq}\tilde{A}_{ij}{}^2 r_{ij}{}^{-6} + S(S+1)\sum_{i,j,m\neq}\tilde{A}_{ij}\tilde{A}_{im}r_{im}{}^{-3}r_{ij}{}^{-3}(-\tfrac{1}{3}+\cos^2\theta_i) + 4S(S+1)\sum_{i,m,j\neq}\tilde{A}_{ij}{}^2 r_{mj}{}^{-6}/3$$

$$-S(S+1)\sum_{i,j,m\neq}\tilde{A}_{ij}{}^2 r_{im}{}^{-3}r_{jm}{}^{-3}(2\cos^2\theta_m-\tfrac{2}{3}) - S(S+1)\sum_{i,j,m\neq}\tilde{A}_{im}\tilde{A}_{ij}r_{jm}{}^{-3}r_{ij}{}^{-3}(2\cos^2\theta_j-\tfrac{2}{3})$$

$$+2S(S+1)\sum_{i,j,m\neq}\tilde{A}_{ij}\tilde{A}_{im}r_{mj}{}^{-6}/3\}/(h^2\sum_{i,j\neq}r_{ij}{}^{-6}). \tag{45}$$

The first bracket of the right-hand side of (45) is the dipole-dipole interaction contribution and the second bracket is the exchange contribution, and $\langle \Delta\nu^2 \rangle$ is the second moment about the mean frequency.

With the usual assumption that the exchange constant A_{ij} is zero except when i and j are neighbors, and is equal to A in that case, we obtain for a simple cubic lattice

$$\langle \Delta\nu^2 \rangle = \{1.08 - 0.038S^{-1}(S+1)^{-1} + [1.07 - 0.13S^{-1}(S+1)^{-1}]A^2 d^6\beta^{-4}g^{-4}\}\Delta\nu_0{}^2, \tag{46}$$

$$\int_0^\infty f(\nu)d\nu = 0.56S^2(S+1)^2 Ng^4\beta^4 d^{-6}H^{-2}, \tag{47}$$

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[4] H. Cheng, IBM Research Rept. RJ–189, San Jose, California, 1961 (unpublished). See also H. Cheng, thesis, California Institute of Technology, Pasadena, 1962, for the most complete treatment.

and for a body-centered cubic lattice,

$$\langle \Delta \nu^2 \rangle = \{1.13 - 0.03 S^{-1}(S+1)^{-1} + [1.14 - 0.08 S^{-1}(S+1)^{-1}] A^2 d^6 \beta^{-4} g^{-4}\} \Delta \nu_0^2, \tag{48}$$

$$\int_0^\infty f(\nu)d\nu = 0.82 S^2 (S+1)^2 N g^4 \beta^4 d^{-6} H^{-2}. \tag{49}$$

The term d is the distance to the nearest neighbors and $\Delta \nu_0^2$ is the second moment of the first Larmor line for perpendicular field,

$$\Delta \nu_0^2 = 3 g^4 \beta^4 h^{-2} S(S+1) \sum_i r_{ij}^{-6}/5. \tag{50}$$

For a simple cubic lattice,

$$\Delta \nu_0^2 = 5.04 g^4 \beta^4 \hbar^{-2} S(S+1) d^{-6}, \tag{51}$$

and for a body-centered-cubic lattice

$$\Delta \nu_0^2 = 7.37 d^4 \beta^4 h^{-2} S(S+1) d^{-6}. \tag{52}$$

It is interesting to note that the cross terms between the dipole-dipole energy and the exchange energy, or the terms linear in A, drop out after averaging over the whole solid angle.

Incidentally, the mean value of ν,

$$\langle \nu \rangle = \int_0^\infty \nu f(\nu)d\nu \Big/ \int_0^\infty f(\nu)d\nu$$

$$= \mathrm{Tr}([\mathcal{H}_{00},[\mathcal{H}_1',S_-]][S_+,\mathcal{H}_{-1}'])/h\ \mathrm{Tr}([\mathcal{H}_1',S_-][S_+,\mathcal{H}_{-1}']),$$

is calculated to be $2g\beta H h^{-1}$, and the absorption is truly of the second Larmor line.

IV. FIRST LARMOR LINE, PARALLEL FIELD

The zeroth and the second moments of this line are given as

$$\int_0^\infty f(\nu)d\nu = \mathrm{Tr}(\mathcal{H}_1'\mathcal{H}_{-1}')/H^2, \tag{53}$$

and

$$\langle \nu^2 \rangle = \mathrm{Tr}([\mathcal{H}_{00},\mathcal{H}_1'][\mathcal{H}_{-1}',\mathcal{H}_{00}])/\hbar^2\ \mathrm{Tr}(\mathcal{H}_1'\mathcal{H}_{-1}'). \tag{54}$$

After averaging with respect to the whole solid angle, we get

$$\langle \Delta \nu^2 \rangle = \left\{ \left[0.6 S(S+1) - \frac{9}{70} \right] \sum_{i,j\neq} r_{ij}^{-12} + S(S+1) \sum_{i,j,m\neq} (\tfrac{1}{2} - \cos^2\theta_i + \tfrac{3}{2}\cos^4\theta_i) r_{ij}^{-6} r_{im}^{-6} \right.$$

$$\left. + S(S+1) \sum_{i,j,m\neq} \left(\frac{1}{2}\cos^2\theta_m + \frac{2\cos^2\theta_i}{7} - \frac{2}{7} - \frac{3\cos^2\theta_i\cos^2\theta_j}{14} \right) r_{ij}^{-6} r_{im}^{-3} r_{jm}^{-3} \right\} (h^2 g^{-4}\beta^{-4} \sum_{i,j\neq} r_{ij}^{-6})^{-1} + \langle \Delta \nu_A^2 \rangle. \tag{55}$$

Here $\langle \Delta \nu_A^2 \rangle$ is the contribution of the exchange energy to the second moment of the second Larmor line for perpendicular field. This means that the contributions of exchange to the second moments for these two cases are equal.

For a simple cubic lattice,

$$\langle \Delta \nu^2 \rangle = \{0.90 - 0.02 S^{-1}(S+1)^{-1} + [1.07 - 0.13 S^{-1}(S+1)^{-1}] A^2 d^6 \beta^{-4} g^{-4}\} \Delta \nu_0^2, \tag{56}$$

$$\int_0^\infty f(\nu)d\nu = 0.56 S^2 (S+1)^2 N g^4 \beta^4 d^{-6} H^{-2}. \tag{57}$$

and for a bcc lattice

$$\langle \Delta \nu^2 \rangle = \{0.77 - 0.01 S^{-1}(S+1)^{-1} + [1.14 - 0.08 S^{-1}(S+1)^{-1}] A^2 d^6 \beta^{-4} g^{-4}\} \Delta \nu_0^2, \tag{58}$$

$$\int_0^\infty f(\nu)d\nu = 0.82 S^2 (S+1)^2 N g^4 \beta^4 d^{-6} H^{-2}. \tag{59}$$

Incidentally, $\langle \nu \rangle$ is evaluated to be $g\nu H h^{-1}$. Notice that the total intensity (57) and (59) are equal to those of the second Larmor line in a perpendicular field.

V. SECOND LARMOR LINE, PARALLEL FIELD

The zeroth and the second moments are given as

$$\int_0^\infty f(\nu)d\nu = \frac{\mathrm{Tr}([g\beta S_z,\mathfrak{IC}_2'][\mathfrak{IC}_{-2}',g\beta S_z])}{4g^2\beta^2 H^2} = H^{-2}\,\mathrm{Tr}(\mathfrak{IC}_2'\mathfrak{IC}_{-2}'), \tag{60}$$

$$\langle \nu^2 \rangle = \frac{\mathrm{Tr}([\mathfrak{IC}_{00},\mathfrak{IC}_2'][\mathfrak{IC}_{-2}',\mathfrak{IC}_{00}])}{h^2\,\mathrm{Tr}(\mathfrak{IC}_2'\mathfrak{IC}_{-2}')}. \tag{61}$$

Averaging over the whole solid angle, we get

$$\langle \Delta\nu^2 \rangle = S(S+1)g^4\beta^4 \Bigg\{ \sum_{i,j,m\neq} r_{ij}^{-6}r_{im}^{-6}\left(\frac{23}{28} - \frac{11\cos^2\theta_i}{14} + \frac{15\cos^4\theta_i}{28}\right)$$

$$+ \sum_{i,j,m\neq} r_{ij}^{-6}r_{im}^{-3}r_{jm}^{-3}\frac{(1+9\cos^2\theta_i\cos^2\theta_j - 10\cos^2\theta_i + 8\cos^2\theta_m)}{7} + [12-9S^{-1}(S+1)^{-1}]\sum_{i,j\neq} r_{ij}^{-12}/35 \Bigg\}$$

$$\times (h^2\sum_{i,j\neq} r_{ij}^{-6})^{-1} + \langle \Delta\nu_A{}^2 \rangle. \tag{62}$$

Again the exchange second moment is the same as that in the previous cases.

The total intensity, $\int_0^\infty f(\nu)d\nu$, is also equal to that in the previous cases, and the dipolar second moment, for simple cubic lattice, is

$$\langle \Delta\nu^2 \rangle = \{1.38 - 0.04S^{-1}(S+1)^{-1}$$
$$+ [1.07-0.13S^{-1}(S+1)^{-1}]A^2 d^6 g^{-4}\beta^{-4}\}\Delta\nu_0{}^2, \tag{63}$$

and for bcc lattice, is

$$\langle \Delta\nu^2 \rangle = \{1.39 - 0.03S^{-1}(S+1)^{-1}$$
$$+ [1.14-0.08S^{-1}(S+1)^{-1}]A^2 d^6 g^{-4}\beta^{-4}\}\Delta\nu_0{}^2. \tag{64}$$

VI. MOMENTS OF THE LOW FREQUENCY LINE IN A PERPENDICULAR FIELD

The absorption operator for this line is

$$O = \frac{[\mathfrak{IC}_1',\frac{1}{2}g\beta S_+]}{g\beta H} - \frac{[\mathfrak{IC}_{-1}',\frac{1}{2}g\beta S_-]}{g\beta H} \tag{65}$$

$$= (-\sum_{i,j\neq} F_{ij}S_{zi}S_{zj} + \frac{1}{2}\sum_{i,j\neq} F_{ij}S_{-i}S_{+j})/H,$$

where $F_{ij} = E_{ij} + E_{ij}{}^*$.

After the averaging process, the second moment for this line is obtained as

$$\langle \Delta\nu^2 \rangle = [3S(S+1)g^4\beta^4 \sum_{i,j,m\neq} r_{im}^{-6}r_{ij}^{-6}$$

$$\times (1+8\cos^2\theta_i - 9\cos^4\theta_i)/14](h^2\sum_{i,j\neq} r_{ij}^{-6})^{-1}$$

$$+ \langle \Delta\nu_A{}^2 \rangle. \tag{66}$$

Again $\langle \Delta\nu_A{}^2 \rangle$ is equal to that of the previous three cases.

For a simple cubic lattice, it is evaluated that

$$\int_0^\infty f(\nu)d\nu = 0.84S^2(S+1)^2 N g^4\beta^4 d^{-6}H^{-2}, \tag{67}$$

and

$$\langle \Delta\nu^2 \rangle = \{0.47 + [1.07-0.13S^{-1}(S+1)^{-1}]$$
$$\times A^2 d^6 g^{-4}\beta^{-4}\}\Delta\nu_0{}^2, \tag{68}$$

and for a bcc lattice, it is

$$\int_0^\infty f(\nu)d\nu = 1.23S^2(S+1)^2 N g^4\beta^4 d^{-6}H^{-2}, \tag{69}$$

and

$$\langle \Delta\nu^2 \rangle = \{0.64 + [1.14-0.08S^{-1}(S+1)^{-1}]$$
$$\times A^2 d^6 g^{-4}\beta^{-4}\}\Delta\nu_0{}^2. \tag{70}$$

APPENDIX

The following calculated results were used extensively in the paper. Expressions (8)–(25) give the mean values after averaging over the direction of the crystal axes with respect to the magnetic field, and therefore are useful for powders. The coordinates used are defined in Fig. 1.

$$\mathrm{Tr}(S_+S_+) = \mathrm{Tr}(S_-S_-) = \mathrm{Tr}(S_z S_- S_-)$$
$$= \mathrm{Tr}(S_z S_+ S_+) = 0, \tag{A1}$$

$$\mathrm{Tr}(S_+ S_-) = \tfrac{2}{3}S(S+1), \tag{A2}$$

$$\text{Tr}(S_+S_-S_z) = -\text{Tr}(S_-S_+S_z) = \tfrac{1}{3}S(S+1), \tag{A3}$$

$$\text{Tr}(S_+{}^2S_-{}^2) = 2S(S+1)[4S(S+1)-3]/15, \tag{A4}$$

$$\text{Tr}(S_+S_-S_+S_-) = 4S(S+1)[2S(S+1)+1]/15, \tag{A5}$$

$$\text{Tr}(S_z{}^2S_+S_-) = S(S+1)[2S(S+1)+1]/15, \tag{A6}$$

$$\text{Tr}(S_zS_-S_zS_+) = 2S(S+1)[S(S+1)-2]/15, \tag{A7}$$

$$\langle \gamma_{im}\gamma_{jm}\rangle_{\text{av}} = \tfrac{1}{3}\cos\theta_m, \tag{A8}$$

$$\langle \gamma_{im}\gamma_{ij}\gamma_{jm}\rangle_{\text{av}} = 0, \tag{A9}$$

$$\langle \gamma_{im}{}^2\gamma_{jm}{}^2\rangle_{\text{av}} = (1+2\cos^2\theta_m)/15, \tag{A10}$$

$$\langle \gamma_{im}{}^2\gamma_{ij}\gamma_{jm}\rangle_{\text{av}} = (\cos\theta_j+2\cos\theta_i\cos\theta_m)/15, \tag{A11}$$

$$\langle \gamma_{im}{}^2\gamma_{ij}{}^2\gamma_{jm}{}^2\rangle_{\text{av}}$$
$$= (2\cos^2\theta_i+2\cos^2\theta_j+2\cos^2\theta_m-1)/35, \tag{A12}$$

$$\langle \gamma_{im}{}^3\gamma_{jm}\rangle_{\text{av}} = \cos\theta_m/5, \tag{A13}$$

$$\langle \gamma_{im}{}^3\gamma_{ij}{}^2\gamma_{jm}\rangle_{\text{av}} = (2\cos^2\theta_i\cos\theta_m+\cos\theta_m$$
$$+2\cos\theta_i\cos\theta_j)/35, \tag{A14}$$

$$\langle \gamma_{im}{}^3\gamma_{jm}{}^3\rangle_{\text{av}} = (2\cos^3\theta_m+3\cos\theta_m)/35, \tag{A15}$$

$$\langle \gamma_{im}{}^4\gamma_{jm}{}^2\rangle_{\text{av}} = (1+4\cos^2\theta_m)/35, \tag{A16}$$

$$\langle \gamma_{im}{}^4\gamma_{ij}\gamma_{jm}\rangle_{\text{av}} = (\cos\theta_j+4\cos\theta_i\cos\theta_m)/35, \tag{A17}$$

$$\langle \gamma_{im}{}^4\gamma_{ij}{}^2\gamma_{jm}{}^2\rangle_{\text{av}} = (12\cos^2\theta_i+12\cos^2\theta_m+10\cos^2\theta_j$$
$$-7+8\cos^2\theta_i\cos^2\theta_m)/315, \tag{A18}$$

$$\langle \gamma_{im}{}^4\gamma_{jm}{}^4\rangle_{\text{av}} = (8\cos^4\theta_m+24\cos^2\theta_n+3)/315, \tag{A19}$$

$$\langle (\alpha_{ij}\beta_{jm}-\beta_{ij}\alpha_{jm})^2\rangle_{\text{av}} = \tfrac{1}{3}\sin^2\theta_j. \tag{A20}$$

$$\langle \gamma_{im}{}^2(\alpha_{ij}\beta_{jm}-\beta_{ij}\alpha_{jm})^2\rangle_{\text{av}} = (\sin^2\theta_j)/15, \tag{A21}$$

$$\langle \gamma_{im}{}^2(\alpha_{im}\beta_{jm}-\beta_{im}\alpha_{jm})^2\rangle_{\text{av}} = (\sin^2\theta_m)/15, \tag{A22}$$

$$\langle \gamma_{im}{}^4(\alpha_{ij}\beta_{jm}-\beta_{ij}\alpha_{im})^2\rangle_{\text{av}} = (\sin^2\theta_j)/35, \tag{A23}$$

$$\langle \gamma_{im}{}^2\gamma_{ij}{}^2(\alpha_{ij}\beta_{jm}-\beta_{ij}\alpha_{im})^2\rangle_{\text{av}}$$
$$= [(1+2\cos^2\theta_i)\sin^2\theta_j]/105, \tag{A24}$$

$$\langle \gamma_{im}{}^2\gamma_{ij}{}^2(\alpha_{ij}\beta_{im}-\beta_{ij}\alpha_{im})^2\rangle_{\text{av}}$$
$$= [(1+2\cos^2\theta_i)\sin^2\theta_i]/105. \tag{A25}$$

Four series over a simple cubic lattice were computed to within one percent of accuracy:

$$\sum_{i,j\neq} r_{ij}{}^{-6} = 8.4Nd^{-6}, \tag{A26}$$

$$\sum_{i,j\neq} r_{ij}{}^{-12} = 6.2Nd^{-12}, \tag{A27}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-6}\cos^2\theta_i = 17.32Nd^{-12}, \tag{A28}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-6}\cos^4\theta_i = 12.16Nd^{-12}, \tag{A29}$$

where N is the total number of lattice points in the lattice and d the lattice constant.

The following four series were obtained by summing all the terms from the lattice points within distance $2d$ from the origin. The error is admittedly large, and is estimated to be around 25%. However, in the calculation of this paper, the contribution from terms involving these sums tends to cancel, and as a result, the over-all accuracy is not greatly affected.

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\cos^2\theta_m = 14.74Nd^{-12}, \tag{A30}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\cos^2\theta_i = 10.37Nd^{-12}, \tag{A31}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\cos^2\theta_i\cos^2\theta_j = 3.17Nd^{-12}, \tag{A32}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3} = 29.9Nd^{-12}. \tag{A33}$$

The following four sums involving the exchange constant were evaluated. With the usual assumption that $\tilde{A}_{ij}$ is zero unless i and j are nearest neighbors, all the four sums were obtained exactly.

$$\sum_{i,j,m\neq} \tilde{A}_{ij}\tilde{A}_{im}r_{mj}{}^{-6} = 3.09A^2Nd^{-6}, \tag{A34}$$

$$\sum_{i,j,m\neq} \tilde{A}_{im}\tilde{A}_{ij}r_{mj}{}^{-3} = 9.23A^2Nd^{-3}, \tag{A35}$$

$$\sum_{i,j,m\neq} \tilde{A}_{im}\tilde{A}_{ij}r_{mj}{}^{-3}\cos^2\theta_j = 4.99A^2Nd^{-3}, \tag{A36}$$

$$\sum_{i,j,m\neq} \tilde{A}_{ij}{}^2 r_{im}{}^{-6} = 44.4NA^2d^{-6}. \tag{A37}$$

The following sum was also computed. Owing to the slowness of convergence the accuracy is not good, but again its contribution to the total sum is small and the over-all accuracy is not greatly affected,

$$\sum_{i,j,m\neq} \tilde{A}_{ij}{}^2 r_{im}{}^{-3}r_{jm}{}^{-3}(3\cos^2\theta_m-1) = 22.8NA^2d^{-6}. \tag{A38}$$

With the same accuracies discussed previously, the following series for a bcc lattice were also computed:

$$\sum_{i,j\neq} r_{ij}{}^{-6} = 12.29Nd^{-6}, \tag{A39}$$

$$\sum_{i,j\neq} r_{ij}{}^{-12} = 9.11Nd^{-12}, \tag{A40}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-6}\cos^2\theta_i = 41.2Nd^{-12}, \tag{A41}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-6}\cos^4\theta_i = 22.3Nd^{-12}, \tag{A42}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\cos^2\theta_m = 36.57Nd^{-12}, \tag{A43}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\cos^2\theta_i = 27.46Nd^{-12}, \tag{A44}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3}\cos^2\theta_i\cos^2\theta_j = 9.78Nd^{-12}, \tag{A45}$$

$$\sum_{i,j,m\neq} r_{ij}{}^{-6}r_{im}{}^{-3}r_{jm}{}^{-3} = 79.27Nd^{-12}, \tag{A46}$$

$$\sum_{i,j,m\neq} \tilde{A}_{ij}\tilde{A}_{im}r_{mj}{}^{-6} = 11.52NA^2d^{-6}, \tag{A47}$$

$$\sum_{i,j,m\neq} \tilde{A}_{im}\tilde{A}_{ij}r_{mj}{}^{-3} = 22.08NA^2d^{-3}, \tag{A48}$$

$$\sum_{i,j,m\neq} \tilde{A}_{im}\tilde{A}_{ij}r_{mj}{}^{-3}\cos^2\theta_j = 9.865NA^2d^{-3}, \tag{A49}$$

$$\sum_{i,j,m\neq} \tilde{A}_{ij}{}^2 r_{im}{}^{-6} = 90.32NA^2d^{-6}, \tag{A50}$$

$$\sum_{i,j,m\neq} \tilde{A}_{ij}{}^2 r_{im}{}^{-3}r_{jm}{}^{-3}(3\cos^2\theta_m-1) = 35.8NA^2d^{-6}. \tag{A51}$$