PHYSICAL REVIEW B

SOLID STATE

THIRD SERIES, VOL. 5, NO. 7 1 APRIL 1972

Z_1^3 Effect in the Stopping Power of Matter for Charged Particles*

J. C. Ashley and R. H. Ritchie
Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

and

Werner Brandt
Department of Physics, New York University, New York, New York 10003
(Received 30 October 1971)

We consider in a classical formulation the interaction of a particle of charge Z_1e incident at a given impact parameter with an electron bound isotropically and harmonically to the origin with a frequency ω. Using a perturbation expansion that assumes that the displacement of the bound electron is small compared to the impact parameter, and integrating over the impact parameter from some minimum value to infinity, we are led to an expression for the stopping power. The leading term in this expansion is proportional to $(Z_1e^2)^2$ and is the usual result for this type of model, while the second term gives us the $(Z_1e^2)^3$ correction. The Z_1^3 correction for the Lenz-Jensen statistical model for the atom is presented. The predictions of this theory are in excellent agreement with available experimental data.

I. INTRODUCTION

According to recent reports,[1,2] the ranges of π^+ mesons in nuclear emulsions are shorter than those of π^- mesons such that the π^+ stopping power is some 14% larger than the π^- stopping power at ~1.2 MeV/amu. Measurements in several metals by Andersen *et al.*[3] yield stopping powers for α particles that are systematically larger than four times the stopping power for protons or deuterons as predicted by Bethe's stopping-power theory. This theory is based on the first Born approximation, in which the stopping power for a particle of velocity v_1, energy E_1, and charge number $Z_1 \ll v_1/v_0$, where $v_0 = e^2/\hbar$, is proportional to $(Z_1e)^2$. This paper extends the stopping-power theory to include the term proportional to $(Z_1e)^3$ in an impulse approximation, i.e., the classical equivalent of the second Born approximation. It accounts for the observed differences between the stopping powers of a particle and its antiparticle, and for differences between the stopping powers of α particles and protons which, so far, have remained unexplained. The Z_1^3 term is important for the proper assessment of shell corrections in stopping powers and of the ranges of charged particles in matter.

II. THEORY

Consider an electron bound isotropically and harmonically with a frequency ω to the origin (i.e., the nucleus of the atom) in a target composed of atoms of atomic number Z_2 at a density n_2. A heavy particle of charge Z_1e approaches the oscillator with nonrelativistic velocity v_1 at an impact parameter b relative to the origin. $\vec{f}(t, b)$ is the classically prescribed force on the electron set up by the moving particle at time t. The displacement of the electron at time t from its equilbrium position (at the origin with zero velocity at $t = -\infty$), $\vec{\xi}(t)$, is then found by solving the equation $\ddot{\vec{\xi}} + \omega^2 \vec{\xi} = \vec{f}/m$, which we write in the form

$$\vec{\xi}(t) = \mathrm{Re}\,\frac{i}{m\omega}\int_{-\infty}^{t} \vec{f}(t', b)\, e^{-i\omega(t-t')}\, dt' \,. \qquad (1)$$

At time t, the electron has thus gained the energy $\frac{1}{2}m(\dot{\xi}^2 + \omega^2\xi^2)$. Using Eq. (1), this energy is given by

$$W_r(t, b) = \frac{1}{2m}\left|\int_{-\infty}^{t} \vec{f}(t', b) e^{i\omega t'}\, dt'\right|^2 . \qquad (2)$$

Thus the energy lost by the incident particle per unit distance dR, i.e., the stopping power of the

fraction F_ω of target electrons bound with a frequency ω, becomes

$$\left(-\frac{dE_1}{dR}\right)_\omega = 2\pi n_2 Z_2 F_\omega \left(\int_0^{a_\omega} b\,db\,W_s(\infty, b) + \int_{a_\omega}^{\infty} b\,db\,W_r(\infty, b)\right), \quad (3)$$

where W_r is the energy transferred to resonance excitations of the electrons and W_s is the energy transferred in close single collisions. The parameter a_ω gives a lower limit of the impact paramet where the electrons can still be viewed as being harmonically bound. For $b < a_\omega$ the electrons are treated as unbound. If the incident particle moves along the line $x = -b$ in the $+y$ direction, the force on the electron can be written in terms of Eq. (1) as

$$\vec{f}(t, b) = -Z_1 e^2 \frac{[b+\zeta_x(t)]\hat{i} - [v_1 t - \zeta_y(t)]\hat{j}}{\{[b+\zeta_x(t)]^2 + [v_1 t - \zeta_y(t)]^2\}^{3/2}}, \quad (4)$$

where $\hat{i}$ and $\hat{j}$ are unit vectors in the $+x$ and $+y$ directions. We expand for small electron displacements such that $\zeta(t)/(b^2+v_1^2t^2)^{1/2} \ll 1$ and retain first-order terms. One can write

$$\vec{f}(t, b) = \vec{f}_0(t, b) + \Delta\vec{f}(t, b), \quad (5)$$

where $\vec{f}_0(t, b)$ is given by the well-known expression

$$\vec{f}_0(t, b) = -Z_1 e^2 \frac{b\hat{i} - v_1 t\hat{j}}{[b^2+(v_1t)^2]^{3/2}}. \quad (6)$$

The new term $\Delta\vec{f}$ becomes

$$\Delta\vec{f}(t, b) = -\frac{Z_1 e^2}{[b^2+(v_1t)^2]^{5/2}} \{[(-2b^2+v_1^2t^2)\zeta_x(t) + 3bv_1 t\,\zeta_y(t)]\hat{i} + [3bv_1 t\,\zeta_x(t) + (b^2-2v_1^2t^2)\zeta_y(t)]\hat{j}\}. \quad (7)$$

We solve Eq. (7) by iteration, replacing $\vec{f}$ by $\vec{f}_0$ when calculating $\vec{\zeta}(t)$ from Eq. (1). We insert the Fourier transforms $\vec{f}_{0\omega}$ and $\Delta\vec{f}_\omega$ into $W_r(\infty, b)$ and obtain by Eq. (3) the stopping-power correction due to $\Delta\vec{f}$:

$$\Delta\left(-\frac{dE_1}{dR}\right)_\omega = \frac{2\pi n_2 Z_2 F_\omega}{m} \int_{a_\omega}^{\infty} b\,db\,(\mathrm{Re}\vec{f}_{0\omega} \cdot \mathrm{Re}\,\Delta\vec{f}_\omega + \mathrm{Im}\vec{f}_{0\omega} \cdot \mathrm{Im}\Delta\vec{f}_\omega). \quad (8)$$

The dipole approximation underlying Eq. (8) restricts its validity range to distances from the nucleus outside the atomic volume in which the electrons responding with frequency ω are bound. This limits a_ω to values larger than the respective shell radius. For smaller impact parameters the momentum transfer becomes so large that the electrons behave as if essentially free and their contribution to the Z_1^3 effect becomes small, because the cross section for Rutherford scattering with free electrons is exactly proportional to Z_1^2. Such a division into glancing collisions and close collisions leading to large momentum transfers is well known from Bohr's early semiclassical treatment of stopping powers.[4,5]

Equation (8) can be written in the form

$$\Delta\left(-\frac{dE_1}{dR}\right)_\omega = \frac{4\pi n_2 Z_2 \omega F_\omega}{m^2 v_1^5} (Z_1 e^2)^3 I\left(\frac{\omega a_\omega}{v_1}\right), \quad (9)$$

where

$$I(\xi) \equiv \int_\xi^\infty \frac{du}{u^2} \left(-K_1(u) \int_{-\infty}^{\infty} dv \frac{\cos uv}{(1+v^2)^{5/2}} [(v^2-2)F_1(u, v) - 3vF_2(u, v)] + K_0(u) \int_{-\infty}^{\infty} dv \frac{\sin uv}{(1+v^2)^{5/2}} [3vF_1(u, v) - (1-2v^2)F_2(u, v)]\right). \quad (10)$$

The functions F_1 and F_2 are defined by

$$F_1(u, v) \equiv \int_{-\infty}^{v} dy \frac{\sin[u(v-y)]}{(1+y^2)^{3/2}},$$

$$F_2(u, v) \equiv \int_{-\infty}^{v} dy \frac{y \sin[u(v-y)]}{(1+y^2)^{3/2}}.$$

K_ν is the modified Bessel function of the second kind of order ν. For $\xi = \omega a_\omega / v_1 \ll 1$, the function $I(\xi)$ can be represented by $I(\xi) \approx -\frac{3}{2}\pi \ln\xi + A$, where $A \approx -2.4$ is a constant. In this limit the function $I(\xi)$ is insensitive to the choice of the minimum impact parameter a_ω. For large arguments $I(\xi)$ vanishes approximately as $I(\xi) \approx (4\pi/\xi^2)e^{-2\xi}$. The computer evaluation of Eq. (10) is presented in Figs. 1 and 2.

If the fraction F_ω of oscillators responding in the frequency range between ω and $\omega + d\omega$ is given by $g(\omega)d\omega$, where $g(\omega)$ is the differential oscillator

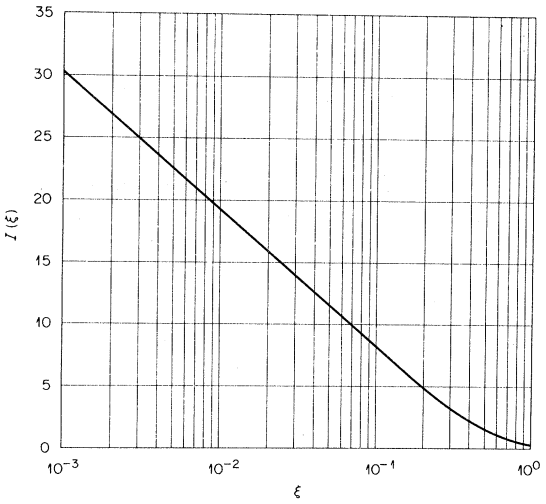

FIG. 1. Function $I(\xi)$, Eq. (10), as a function of ξ for $\xi \leq 1$.

strength normalized such that $\int_0^\infty g(\omega)\,d\omega = 1$, the total stopping-power correction becomes

$$\Delta\left(-\frac{dE_1}{dR}\right) = \frac{4\pi n_2 Z_2}{m^2 v_1^5}(Z_1 e^2)^3 \int_0^\infty d\omega\, g(\omega)\omega\, I\left(\frac{\omega a_\omega}{v_1}\right). \tag{11}$$

For a comprehensive study of its v_1 and Z_2 dependence, we evaluate the Z_1^3 term for the statistical model of the target atoms in the Lenz-Jensen (LJ) approximation[6] for the electron density distribution $\rho(r)$. Then

$$g(\omega) = Z_2^{-1} \int d^3r\, \rho(r)\delta(\omega_0(r) - \omega)\,,$$

where $\omega_0(r) = \chi[4\pi\rho(r)e^2/m]^{1/2}$; the constant χ is a number of $\sim\sqrt{2}$.[7,8] The minimum impact parameter is taken to be approximately the same size as the radius of the shell of charge associated with the frequency $\omega_0(r)$, i.e., we take $a_\omega = \eta r$ where η is a number of order one. The total stopping power can then be written in the reduced form

$$\frac{mv_1^2}{4\pi(Z_1 e^2)^2 n_2 Z_2}\left(-\frac{dE_1}{dR}\right) = xS(x, Z_2) = L(x) + \frac{Z_1}{Z_2^{1/2}}\,\frac{F(\eta\chi Z_2^{1/6}/x^{1/2})}{x^{3/2}}\,, \tag{12}$$

where $S(x, Z_2)$ is a dimensionless stopping cross section in terms of the reduced variable $x = v_1^2/v_0^2 Z_2$. Numerically, $v_1^2/v_0^2 = 40.2\,E_1$(in MeV/amu). For $x \gg 1$, $L(x)$ approaches the Bethe-Bloch formula $L(x) = \ln(4\mathcal{R}/K_B)x$, where $\mathcal{R} = \frac{1}{2}mv_0^2 = 13.6$ eV; $K_B = I/Z_2$ is Bloch's constant as determined by the mean excitation potential I of the target. We calculate $L(x)$ by setting

$$L(x) = \ln\left(\frac{4\mathcal{R}x}{\chi K_{\mathrm{LJ}}}\right) - \frac{4\pi}{Z_2}\int_0^{r_c} dr\, r^2\rho(r) \times \ln\left(\frac{4\mathcal{R}x}{\hbar\omega_0(r)/Z_2}\right). \tag{13}$$

The second term is the statistical approximation to the inner-shell correction; the integration limit r_c is given by the relation $\hbar\omega_0(r)/Z_2 = 4\mathcal{R}x/C$ with a cutoff constant $C \simeq 1$.[9,10] With $K_{\mathrm{LJ}} = 7.583$ eV[11] and $K_B = 9.76$ eV,[12] one obtains $\chi = 1.29$. The function $L(x)$ was evaluated on a computer for $C = 1$. The result is shown in Fig. 3.

The function F is given by

$$F = \frac{4\pi a_0}{v_0 Z_2^2}\int_0^\infty dr\, r^2\rho(r)\omega_0(r) I\left(\frac{\eta r\omega_0(r)}{v_1}\right), \tag{14}$$

where a_0 is the Bohr radius. We omit an inner-shell correction term analogous to the second term in Eq. (13) because it makes a negligible contribution to the total stopping power in the validity range of Eq. (14). We have calculated the function $F(w)$, where $w \equiv \eta\chi Z_2^{1/6}/x^{1/2}$, by numerical integration with the result shown in Fig. 3.

The relative change in the stopping power can be calculated from the expression

$$\frac{xS(x, Z_2) - L(x)}{L(x)} = \frac{Z_1}{Z_2^{1/2}}\,\frac{F(\eta\chi Z_2^{1/6}/x^{1/2})}{x^{3/2}L(x)}\,. \tag{15}$$

The dominant dependence of this correction on particle velocity is proportional to $x^{-3/2}$.

III. COMPARISON WITH EXPERIMENT

We compare the theory with the experimental results of Ref. 3. In Fig. 4 we plot Eq. (15) as a

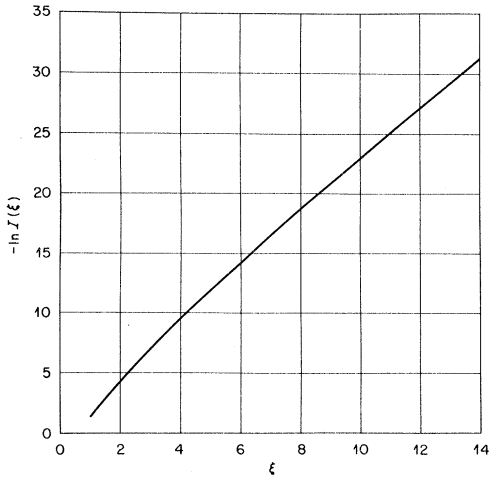

FIG. 2. Function $I(\xi)$, Eq. (10), plotted as $-\ln(\xi)$ as a function of ξ for $\xi \geq 1$.

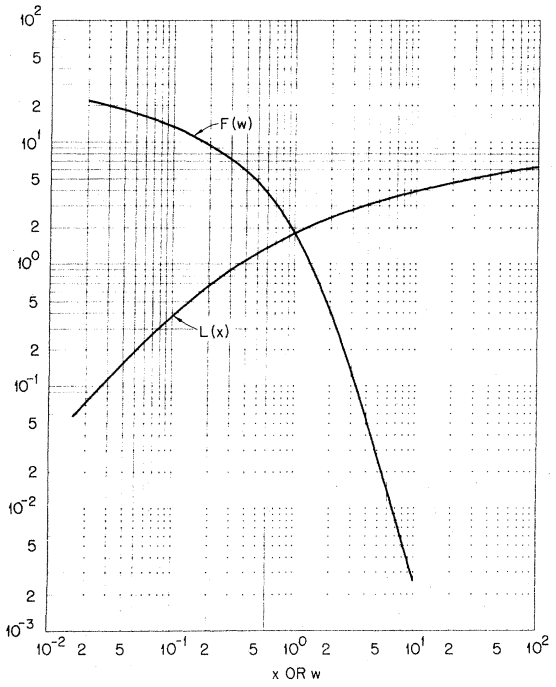

FIG. 3. Functions $L(x)$ and $F(w)$ in the statistical approximation of the Z_1^3 stopping-power correction, Eq. (15), as a function of their respective arguments.

function of $\beta = v_1/c$ for $Z_1 = +1$ particles in tantalum ($Z_2 = 73$) with the trial values $\eta = \frac{3}{4}$ and $\eta = \frac{2}{3}$. The experimental points are taken from Fig. 1 of Ref. 3. A parameter value close to $\eta = 0.7$ gives excellent agreement for the high-Z_2 material over the v_1 range investigated.

Figure 5 shows the comparison for $Z_1 = +1$ particles in aluminum ($Z_2 = 13$) with the trial values $\eta = \frac{2}{3}$, $\eta = \frac{3}{4}$, and $\eta = 1$. The experimental points are taken from Fig. 2 of Ref. 3. A value close to $\eta = 0.9$ provides an excellent fit.

The trend of η with Z_2 may in part reflect the approximate nature of the universal function $L(x)$ as given in Eq. (13),[13] but it is also understandable in terms of the deviations of $g(\omega)$ of real atoms with small Z_2 from that of the statistical atom. In fact, if we rewrite the argument of $F(w)$, Eq. (14), with a constant α as

$$w \equiv \eta\chi Z_2^{1/6}/x^{1/2} = \alpha a_{TF} K_B Z_2/\hbar v_1 , \qquad (16)$$

where $a_{TF} = 0.885 a_0/Z_2^{1/3}$ is the Thomas-Fermi screening radius, we can accommodate these deviations in an average way by inserting for K_B the empirical relation[12] $K_B = 9.76(1 + 6.02 Z_2^{-1.19})$ eV, which represents the over-all trend for $Z_2 \geq 13$. In other words, if in Eq. (12) we set $\eta\chi = \eta_0\chi \times (1 + 6.02 Z_2^{-1.19})$, a single value of the constant $\eta_0\chi \approx 0.8$ brings our theory and the experiments on widely different targets into close agreement. When

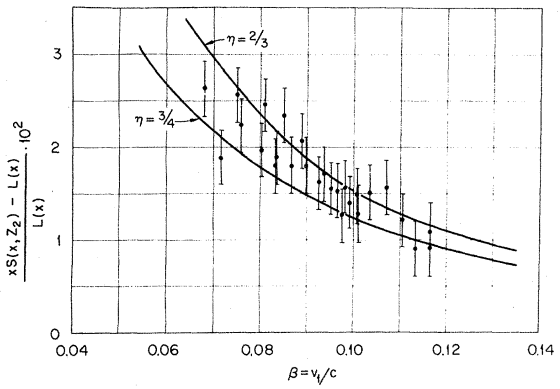

FIG. 4. Theoretical predictions for the stopping-power correction due to the Z_1^3 effect, for $Z_1 = +1$ and values of the parameter $\eta = \frac{2}{3}$, $\frac{3}{4}$, as a function of $\beta = v_1/c$, compared with experimental values from Ref. 3 for Ta($Z_2 = 73$).

$w \ll 1$, the function $F(w)$ [Eq. (14)] depends only logarithmically on w (cf. Fig. 3). That is, in the limit of high-particle velocities our Z_1^3 correction becomes insensitive, as it should, to the choice of η.

We have made a calculation of the difference in stopping power due to the Z_1^3 effect between $Z_1 = +1$ and $Z_1 = -1$ particles in emulsion and find a 8–9% difference at 1.2 MeV/amu, in fair agreement with

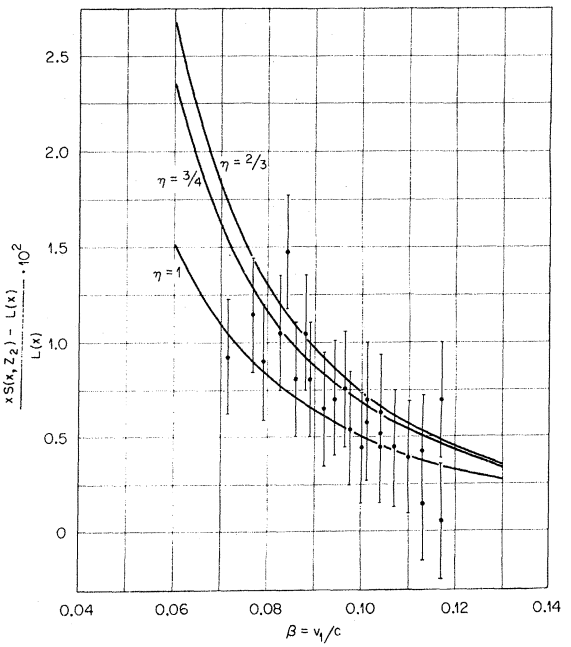

FIG. 5. Theoretical predictions for the stopping-power correction due to the Z_1^3 effect, for $Z_1 = +1$ and values of the parameter $\eta = \frac{2}{3}$, $\frac{3}{4}$, 1, as a function of $\beta = v_1/c$, compared with experimental values from Ref. 3 for Al($Z_2 = 13$).

the estimate derived from π^+ and π^- range data.[2]

Note added in proof. A tabular presentation of the functions I, F, and L is given in a paper to be submitted to Atomic Data by the authors, as yet unpublished.

ACKNOWLEDGMENT

We want to thank V. E. Anderson for his work on the computer evaluation of the various functions required in this theory.

*Research sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corp.

[1]W. H. Barkas, J. W. Dyer, and H. H. Heckman, Phys. Rev. Letters 11, 26 (1963); 11, 138(E) (1963).

[2]H. H. Heckman and P. J. Lindstrom, Phys. Rev. Letters 22, 871 (1969), and references cited therein.

[3]H. H. Andersen, H. Simonsen, and H. Sørensen, Nucl. Phys. A125, 171 (1969).

[4]N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 18, No. 8 (1948).

[5]If the impact parameter separating these two types of collisions is chosen by the criterion that the energy to a free electron at this impact parameter is equal to the binding energy of the struck electron, one obtains a stopping-power formula which is in disagreement with Bethe's quantum-mechanical formula. The disagreement arises from the fact that usually such a choice of intermediate impact parameter violates the uncertainty principle. Proper consideration of the uncertainty principle in our problem requires that we limit the range of a_ω to values larger than $\hbar/mv_1$.

[6]See, e.g., P. Gombas, *Die statishische Theorie des Atoms und ihre Anwendungen* (Springer-Verlag, Berlin, 1949), p. 74.

[7]J. Lindhard and M. Scharff, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 27, No. 15 (1953).

[8]W. Brandt and S. Lundqvist, Phys. Rev. 139, A612 (1965).

[9]J. Lindhard and M. Scharff, in *Penetration of Charged Particles in Matter*, edited by E. A. Uchling, National Academy of Sciences–National Research Council Publication No. 752 (U. S. GPO, Washington, D. C., 1960), p. 49.

[10]W. Brandt, J. Lindhard, and M. Scharff, in *Proceedings of the Second International Conference on the Physics of Electronic and Atomic Collisions, Boulder*, 1961 (Benjamin, New York, 1961), p. 11.

[11]W. Brandt, L. Eder, and S. Lundqvist, J. Quant. Spectry. Radiative Transfer 7, 185 (1967).

[12]R. M. Sternheimer, Phys. Rev. 145, 247 (1966).

[13]See, e.g., E. Bonderup, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 35, No. 17 (1967) for a more detailed treatment of shell corrections using statistical models of the atoms.

PHYSICAL REVIEW B VOLUME 5, NUMBER 7 1 APRIL 1972

Temperature-Independent Spin-Lattice Relaxation Time in Metals at Very Low Temperatures*

F. Bacon, J. A. Barclay,† W. D. Brewer,‡ D. A. Shirley, and J. E. Templeton§
Department of Chemistry and Lawrence Radiation Laboratory, University of California, Berkeley, California 94720
(Received 10 June 1971)

The formalism of nuclear spin-lattice relaxation at low temperatures is developed, leading to a new relaxation time T_μ and a straightforward method of interpreting very-low-temperature relaxation data. Data for ^{60}Co in Fe, Ni, and Co hosts and for ^{56}Co in Fe are summarized. The use of NMR in oriented nuclei for determining relaxation times is discussed, and some comments are made on the role of frequency modulation in NMR experiments with oriented nuclei.

I. INTRODUCTION

Nuclear magnetic resonance in oriented nuclei (NMR/ON), in which resonance is detected through the distribution of nuclear radiations, was suggested by Bloembergen and Temmer[1] and first observed in nuclei oriented by thermal-equilibrium methods by Matthias and Holliday.[2] It was used to study relaxation in ferromagnetic metals,[3] a phenomenon that has also been studied by nonresonant methods.[4]

In 1964 Cameron *et al.*[5] suggested that, for nuclei relaxing in a metal through interaction with conduction electrons, the spin-lattice relaxation time T_1 will approach a constant value at temperatures low enough that the magnetic quantum γH is larger than kT. This effect was observed by Brewer *et al.*, who reported it in abbreviated form in 1968.[6] These authors made a detailed interpretation of their relaxation data in terms of simple rate equations, finding multiexponential decay of the orientation parameters.[7,8] They found that T_1 was no longer a useful relaxation time at very low temperatures, however, and their data in ^{60}Co*Fe*