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Nuclear Spin Diffusion Induced by Paramagnetic Impurities in Nonconducting Solids*

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It is shown that paramagnetic impurities can induce nuclear spin diffusion in nonconducting solids. The component of the impurity spin along the external magnetic field (assumed to be the z axis), because of its interaction with the lattice, fluctuates. The resulting spectral intensity of the magnetic moment has components at all frequencies. The component at zero frequency creates a static magnetic field which is different at two neighboring nuclei, thereby splitting the levels $|\frac{1}{2}, -\frac{1}{2}\rangle$ and $|-\frac{1}{2}, \frac{1}{2}\rangle$, where the first and second quantum numbers refer, respectively, to the z components of two neighboring spins. These are exact two-spin eigenfunctions if the interaction of the nuclear spins is neglected. When this is taken into account, the correct eigenfunctions to first order are $\psi_1 = |\frac{1}{2}, -\frac{1}{2}\rangle + \epsilon |-\frac{1}{2}, \frac{1}{2}\rangle$ and $\psi_2 = |-\frac{1}{2}, \frac{1}{2}\rangle - \epsilon |\frac{1}{2}, -\frac{1}{2}\rangle$, where ϵ is a small number. The Fourier component of the impurity spin at the frequency corresponding to the energy difference of ψ_1 and ψ_2 causes transitions between these states. This is a spin-diffusion process because ϵ is small. This means that Bloembergen's differential equation for nuclear spin-lattice relaxation in nonconducting solids must be generalized to include nuclear spin diffusion inside the critical radius.

I. INTRODUCTION

Bloembergen[1] first showed the importance of paramagnetic impurities in nuclear spin-lattice relaxation in nonconducting solids. His theory states that nuclear relaxation is the result of two mechanisms. One is the direct relaxation of nuclei by paramagnetic impurities and the other is nuclear spin diffusion,[2] which arises from the nuclear dipole-dipole interaction and is on the order of 10^{-12} $\mathrm{cm^2/sec}$.

The spins closest to the impurity feel the greatest direct relaxation rates and this creates a gradient in the magnetization. Spin diffusion transports the magnetization throughout the sample, thereby smoothing out the gradient and increasing the relaxation rate. Bloembergen set up the differential equation that describes this relaxation process:

$$\frac{\partial m}{\partial t} = D\nabla^2 m - \frac{C}{r^6}\, m \,. \tag{1}$$

m is the difference between the instantaneous value of the magnetization and the equilibrium value of the magnetization, D is the nuclear-spin-diffusion coefficient, r is the distance of the nuclear spins from the paramagnetic impurity, and C is a coefficient that describes the effect of direct relaxation. He introduced the boundary condition that spin diffusion vanishes inside a critical radius r_c. This radius is where the static field of the impurity spin splits adjacent nuclear spin levels by an amount greater than their linewidth, thereby making the spin-diffusion process nonenergy conserving, and causing it to vanish.

In this paper, a spin-diffusion mechanism that is important inside the critical radius is derived. The mechanism is the resonant field created by the impurity spin at the frequency necessary to induce mutual spin flips between adjacent nuclear spins. This means that Bloembergen's differential equation (1) must be generalized to include spin diffusion inside the critical radius.

The correlation function and the spectral intensity of the expectation value of the impurity spin are needed for the calculation of the spin-diffusion mechanism, and are derived in the Appendix.

II. NUCLEAR SPIN DIFFUSION INSIDE CRITICAL RADIUS

Consider a nonconducting solid consisting of molecules of nuclear spin $\frac{1}{2}$, and having a small number of paramagnetic impurities of magnetic moment μ_p. The sample is in an external magnetic field H_0 along the z axis.

Spin diffusion in the nuclear spin system occurs when two adjacent nuclear spins undergo transitions from the state $|\frac{1}{2}, -\frac{1}{2}\rangle$ to the state $|-\frac{1}{2}, \frac{1}{2}\rangle$, or vice versa. The first and second quantum numbers refer, respectively, to the z component of two neighboring spins i and j.

The two nuclear spins interact with each other through the nuclear dipole-dipole interaction

$$\mathcal{H}_d = (\vec{\mu}_i \cdot \vec{\mu}_j)/a^3 - 3(\vec{\mu}_i \cdot \vec{a})(\vec{\mu}_j \cdot \vec{a})/a^5,$$

where μ_i and μ_j are the nuclear magnetic moments of i and j and $\vec{a}$ is the radius vector between the two nuclei. The effect of the other nuclear spins is to cause a local variation in the nuclear Larmor frequency.

The spins i and j also interact with the paramagnetic impurity. The impurity spin is assumed to be so tightly coupled to the lattice that the nuclear spin system has no effect on its behavior. This allows us to treat the fields created by the fluctuating impurity spin as external fields. (A Fourier analysis of the magnetic fields created by the impurity spin is in the Appendix.) We denote the static field created by the paramagnetic impurity at the nuclear spin i, for example, as $\vec{H}_i(0)$ and the Fourier component at ω as $\vec{H}_i(\omega)$. The scalar quantities $H_i(0)$ and $H_i(\omega)$ denote components along the z axis.

The Hamiltonian incorporating the interaction of the two nuclear spins with the external magnetic field, with each other and with the paramagnetic impurity is

$$\mathcal{H} = -\mu_{iz}[H_0 + H_i(0)] - \mu_{jz}[H_0 + H_j(0)] + \mathcal{H}_d - [\vec{\mu}_i \cdot \vec{H}_i(\omega) + \vec{\mu}_j \cdot \vec{H}_j(\omega)] \cos\omega t. \tag{2}$$

The Zeeman term

$$\mathcal{H}_0 = -\mu_{iz}[H_0 + H_i(0)] - \mu_{jz}[H_0 + H_j(0)],$$

which is the sum of the energy of spins i and j in their respective total static magnetic fields, is the zeroth-order part of the Hamiltonian. $|\frac{1}{2}, -\frac{1}{2}\rangle$ and $|-\frac{1}{2}, \frac{1}{2}\rangle$ are eigenfunctions of $\mathcal{H}_0$. In the absence of $H_i(0)$ and $H_j(0)$ they would be degenerate eigenfunctions.

$\mathcal{H}_d$ will affect these two-spin eigenfunctions to first order by mixing them. To first order, the eigenstates are found to be

$$\psi_1 = |\tfrac{1}{2}, -\tfrac{1}{2}\rangle - \epsilon|-\tfrac{1}{2}, \tfrac{1}{2}\rangle,$$
$$\psi_2 = |-\tfrac{1}{2}, \tfrac{1}{2}\rangle + \epsilon|\tfrac{1}{2}, -\tfrac{1}{2}\rangle, \tag{3}$$

where $\epsilon = \gamma_n^2 \hbar\,(1 - 3\cos^2\theta_{ij})/4a^3(\omega_i - \omega_j)$. θ_{ij} is the angle between the z direction and $\vec{a}$. ω_i and ω_j are the Larmor frequencies of spins i and j in their respective instantaneous total static magnetic fields. The total static field at any nucleus is the sum of H_0 and the static field created at the nucleus by the neighboring spins. At its largest, $\epsilon \simeq \frac{1}{5}$. As r decreases to zero, ϵ decreases.

Because ϵ is small, diffusion is essentially transitions between the states ψ_1 and ψ_2. The perturbing term in the Hamiltonian

$$\mathcal{H}_{\text{pert}} = -[\vec{\mu}_i \cdot \vec{H}_i(\omega) + \vec{\mu}_j \cdot \vec{H}_j(\omega)] \cos\omega t$$

can induce such transitions. This term is the interaction of the nuclear spins with the time-dependent fields created by the impurity spin.

According to first-order time-dependent perturbation theory, a Fourier component of $\mathcal{H}_{\text{pert}}$ at a frequency ω will create a transition probability W:

$$W = \frac{\Pi}{2\hbar^2}|\langle\psi_2|\mathcal{H}_{\text{pert}}(\omega)|\psi_1\rangle|^2\,\delta(\omega_j - \omega_i - \omega) = \frac{\Pi\gamma_n^6\hbar^2(1 - 3\cos^2\theta_{ij})^2[H_j(\omega) - H_i(\omega)]^2\,\delta(\omega_j - \omega_i - \omega)}{32a^6(\omega_j - \omega_i)^2}. \tag{4}$$

Let us express, using (A10) of the Appendix:

$$[H_j(\omega) - H_i(\omega)]^2 = \frac{J_z(\omega)}{a^6}\left(\frac{1}{n_j^3} - \frac{1}{n_i^3}\right)^2 = \frac{2\tau_c}{a^6}\left(\frac{1}{n_j^3} - \frac{1}{n_i^3}\right)^2 \frac{\langle\mu_p^2\rangle - \langle\mu_p\rangle^2}{1 + \omega^2\tau_c^2}, \tag{5}$$

where n_i and n_j are the number of lattice spacings, nuclei i and j are away from the paramagnetic center, $J_z(\omega)$ is the intensity of the expectation value of the impurity spin at ω, and τ_c is the spin-lattice relaxation time of the impurity spin.

The nuclei neighboring i and j cause the local static field to change over time. This means that Eq. (4) must be integrated over the line shape $f(\omega)$ of nuclei i and j. After taking the line shape into account and using (5), we find for the total transition probability W_T:

$$W_T=\frac{\Pi\gamma_n^6\hbar^2(1-3\cos^2\theta_{ij})^2\tau_c(\langle\mu_p^2\rangle-\langle\mu_p\rangle^2)}{16a^{12}}\left(\frac{1}{n_j^3}-\frac{1}{n_i^3}\right)^2$$
$$\times\int_0^\infty d\omega_i f(\omega_i)\int_0^\infty\frac{d\omega_j f(\omega_j)}{(\omega_j-\omega_i)^2[1+(\omega_j-\omega_i)^2\tau_c^2]}\,. \tag{6}$$

It is sufficient to evaluate (6) using a square shape of width ΔH for $f(\omega)$, because the difference between the contributions of various line shapes to W_T is on the order of unity. The line shape for nucleus i is centered at $\omega_{i0}=\gamma_n[H_0+H_i(0)]$, and for nucleus j it is centered at $\omega_{j0}=\gamma_n[H_0+H_j(0)]$. After evaluating (6) with a square line shape and averaging over angles, whereby $\langle(1-3\cos^2\theta_{ij})^2\rangle_{\text{av}}=\frac{4}{5}$, we obtain for the induced spin-diffusion coefficient D_p:

$$D_p=W_Ta^2=\frac{\Pi\gamma_n^6\hbar^2\tau_c(\langle\mu_p^2\rangle-\langle\mu_p\rangle^2)}{20a^{10}}\left(\frac{1}{n_j^3}-\frac{1}{n_i^3}\right)^2 I, \tag{7}$$

where

$$I=\left(\frac{1}{\gamma_n\Delta H}\right)^2\left[2\alpha\tan^{-1}\alpha-\beta\tan^{-1}\beta-\gamma\tan^{-1}\alpha\right.$$
$$\left.+\ln\left(\frac{\alpha^2}{1+\alpha^2}\right)+\frac{1}{2}\ln\left(\frac{1+\beta^2}{\beta^2}\right)+\frac{1}{2}\ln\left(\frac{1+\gamma^2}{\gamma^2}\right)\right]. \tag{8}$$

In the above expression $\alpha=\gamma_n\tau_c[H_j(0)-H_i(0)]$, $\beta=\gamma_n\tau_c[H_j(0)-H_i(0)+\Delta H]$, and $\gamma=\gamma_n\tau_c[H_j(0)-H_i(0)-\Delta H]$.

This result is less complicated than it looks, for often one of two limits applies. One is the short-correlation time region $\alpha,\beta,\gamma\ll 1$, where I reduces to $I\simeq 1/(\omega_{j0}-\omega_{i0})^2$. (Generally, this limit applies when $\tau_c\lesssim 10^{-5}$ sec.) The other limit is the region of long-correlation times $\alpha,\beta,\gamma\gg 1$, where $I\simeq 1/\tau_c^2(\omega_{j0}^2-\omega_{i0}^2)^4$. (This limit usually applies when $\tau_c>10^{-5}$ sec.)

TABLE I. Table of D_p/D, the ratio of spin diffusion induced inside r_c divided by Bloembergen's spin-diffusion coefficient. The critical radius for these experiments is $r_c=4a\simeq 10$ Å. The τ_c data used to calculate D_p are taken from Ref. 3. The numbers labeled r represent the number of lattice spacings, na, the nucleus is from the Ce^{3+} ion.

$r=na$	D_p/D	T(°K)	τ_c(sec)
$4a$	0.02	2	1.5×10^{-3}
$3a$	0.2	2	1.5×10^{-3}
$2a$	0.2	2	1.5×10^{-3}
a	0.2	2	1.5×10^{-3}
$4a$	4.3	5	7×10^{-4}
$3a$	15.0	5	7×10^{-4}
$2a$	19.3	5	7×10^{-4}
a	0.1	5	7×10^{-4}
$4a$	1.3	10	2×10^{-4}
$3a$	1.4	10	2×10^{-4}
$2a$	1.7	10	2×10^{-4}
a	4.4	10	2×10^{-4}

To obtain some idea of the magnitude of D_p, it is helpful to compare Eq. (7) with Bloembergen's spin-diffusion coefficient D which Lowe and Gade[2] found to be

$$D=0.15\,\gamma_n^2\hbar/a. \tag{9}$$

We will take as an example spin diffusion induced in CaF_2 by Ce^{3+} paramagnetic impurities with a magnetic moment $\mu_p=2\times10^{-20}$ cgs. The linewidth of the fluorine nuclei is approximately $\Delta H=3$ G, the lattice constant is $a=2.7$ Å, and $\gamma_n=2.5\times10^4$ cgs. Bierig, Weber, and Warshaw[3] measured τ_c of the Ce^{3+} ions in the temperature range $2<T<15$ °K in an external magnetic field of 1500 G.

The so-called barrier radius inside of which Bloembergen's diffusion coefficient vanishes, is determined by equating $H_j(0)-H_i(0)$ to ΔH. For these experiments this gives a value of $r_c\simeq 10$ Å, which corresponds to n_j or n_i equal to 4. Thus, Bloembergen's spin-diffusion coefficient between molecules 4-3, 3-2, and 2-1 vanishes (where the notation n_i-n_j means adjacent fluorine nuclei n_ia and n_ja away from the Ce^{3+} impurity).

In Table I we show that such spin-diffusion transitions can be induced by the Ce^{3+} ion. The results, which are calculated using the data of Ref. 3, are presented in the form of the ratio of D_p/D. We note that a nucleus can have spin-diffusion transitions with nuclei other than its nearest neighbor. It can also have mutual spin flips with its next-nearest neighbor, for example, or with nuclei even further away. Thus, in the column labeled D_p/D, the results correspond to the most important D_p of all spin-diffusion transitions that a nucleus can make inside r_c.[4] For example, for the nucleus at $n_j=4$, the ratio D_p/D is calculated using the largest D_p among the possible spin-diffusion transitions 4-3, 4-2, and 4-1. (Though a more exact answer would be obtained by summing up the D_p for all the transitions, generally one of the transitions is so much larger than the others that only it need be considered.)

The data in Table I show that at $T=5$ and 10 °K the spin diffusion D_p is greater inside r_c than outside r_c. At $T=2$ °K there is spin diffusion inside r_c but it is smaller than that outside r_c and on the order of $D/5$.

As another example, consider Leifson and Jeffries's data[5] on the spin-lattice relaxation of Ce^{3+} in $(Ce, La)_2Mg_3(NO_3)_{12}\cdot 24H_2O$ in a magnetic field of 3600 G and in the temperature range $2<T<2.7$ °K. The linewidth of the protons is $\Delta H=3$ G, the lattice constant of the protons is $a = 3$ Å, $\gamma_n = 1.66 \times 10^4$ cgs, and the magnetic moment of the Ce^{3+} ions is $\mu_p = 1.62 \times 10^{-20}$ cgs. Under these experimental conditions, $r_c \simeq 12$ Å, which corresponds to n_j or n_l equal to 4.

In Table II we present the ratio D_p/D calculated using these data. These calculations show that at $T = 2$ °K, except for the nucleus at $2a$, the spin diffusion inside r_c is greater than it is outside. At $T = 2.7$ °K, there is spin diffusion inside r_c; however, it is smaller than at 2.0 °K and is on the order of $D/5$.

These results show that a paramagnetic impurity induces spin diffusion inside the critical radius. This means that Bloembergen's differential equation describing the transportation of the magnetization, Eq. (1), must be evaluated with the boundary condition that there is one spin-diffusion coefficient outside the critical radius and another one inside.

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APPENDIX

In this Appendix[6] we derive the correlation function and the spectral intensity of the expectation value of the z component of the impurity spin. The spectral intensity is needed to find the total static and time-dependent magnetic fields created by the impurity spin.

Consider a spin-$\frac{1}{2}$ impurity with magnetic moment μ_p in an external magnetic field H along the z axis. We assume the evolution of the z component of the spin to be a Markov process.[7] Thus, the correlation function $G(\tau)$ of the expectation value of the z component can be expanded:

$$G(\tau) = \overline{\langle\mu_p(0)\rangle\,\langle\mu_p(\tau)\rangle} = p_1(0)\langle\mu_p(0)\rangle_1\langle\mu_p(\tau)\rangle_1 + p_2(0)\langle\mu_p(0)\rangle_2\langle\mu_p(\tau)\rangle_2 \,. \tag{A1}$$

The bar represents an ensemble average, the brackets represent expectation values, and $\langle\mu_p(\tau)\rangle_1$ and $\langle\mu_p(\tau)\rangle_2$ are $\langle\mu_p(\tau)\rangle$ with the initial conditions $\langle\mu_p(0)\rangle_1 = -\mu_p$ and $\langle\mu_p(0)\rangle_2 = +\mu_p$. The quantities $p_1(0)$ and $p_2(0)$ are the probabilities that at $t = 0$, the spin is in state 1 $(+\frac{1}{2})$ or state 2 $(-\frac{1}{2})$. For a solid, the spins obey Boltzmann statistics. Therefore, $p_1(0) = e^{\mu_p H/kT}/Z$ (where Z is the partition function and k is Boltzmann's constant) and $p_2(0) = e^{-\mu_p H/kT}/Z$, and the correlation function can be written

$$G(\tau) = \frac{e^{-\mu_p H/kT}}{Z}\langle\mu_p(\tau)\rangle_1 - \frac{e^{\mu_p H/kT}}{Z}\langle\mu_p(\tau)\rangle_2 \,. \tag{A2}$$

$\langle\mu_p(\tau)\rangle_1$ and $\langle\mu_p(\tau)\rangle_2$ are evaluated from the rate equations for the two states,

$$\frac{dp_1}{dt} = -p_1\, W\!\downarrow + p_2\, W\!\uparrow \,, \quad \frac{dp_2}{dt} = -p_2\, W\!\uparrow + p_1\, W\!\downarrow \,. \tag{A3}$$

TABLE II. Table of D_p/D, the ratio of spin diffusion induced inside r_c divided by Bloembergen's spin-diffusion coefficient. The critical radius for these experiments is $r_c = 4a \simeq 12$ Å. The τ_c data used to calculate D_p are taken from Ref. 5. The numbers in the column labeled r represent the number of lattice spacings, na, the nucleus is from the Ce^{3+} ions.

$r = na$	D_p/D	T(°K)	τ_c(sec)
$4a$	0.1	2.7	10^{-2}
$3a$	0.2	2.7	10^{-2}
$2a$	0.3	2.7	10^{-2}
a	0.01	2.7	10^{-2}
$4a$	2.1	2.0	10^{-4}
$3a$	3.2	2.0	10^{-4}
$2a$	0.01	2.0	10^{-4}
a	5.1	2.0	10^{-4}

$W\!\uparrow$ and $W\!\downarrow$ are, respectively, the probabilities that a spin in state 2 will go to state 1 and that a spin in state 1 will go to state 2. p_1 and p_2 are the probabilities that states 1 and 2 are occupied.

The lattice is in thermal equilibrium. In order that the relaxing spins also reach thermal equilibrium, it is necessary that the relaxation rate from the higher energy levels to the lower ones be greater than the transition rates from the lower levels to the higher ones. For a spin-$\frac{1}{2}$ system this amounts to

$$W\!\uparrow / W\!\downarrow = e^{-2\mu H/kT} \,. \tag{A4}$$

Equations (A3) can be rewritten using (A4):

$$\frac{dp_1}{dt} = -Wp_1 + Wp_2 e^x \,, \quad \frac{dp_2}{dt} = Wp_1 - We^x p_2 \,, \tag{A5}$$

where $x = -2\mu_p H/kT$ and $W\!\downarrow = W$. The solutions of these equations are of the form $p_1 = A\, e^{\lambda t}$ and $p_2 = B\, e^{\lambda t}$ with the conditions $p_1(t) + p_2(t) = 1$. This leads to a complete solution:

$$\begin{aligned} p_1(t) &= \frac{1}{1+e^{-x}} + A_1 e^{-W(e^x+1)t} \,, \\ p_2(t) &= \frac{e^{-x}}{1+e^{-x}} - A_1 e^{-W(e^x+1)t} \,. \end{aligned} \tag{A6}$$

$\langle\mu_p(\tau)\rangle_1$ and $\langle\mu_p(\tau)\rangle_2$ are evaluated using these solutions with the appropriate initial conditions. For example, since initially $\langle\mu_p(0)\rangle_1 = -\mu_p$,

$$\langle\mu_p(\tau)\rangle_1 = \mu_p p_1(\tau) - \mu_p p_2(\tau). \tag{A7}$$

The initial conditions are $p_1(0) = 1$ and $p_2(0) = 0$. Consequently, $A_1 = e^{-x}/(1+e^{-x})$. Equations (A6) and (A7), evaluated with this value of A_1, yield

$$\begin{aligned} \langle\mu_p(\tau)\rangle_1 &= \frac{\mu_p(1-e^{-x})}{1+e^{-x}} + \frac{2\mu_p e^{-x} e^{-W(1+e^x)t}}{1+e^{-x}} \,, \\ \langle\mu_p(\tau)\rangle_2 &= \frac{\mu_p(e^{-x}-1)}{1+e^{-x}} + \frac{2\mu_p e^{-W(e^x+1)t}}{1+e^{-x}} \,. \end{aligned} \tag{A8}$$

After Eq. (A2) has been evaluated with the results of Eqs. (A8) and separated into constant and time-dependent parts, the correlation function can be written

$$G(\tau)=\langle\mu_p\rangle^2+(\langle\mu_p^2\rangle-\langle\mu_p\rangle^2)e^{-\tau/\rho}\,. \tag{A9}$$

The brackets $\langle\,\rangle$ in Eq. (A9) signify the statistical mechanical expectation value. The relationship $1/\rho=W\uparrow+W\downarrow=W(e^{-x}+1)$ was used in obtaining (A9). It is evident from (A9) that the correlation time τ_c is equal to the spin-lattice relaxation time ρ. The spectral density is defined as

$$J_z(\omega)=\int_{-\infty}^{\infty}d\tau\;e^{i\omega\tau}\,G(\tau).$$

Thus

$$J_z(\omega)=\langle\mu_p\rangle^2\;2\pi\delta(\omega)+\frac{(\langle\mu_p^2\rangle-\langle\mu_p\rangle^2)2\tau_c}{1+\omega^2\tau_c^2}\;. \tag{A10}$$

The methods used here to find $G(\tau)$ and $J_z(\omega)$ for spin-$\frac{1}{2}$ particles can be applied to any spin. However, the solutions for higher spins become more complicated as more equations are involved [e.g., Eq. (A3) would have $2S+1$ equations].

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Charge Screening of Impurities in Gold Observed by the Mössbauer Isomer Shift

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The isomer shift of Au^{197} in Au-Hg alloys containing up to 12% Hg has been measured. The change produced by the Hg is extremely small, five times smaller than that produced by any solute studied previously. This result is compared with those in other gold alloys and compounds. A microscopic theoretical calculation based on a partial-wave model is successful in explaining this anomaly.

I. INTRODUCTION

The isotope Au^{197} has been extensively used in Mössbauer-effect research in a wide range of systems. The two areas of research that have been investigated most intensively are the magnetic hfs of the gold nucleus produced by the hyperfine field (coming primarily from a contact interaction resulting from conduction-electron polarization) occurring in compounds of gold with magnetic atoms[1-3] and gold dissolved in magnetic metals,[4-6] and the "isomer shift," which determines the total electronic density at the gold nucleus.

Gold has been a widely used material for isomer-shift studies for two reasons: First, there is a large body of information on gold chemistry and metallurgy, so that there are large families of well-established materials which can be studied. Second, the strong Mössbauer absorption and large range of observed isomer shifts (about five times the experimental linewidth) make it relatively easy to make meaningful measurements on all of these gold-containing systems.

A detailed analysis of the isomer shift in insulating gold compounds has recently been given.[7] The extensive work done previously in metallic systems can perhaps best be characterized in terms of increasing sophistication as both experimental and theoretical techniques developed. Almost ten years ago, an extensive study was performed on the isomer shift of gold dissolved as a dilute impurity in a wide range of metallic hosts.[8] The conclusion of that work was that a correlation could be obtained between isomer shift and electronegativity of the host, suggesting that the increased isomer shift (corresponding to increased s-electron density at the gold nucleus) resulted from the less-electronegative hosts giving up electrons to the gold ion.