Proposed Detection of Dynamically Oriented Co^{57} by the Mössbauer Effect*

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The Mössbauer effect may be a useful probe of the static and dynamic mechanisms coupling a parent nucleus to a paramagnetic environment in a static or time-varying magnetic field. A theoretical expression is developed for the relative-intensity ratio $R(v)$ for a transmission-type Mössbauer spectrum of the gamma radiation obtained in the decay of oriented Co^{57}. This expression explicitly exhibits a sensitive dependence on the occupation numbers of the Zeeman levels of the parent nucleus. The ratio is calculated numerically for the cases of static and dynamic orientation in a field of 100 kG with the source held at 1°K and a warm absorber. Dynamic orientation is assumed to be achieved by the solid effect in which microwave pumping radiation induces transitions which flip the electron spin by one unit and the nuclear spin by two units. The value of $R(v)$ for the dynamic case is mostly $\sim 10^{-7}$, but is $\sim 10^2$ when v is chosen to provide overlap of the $\frac{3}{2}\rightarrow\frac{1}{2}$ and $-\frac{1}{2}\rightarrow\frac{1}{2}$ emission lines with the $-\frac{1}{2}\rightarrow\frac{3}{2}$ and $-\frac{1}{2}\rightarrow\frac{1}{2}$ absorption lines, respectively. This differs significantly from $R(v)$ for the static case. which is ~ 1 for the same two overlaps.

I. INTRODUCTION

IT is the purpose of this paper to suggest one more application of the Mössbauer effect[1–5]: the detection of nuclear orientation of the parent nuclei, the daughter states of which emit Mössbauer radiation.[6] What could be learned from such an experiment? Hopefully, one might expect to obtain details regarding the static and dynamic interactions between unstable nuclei and their local environment.

To make this suggestion specific, we present a calculation relating to a *Gedankenexperiment* on Co^{57} nuclei. We suppose that these nuclei are placed in a paramagnetic host and subjected to an external magnetic field which has time-varying as well as static components. We assume that the hyperfine and quadrupole interactions experienced by the nuclei will permit dynamic nuclear orientation by the Jeffries-Abragam effect.[7] If this occurs, the resulting non-Boltzmann population distribution achieved among the magnetic sublevels of the Co^{57} parent nuclei will determine the intensity of the Zeeman components of the 14.4-keV radiation from the Fe^{57} daughter nuclei. This radiation would be monitored by a slowly moving absorber consisting of Fe^{57} nuclei in the same host material as the source and in an external magnetic field with the same static but no time-varying components.

II. DYNAMIC NUCLEAR ORIENTATION OF Co^{57}

The relevant properties of Co^{57} and Fe^{57} are well known.[8–13] In an external static magnetic field the magnetic substates of Co^{57} and of the three states of Fe^{57} are split into Zeeman levels. In Fig. 1 the decays and Zeeman splittings of Co^{57} and Fe^{57} are represented schematically.

One of the essential requirements in the choice of a host for the Co^{57} nuclei is that the linewidth of the 14.4-keV radiation be narrow enough so that the Zeeman components of the Mössbauer pattern are resolvable. This can be achieved by using sufficiently strong magnetic fields.[12] A second requirement of the host material is that it be paramagnetic. The analysis is simplified if one chooses free electrons or paramagnetic ions for which the electron spin $S=\frac{1}{2}$.

We now apply a time-varying magnetic field, $H_1 \cos\omega t$, oriented at right angles to the static field H_0 with the frequency ω chosen to induce simultaneously single-spin flips of the electron and double-spin flips of the Co^{57}

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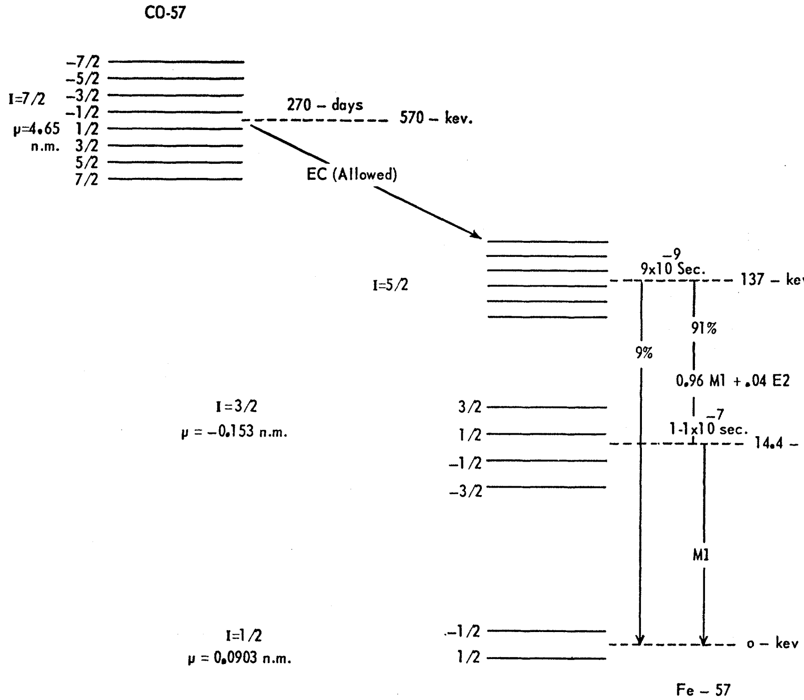

FIG. 1. Decay scheme and Zeeman-level splittings of Co^{57} and Fe^{57}.

nucleus:

$$\hbar\omega=\hbar(\omega_e+2\omega_n)=g|\beta|H_0+2|g_n|\beta_nH_0. \quad (1)$$

We assume that the conditions for such a magnetic resonance experiment are satisfied. Explicit expressions for the radiative induced transition probabilities in an electron-nuclear coupled system have been calculated by Jeffries.[14] Let W_2, W_1, and W_0 denote, respectively, the transition probabilities for which two, one, or zero nuclear spin flips ($\Delta m=\pm2$, ±1, or 0) are associated with a single electron spin flip ($\Delta M=\pm1$). Jeffries shows that the "forbidden" transitions W_2 and W_1 are equally probable and that both are smaller than the "allowed" transitions W_0 by a factor proportional to the square of the ratio of the quadrupole coupling constant to the hyperfine interaction constant $(P/A)^2$. A nonzero P requires a nucleus of spin $I\geq1$. This is satisfied by Co^{57} for which $I=\frac{7}{2}$. A large P requires that the electric field gradient tensor at the nuclear site be large. This may be satisfied by a suitable choice of the crystalline host.

The steady-state population distribution of this pumped electron-nuclear coupled system now depends on the relaxation mechanisms which are assumed to be dominant. We do not know what these are, but we make a guess, suggested by experience with similar systems, that three types of relaxations are fast and that all others are slow enough to be neglected. The fast relaxations are assumed to be those for which (i) $\Delta M=\pm1$, $\Delta m=0$; (ii) $\Delta M=\pm1$, $\Delta m=\mp1$; and (iii) $\Delta M=\pm1$, $\Delta m=\pm1$. We refer to these fast relaxations as paramagnetic, flip-flop, and flip-flip, respectively.

A number of features described in the preceding paragraphs may be visualized by reference to Fig. 2. There are 16 energy levels. Because of the large static field assumed, the level spacing is assumed to be negligibly affected by the hyperfine and quadrupole coupling. Sufficient admixing of states is, however, assumed to be provided by A and P so that with moderate power to pump, the rate W_2 is large compared to all the thermal relaxation rates.

The steady-state population distribution of this system may now be calculated from the relevant rate equations and normalization condition. On the other hand, an "inspection method" for the solution of the population distribution in multilevel quantum-mechanical systems has been developed by Keating and Barker.[15–17] This method has been applied to the system under consideration to determine the normalized population distribution N_i $(i=1, 2, \cdots, 16)$. The solution represents an important intermediate result, essential to the calculations which follow. We write, for illustration, the occupation numbers of some of the levels in

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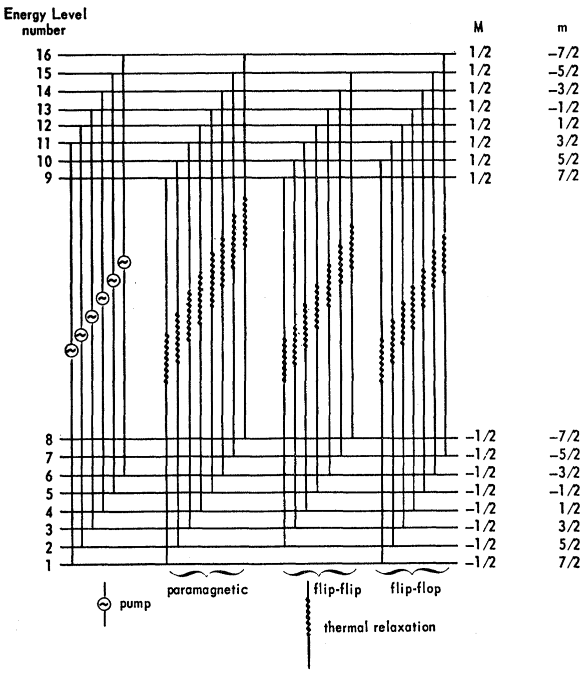

FIG. 2. Co^{57} nuclear spin-unpaired electron-spin coupled energy-level diagram in a static magnetic field. The pumping scheme and dominant relaxation mechanisms proposed for nuclear orientation are indicated. The nuclear-magnetic quantum number is m. The electronic magnetic quantum number is M.

terms of the total number N of spin pairs, the electron spin Boltzmann factor x, the nuclear Boltzmann factor y, and a parameter η defined as the ratio of flip-flop to the flip-flip thermal relaxation rates. Explicitly,

$$N_1=N_{11}=N[(x^7/y+x^7+x^6/y+x^6+x^5/y+x^5+x^4/y)+\eta(x^7+x^7y+x^6+x^6y+x^5+x^5y+x^4)]/d, \quad (2a)$$

$$N_2=N_{12}=N[(x^7+x^6/y+x^6+x^5/y+x^5+x^4/y+x^4)+\eta(x^7y+x^6+x^6y+x^5+x^5y+x^4+x^4y)]/d, \quad (2b)$$

$$N_7=N[(x^4/y+x^4+x^3/y+x^3+x^2/y+x^2+x/y)+\eta(x^4+x^4y+x^3+x^3y+x^2+x^2y+x)]/d, \quad (2c)$$

$$N_8=N[(x^4+x^3/y+x^3+x^2/y+x^2+x/y+x)+\eta(x^4y+x^3+x^3y+x^2+x^2y+x+xy)]/d; \quad (2d)$$

where d is a normalization factor which is not important for the present considerations.

The Boltzmann population distribution N_i^0 has an algebraic structure which differs vastly from Eqs. (2). For example, $N_{11}^0/N_1^0=xy^2$ and $N_2^0/N_1^0=y$. It is very interesting to compare the relative non-Boltzmann population distribution r_i^0, which we obtained in an IBM-1620 evaluation in which the temperature T and the magnetic field H_0 were taken as 1°K and 100 kG, respectively. It is clear from the results, presented in Table I, that the effect of the assumed pump and relaxation mechanisms is essentially to depopulate all the levels quite dramatically in favor of levels 7 and 8. This can be understood physically by examining the pumping scheme and thermal relaxations as shown in Fig. 2.

III. MÖSSBAUER EFFECT DETECTION OF Co^{57} NUCLEAR ORIENTATION

We now focus our attention on the calculation of a quantity which is of interest in a transmission-type Mössbauer effect experiment[18,19]:

$$R(v)\equiv\left[\frac{I_\infty-I_t(v)}{I_\infty-I_t(-v)}\right]. \quad (3)$$

The parameter $R(v)$ is useful in that it serves to cancel out unknown factors and eliminates formally the nonresonant background of gamma radiation. There are two differences between the proposed gamma source and the absorber. The source is assumed to be subjected to an rf pumping field and is kept cold. The absorber is assumed to be at the same static field as the absorber and is kept sufficiently warm ($\sim$300°K) so that we can neglect the difference between Zeeman sublevel populations in the absorber. In Eq. (3), $I_t(v)$ and $I_t(-v)$ represent the intensities transmitted when the absorber is moving away from and towards the source with a speed v, respectively; I_∞ represents the transmitted intensity when this relative motion is fast enough so that no lines overlap.

The 14.4-keV gammas emitted in the decay from the first excited state of Fe^{57} to its ground state are split into six Zeeman components in accordance with the dipole selection rules $M=m_j-m_k=\pm1, 0$. The resonance emission spectrum is thus composed of several lines of Lorentzian shape, each having the width Γ. The hyperfine emission spectrum of the source can be analyzed by filtering the radiation through the absorber. Dash *et al.*[19] have developed the expression for $R(v)$ valid for a thin source and a thin absorber:

$$R(v)=\frac{\sum_{(jk)(k'j')}\Delta_v K_{k'j'}w_{jk}\sum_l p_l Q_{lj}}{\sum_{(jk)(k'j')}\Delta_v K_{k'j'}w_{jk}\sum_l p_{-l} Q_{lj}}. \quad (4)$$

The symbols of Eq. (4) are defined, in the notations of Ref. 19, as follows:

$K_{k'j'}=1-J_0(iW_{k'j'}x/2)\exp(-W_{k'j'}x/2)$,
$x=naf\sigma$,
$\Delta_v=1$ when $E_{jk}(1-v/c)=E_{k'j'}$,
$\Delta_v=0$ when $|E_{jk}(1-v/c)-E_{k'j'}|>5\Gamma$,
w_{jk} = dipole transition probability for the transition $m_j\rightarrow m_k$. We use the results calculated by Frauenfelder *et al.*[20]

[18] H. Frauenfelder, D. R. F. Cochran, D. E. Nagle, and R. D. Taylor, Nuovo Cimento **19**, 183 (1961).
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TABLE I. Comparison of relative (unnormalized) populations of states in the Co^{57} nuclear electron spin-coupled system for the non-Boltzmann and Boltzmann cases at $T=1°K$, $H_0=100$ kG.

r_i	r_i^0	
$r_1=r_{11}=0.478(1+\eta)\times10^{-23}$;	$r_1^0=1.000$,	$r_{11}^0=0.136\times10^{-5}$
$r_2=r_{12}=[0.934+\eta(0.889)]\times10^{-23}$;	$r_2^0=0.953$,	$r_{12}^0=0.126\times10^{-5}$
$r_3=r_{13}=[0.327+\eta(0.312)]\times10^{-17}$;	$r_3^0=0.907$,	$r_{13}^0=0.120\times10^{-5}$
$r_4=r_{14}=[0.639+\eta(0.609)]\times10^{-17}$;	$r_4^0=0.864$,	$r_{14}^0=0.115\times10^{-5}$
$r_5=r_{15}=[0.224+\eta(0.213)]\times10^{-11}$;	$r_5^0=0.823$,	$r_{15}^0=0.109\times10^{-5}$
$r_6=r_{16}=[0.437+\eta(0.417)]\times10^{-11}$;	$r_6^0=0.784$,	$r_{16}^0=0.103\times10^{-5}$
$r_7=[0.153+\eta(0.146)]\times10^{-5}$;	$r_7^0=0.747$	
$r_8=[0.299+\eta(0.285)]\times10^{-5}$;	$r_8^0=0.712$	
$r_9=[0.698+\eta(0.665)]\times10^{-29}$;	$r_9^0=0.146\times10^{-5}$	
$r_{10}=[0.136+\eta(0.130)]\times10^{-28}$;	$r_{10}^0=0.139\times30^{-5}$	

p_l = population of the lth level of parent Co^{57} nuclei,

Q_{lj} = matrix element of the matrix obtained by the multiplication of the two Clebsch-Gordan matrices for the two transitions preceding the arrival at the 14.4-keV state of Fe^{57} (tabulated in Ref. 19).

$W_{k'j'}$ = normalized relative absorption probabilities for the transitions $m_{k'}\rightarrow m_{j'}$. They are discussed in Appendix I.

Equation (4) is valid provided the nuclear-spin relaxation times of the two excited states of Fe^{57} are long in comparison to the lifetimes of the states. This condition is probably well satisfied since for a cold source the nuclear T_1 may easily be longer than 10^{-2} sec and the gamma half-life in our case is $\sim10^{-7}$ sec.

In Eq. (4) the substitution $Q_{lj}=Q_{lj}^{M1}\times0.96+Q_{lj}^{E2}\times0.04$ is required to take into account the admixture of magnetic-dipole and electric-quadrupole gamma radiation in the decay from the second to the first excited state of Fe^{57}. The interference term between the $M1$ and $E2$ radiations is neglected taking $0.04\ll0.96$.

IV. NUMERICAL EXAMPLES

We consider the different velocities and evaluate $R(v)$ for the three following cases:

Case (a) line $\frac{3}{2}\rightarrow\frac{1}{2}$ overlaps line $-\frac{1}{2}\rightarrow-\frac{3}{2}$.
Case (b) line $-\frac{1}{2}\rightarrow\frac{1}{2}$ overlaps line $-\frac{1}{2}\rightarrow\frac{1}{2}$.
Case (c) lines $-\frac{1}{2}\rightarrow-\frac{1}{2}$, $\frac{1}{2}\rightarrow-\frac{1}{2}$, $\frac{1}{2}\rightarrow\frac{1}{2}$, and $\frac{3}{2}\rightarrow\frac{1}{2}$ overlap lines $-\frac{1}{2}\rightarrow-\frac{3}{2}$, $-\frac{1}{2}\rightarrow-\frac{1}{2}$, $\frac{1}{2}\rightarrow-\frac{1}{2}$, and $\frac{1}{2}\rightarrow\frac{1}{2}$, respectively.

We take $\beta=30°$, $\beta'=60°$, and $(\alpha-\alpha')=90°$. The total resonance cross section,[18] $\sigma=1.48\times10^{-18}$ cm^2. A reasonable value [20] for $naf/2=10^{19}$ cm^{-2}.

Assume first that the Co^{57} parent nuclei have a Boltzmann population distribution in a magnetic field of 100 kG and at a temperature $T=1°K$. The values of $R(v)$ are 1.295 for case (a), 0.917 for case (b), and 1.173 for case (c).

Assume now that the Co^{57} parent nuclei have the particular steady-state non-Boltzmann populations calculated in Sec. II. The values of $R(v)$ are 0.248×10^{-7} for case (a), 0.355×10^2 for case (b), and 0.479×10^{-1} for case (c). These results were found to be insensitive to values of η, the ratio of flip-flop to flip-flip relaxation rates for the range $10^{-4}\leq\eta\leq10^8$.

V. DISCUSSION OF RESULTS

The numerical values for $R(v)$ for all the cases for Boltzmann distribution are of the order of 1 whereas, for the non-Boltzmann distribution, $R(v)$ changes by several orders of magnitude. This striking behavior can be explained by a close examination of Eq. (4). Because of the zero values of some of the Q_{lj} the populations of levels 7 and 8 do not contribute to the numerator of (4) whereas they do contribute to the denominator of (4) for case (a). Since levels 7 and 8 carry almost all the spin pairs the low value of $R(v)$ for case (a) is explained. The values of $R(v)$ for cases (b) and (c) can be similarly explained.

VI. CONCLUDING REMARKS

The principal results of this paper are embodied in Eq. (4) which provides the theoretical framework for calculating the quantities of interest in a transmission-type Mössbauer experiment in which the parent nuclei have been dynamically oriented. The numerical results obtained clearly indicate that the Mössbauer effect may be used to distinguish dynamic from static orientation. It may also be safely inferred that various types of dynamic nuclear orientation should be distinguishable from one another. A different pumping scheme than the one chosen could readily result in a vastly different population distribution.

The particular numerical calculations are made for illustrative purposes. We are not seriously recommending an experiment which would require two 100-kG superconducting solenoids with a $T=1°K$ environment for the emitter and room temperature for the absorber. All that is required by way of a static external magnetic field is that the Zeeman components of the recoilless gamma radiation be resolved. In Zn^{67} a field as low as 100 G is sufficient for this purpose.[21] Consequently, a

[21] P. P. Craig, D. E. Nagle, and D. R. F. Cochran, Phys. Rev. Letters 4, 561 (1960).

moderate field of 3300 G with a conventional 10-Gc klystron might easily demonstrate this effect in Zn^{67} providing the paramagnetic host is suitably chosen.

The calculations do rely on simplifying assumptions which are not always satisfied in dynamic nuclear orientation experiments: namely, an EPR linewidth which is narrower than the nuclear Zeeman splitting as well as some mechanism for diffusing the solid effect to nuclei which are remote from a given paramagnetic center. However, the experimental and theoretical work of Jeffries,[22] Abragam,[23] Motchane,[24] Khutsishvili,[25] Krebs and Thompson,[26] and others is beginning to bring understanding to these problems. In fact, we believe the Mössbauer effect could profitably be used to add much needed experimental data on these more complicated aspects of dynamic nuclear orientation.

ACKNOWLEDGMENTS

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APPENDIX I

The normalized relative absorption transition probabilities, $W_{k'j'}$, can be expressed, from the results of the calculations of Frauenfelder *et al.*,[20] as

$$W_{k'j'}=\frac{w_{k'j'}}{\sum_{k'j'} I_{k'j'} \sum_{jk} \cos^2\Theta(jk,k'j')}. \tag{5}$$

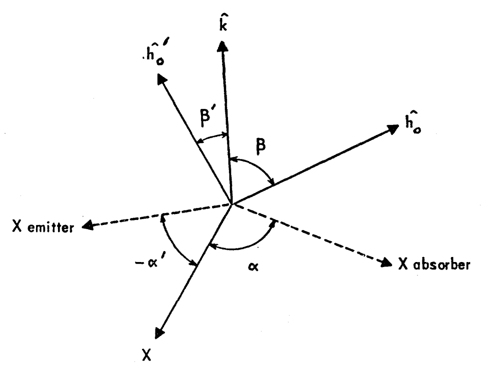

FIG. 3. Euler angles α,α' and β,β' of the quantization axes $\hat{h}_0, \hat{h}_0'$ for the emitter (unprimed) and absorber (primed). The third Euler angle γ,γ' is ignorable because of the symmetry observed by each system about its quantization axis. $\hat{k}$ is the direction of propagation of the γ ray.

TABLE II. Fractional overlap factors for dipole radiation.

M	M'	$\cos^2\Theta$
1	±1	$\dfrac{(\cos\beta\pm\cos\beta')^2+\sin^2\beta\sin^2\beta'\cos^2(\alpha-\alpha')}{(1+\cos^2\beta)(1+\cos^2\beta')}$
±1	0	$\dfrac{1-\sin^2\beta\cos^2(\alpha-\alpha')}{1+\cos^2\beta}$
0	±1	$\dfrac{1-\sin^2\beta'\cos^2(\alpha-\alpha')}{1+\cos^2\beta'}$
0	0	$\cos^2(\alpha-\alpha')$

The structure of Eq. (5) differs in two ways from the analogous relation for the source given by Dash *et al.*[19] There are no $p_{j'}$ in Eq. (5) as we consider the absorber to be sufficiently warm so that all the Zeeman levels of the ground and first excited states of Fe^{57} are equally populated. Further we use the reasoning and results of Frauenfelder *et al.*[20] to take into account the relative directions of the magnetic fields acting on the emitter and absorber nuclei. The factor $\cos^2\Theta(jk,k'j')$ refers to the overlap which occurs when the source line $m_j\to m_k$ coincides with the absorber line $m_{k'}\to m_{j'}$. In Fig. 3, the direction in which the gamma rays are emitted from the source is taken along the z axis; the axes of quantization from the emitter and absorber are denoted by $\hat{h}_0$ and $\hat{h}_0'$ and are taken along the directions of the external magnetic fields applied to each. The dependence of the fractional overlap factor $\cos^2\Theta$ on the Euler angles $\alpha,\beta;\alpha',\beta'$ are given in Table II for dipole radiation: $M,M'=\pm1,0$. The third Euler angles γ,γ' may be ignored because of symmetry.[20] The relative absorption transition probability

$$w_{k'j'}=g_{M'}{}^L(\beta')\begin{pmatrix} I_f & L & I_i' \\ -m_k & M' & m_j' \end{pmatrix}^2\cos^2\Theta = I_{k'j'}\cos^2\Theta. \tag{6}$$

If we take advantage of the property of the Wigner $3-j$ symbol that

$$\begin{pmatrix} I_i' & L & I_f' \\ m_j' & M' & -m_{k'} \end{pmatrix}^2=\begin{pmatrix} I_f' & L & I_i' \\ -m_{k'} & M' & m_{j'} \end{pmatrix}^2,$$

we may write a simple relationship between $w_{k'j'}$ and w_{jk}:

$$w_{k'j'}(\beta')=w_{jk}(\beta')\cos^2\Theta.$$

[22] C. D. Jeffries, *Dynamic Nuclear Orientation* (Interscience Publishers, Inc., New York, 1963).
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