

Electron Spin Density and Internal Conversion Process

Masato MORITA, Kenzo SUGIMOTO, Masami YAMADA*
and Yoshimatsu YOKOO

Department of Physics, Osaka University, Toyonaka, Osaka

**Science and Engineering Research Laboratory, Waseda University, Tokyo*

(Received November 8, 1968)

We have studied a possibility to explore the spin density of orbital electrons by utilizing the spin dependence of internal conversion processes. We have calculated the relative intensity of internal conversion electrons for the case where the radial wave functions of the orbital electrons are different for spin up and spin down. As a particular example, we have studied the internal conversion process in the decay of the 14.4 keV level of Fe^{57} , using the radial wave functions of the orbital electrons of Fe obtained in the unrestricted Hartree-Fock method which have a reasonable fit to the experimental data on the internal magnetic field. It is shown that the L_I and M_I conversion electrons distribute almost isotropically, and the intensity of conversion electrons should change by about 0.5% for L_I and 2% for M_I for different spin polarizations.

§ 1. Introduction

Fermi and Segè showed¹⁾ that atomic hyperfine fields arise from the interaction of the magnetic moment of the nucleus with electronic spin and orbital moments. The electron spin interaction is also divided into two parts: the Fermi contact interaction and the dipolar coupling term. The internal magnetic field due to the Fermi contact interaction is given by

$$H_i = - (16\pi/3) \mu_B [\rho_{\uparrow}(0) - \rho_{\downarrow}(0)],$$

where μ_B is the Bohr magneton, $\rho_{\uparrow}(0)$ is the electron density with spin up per unit volume at nucleus, $\rho_{\downarrow}(0)$ is the electron density with spin down. Originally, the density at the nucleus of an outer unpaired s electron is considered to be responsible for the observed effective magnetic field. $\text{Mn}^{++}(3d^5, 4s^0)$ has no unpaired $4s$ electrons. Therefore no hyperfine field from the contact interaction is expected to exist. Experimentally, the internal field is rather high²⁾ and it is -650 kgauss. Since $\rho(0)$ has nonzero values for s electrons only, the nonvanishing values of $[\rho_{\uparrow}(0) - \rho_{\downarrow}(0)]$ may originate from combinations of $1s$, $2s$ and $3s$ electrons. The origins of spin polarization of the s electrons in the closed shells are, in the case of the transition elements, the exchange forces between the unpaired $3d$ electrons and these s electrons. The Coulomb repulsion between the $3d$ and s electrons is weaker for parallel spins than antiparallel spins. Therefore, if their spins are parallel, the s electron is attracted to the $3d$ shell and the electron density with antiparallel spin is dominant at the nucleus,

The theory of the atomic hyperfine fields is a very popular problem and has been studied by many authors. Among them, Watson and Freeman³⁾ calculated the Fermi contact interaction with the Hartree-Fock method. For example, the contribution from the core electrons to the internal field in Mn^{++} is estimated to be -690 kgauss in agreement with the observed -650 kgauss.^{2),3)}

We have studied a possibility to explore the spin density of orbital electrons by utilizing the internal conversion processes which depend on the relative spin polarization between nucleus and electron—the spin dependence of conversion process.

Our problem starts from the assumption that the s electrons of the closed shell are spin-polarized relative to the nuclear spin. That is, if the radial wave functions of the s electrons are different for two states with spin up and down, there must be certain observable effects in the internal conversion processes. Since we can measure the conversion electrons from different shells separately, we should also find the spin polarization of each shell. In § 2, we demonstrate that the relative intensity of conversion electron is suitable for our purpose. As a particular example, we study, in § 3, the internal conversion process in the decay of the 14.4 keV level of Fe^{57} , since the state decays dominantly through this process and the radial wave functions of the orbital electrons of Fe are given by Watson and Freeman and by Bagus and Liu.⁴⁾ It is shown that the conversion electrons distribute almost isotropically in this case, and the intensity of conversion electrons changes a few percent for different spin polarization.

§ 2. Spin-polarization dependence in relative intensity of conversion electrons

The conversion processes depend on the relative spin polarization between nucleus and electron. Therefore, it is easily understood that the probability of the conversion process is different for two spin states of electrons, if the nuclear spins are polarized, if the radial wave functions for electrons with spin up and down are not equal, and if we can measure the conversion probability of the orbital electrons with spin up and down separately.

In order to simplify the discussion, we assume that 1) the conversion process is a magnetic dipole transition of the nuclear decay scheme, $1^+ \rightarrow 0^+$, 2) the initial nuclear state is completely polarized*) ($J=M=1$) and the orbital electrons are in the ns state, 3) the conversion electron is emitted in the s state and the effect of the d state is neglected. From these assumptions, the conservation law of the magnetic quantum number requires a relation,

$$1 - 0 = m_s^{\text{final}} - m_s^{\text{initial}}, \quad (1)$$

where m_s is the magnetic quantum number of the intrinsic electronic spin which

*) The z -axis of the quantization is chosen to be the direction of the nuclear spin \mathbf{J} .

takes only two values, $\pm 1/2$. Equation (1) is satisfied with the unique solution,

$$m_s^{\text{final}} = 1/2 \quad \text{and} \quad m_s^{\text{initial}} = -1/2. \quad (2)$$

That is, in this particular decay scheme, the conversion process can proceed only if the spin of the orbital electron is antiparallel to the nuclear spin \mathbf{J} .

Now, let us suppose the transition metal like iron, and let us apply a magnetizing field \mathbf{H} in the direction of \mathbf{J} . The total spin of the orbital d electrons is antiparallel to \mathbf{J} and \mathbf{H} . In this geometry, the conversion takes place from the ns electron whose spin is parallel to that of the d electrons. (This is denoted by $\tau' = \uparrow$ in § 3.) We call this the *case I*. On the other hand, if we apply \mathbf{H} in the opposite direction of \mathbf{J} , the conversion takes place from the ns electron whose spin is antiparallel to that of the d electrons. (This is denoted by $\tau' = \downarrow$ in § 3.) We call this the *case II*. The radial wave functions for $\tau' = \uparrow$ and \downarrow are different in the unrestricted Hartree-Fock method so that the intensities, $W_I(\theta)$ and $W_{II}(\theta)$, of conversion electrons in these two cases are different. Here θ is the angle between the nuclear spin \mathbf{J} and the direction of the emission of the electron. The magnitude of $[W_I(\theta) - W_{II}(\theta)]/W_I(\theta)$ becomes a measure of the difference of the radial wave functions of the ns electrons with different spin polarizations.

The nuclear polarization can be achieved in several ways; e.g. in the Mössbauer-type experiments, in the nuclear reactions, or in the beta decays. In the next section, we will study $W(\theta)$ for a particular example of Fe^{57} .

§ 3. Internal conversion process in Fe^{57}

In § 2, we have discussed the relative intensity of the conversion electrons corresponding to the nuclear de-excitation, $J=M=1 \rightarrow J=M=0$ with no parity change. Since the orbital electrons are in the ns states, the conversion electrons are allowed only in the states with^{*)} $\kappa = -1(s_{1/2})$ and $\kappa = 2(d_{3/2})$. The angular distribution function (or relative intensity including angular dependence) for this transition is given by the function $F_L^M(\theta)$ with $L=M=1$, from Eq. (44) of a paper by Rose, Biedenharn and Arfken.⁶⁾ The explicit form of $F_1^1(\theta)$ is obtained as Eq. (3) below.

Now we study the decay of the 14.4 keV level of Fe^{57} through the internal conversion process. The conversion coefficient is known to be $\alpha = 9.7$ in this case. We choose a subtransition, $J=3/2^+, M=3/2 \rightarrow J=1/2^+, M=1/2$ which is equivalent to 100% polarization of the 14.4 keV state. The angular distribution of the conversion electrons is again given by $F_1^1(\theta)$ with $\kappa = -1$ and 2. We denote it as $W(\theta)$. We have directly^{**)}

^{*)} The quantum number κ specifies the values of l and j of the Dirac particle, simultaneously.⁵⁾

^{**)} The units, $\hbar = c = m = 1$, are used throughout this paper. Here m is the electron mass.

$$W(\theta) \equiv F_1^1(\theta) = 3|R_{\kappa=2, ns\uparrow}|^2(1 - P_2) + 8|R_{\kappa=-1, ns\downarrow}|^2 + 8 \operatorname{Re} \{ \exp(i(\Delta_{\kappa=-1} - \Delta_{\kappa=2})) R_{\kappa=2, ns\downarrow}^* R_{\kappa=-1, ns\downarrow} \} P_2 + |R_{\kappa=2, ns\downarrow}|^2(1 + P_2) \quad (3)$$

with

$$P_2 = (1/2)(3 \cos^2 \theta - 1), \quad (4)$$

$$R_{\kappa, ns\tau} = \int_0^\infty [F_\kappa(pr) G_{ns\tau'}(r) + G_\kappa(pr) F_{ns\tau'}(r)] h_1^{(1)}(kr) dr, \quad (5)$$

$$G = rg \quad \text{and} \quad F = rf. \quad (6)$$

Here g and f are large and small components of the radial wave function of the electron. The conversion electron has a momentum p and an energy W . $h_1^{(1)}(kr)$ comes from the electromagnetic interaction, and it is the spherical Hankel function of the first kind and of order one. The wave number k is numerically equal to the excitation energy of the nuclear level over mc^2 . τ is either \uparrow or \downarrow , corresponding to the spin up or down of the ns electron with respect to the nuclear spin \mathbf{J} , while τ' is also \uparrow or \downarrow , corresponding to the spin up or down of the ns electron with respect to the total spin of $3d$ electrons. Therefore, we have two cases,

$$\text{I. } \tau' = -\tau,$$

$$\text{II. } \tau' = \tau. \quad (7)$$

The angular distributions $W_I(\theta)$ and $W_{II}(\theta)$ (or equivalently, the relative intensities of conversion electrons at an angle θ in cases I and II) are different since the radial integrals (5) are different. In the following we make numerical calculations of $W_I(\theta)$ and $W_{II}(\theta)$ for the L_I and M_I conversion electrons. These are conversion electrons from the $2s$ and $3s$ orbits, respectively.

Radial wave functions for the $2s$ and $3s$ states

Nonrelativistic wave functions for all 26 bound electrons in the 5D state of Fe were obtained from the unrestricted Hartree-Fock calculation by Bagus and Liu.⁴⁾ For $2s$ and $3s$ electrons, these wave functions are given by

$$\varphi_{n\tau'}(\mathbf{r}) = \sum_i C_{i, n\tau'} X_{i, \tau'}, \quad n=2 \quad \text{or} \quad 3 \quad (8)$$

where

$$X_{i, \tau'} = (4\pi)^{-1/2} R_i(r) S_{\tau'}, \quad (9)$$

and where the radial functions $R_i(r)$ are normalized nodeless Slater-type orbits,

$$R_i(r) = \alpha^{3/2} [(2n_i)!]^{-1/2} (2Z_i)^{n_i+1/2} (\alpha r)^{n_i-1} e^{-Z_i \alpha r} \quad (10)$$

with

$$\alpha = \text{fine structure constant.}$$

Table I. Radial wave functions for the 2s and 3s electrons of Fe in the unrestricted Hartree-Fock method.⁴⁾ C_i , n_i and Z_i are defined in Eqs. (8)~(10).

i	n_i	Z_i	C_i			
			2s \uparrow	2s \downarrow	3s \uparrow	3s \downarrow
1	1	38.73300	0.00644	0.00629	-0.00290	-0.00274
2	1	25.87820	-0.31094	-0.31144	0.11682	0.11535
3	2	21.33300	-0.18016	-0.18129	0.07296	0.07273
4	2	11.01230	1.03806	1.04258	-0.42236	-0.41937
5	3	9.50024	0.14489	0.14567	-0.30850	-0.30949
6	3	6.99020	0.00437	-0.00182	0.36701	0.38109
7	3	4.63797	0.00415	0.00475	0.77597	0.75070
8	4	3.92345	-0.00023	-0.00079	0.16592	0.18267
9	4	1.95370	0.00006	0.00015	0.00769	0.00796
10	4	1.15237	-0.00002	-0.00009	-0.00393	-0.00415
11	4	0.80019	0.00001	0.00004	0.00166	0.00175

The spin functions are

$$\begin{aligned}
 S_{\tau'} &= \begin{pmatrix} 1 \\ 0 \end{pmatrix} \text{ for } \tau' = \uparrow, \\
 &= \begin{pmatrix} 0 \\ 1 \end{pmatrix} \text{ for } \tau' = \downarrow.
 \end{aligned} \tag{11}$$

The numerical values of C_i , n_i , Z_i are given in Table I, which has been taken from reference 4).

In our calculation, we assume that the large component of the radial wave function is equal to the radial part given by Bagus and Liu,

$$g_{n\tau'}(r) = \sum_i C_{i,n\tau'} R_i(r). \tag{12}$$

We also assume that the small component f is the solution of the Dirac wave equation,

$$\frac{dg(r)}{dr} = (W + 1 - V)f(r), \tag{13}$$

if we replace $g(r)$ by Eq. (12). For $V(r)$, we adopt the following electrostatic potential:⁷⁾

$$V(r) = -[25\varphi(x) + 1](\alpha/r),$$

with

$$\varphi(x) = [1 + 1.5x - 0.7x^2 + 0.38x^3]^{-1} \text{ and with } x = 4(50/9\pi^2)^{1/3}\alpha r, \tag{14}$$

which includes the screening effect with sufficient accuracy. As an example, $G_{2s\uparrow}$ and $F_{2s\uparrow}$ are given in Fig. 1. Also, $r^2[g_{n\tau'}^2(r) - g_{n\tau'}^2(r)]$ is given in Fig. 2.

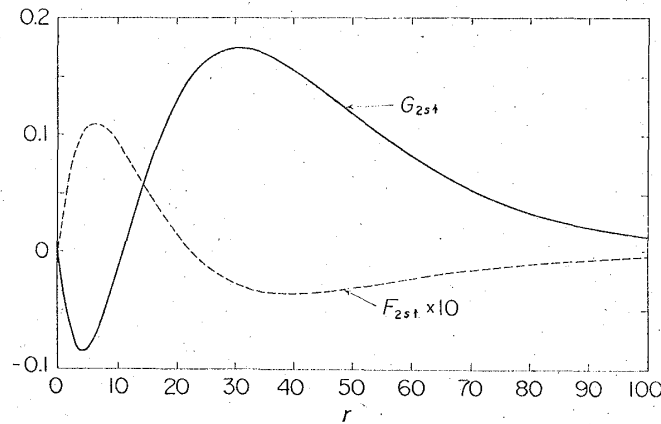


Fig. 1. The Coulomb wave functions, G and F , are given for the $2s$ electron with spin up. The solid and broken curves represent G in Eq. (12) and F in Eq. (13), respectively.

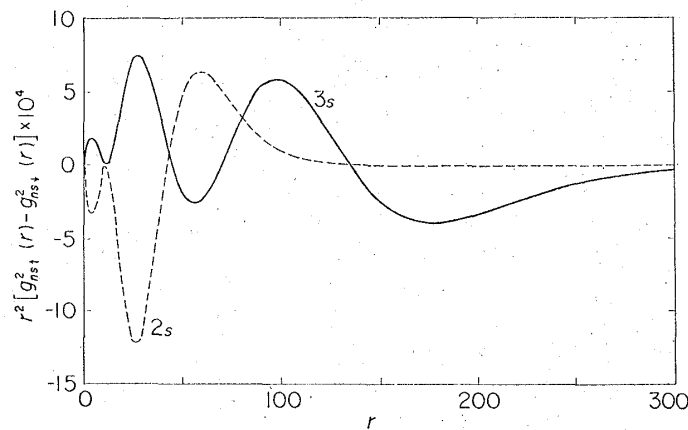


Fig. 2. $r^2[g_{ns↑}^2(r) - g_{ns↓}^2(r)]$'s are given by the solid and broken curves for the $3s$ and $2s$ electrons, respectively.

Radial wave functions for the conversion electrons with $\kappa = -1$ and 2

The radial wave functions for the conversion electrons are obtained as the numerical solutions of the Dirac Coulomb field with screening effect. The computer program for this calculation is that used by Yamada and Kodama^{7),8)} to study beta decay. The calculation has been done for $\kappa = -1$ and 2 and electron energies, $W = 1.0265$ and 1.0280 . These two values of W correspond to the conversions from $2s$ and $3s$ orbits, respectively. As an example, we show G_{-1} and F_{-1} for $W = 1.0265$ in Fig. 3. A normalization is adopted so that the average of $(G_{-1}^2 + F_{-1}^2)$ in a period is unity at $r = \infty$.

Radial integrals $R_{\kappa, ns\tau}$

The radial integrals $R_{\kappa, ns\tau}$ are computed with the above described radial wave functions and the results are summarized in Table II with the following definitions:

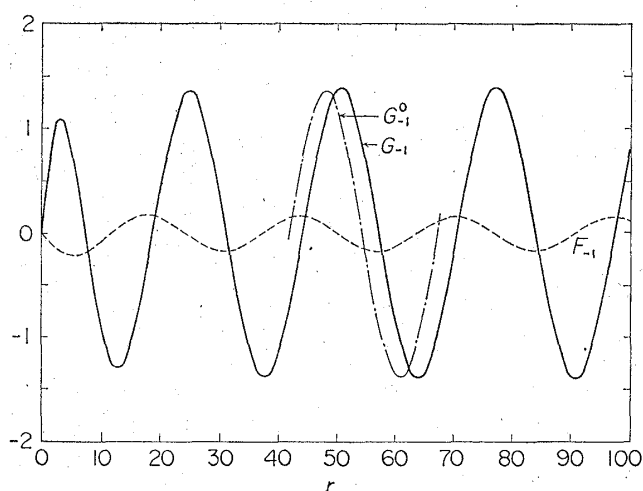


Fig. 3. The Coulomb wave functions G_{-1} and F_{-1} , are given by the solid and broken curves for $W=1.0265$. The screening correction is properly taken into account. The pure Coulomb wave function G_{-1}^0 is also given by the dash-dot curve. A normalization is adopted so that the average of $(G_{-1}^2 + F_{-1}^2)$ in a period is unity at $r=\infty$.

Table II. Radial integrals, Eq. (15), in case II. The table holds in case I if the sign of τ is reversed. Numerical values are given at the upper limits of the integration, $r=183.98$ and 178.09 for $n=2$ and 3 , respectively. Those for $R^{(B)}$ are still oscillating in this range of r . We give, however, the following numerical values so as to show $R^{(B)}$ be negligibly small in comparison with $R^{(N)}$. $W=1.0265$ and 1.0280 for $2s$ and $3s$, respectively.

n	κ	τ	$R_{\kappa, n\sigma\tau}^{(B)}$	$R_{\kappa, n\sigma\tau}^{(N)}$
2	-1	↑	-0.33×10^{-3}	-0.2166×10^2
2	-1	↓	-0.33×10^{-3}	-0.2171×10^2
2	2	↑	0.51×10^{-4}	0.2287×10^1
2	2	↓	0.49×10^{-4}	0.2293×10^1
3	-1	↑	-0.34×10^{-2}	0.8207×10^1
3	-1	↓	-0.34×10^{-2}	0.8120×10^1
3	2	↑	0.14×10^{-2}	-0.8704
3	2	↓	0.14×10^{-2}	-0.8606

$$R_{\kappa, n\sigma\tau} = R_{\kappa, n\sigma\tau}^{(B)} + iR_{\kappa, n\sigma\tau}^{(N)},$$

$$R_{\kappa, n\sigma\tau}^{(B)} \equiv \int_0^{\infty} [F_{\kappa}(pr) G_{n\sigma\tau}(r) + G_{\kappa}(pr) F_{n\sigma\tau}(r)] j_1(kr) dr,$$

$$R_{\kappa, n\sigma\tau}^{(N)} \equiv \int_0^{\infty} [F_{\kappa}(pr) G_{n\sigma\tau}(r) + G_{\kappa}(pr) F_{n\sigma\tau}(r)] n_1(kr) dr, \quad (15)$$

where j_1 and n_1 are the spherical Bessel and spherical Neumann functions of order one, respectively.

Results

As we can see in Table II, the conversion electrons are almost in the s state so that the angular distribution is nearly *isotropic*. The Coulomb phase shifts are obtained graphically from the radial wave function, and the results are

$$\begin{aligned} \Delta_{\kappa=-1} - \Delta_{\kappa=2} &= 1.13 \text{ rad for } W=1.0265 (2s), \\ &= 1.10 \text{ rad for } W=1.0280 (3s). \end{aligned} \quad (16)$$

Finally, the relative intensities for the cases I and II have the following difference:

$$\begin{aligned} \frac{W_I(\theta) - W_{II}(\theta)}{W_I(\theta)} &= -0.5\% \text{ for } L_I \text{ (from } 2s), \\ &= 2.1\% \text{ for } M_I \text{ (from } 3s), \end{aligned} \quad (17)$$

at $\theta=0$.

We did not compute the effect for the 1s electron. This is because the s - d interaction is much smaller and the effect in conversion process is negligible. Our calculation does not include interactions with ns electrons of neighboring nuclei as well as their orbital electrons.

Acknowledgements

We would like to express our sincere thanks to Professor J. Kanamori and Professor H. Miwa for stimulating discussions. One of us (M. M.) is indebted to Professor N. Koller, the Rutgers University, for a discussion on possible use of the Mössbauer effect for this problem.

References

- 1) E. Fermi, *Z. Phys.* **60** (1930), 320.
E. Fermi and E. Segrè, *Z. Phys.* **82** (1933), 729.
- 2) A. Abragam, J. Horowitz and M. H. L. Pryce, *Proc. Roy. Soc.* **A230** (1955), 169.
- 3) R. E. Watson and A. J. Freeman, *Phys. Rev.* **123** (1961), 2027; *Phys. Rev.* **120** (1960), 1125, 1134.
- 4) A calculation of the internal field with radial wave functions in the unrestricted Hartree-Fock method was done by R. E. Watson and A. J. Freeman, *Phys. Rev.* **123** (1961), 2027. Since the explicit forms of these wave functions are not given there, we adopt those given by P. S. Bagus and B. Liu, *Phys. Rev.* **148** (1966), 79.
- 5) See, e.g., M. E. Rose, *Relativistic Electron Theory* (John Wiley & Sons, Inc., Publishers, New York, 1960), p. 27.
- 6) M. E. Rose, L. C. Biedenharn and G. B. Arfken, *Phys. Rev.* **85** (1952), 5. In Eq. (44) of this paper, a Clebsch-Gordan coefficient, $C_{M+\tau, -M-\tau}^{j'j}$ is dropped.
- 7) M. Yamada, *Bull. Sci. Eng. Research Lab., Waseda Univ.* No. 36 (1966), 67.
- 8) M. Yamada and T. Kodama, *J. Phys. Soc. Japan Suppl.* **24** (1968), 729.