field on the basis of the decay-time behavior. Equation (B1) is convenient because it does not involve the geometry of the sample, but it is not rigorous for intensity, because the distribution of power between the front and back surfaces of the sample is dependent on kL and on the excitation distribution.

APPENDIX C: INTENSITIES OF ZEEMAN COMPONENTS

The variation of reabsorption effects with angle occurs largely through the amplitudes a_i in Table IV. The method of obtaining these values from Refs. 7 and 8 is outlined in the text and more fully described elsewhere.[21] The component notation is given in Fig. 3.

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[1]T. Holstein, Phys. Rev. 72, 1212 (1947). $T(\rho)$ and k_0 in this reference are written as $T_k(\rho)$ and k, respectively, in the present paper.

[2]J. Heber, K. H. Hellwege, and U. Köbler, Helv. Phys. Acta 41, 879 (1968).

[3]Charles W. Scherr, preceding paper, Phys. Rev. B 4, 3885 (1971).

[4]F. Varsanyi, D. L. Wood, and A. L. Schawlow, Phys. Rev. Letters 3, 544 (1959).

[5]Charles W. Scherr (private communication).

[6]*Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U.S. GPO, Washington, D.C., 1964), Appl. Math. Ser. 55.

[7]D. F. Nelson and M. D. Sturge, Phys. Rev. 137, A1117 (1965).

[8]S. Sugano and Y. Tanabe, J. Phys. Soc. Japan 13, 880 (1958).

[9]S. Sugano, A. L. Schawlow, and F. Varsanyi, Phys. Rev. 120, 2045 (1960), see Sec. III.

[10]S. Sugano and I. Tsujikawa, J. Phys. Soc. Japan 13, 899 (1958).

[11]J. P. Van der Ziel and N. Bloembergen, Phys. Rev. 138, A1287 (1965).

[12]E. O. Schulz-Du Bois, Bell System Tech. J. 38, 271 (1959).

[13]D. W. Posener, Australian J. Phys. 12, 184 (1959).

[14]G. F. Imbusch, W. M. Yen, A. L. Schawlow, G. E. Devlin, and J. P. Remeika, Phys. Rev. 136, A481 (1964).

[15]D. M. Dodd, D. L. Wood, and R. L. Barns, Phys. Rev. 35, 1183 (1964).

[16]B. B. Boiko, V. V. Valyavko, N. I. Insarova, and N. S. Petrov, Zh. Priklad. Spektrosk. (USSR) 11, 933 (1969).

[17]P. Kisliuk, N. C. Chang, P. L. Scott, and M. H. L. Pryce, Phys. Rev. 184, 367 (1969).

[18]M. J. Berggren, G. F. Imbusch, and P. L. Scott, Phys. Rev. 188, 675 (1969).

[19]G. F. Imbusch, Phys. Rev. 153, 326 (1967).

[20]R. J. Birgeneau, J. Chem. Phys. 50, 4282 (1969).

[21]N. S. K. Menon, dissertation (The University of Texas at Austin, 1971) (unpublished). (Available from University Microfilm in Ann Arbor, Mich.)

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Theoretical Magnetic Form Factors of Co^{++} and Fe^{++} in Their Monoxides

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The spherical magnetic form factors of Co^{++} in CoO and Fe^{++} in FeO are calculated, including the effect of unquenched orbital magnetic moment. Expansions relative to the "spin-only" case are found to be 11% for Co^{++} and 9% for Fe^{++}. For CoO this is a major step in the quantitative explanation of the 15–17% expansion of the experimental curve compared to the Freeman-Watson "spin-only" curve. For FeO no good experimental data are yet available for comparison.

I. INTRODUCTION

The spherical magnetic form factor of a transition-metal ion is a measure of the charge-density distribution of the unpaired electrons of the unfilled $3d$ shell, and as such is of great importance in the study of physical properties of these ions. Halpern and Johnson,[1] in their theory of scattering of neutrons by these ions, derived an expression for the magnetic form factor. Weiss and Freeman[2] calculated the effects of the nonspherical charge distribution of the $3d$ electrons in various crystalline fields and thus made it possible to isolate the spherically symmetric form factor. The assumption in both these cases has been that the orbital magnetic moment is completely quenched by the crystalline field, and its associated magnetic moment does not contribute to neutron scattering.

However, an examination of the g factors of the transition-metal ions in different salts shows that there may be sizable residual orbital moments present.[3] In Co^{++} and Fe^{++} this would be expected, since the orbital degeneracy of the ground state is not completely lifted by the cubic crystalline field.[4] In Ni^{++} the degeneracy is completely lifted, but the spin-orbit coupling causes the admixture of a higher

TABLE I. van Laar multispin model for CoO.

	Origin of the submotive	Direction cosines of spins α	β	γ
1.	000	−0.325	−0.325	+0.888
2.	$\frac{1}{4}0\frac{1}{4}$	+0.325	−0.325	−0.888
3.	$\frac{1}{4}\frac{3}{4}\frac{1}{4}$	+0.325	+0.325	+0.888
4.	$0\frac{3}{4}\frac{3}{4}$	−0.325	+0.325	−0.888

state into the ground state, giving rise to an orbital moment.

Recently the spherically symmetric form factor of Co^{++} in antiferromagnetic cobaltous oxide was determined experimentally by Khan and Erickson.[5] They found that the form factor is expanded by 15–17% compared to that calculated for the "spin-only" case following Halpern and Johnson. The present work is undertaken to determine the effect of orbital contribution to this expansion. Since the case of Fe^{++} is similar, this was also studied, though no suitable experimental data are available as yet for comparison.

Scattering by orbital moments has been considered previously by Trammell[6] and in an elaboration of his work by Odiot and Saint-James.[7] They were concerned with rare-earth ions, in which the orbital moments were completely unquenched and the effects of the crystalline fields are secondary. The transition-metal ion worked out in the framework of Trammell's theory is Ni^{++} in antiferromagnetic NiO. This was done by Blume,[8] who accounted for 4% of the 17% expansion of the experimental curve of Alperin[9] relative to the "spin-only" case as due to orbital effect. The case of NiO is simpler compared to that of CoO because of the nondegeneracy of the ground state and the uniaxiality of the spin structure in the antiferromagnetic state (Li's model A). In the present work Trammell's[6] expression for the orbital contribution to the neutron-ion interaction is used. The ground states of Co^{++} and Fe^{++} in their monoxides below the Néel temperature as determined by Kanamori[4] have also been useful.

II. SPIN STRUCTURES OF CoO AND FeO

Calculation of the theoretical magnetic form factor requires a knowledge of the spin structure in the crystal. The magnetic unit cell of CoO and FeO in the antiferromagnetic state, i.e., below the Néel temperature, is eight times the crystallographic unit cell and contains 32 magnetic ions. The magnetic structure (spin structure) of these crystals is such that each spin has antiparallel partners at vector distances $(0, 0, \frac{1}{2})$, $(0, \frac{1}{2}, 0)$, $(\frac{1}{2}, 0, 0)$, parallel partners at $(\frac{1}{2}, \frac{1}{2}, 0)$, $(\frac{1}{2}, 0, \frac{1}{2})$, $(0, \frac{1}{2}, \frac{1}{2})$, and an antiparallel one at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. If we call such an antiferromagnetic set of spins a submotive, the 32 spins in a magnetic unit cell form four submotives. Thus the spin structure can be described fully by giving the orientation of four spins each belonging to a different submotive. At present the most accepted model of the spin arrangement of CoO is the van Laar multispin model[10,11] as given in Table I and Fig. 1.

The spin structure of FeO is collinear as given by Li's model A[12] (Fig. 2), which is characterized by the fact that crystallographically unique (111) planes are ferromagnetic sheets, and the alternate (111) planes have parallel and antiparallel arrangement of their spins.

III. THEORY

The differential cross section for the elastic magnetic scattering of unpolarized neutrons into solid angle $d\Omega$ is[1,8]

$$\frac{d\sigma}{d\Omega}=\left(\frac{\gamma e^2}{mc^2}\right)^2\sum_{qq'}p_q\left|\sum_{\vec{n}}e^{i\vec{K}\cdot\vec{n}}\langle q'|\vec{T}_{\vec{n}}|q\rangle\right|^2, \qquad (1)$$

where $|q\rangle$ and $|q'\rangle$ are the initial and final states of the crystal assumed to have the same energy, $\vec{K}=\vec{k}-\vec{k}'$ is the difference between the initial and final wave vectors $\vec{k}$ and $\vec{k}'$, respectively, of the neutron, p_q is the probability that the state $|q\rangle$ is occupied, $\vec{n}$ is the lattice vector, and $\gamma=-1.91$ is the gyromagnetic ratio of neutron. $\vec{T}_{\vec{n}}$ represents the interaction of the neutron with the electrons of the ion at site $\vec{n}$, and is given by

$$\vec{T}_{\vec{n}}=\sum_j e^{i\vec{K}\cdot\vec{r}_j}\hat{K}\times(\vec{s}_j\times\hat{K})-\frac{i}{\hbar K}\sum_j e^{i\vec{K}\cdot\vec{r}_j}(\hat{K}\times\vec{p}_j), \qquad (2a)$$

where $\vec{s}_j$ and $\vec{p}_j$ are the spin and linear momentum of the jth electron, $\vec{r}_j$ is the position of the electron relative to the lattice point $\vec{n}$, $\hat{K}$ is the unit vector in the direction of $\vec{K}$, and the summation is over all electrons of the ion at the lattice site $\vec{n}$. It was shown by Trammell[6] that

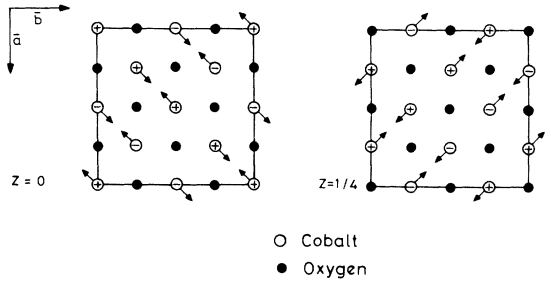


FIG. 1. Multispin-axis structure of CoO. The + and − signs designate the up and down directions into the paper of the z components of the spins. The arrows represent the projection of the spins on the ab plane. The layers $z=\frac{1}{2}$ and $z=\frac{3}{4}$ can be constructed by reversing the spin directions in the layers $z=0$ and $z=\frac{1}{4}$, respectively.

$$\vec{T}_{\vec{n}} = \hat{K} \times \vec{Q}_{\vec{n}} \times \hat{K} \ , \tag{2b}$$

where

$$\vec{Q}_{\vec{n}} = \sum_j \{ e^{i\vec{K}\cdot\vec{r}_j} \vec{s}_j + \tfrac{1}{4} [\vec{l}_j f(\vec{K}\cdot\vec{r}_j) + f(\vec{K}\cdot\vec{r}_j) \vec{l}_j] \} \ . \tag{3}$$

Equation (3) is exact for elastic scattering and holds to a good approximation for inelastic scattering as well:

$$f(\vec{K}\cdot\vec{r}_j) = 2 \left[\frac{d}{dx} \left(\frac{e^x - 1}{x} \right) \right]_{x = i\vec{K}\cdot\vec{r}_j} \ , \tag{4}$$

and $\vec{l}_j$ is the orbital angular momentum of the jth electron of the ion. It should be noted that

$$\lim f(\vec{K}\cdot\vec{r}_j) \to 1 \quad \text{as} \quad \vec{K} \to 0 \ . \tag{5}$$

Clearly, $\vec{Q}_{\vec{n}}$ is arbitrary by $\alpha\vec{K}$, where α is a constant.[13] The separation of the scattering cross section into form factor and structure factor is highly dependent on the choice of $\vec{Q}_{\vec{n}}$. The most convenient and physically important choice of $\vec{Q}_{\vec{n}}$ is (3), i.e., for $\alpha = 0$.

IV. DERIVATION OF FORM FACTOR

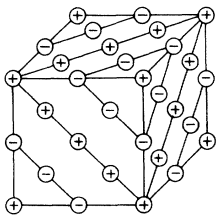
A. For Co^{++} in CoO

The derivation of the form factor requires a knowledge of the ground-state wave function of the magnetic ions in the crystal. Kanamori[4] determined the ground state of Co^{++} in CoO. He assumed the Li's model A for the spin structure and solved self-consistently for the lowest-energy state of the Hamiltonian

$$\mathcal{H}_0 = -2J_1 Z_1 \langle S \rangle S_z + \lambda' \vec{l}\cdot\vec{S} \tag{6}$$

in a triply degenerate ground state under the cubic crystalline field. $\vec{l}$ is the pseudo-angular-momentum in the crystalline field and $\vec{S}$ is the spin of the ground state of Co^{++}. Z_1 is the member of next nearest neighbors as the nearest neighbors's effect cancels out in the exchange term by virtue of an equal number of them being parallel and antiparallel; the Z axis is the fourfold symmetry axis of the crystal, which was also shown to be the spin direction. Thus, Kanamori's ground state for Co^{++} is

$$|\psi_{Co^{++}}\rangle = 0.875 \left|\tfrac{3}{2}, -1\right\rangle - 0.446 \left|\tfrac{1}{2}, 0\right\rangle + 0.188 \left|-\tfrac{1}{2}, 1\right\rangle \ , \tag{7}$$



Li's Model A

FIG. 2. Circles with a + sign represent the Fe^{++} ion with spin up. Circle with a − sign represent that with spin down.

where the notation $|M_S, M_l\rangle$ is used to specify the wave functions on the right-hand side ($S = \frac{3}{2}$, $l = 1$ is implied).

But, in view of the fact that the van Laar model has replaced Li's model A for the spin structure of CoO, we should check the validity of (7). We note that in van Laar's model the relative arrangement of next nearest neighbors is the same as in Li's model A; so the exchange term is unchanged provided the axis, i.e., the axis of quantization, is taken along the spin direction of the submotive. Also, since $\lambda' \vec{l}\cdot\vec{S}$ is isotropic,[4] the Hamiltonian (6) is unaltered. Hence, it follows that (7) can represent the ground state of the Co^{++} ions belonging to any submotive, with the axis of quantization along the spin direction of that ion. But (6) is not the complete Hamiltonian, as it neglects the interactions responsible for the relative orientation of submotives. Such an interaction, as calculated by Kanamori for Li's model A, is $-Cl_z^2$ ($C \approx 100\ \text{cm}^{-1}$), arising from the magnetostriction. The ground-state wave function (7) is changed due to this additional interaction as follows:

$$|\psi'_{Co^{++}}\rangle = 0.900 \left|\tfrac{3}{2}, -1\right\rangle - 0.401 \left|\tfrac{1}{2}, 0\right\rangle + 0.169 \left|-\tfrac{1}{2}, 1\right\rangle \ . \tag{8}$$

The notations are the same as used in (7). It is shown in Sec. V that the form factor is insensitive to this change in the ground-state wave function; hence the neglect of $-Cl_z^2$ in (6) is justified as far as the calculation of the form factor is concerned. Recently it was indicated by Bertaut[14] from symmetry considerations that, to some extent, the Dzialoshinski-Moriya interaction[15,16] $\vec{D}\cdot(\vec{S}_1 \times \vec{S}_2)$ is responsible for the orientation of the submotives in the van Laar model. But this interaction is expected to be only a few percent of the isotropic exchange interaction; while it may be effective in relative orientation of the submotive directions, one expects little change in the ionic wave functions except, of course, for the axis of quantization. The calculation of the ground-state wave functions of Co^{++} in CoO, taking in account all plausible interactions which give rise to the van Laar model, is a problem in itself. But, in view of the above arguments, it is certain that the ground state of the Hamiltonian (6), i.e., $|\psi_{Co^{++}}\rangle$, is accurate to a good approximation for the calculation of the spherical form factor. Therefore we shall use $|\psi_{Co^{++}}\rangle$ in the present work.

The pseudo-angular-momentum eigenfunctions in (7) can be transformed to ^4F- and ^4P-state functions of Co^{++},[17]

$$|\psi_{Co^{++}}\rangle = (1 - \beta^2)^{1/2} |{}^4F\rangle + \beta |{}^4P\rangle \ , \tag{9}$$

where

$$\beta = 0.185 \ ,$$

$$|{}^4F\rangle = 0.875(\sqrt{\tfrac{3}{8}}\,|M_S=\tfrac{3}{2},\ M_L=-1\rangle+\sqrt{\tfrac{5}{8}}\,|M_S=\tfrac{3}{2},\ M_L=3\rangle)-0.446\,|M_S=\tfrac{1}{2},\ M_L=0\rangle$$
$$+0.188(\sqrt{\tfrac{3}{8}}\,|M_S=-\tfrac{1}{2},\ M_L=1\rangle+\sqrt{\tfrac{5}{8}}\,|M_S=-\tfrac{1}{2},\ M_L=-3\rangle)\ ,$$

and

$$|{}^4P\rangle = 0.875\,|M_S=\tfrac{3}{2},\ M_L=-1\rangle - 0.446\,|M_S=\tfrac{1}{2},\ M_L=0\rangle + 0.188\,|M_S=-\tfrac{1}{2},\ M_L=1\rangle\ .$$

The notation used above is the same as in Ref. 17. If we have $|\psi_{Co^{++}}\rangle$, i.e., if (7) is in the crystal-fixed coordinate system whose z axis is along the four-fold symmetry axes of CoO, the wave function of the ith ion is given by

$$|q_i\rangle = \mathcal{R}_i\,|\psi_{Co^{++}}\rangle\ , \tag{10}$$

where $\mathcal{R}_i$ is the rotation operator which corresponds to the rotation of the crystal z axis to make it coincide with the direction of the magnetic moment of the ith ion.

To simplify (1) for the case of Co^{++} in CoO we write $\vec{r}_j = \vec{a}_j + \vec{r}_j{}'$, where $\vec{a}_j$ is the position of the nucleus of the jth magnetic ion in the magnetic unit cell and $\vec{r}_j{}'$ is the coordinate of the j'th electron of the jth atom with origin at $\vec{a}_j$. Since we are concerned with the elastic scattering, we have in (1), $|q\rangle = |q'\rangle$, $p_q = 1$, so $\sum_{q,q'} p_q$ drops out. We assume that $|q\rangle$, the wave function of the magnetic unit cell, can be written as a product of state vectors referring to the individual ions (Heitler-London approximation), i.e.,

$$|q\rangle = |q_1\rangle\,|q_2\rangle\cdots|q_{32}\rangle\ . \tag{11}$$

Now, it can be readily seen that all ions belonging to the same submotive give rise to identical contributions, which are simply added up. This is due to the fact that the negative sign arising from the reversal of the direction of the magnetic moment of ions[8] is counterpoised by another negative sign arising from the exponential factor which depends on the difference of their positions. For the orbital part of the magnetic interaction operator, the same exponential factor as in the spin part has been obtained by going back to (2a), which is converted to (2b) by taking out the exponential factor. Since each submotive consists of eight magnetic atoms in a unit cell, (1) can be simplified to yield

$$\frac{d\sigma}{d\Omega} = \left(\frac{8\gamma e^2}{mc^2}\right)^2 \sin^2\omega\,|\vec{Q}|_{\vec{K}=\vec{G}}{}^2\ , \tag{12}$$

where

$$(\vec{Q})_{\vec{K}=\vec{G}} = [\langle q_1|\vec{H}|q_1\rangle + \langle q_2|\vec{H}|q_2\rangle e^{2\pi i(h+l)/4} + \langle q_3|\vec{H}|q_3\rangle \times e^{2\pi i(h+3k+2l)/4} + \langle q_4|\vec{H}|q_4\rangle e^{2\pi i(3k+3l)/4}]_{\vec{K}=\vec{G}}\ , \tag{13}$$

$$\vec{H} = \sum_j \{e^{i\vec{K}\cdot\vec{r}_j}\vec{s}_j + \tfrac{1}{4}[\vec{l}_j f(\vec{K}\cdot\vec{r}_j) + f(\vec{K}\cdot\vec{r}_j)\vec{l}_j]\}\ . \tag{14}$$

j runs over all unpaired electrons of a magnetic ion. ω is the angle between $\vec{K}$ and $\vec{Q}$ at $\vec{K}=\vec{G}$, and $|q_1\rangle$, $|q_2\rangle$, $|q_3\rangle$, and $|q_4\rangle$ are the ground states of the ions belonging to the four submotives. $\vec{G}$ is the reciprocal lattice vector.

In the present case it is useful to keep the wave functions the same and transform $\vec{H}$ through similarity transformation by $\mathcal{R}_i$. Therefore, we have

$$\frac{d\sigma}{d\Omega} = \left(\frac{8\gamma e^2}{mc^2}\right)^2 \sin^2\omega\,|\langle\psi_{Co^{++}}|\,\mathcal{R}_1^{-1}\vec{H}\mathcal{R}_1 + \mathcal{R}_2^{-1}\vec{H}\mathcal{R}_2 e^{2\pi i(h+l)/4} + \mathcal{R}_3^{-1}\vec{H}\,\mathcal{R}_3 e^{2\pi i(h+3k+2l)/4} + \mathcal{R}_4^{-1}\vec{H}\,\mathcal{R}_4 e^{2\pi i(3k+3l)/4}|\psi_{Co^{++}}\rangle|^2\cdots\ . \tag{15}$$

Now, making use of the standard identity for the vector operator[18] $\vec{H}$, we have

$$\mathcal{R}^{-1}H_j\mathcal{R} = \sum_i R^{(3)}_{ji}H_i\ , \tag{16}$$

where $R^{(3)}$ is the corresponding rotation matrix in Cartesian space; it is shown in Appendix A that

$$\langle\psi_{Co^{++}}|\,\mathcal{R}_i^{-1}(\textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j}\vec{s}_j)\mathcal{R}_i\,|\psi_{Co^{++}}\rangle = \hat{k}_i\,\langle\psi_{Co^{++}}|\textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j}s_{jx}|\psi_{Co^{++}}\rangle\ . \tag{17}$$

We also have

$$\langle\psi_{Co^{++}}|\,\mathcal{R}_i^{-1}\{\tfrac{1}{4}\textstyle\sum_j[\vec{l}_j f(\vec{K}\cdot\vec{r}_j) + f(\vec{K}\cdot\vec{r}_j)\vec{l}_j]\}\mathcal{R}_i\,|\psi_{Co^{++}}\rangle = \hat{k}_i\,\langle\psi_{Co^{++}}|\,\tfrac{1}{4}\textstyle\sum_j(l_{jx}f_j + f_j l_{jx})\,|\psi_{Co^{++}}\rangle + \hat{k}_i'\langle\psi_{Co^{++}}|\,\tfrac{1}{4}\textstyle\sum_j(l_{jx}f_j + f_j l_{jx})\,|\psi_{Co^{++}}\rangle + \hat{k}_i''\langle\psi_{Co^{++}}|\,\tfrac{1}{4}\textstyle\sum_j(l_{jy}f_j + f_j l_{jy})\,|\psi_{Co^{++}}\rangle\ , \tag{18}$$

where $f_j \equiv f(\vec{K}\cdot\vec{r}_j)$, and where $\hat{k}_i$, $\hat{k}_i'$, and $\hat{k}_i''$ are the unit vectors in the direction of new z, x, and y axes, respectively.

We note the following points in connection with (18):

(i) $\hat{k}_i'$ and $\hat{k}_i''$ are not unique because there is an arbitrary rotation about $\hat{k}_i$ which will change $\hat{k}_i'$ and $\hat{k}_i''$.

(ii) The first term of (18) is predominant for small values of $|\vec{K}|$. For an extreme case, $\vec{K}=0$, only the first term exists.

Since

$$\langle\psi_{Co^{++}}|\tfrac{1}{4}\textstyle\sum_j(l_{jx}f_j+f_jl_{jx})|\psi_{Co^{++}}\rangle \tag{19}$$

and

$$\langle\psi_{Co^{++}}|\tfrac{1}{4}\textstyle\sum_j(l_{jy}f_j+f_jl_{jy})|\psi_{Co^{++}}\rangle \tag{20}$$

do not contain spherically symmetric parts (Appendix B), the spherical form factor which we aim at would not have any contribution from the second and third terms of (18). Thus, the omission of these terms does not affect the spherical form factor.

After dropping out the second and third terms of (18), we use (18) together with (17) to obtain the following expression for (15):

$$\left(\frac{8\gamma e^2}{mc^2}\right)^2\sin^2\omega\,|\langle\psi_{Co^{++}}|H_z|\psi_{Co^{++}}\rangle|^2\,|\hat{k}_1+\hat{k}_2e^{2\pi i(h+l)/4}+\hat{k}_3e^{2\pi i(h+3k+2l)/4}+\hat{k}_4e^{2\pi i(3k+3l)/4}|^2 . \tag{21}$$

Thus, starting with the fundamentals and giving proper consideration to the orbital contribution, we arrive at an expression for the differential cross section similar to (22) quoted below, which was used by van Laar[11] and Khan and Erickson[5] to analyze the experimental results for CoO.

We get

$$\frac{d\sigma}{d\Omega}=\left(\frac{8\gamma e^2}{mc^2}\right)^2\sin^2\omega\left(\frac{\mu_{Co^{++}}f_{Co^{++}}}{2}\right)^2|\,\hat{k}_1+\hat{k}_2e^{2\pi i(h+l)/4}+\hat{k}_3e^{2\pi i(h+3k+2l)/4}+\hat{k}_4e^{2\pi i(3k+3l)/4}|^2 , \tag{22}$$

where $\mu_{Co^{++}}$ and $f_{Co^{++}}$ are, respectively, the magnetic moment and magnetic form factor of Co^{++}. Equation (22) was obtained from the "spin-only" case by treating the spins classically and replacing the spin moment by the total magnetic moment of the ion. Comparing (22) with (21), we can identify the spherical form factor as follows:

$$(\mu^2_{Co^{++}}f^2_{Co^{++}})_{\text{sph. part}}=4\,|\langle\psi_{Co^{++}}|H_z|\psi_{Co^{++}}\rangle|^2_{\text{sph. part}} . \tag{23}$$

It should be noted that comparison of (21) with (22) is valid only for the spherical symmetric part of the form factor due to the approximations involved in neglecting (19) and (20). Taking the limits of both sides for $\vec{K}\to 0$, we get

$$\mu_{Co^{++}}=\langle\psi_{Co^{++}}|\textstyle\sum_j(2s_{jz}+l_{jz})|\psi_{Co^{++}}\rangle = 2\langle\psi_{Co^{++}}|H_z|\psi_{Co^{++}}\rangle_{\vec{K}=0} , \tag{24}$$

as $f_{Co^{++}}(0)=1$ by the normalization condition. Hence we have

$$|f_{Co^{++}}(\vec{K})|_{\text{sph.}}=\left|\frac{\langle\psi_{Co^{++}}|H_z|\psi_{Co^{++}}\rangle}{\langle\psi_{Co^{++}}|H_z|\psi_{Co^{++}}\rangle_{\vec{K}=0}}\right|_{\text{sph.}} . \tag{25}$$

B. For Fe^{++} in FeO

The ground state of Fe^{++} is[4]

$$|\psi_{Fe^{++}}\rangle=0.909\,|2,-1\rangle-0.395\,|1,0\rangle+0.135\,|0,1\rangle . \tag{26}$$

Only M_S and M_l values are specified for the kets on the right-hand side of (26); $S=2$, $l=1$ is implied. As before, the pseudo-angular-momentum eigenfunctions in (26) are transformed to ^5D-state functions of Fe^{++} to yield

$$|\psi_{Fe^{++}}\rangle=-0.909\,|M_S=2,\ M_L=1\rangle-(0.395/\sqrt{2})\times(|M_S=1,\ M_L=2\rangle-|M_S=1,\ M_L=-2\rangle)+0.135\,|M_S=0,\ M_L=-1\rangle . \tag{27}$$

The notation is similar to that used for $|\psi_{Co^{++}}\rangle$.

Proceeding as we did for Co^{++}, we can easily derive the form factor for Fe^{++} in FeO. Due to the collinear spin structure of FeO, the derivation of the form factor is less involved, as simply an additional negative sign occurs with the magnetic interaction operator $\vec{H}$ for the ions with antiparallel spins.[8] As a further consequence of collinearity of the spin structure, we do not need to make approximations similar to those involved in neglecting (19) and (20). Thus we obtain the following expression for the differential cross section:

$$\frac{d\sigma}{d\Omega}=\left(\frac{8\gamma e^2}{mc^2}\right)^2\sin^2\omega|\langle\psi_{Fe^{++}}|\vec{H}|\psi_{Fe^{++}}\rangle|^2\,|1-e^{2\pi i(h+l)/4}+e^{2\pi i(h+3k+2l)/4}-e^{2\pi i(3k+3l)/4}|^2 . \tag{28}$$

Adopting a procedure similar to the one which led to (22) for Co^{++}, we obtain for Fe^{++} the following expression for the differential cross section:

$$\frac{d\sigma}{d\Omega}=\left(\frac{8\gamma e^2}{mc^2}\right)^2\sin^2\omega\left(\frac{\mu_{Fe^{++}}f_{Fe^{++}}}{2}\right)^2|1-e^{2\pi i(h+l)/4}+e^{2\pi i(h+3k+2l)/4}-e^{2\pi i(3k+3l)/4}|^2 . \tag{29}$$

Comparing (28) and (29), we obtain

$$\mu_{Fe^{++}}f_{Fe^{++}}(\vec{K})=2|\langle\psi_{Fe^{++}}|\vec{H}|\psi_{Fe^{++}}\rangle| . \tag{30}$$

Since $f_{Fe^{++}}(0)=1$ by normalization, $\mu_{Fe^{++}}$ is given by $2|\langle\psi_{Fe^{++}}|\vec{H}|\psi_{Fe^{++}}\rangle|_{\vec{K}=0}$, which follows from the limit of (30) for $\vec{K}\to 0$. Hence, we have

$$f_{Fe^{++}}(\vec{K})=\frac{|\langle\psi_{Fe^{++}}|\vec{H}|\psi_{Fe^{++}}\rangle|}{|\langle\psi_{Fe^{++}}|\vec{H}|\psi_{Fe^{++}}\rangle|_{\vec{K}=0}} , \tag{31}$$

which is the complete form factor, unlike (25).

V. CALCULATION OF FORM FACTOR

A. For Co^{++} in CoO

We substitute in (25) the explicit form of $|\psi_{Co^{++}}\rangle$ and $\vec{H}$ from (9) and (14), respectively. Since $\vec{H}$ is the sum of one-electron operators, we must express $|\psi_{Co^{++}}\rangle$ in terms of one-electron wave func-

tions. There are three holes in the $3d$ shell of Co^{++}; $|\psi_{Co^{++}}\rangle$ is expressed in terms of these hole wave functions in Appendix C. Now, (25) is easily calculated and yields the following result:

$$f_{Co^{++}}(\vec{K}) = (1/1.746)[0.491\langle 2|e^{i\vec{K}\cdot\vec{r}}|2\rangle + 0.391\langle 1|e^{i\vec{K}\cdot\vec{r}}|1\rangle + 0.348\langle 0|e^{i\vec{K}\cdot\vec{r}}|0\rangle + 0.272\langle 2|f(\vec{K}\cdot\vec{r})|2\rangle + 0.244\langle 1|f(\vec{K}\cdot\vec{r})|1\rangle] . \quad (32)$$

Using the results of Appendix E for $\langle dm|e^{i\vec{K}\cdot\vec{r}}|dm\rangle$ and $\langle dm|f(\vec{K}\cdot\vec{r})|dm\rangle$, we have for (32),

$$f_{Co^{++}}(\vec{K}) = (1/1.746)[1.230\langle j_0\rangle + 0.516\langle g_0\rangle + \tfrac{2}{7}(5\pi)^{1/2}Y_{20}(\hat{K})(-0.105\langle j_2\rangle + 0.300\langle g_2\rangle) + \tfrac{2}{7}\pi^{1/2}Y_{40}(\hat{K})(1.011\langle j_4\rangle - 0.704\langle g_4\rangle)] . \quad (33)$$

To separate out the spherically symmetric part of $f_{Co^{++}}(\vec{K})$, we retain in (33) only the spherical symmetric part of $Y_{lm}(\hat{K})$ given in Appendix E. Thus, we have

$$f^s_{Co^{++}}(K) = (1/1.746)[1.230\langle j_0\rangle + 0.037\langle j_2\rangle + 0.162\langle j_4\rangle + 0.516\langle g_0\rangle - 0.107\langle g_2\rangle - 0.113\langle g_4\rangle]$$
$$= (1/1.746)[1.230\langle j_0\rangle + 0.037\langle j_2\rangle + 0.162\langle j_4\rangle + 0.516(\langle g_0\rangle - \tfrac{1}{2}\langle g_2\rangle) + 0.151(\langle g_2\rangle - \tfrac{3}{4}\langle g_4\rangle)] , \quad (34)$$

where

$$f^s_{Co^{++}}(K) \equiv [f_{Co^{++}}(\vec{K})]_{\text{sph. part}} . \quad (35)$$

Now, using the relation between $\langle g_L\rangle$ and $\langle j_L\rangle$,[13]

$$\langle g_{L-1}\rangle - \frac{L}{L+1}\langle g_{L+1}\rangle = \frac{2}{L+1}(\langle j_{L-1}\rangle + \langle j_{L+1}\rangle) , \quad (36)$$

(35) can be written in terms of $\langle j_L\rangle$ only:

$$f^s_{Co^{++}}(K) = \langle j_0\rangle + 0.360\langle j_2\rangle + 0.136\langle j_4\rangle . \quad (37)$$

$\langle j_0\rangle$, $\langle j_2\rangle$, and $\langle j_4\rangle$ are tabulated as functions of $(\sin\theta)/\lambda$ by Watson[19] [$K = (4\pi\sin\theta)/\lambda$]. Using Watson's $\langle j_L\rangle$ values for Co^{++}, $f^s_{Co^{++}}$ is plotted against $(\sin\theta)/\lambda$ in Fig. 3. It is found to be in agreement within 6% with the experimentally determined form factor for Co^{++}. The free-ion form factor $\langle j_0\rangle$ is also plotted for comparison. The spherically symmetric form factor calculated with $|\psi'_{Co^{++}}\rangle$, i.e., (8), as the ground-state wave function of Co^{++} is found to be

$$f^{s'}_{Co^{++}}(K) = \langle j_0\rangle + 0.362\langle j_2\rangle + 0.161\langle j_4\rangle , \quad (38)$$

which almost coincides with $f^s_{Co^{++}}(K)$. This endorses the arguments, given in Sec. III, in favor of the use of $|\psi_{Co^{++}}\rangle$.

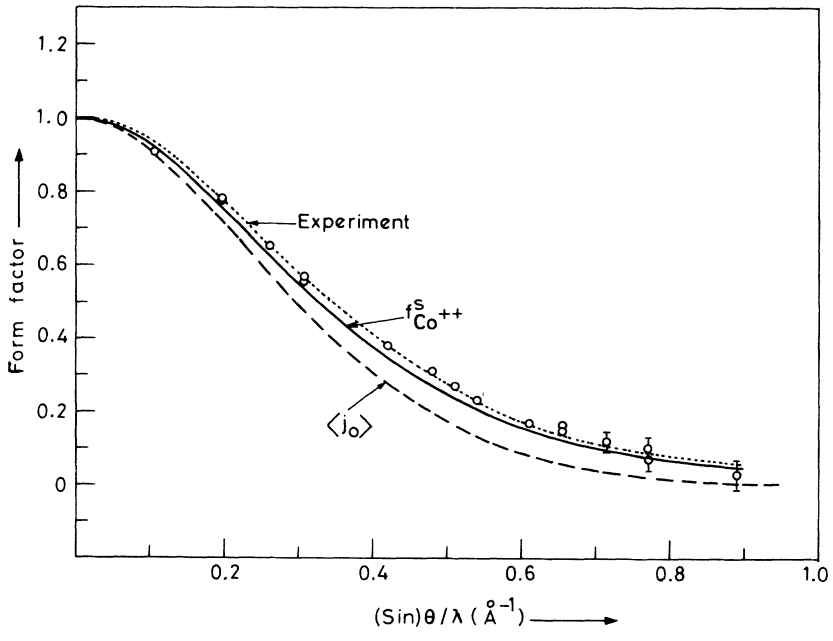


FIG. 3. Plot of the theoretically determined spherically symmetric magnetic form factor for Co^{++} vs $(\sin\theta)/\lambda$. For comparison the experimental form factor and the free-ion form factor $\langle j_0\rangle$ for Co^{++} are plotted.

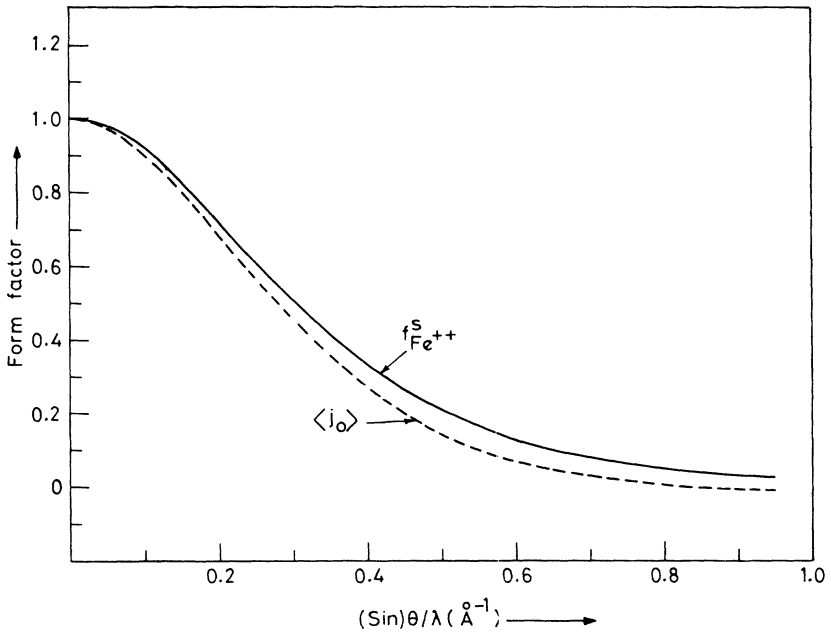


FIG. 4. Plot of theoretically predicted spherically symmetric magnetic form factor for Fe^{++} vs $(\sin\theta)/\lambda$. For comparison, the form factor for the free Fe^{++} ion, i.e., $\langle j_0 \rangle$, is plotted.

B. For Fe^{++} in FeO

There are four holes in the $3d$ shell of Fe^{++}. The ^5D functions occurring in (27) are expressed in terms of these four-hole wave functions in Appendix D. Since we are concerned with the spherically symmetric part of the form factor, we shall omit the $\langle \psi_{Fe^{++}} | H_x | \psi_{Fe^{++}} \rangle$ and $\langle \psi_{Fe^{++}} | H_y | \psi_{Fe^{++}} \rangle$ terms in (31) (Appendix B). Now by straightforward simplification we obtain

$$\frac{\langle \psi_{Fe^{++}} | H_z | \psi_{Fe^{++}} \rangle}{[\langle \psi_{Fe^{++}} | H_z | \psi_{Fe^{++}} \rangle]_{\vec{K}=0}} = \frac{1}{2.212}\{(0.909)^2[\langle 2|e^{i\vec{K}\cdot\vec{r}}|2\rangle + \tfrac{1}{2}\langle 1|e^{i\vec{K}\cdot\vec{r}}|1\rangle + \tfrac{1}{2}\langle 0|e^{i\vec{K}\cdot\vec{r}}|0\rangle + \tfrac{1}{2}\langle 1|f(\vec{K}\cdot\vec{r})|1\rangle] + (0.395)^2\langle 2|f(\vec{K}\cdot\vec{r})|2\rangle + (0.135)^2(-\tfrac{1}{2})\langle 1|f(\vec{K}\cdot\vec{r})|1\rangle\} . \quad (39)$$

Using the results of Appendix E, (39) becomes

$$(1/2.212)[1.652\langle j_0\rangle + 0.560\langle g_0\rangle + Y_{20}(\hat{K})\tfrac{1}{7}(5\pi)^{1/2}(0.826\langle j_2\rangle - 0.184\langle g_2\rangle) + Y_{40}(\hat{K})\tfrac{1}{7}\pi^{1/2}(3.305\langle j_4\rangle - 2.920\langle g_4\rangle)] . \quad (40)$$

Using the explicit forms of $Y_{20}(\hat{K})$ and $Y_{40}(\hat{K})$ in (40) from Appendix E, we can separate out the spherical form factor,

$$f^s_{Fe^{++}}(K) = (1/2.212)[1.652\langle j_0\rangle + 0.560\langle g_0\rangle - \tfrac{5}{28}(0.826\langle j_2\rangle - 0.184\langle g_2\rangle) + \tfrac{9}{112}(3.305\langle j_4\rangle - 2.920\langle g_4\rangle)] \quad (41)$$

$$= (1/2.212)[1.652\langle j_0\rangle - 0.147\langle j_2\rangle + 0.265\langle j_4\rangle + 0.560(\langle g_0\rangle - \tfrac{1}{2}\langle g_2\rangle) + 0.313(\langle g_2\rangle - \tfrac{3}{4}\langle g_4\rangle)] . \quad (42)$$

Now, using relation (36), we have for (42),

$$f^s_{Fe^{++}} = \langle j_0\rangle + 0.257\langle j_2\rangle + 0.190\langle j_4\rangle . \quad (43)$$

Using the values of $\langle j_0\rangle$, $\langle j_2\rangle$, and $\langle j_4\rangle$ as tabulated by Watson,[19] $f^s_{Fe^{++}}$ is plotted against $(\sin\theta)/\lambda$ in Fig. 4. For comparison the free-ion form factor $\langle j_0\rangle$ is also plotted. Unlike CoO, no experimental results of neutron scattering by vacancy-free FeO are available for comparison with the present theoretical form factor.

VI. CONCLUSION

As shown in Fig. 3, the 11% of the 15–17% expansion of the experimental form factor for Co^{++} in CoO from that of the free Co^{++} ion can be explained by inclusion of the orbital effect. There are two small corrections which must also be considered.

(a) The covalency effect which was studied by

Hubbard and Marshall[20] for Mn^{++}, Ni^{++}, etc., in the strong-field representation using the "spin-only" form factor. Such studies are difficult for CoO and FeO because of the degenerate ground state and sizable residual orbital moments.

(b) The spin polarization of fully occupied shells as discussed by Watson and Freeman.[21] It was found to expand the form factor of Ni^{++} by 4% relative to the "restricted" Hartree-Fock calculation. A similar expansion in the form factor of Co^{++} is expected.

Thus, the 4–6% discrepancy in the theoretical and experimental form factors for Co^{++} necessitates the quantitative estimation of the above-mentioned corrections, which is beyond the scope of the present work.

ACKNOWLEDGMENTS

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APPENDIX A

If the magnetic moment $\vec{\mu}_j$ of the jth ion is oriented at (θ_j, ϕ_j) with respect to the crystal-fixed coordinate system (Fig. 5), the corresponding rotation operator $\mathcal{R}_j$ is given by

$$\begin{aligned}\mathcal{R}_j &\equiv \mathcal{R}(\theta_j, \phi_j)\\ &= \mathcal{R}_{L'j}\mathcal{R}_{S'j}\\ &= e^{-i\phi_j L'_z} e^{-i\theta_j L'_y} e^{-i\phi_j S'_z} e^{-i\theta_j S'_y}\,, \end{aligned} \tag{A1}$$

where $\vec{L}' = \sum \vec{l}_i$ and $\vec{S}' = \sum \vec{s}_i$ with i running over all the electrons of the ion.

Clearly, $\mathcal{R}(\theta_j, \phi_j)$ is arbitrary by a rotation about $\vec{\mu}_j$. Now, the rotation matrix corresponding to (A1) in ordinary space is

$$R^{(3)}(\theta_j, \phi_j) = R^{(3)}_{L'}(\theta_j, \phi_j)\, R^{(3)}_{S'}(\theta_j, \phi_j)\,, \tag{A2}$$

where

$$R^{(3)}_{L'}(\theta_j, \phi_j) \equiv R^{(3)}_{L'j} = \begin{pmatrix} \cos\theta_j\cos\phi_j & -\sin\phi_j & \sin\theta_j\cos\phi_j \\ \cos\theta_j\sin\phi_j & \cos\phi_j & \sin\theta_j\sin\phi_j \\ -\sin\theta_j & 0 & \cos\theta_j \end{pmatrix} \tag{A3}$$

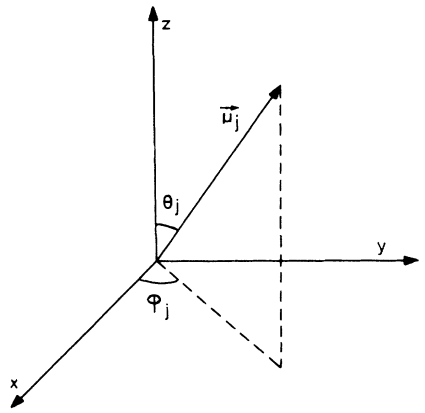

FIG. 5. Orientation of the magnetic moment of the jth ion in the crystal-fixed coordinate system; θ_j and ϕ_j are polar and azimuthal angles of the magnetic moment $\vec{\mu}_j$.

is the rotation matrix for orbital moments; a rotation matrix identical to (A3) can be written for spin moments as well.

Now, we have for the spin part of (16),

$$\begin{aligned}\mathcal{R}^{-1}_{S'j} s_x \mathcal{R}_{S'j} &= \textstyle\sum_i (R^{(3)}_{S'j})_{1i} s_i \\ &= s_x\cos\theta_j\cos\phi_j - s_y\sin\phi_j + s_z\sin\theta_j\cos\phi_j\,, \end{aligned} \tag{A4}$$

$$\begin{aligned}\mathcal{R}^{-1}_{S'j} s_y \mathcal{R}_{S'j} &= \textstyle\sum_i (R^{(3)}_{S'j})_{2i} s_i \\ &= s_x\cos\theta_j\sin\phi_j + s_y\cos\phi_j + s_z\sin\theta_j\sin\phi_j\,, \end{aligned} \tag{A5}$$

and

$$\begin{aligned}\mathcal{R}^{-1}_{S'j} s_z \mathcal{R}_{S'j} &= \textstyle\sum_i (R^{(3)}_{S'j})_{3i} s_i \\ &= -s_x\sin\theta_j + s_z\cos\theta_j \end{aligned} \tag{A6}$$

(where indices 1, 2, and 3 denote x, y, and z components, respectively), and similar expressions for $\mathcal{R}^{-1}_{L'j}\vec{l}\,\mathcal{R}_{L'j}$ can be written down. It can be readily seen by using the explicit form of $|\psi_{Co^{++}}\rangle$ given in Appendix C that

$$\begin{aligned}&\langle \psi_{Co^{++}} | \textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j} s_{jx} | \psi_{Co^{++}} \rangle \\ &\quad = \langle \psi_{Co^{++}} | \textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j} s_{jy} | \psi_{Co^{++}} \rangle = 0\,. \end{aligned} \tag{A7}$$

Now,

$$\begin{aligned}\langle q_i | \textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j} \vec{s}_j | q_i \rangle &= \langle \psi_{Co^{++}} | \textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j} \mathcal{R}_i^{-1} \vec{s}_j \mathcal{R}_i | \psi_{Co^{++}} \rangle \\ &= \langle \psi_{Co^{++}} | \textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j} (\hat{x}\, s_{jx}\sin\theta_i\cos\phi_i + \hat{y}\, s_{jx}\sin\theta_i\sin\phi_i + \hat{z}\, s_{jx}\cos\theta_i) | \psi_{Co^{++}} \rangle \\ &= \hat{k}_i \langle \psi_{Co^{++}} | \textstyle\sum_j e^{i\vec{K}\cdot\vec{r}_j} s_{jx} | \psi_{Co^{++}} \rangle\,, \qquad i = 1, 2, 3, 4 \end{aligned} \tag{A8}$$

where $\hat{k}_i$ is the unit vector in the direction of the magnetic moment of the ith ion. For the orbital part of $\vec{H}$, viz., $\frac{1}{4}\sum_j(\vec{l}_j f_j + f_j \vec{l}_j)$, we can proceed parallel to the spin part, but the difference is that

$$\langle \psi_{Co^{++}} | \tfrac{1}{4}\textstyle\sum_j (l_{jx} f_j + f_j l_{jx}) | \psi_{Co^{++}} \rangle \neq 0 \quad \text{and} \quad \langle \psi_{Co^{++}} | \tfrac{1}{4}\textstyle\sum_j (l_{jy} f_j + f_j l_{jy}) | \psi_{Co^{++}} \rangle \neq 0 \ .$$

So we get

$$\langle q_i | \textstyle\sum_j \frac{1}{4}(\vec{l}_j f_j + f_j \vec{l}_j) | q_i \rangle = \langle \psi_{Co^{++}} | \mathcal{R}_{L'j}^{-1} [\sum_j \tfrac{1}{4}(\vec{l}_j f_j + f_j \vec{l}_j)] \mathcal{R}_{L'j} | \psi_{Co^{++}} \rangle = \hat{k}_i \langle \psi_{Co^{++}} | \tfrac{1}{4}\textstyle\sum_j (l_{jz} f_j + f_j l_{jz}) | \psi_{Co^{++}} \rangle$$
$$+ \hat{k}_i' \langle \psi_{Co^{++}} | \tfrac{1}{4}\textstyle\sum_j (l_{jx} f_j + f_j l_{jx}) | \psi_{Co^{++}} \rangle + \hat{k}_i'' \langle \psi_{Co^{++}} | \tfrac{1}{4}\textstyle\sum_j (l_{jy} f_j + f_j l_{jy}) | \psi_{Co^{++}} \rangle \ , \qquad \text{(A9)}$$

where

$$\hat{k}_i = \hat{x}\sin\theta_i \cos\phi_i + \hat{y}\sin\theta_i \sin\phi_i + \hat{z}\cos\theta_i \ ,$$

$$\hat{k}_i' = \hat{x}\cos\theta_i \cos\phi_i + \hat{y}\cos\theta_i \sin\phi_i - \hat{z}\sin\theta_i \ ,$$

$$\hat{k}_i'' = -\hat{x}\sin\phi_i + \hat{y}\cos\phi_i \ ,$$

which are unit vectors in the direction of new z, x, and y axes, respectively.

APPENDIX B

$$\langle q_f | \tfrac{1}{4}\textstyle\sum_j (l_{jx} f_j + f_j l_{jx}) | q_f \rangle$$

and

$$\langle q_f | \tfrac{1}{4}\textstyle\sum_j (l_{jy} f_j + f_j l_{jy}) | q_f \rangle$$

(where $|q_f\rangle = |\psi_{Co^{++}}\rangle$ or $|q_f\rangle = |\psi_{Fe^{++}}\rangle$) will consist of the matrix elements of the following type:

$$\langle dm | f(\vec{K}\cdot\vec{r}) | dm' \rangle \ ,$$

with

$$|m - m'| = 1 \ . \qquad \text{(B1)}$$

It is obvious from (E1) that only $Y^*_{L\pm1}(\hat{K})$ ($L = 2$ and 4) will occur in (B1); and, as shown in Appendix E, these do not contribute to the spherical symmetric part.

APPENDIX C

Since $|\psi_{Co^{++}}\rangle$, i.e., Eq. (9), is made up of $|^4F\rangle$ and $|^4P\rangle$, we expand all the wave functions occurring in these states in terms of three-hole functions of the d shell as follows[22]:

^4F-state function:

$$|M_S = \tfrac{3}{2},\ M_L = 3\rangle = (2^+, 1^+, 0^+) \ ,$$

$$|M_S = \tfrac{3}{2},\ M_L = -1\rangle = (1/\sqrt{10})[(\sqrt{6})(1^+, 0^+, -2^+) + 2(2^+, -1^+, -2^+)] \ ,$$

$$|M_S = \tfrac{1}{2},\ M_L = 0\rangle = (1/\sqrt{15})\{(1^-, 0^+, -1^+) + (1^+, 0^-, -1^+) + (1^+, 0^+, -1^-) + 2[(2^-, 0^+, -2^+) + (2^+, 0^-, -2^+) + (2^+, 0^+, -2^-)]\} \ ,$$

$$|M_S = -\tfrac{1}{2},\ M_L = -3\rangle = (1/\sqrt{3})[(0^-, -1^-, -2^+) + (0^+, -1^-, -2^-) + (0^-, -1^+, -2^-)] \ ,$$

$$|M_S = -\tfrac{1}{2},\ M_L = 1\ \rangle = (1/\sqrt{30})\{(\sqrt{6})[(2^-, 0^-, -1^+) + (2^-, 0^+, -1^-) + (2^+, 0^-, -1^-)]$$
$$+ 2[(2^-, 1^-, -2^+) + (2^-, 1^+, -2^-) + (2^+, 1^-, -2^-)]\} \ ;$$

^4P-state functions:

$$|M_S = \tfrac{3}{2},\ M_L = -1\rangle = (1/\sqrt{10})[-2(1^+, -2^+, 0^+) + (\sqrt{6})(2^+, -2^+, -1^+)] \ ,$$

$$|M_S = \tfrac{1}{2},\ M_L = 0\rangle = (1/\sqrt{15})\{-2[(1^+, -1^-, 0^+) + (1^-, -1^+, 0^+)$$
$$+ (1^+, -1^+, 0^-)] + (2^-, -2^+, 0^+) + (2^+, -2^-, 0^+) + (2^+, -2^+, 0^-)\} \ ,$$

$$|M_S = -\tfrac{1}{2},\ M_L = 1\rangle = (1/\sqrt{30})\{-2[(2^-, -1^-, 0^+) + (2^+, -1^-, 0^-)$$
$$+ (2^-, -1^+, 0^-)] + (\sqrt{6})[(2^-, -2^-, 1^+) + (2^-, -2^+, 1^-) + (2^+, -2^-, 1^-)]\} \ .$$

Since $l = 2$ and $s = \frac{1}{2}$ for each single-particle wave function, only m_l and m_s are specified. + and − superscripts over m_l indicate $m_s = \frac{1}{2}$ and $-\frac{1}{2}$, respectively. The parentheses denote the antisymmetric product function, i.e., the Slater determinant made up of the single-particle functions given inside it.

APPENDIX D

To express $|\psi_{Fe^{++}}\rangle$, i.e., Eq. (27), in terms of single-particle wave functions (there are four holes in the $3d$ shell of Fe^{++}) we need the expansion of ^5D states as follows[22]:

$$|M_S = 2,\ M_L = -1\rangle = (2^+,\ 0^+,\ -2^+,\ -1^+)\ ,$$

$$|M_S = 1,\ M_L = 2\rangle = \tfrac{1}{2}[(2^-,\ 1^+,\ -1^+,\ 0^+) + (2^+,\ 1^-,\ -1^+,\ 0^+) + (2^+,\ 1^+,\ -1^-,\ 0^+) + (2^+,\ 1^+,\ -1^+,\ 0^-)]\ ,$$

$$|M_S = 1,\ M_L = -2\rangle = \tfrac{1}{2}[(1^-,\ 0^+,\ -2^+,\ -1^+) + (1^+,\ 0^-,\ -2^+,\ -1^+) + (1^+,\ 0^+,\ -2^-,\ -1^+) + (1^+,\ 0^+,\ -2^+,\ -1^-)]\ ,$$

$$|M_S = 0,\ M_L = 1\rangle = (1/\sqrt{6})[(2^-,\ 1^-,\ -2^+,\ 0^+) + (2^-,\ 1^+,\ -2^-,\ 0^+) + (2^-,\ 1^+,\ -2^+,\ 0^-) + (2^+,\ 1^-,\ -2^-,\ 0^+) + (2^+,\ 1^-,\ -2^+,\ 0^-) + (2^+,\ 1^+,\ -2^-,\ 0^-)]\ .$$

The notation is the same as in Appendix C.

APPENDIX E

We have the following relation[8,2]:

$$\langle dm|\,\Phi(\vec{K}\cdot\vec{r})\,|dm'\rangle = 4\pi \sum_{L,M} i^L \langle h_L\rangle\, Y^*_{LM}(\hat{K}) \int Y^*_{2m}(\hat{r})\, Y_{LM}(\hat{r})\, Y_{2m'}(\hat{r})\, d\Omega$$

$$= \sum_L 2[(2L+1)\pi]^{1/2} i^L \langle h_L\rangle\, C^L(2, m; 2, m')\, Y^*_{L m-m'}(\hat{K})\ , \tag{E1}$$

where $\langle h_L\rangle = \int_0^\infty R^2(r)\, h_L(Kr)\, r^2\, dr$, and the coefficients $C^L(l,\ m;\ l',\ m')$ are tabulated by Condon and Shortley[22] [nonzero terms in (E1) are for $L = 0,\ 2,$ and 4 only]. If $\Phi(\vec{K}\cdot\vec{r}) = e^{i\vec{K}\cdot\vec{r}}$, $h_L(Kr)$ is the spherical Bessel function $j_L(Kr)$, while for $\Phi(\vec{K}\cdot\vec{r}) = f(\vec{K}\cdot\vec{r})$, $h_L(Kr) = g_L(Kr)$, where $g_L(Kr) = \frac{1}{2} i^{-L} \int_{-1}^{+1} f(Kr\mu) \times P_L(\mu)\, d\mu$, and $P_L(\mu)$ are Legendre polynomials.

The following matrix elements are needed:

$$\langle 2, 2|\,\Phi(\vec{K}\cdot\vec{r})\,|2, 2\rangle$$

$$= \langle 2, -2|\,\Phi(\vec{K}\cdot\vec{r})\,|2, -2\rangle$$

$$= \langle h_0\rangle + \tfrac{4}{7}(5\pi)^{1/2} Y_{20}(\hat{K})\langle h_2\rangle + \tfrac{2}{7}\pi^{1/2} Y_{40}(\hat{K})\langle h_4\rangle\ , \tag{E2}$$

$$\langle 2, 1|\,\Phi(\vec{K}\cdot\vec{r})\,|2, 1\rangle$$

$$= \langle 2, -1|\,\Phi(\vec{K}\cdot\vec{r})\,|2, -1\rangle$$

$$= \langle h_0\rangle - \tfrac{2}{7}(5\pi)^{1/2} Y_{20}(\hat{K})\langle h_2\rangle - \tfrac{8}{7}\pi^{1/2} Y_{40}(\hat{K})\langle h_4\rangle\ , \tag{E3}$$

$$\langle 2, 0|\,\Phi(\vec{K}\cdot\vec{r})\,|2, 0\rangle$$

$$= \langle h_0\rangle - \tfrac{4}{7}(5\pi)^{1/2} Y_{20}(\hat{K})\langle h_2\rangle + \tfrac{4}{7}(9\pi)^{1/2} Y_{40}(\hat{K})\langle h_4\rangle\ . \tag{E4}$$

We have used the fact that $Y_{20}(\hat{K})$ and $Y_{40}(\hat{K})$ are real. We also have[23]

$$Y_{00}(\hat{K}) = \left(\frac{1}{4\pi}\right)^{1/2}\ , \tag{E5}$$

$$Y_{20}(\hat{K}) = \left(\frac{5}{4\pi}\right)^{1/2}\left(\frac{1}{4}\right)^{1/2} \frac{3K_z^2 - K^2}{K^2}\ , \tag{E6}$$

$$Y_{40}(\hat{K}) = \left(\frac{9}{4\pi}\right)^{1/2}\left(\frac{1}{64}\right)^{1/2} \frac{35K_z^4 - 30K_z^2K^2 + 3K^4}{K^4}\ , \tag{E7}$$

$$Y_{2\pm1}(\hat{K}) = \mp\left(\frac{15}{8\pi}\right)^{1/2} \frac{(K_x \pm iK_y)K_z}{K^2}, \tag{E8}$$

$$Y_{4\pm1}(\hat{K}) = \mp\left(\frac{9}{4\pi}\right)^{1/2}\left(\frac{5}{16}\right)^{1/2} \times\left(\frac{K_x \pm iK_y}{K}\right)\left(\frac{7K_z^3 - 3K^2K_z}{K^3}\right). \tag{E9}$$

Clearly, the spherical parts of (E5), (E6), and (E7) are as follows:

$$[Y_{00}(\hat{K})]_{\text{sph.}} = (1/4\pi)^{1/2},$$

$$[Y_{20}(\hat{K})]_{\text{sph.}} = -\tfrac{1}{4}(5/\pi)^{1/2},$$

$$[Y_{40}(\hat{K})]_{\text{sph.}} = 9/16\pi^{1/2},$$

while the spherical part of (E8) and (E9) is zero.

[1]O. Halpern and M. H. Johnson, Phys. Rev. 55, 898 (1939).

[2]R. J. Weiss and A. J. Freeman, J. Chem. Phys. Solids 10, 147 (1959).

[3]W. Low, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1960), Suppl. 2.

[4]J. Kanamori, Progr. Theoret. Phys. (Kyoto) 17, 177 (1957).

[5]D. C. Khan and R. A. Erickson, Phys. Rev. B 1, 2243 (1970).

[6]G. T. Trammell, Phys. Rev. 92, 1387 (1953).

[7]S. Odiot and D. Saint-James, J. Phys. Chem. Solids 17, 117 (1960).

[8]M. Blume, Phys. Rev. 124, 96 (1961).

[9]H. Alperin, Phys. Rev. Letters 6, 55 (1961).

[10]B. van Laar, Phys. Rev. 138, A584 (1965).

[11]B. van Laar, Ph. D. thesis (University of Leiden, 1968) (unpublished).

[12]Y. Y. Li, Phys. Rev. 100, 627 (1955).

[13]S. W. Lovesey, J. Phys. C 2, 470 (1969).

[14]E. F. Bertaut, J. Phys. Chem. Solids 30, 763 (1969).

[15]I. Dzialoshinski, J. Phys. Chem. Solids 4, 241 (1958).

[16]T. Moriya, Phys. Rev. 120, 91 (1960).

[17]S. W. Lovesey and D. E. Rimmer, Rept. Progr. Phys. 32, 333 (1969).

[18]E. Merzbacher, *Quantum Mechanics* (Wiley, New York, 1962).

[19]R. E. Watson and A. J. Freeman, Acta Cryst. 14, 27 (1961).

[20]J. Hubbard and W. Marshall, Proc. Phys. Soc. (London) 86, 561 (1965).

[21]R. E. Watson and A. J. Freeman, Phys. Rev. 120, 1125 (1960).

[22]E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge U.P., New York, 1935).

[23]C. J. Ballhausen, *Introduction to Ligand Field Theory* (McGraw-Hill, New York, 1962).