Internal Magnetic Fields at Pt Nuclei in Pt-Fe Alloys

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The Mössbauer effect of the 99-keV, $\frac{3}{2}^-$-to-$\frac{1}{2}^-$ transition in Pt^{195} has been measured, using a single-line source of Au^{195} in Cu and ferromagnetic Fe-Pt alloy absorbers. The internal field at the Pt nucleus, $H_{int}(Pt)$, in the alloy absorbers was determined and found to be nearly constant, about 1.26×10^6 Oe, over a range of composition from 3 to 30 at.% Pt. $H_{int}(Pt)$ for an $Fe_{0.50}Pt_{0.50}$ alloy absorber and for a source ($Au^{195}\rightarrow Pt^{195}$ in Fe) are smaller, $H_{int}(Pt)\simeq1.0\times10^6$ and 0.85×10^6 Oe, respectively. The g factor of the state is found to be -0.40 ± 0.10 nm.

INTRODUCTION

THE internal field at the nucleus of a diamagnetic solute, $H_{int}(sol)$, in ferromagnetic environments has been the subject of considerable recent study; a summary of results for dilute concentrations in Fe, Co, and Ni hosts is contained in the papers of Shirley and Westenbarger[1] and of Shirley.[2] The effect of solute concentration on $H_{int}(sol)$ has, however, been investigated in only a few systems, for example, Au-Ni[3] and W-Fe.[4] The Pt_xFe_{1-x} alloys are particularly convenient for such studies since they are easily formed over a wide range of x; and the Mössbauer effect of the 99 keV, $\frac{3}{2}^-$ to $\frac{1}{2}^-$ transition in Pt^{195} provides a convenient probe for the hyperfine interaction even though the lifetime of the level is short, $\tau=230\pm20$ psec, and the hyperfine pattern is unresolved.

The Mössbauer effect using the 99-keV transition in Pt^{195} in various diamagnetic source and absorber materials has been described in a separate paper.[5] The emphasis there was on Debye temperatures, linewidths, and the "recoilless" fractions for various environments; some results and expectations for the 129-keV transition were also discussed. Sources of Au^{195} in Cu and Ir, also used in the present work, gave natural linewidths and resonant absorption spectra of the order of several percent at 4.2°K. In this paper we report on $H_{int}(Pt)$ in the PtFe alloy absorbers ranging in composition from $Pt_{0.03}Fe_{0.97}$ to $Pt_{0.50}Fe_{0.50}$. A measurement of $H_{int}(Pt)$ using a source of carrier free Au^{195} diffused in iron is briefly described. Our analysis of the Mössbauer spectra also yields the gyromagnetic ratio g_1 of the $\frac{3}{2}^-$, 99-keV state. Following a brief description of the alloy preparation and the apparatus, we discuss the results of the alloy absorbers (1–5), the results with the Au^{195} source, and finally the magnetic moment of the $\frac{3}{2}^-$ state. The internal fields at the Pt nuclei have been used for angular correlation precession measurements of the g factor of collective states in $Pt^{192,194,196}$. The work will be reported in a subsequent paper.[6] Those aspects of the Mössbauer studies analysis which bear on the precession measurement will be commented on here.

The present analysis of the observed hyperfine spectra differs little from a preliminary report of this work.[7] A similar study of $H_{int}(sol)$ has also been described by Benczer-Koller *et al.*,[8] and more recently by Agresti *et al.*[9]

ALLOY PREPARATION AND METHOD

Five alloys were prepared using iron and platinum wire of high purity, these were melted together in a water-cooled copper hearth argon arc furnace. The samples were remelted several times until no gross segregation could be found metallographically. Table I presents the relevant history of each sample. The $Pt_{0.50}Fe_{0.50}$ sample was cold-pressed, machined, and ground to the shape of a disc. It was then heat-treated for 1 h in purified helium at 800°C and quenched to

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TABLE I. Characteristics of the specimens.

Specimen[a]	Wt% Pt	at. % Pt	Thickness of Pt (mg/cm²)	No. of remelts	Treatment before machining and grinding	Heat treatment in purified He[b]	H_{int} (Pt) (10^6kOe) at 4°K
1, A	8	3	191	4	Hot-forged to disc shape	1300°C for 1 h; slow cooled to R.T.	1.26±0.10
2, A	27	10	131	15	Hot-forged to disc shape	1300°C for 1 h; slow cooled to R.T.	1.26±0.10
3, A	54	25	268	15	Cold-pressed to disc shape	800°C for 1 h and quenched to R.T.	1.24±0.10
4, A	60	30	295	15	Cold-pressed to disc shape	1000°C for 1 h; slow cooled to R.T.	1.38±0.12
5, A	78	50	655	50	Hot-forged to disc shape	800°C for 1 h and quenched to R.T.	~1.1
6, S		Carrier free Au^{195} in Fe	Annealed for 8 h at 740°C in H_2				0.85±0.12
6a, S		Sample 6, reannealed for 50 h at 720°C in argon					0.72±0.12

[a] A, Absorber; S, Source. [b] R.T. = room temperature.

room temperature. Further analysis for homogeneity was performed on these alloys using the electron-microbeam-probe technique; no concentration gradients, precipitates, or inclusions were detected. The Mössbauer spectra of these absorbers were obtained for sample temperatures near 4.2°K. At this temperature, any fcc γ phase fraction present in samples 3 and 4 should all have transformed into the bcc α phase.[10]

The data were accumulated on a multichannel analyzer using a constant acceleration spectrometer. Earlier runs[7] on these samples using a mechanical, constant velocity drive system yielded similar spectra.

RESULTS

The Mössbauer absorption spectra for samples 1 through 4 are shown in Fig. 1. The spectra are similar, showing a double-line pattern with a splitting nearly independent of composition. The splitting is dominated by the large g value of the ground state,[11] $g_0=1.20$, so that the value of H_{int}(Pt) is determined with greater precision than is the value of g_1.

Using the fact that g_1 is negative (see below), six-line spectra were simulated by computer for various values of H_{int}(Pt), g_1, and line width; each line was assumed to have Lorentzian shape. The simulated spectra were compared with the 3 at.% Pt alloy curve of Fig. 1(a) to determine the best values of H_{int}(Pt) and g_1. Figure 2 shows the data of Fig. 1(a) together with the best computer curve (solid line) and the two extreme fits (dashed lines). Since both H_{int} and g_1 must be found from an only partially resolved spectrum, a graphical method for determining the best fit and the limits of fit was employed. Two experimental parameters were used, the apparent splitting Δ, and the ratio of trough to effect, $R=[N(A)-N(B)]/[N(C)-N(B)]$, where A, B, and C are the points indicated on Fig. 2. Two plots of R versus Δ for a wide range of values of g_1 and H are shown in Fig. 3 for individual line widths of 1.88 and 2.00 cm/sec. The smaller line width corresponds to a Debye temperature for the alloy absorber equal to that of Pt metal, about 240°K. The larger width corresponds to a Debye temperature for the alloy of 350°K. The experimental limits on R and Δ yield the following values:

$$g_1=(-0.40\pm0.10)\,\text{nm},$$

$$H=(1.26\pm0.10)\times10^6\ \text{G}.$$

The values for H_{int}(Pt) for the other alloys are listed in Table I; the sign is determined experimentally and is in agreement with systematics.[1,2]

Our conclusion for $g(\frac{3}{2}^-)$ agrees, within the rather large errors, with that obtained by Benczer-Koller *et al.*, who obtained $-0.60\leq g\leq-0.47$ by a Mössbauer experiment on $Fe_{0.90}Pt_{0.10}$. Their upper limit (-0.47) was set by the Ho and Phillips[12] calorimetric determination of the hyperfine field on Pt dissolved in Fe in a $Pt_{0.03}Fe_{0.97}$ sample: $H=1.39\times10^6$ Oe. Examination of the Benczer-Koller curve shows a larger splitting, greater width, and higher resolution than we obtained for a similar sample [Fig. 1(b)]. This indicates that the internal field in their sample was greater than the internal field in our sample. We cannot explain this discrepancy in internal field.

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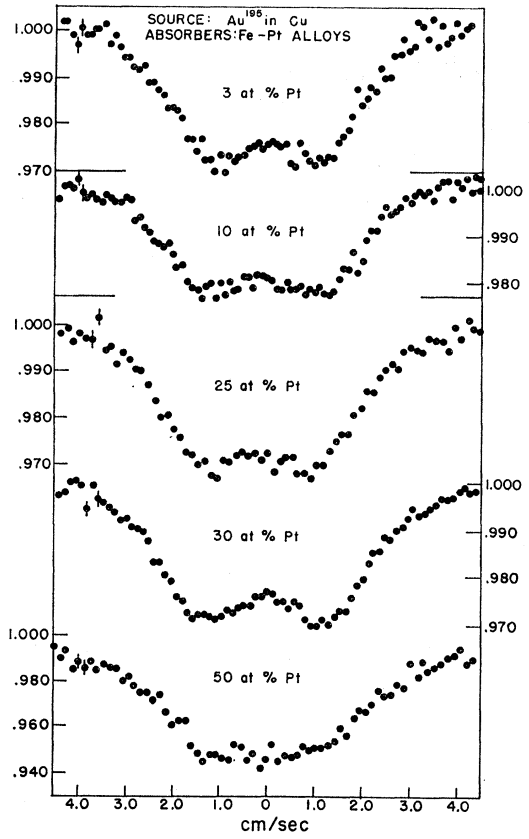

FIG. 1. Mössbauer absorption spectra for Fe-Pt alloy absorbers and Au^{195} in Cu source, taken at 4.2°K.

To check the interpretation of the spectra, the absorber was magnetized parallel to the propagation direction of the γ rays (perpendicular to the absorber foil plane) by an external field of 45 kOe produced by a Bitter-type solenoid. The 99-keV radiation was detected with a $1\times1.5\times0.4$ cm Ge(Li) detector having a resolution of 9 keV at 100 keV. (The poor resolution was due to cable capacitance occasioned by the necessity for keeping the low noise, vacuum tube, preamplifier

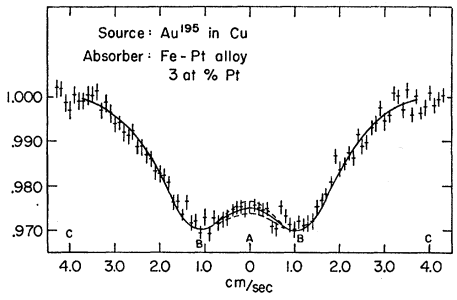

FIG. 2. The Mössbauer absorption spectrum taken at 4.2°K for the Au^{195} in Cu source and the 3 at.% Pt Fe-Pt alloy absorber [data of Fig. 1(a)], showing best computer fit (solid curve) and two extreme fits (dashed lines).

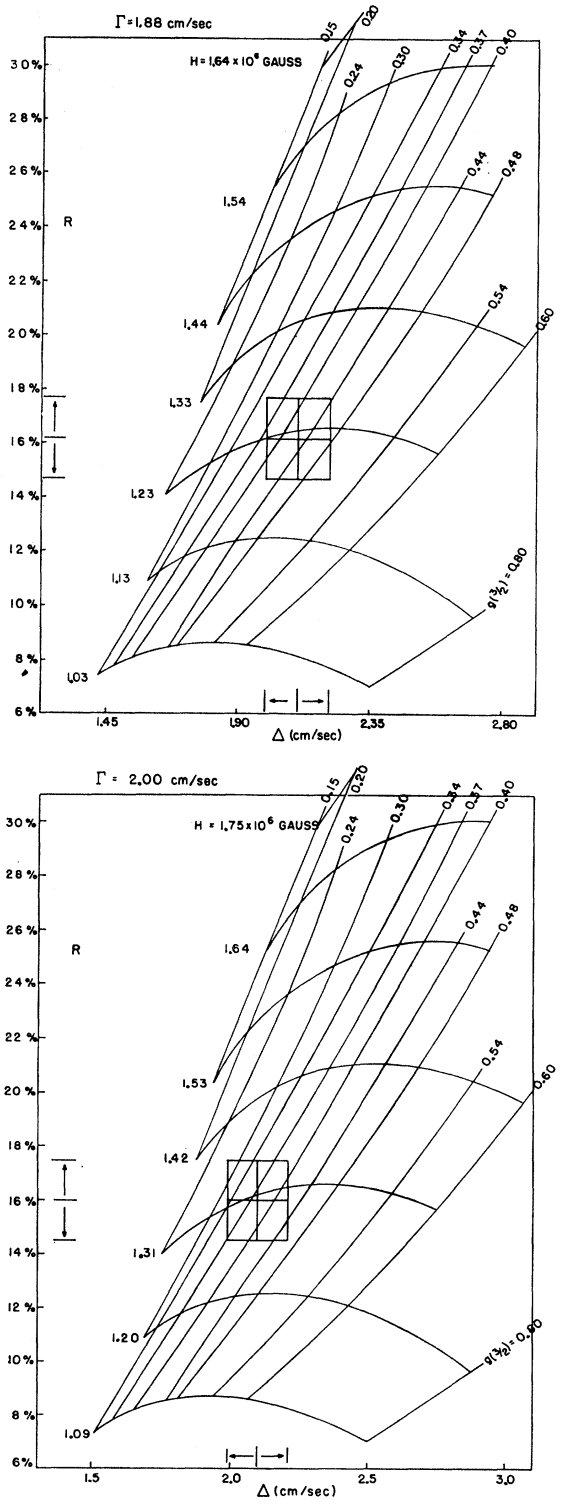

FIG. 3. Resolution $R=[N(A)-N(B)]/[N(C)-N(B)]$ (see Fig. 2) versus apparent splitting Δ for a wide range of g_1 and H_{int}(Pt) for (a) individual line widths, $\Gamma=1.88$ cm/sec. (b) individual line widths, $\Gamma=2.00$ cm/sec. The boxes show the limits of fit for the data of Fig. 1(a).

more than 1 m from the solenoid in order to avoid the fringing field of the magnet, and partly from microphonic noise from vibrations arising from cooling water flow.) Tests of the resolution of the detector (the plane of which was perpendicular to the field direction) as a function of magnetic field, showed that the resolution did not deteriorate and may have improved as the magnetic field on the sample was raised to about 70 kOe. The longitudinal polarization eliminates the $\Delta m=0$ transitions; the intensity pattern for $g_1/g_0>0$ should be 1:3:3:1, while for $g_1/g_0<0$ it should be 3:1:1:3. The spectrum shown in Fig. 4, is unresolved, but the splitting is increased by 18% upon polarization, indicating that the g values have opposite signs; this result is expected from the single particle assignments of the levels. The spectrum is consistent with the values for g_1 and $H_{\text{int}}(\text{Pt})$ states above.

Spehl *et al.*[13] have determined the $g(2+)$ value for the 328-keV state of Pt^{194} by precessing a Coulomb excitation angular distribution with an external magnetic field; they find that $g(2+)=+0.33_{-0.10}^{+0.15}$. The precession of a γ-γ angular correlation through this same state was measured[6,14] using the internal field corresponding to alloy No. 1. The product $g(2+)H_{\text{int}}(\text{Pt})$ is negative, hence $H_{\text{int}}(\text{Pt})$ is also negative. (We obtain[6] $g(2+)\text{Pt}^{194}=0.32\pm0.05$.) The sign of the precession using internal fields has been confirmed by measurements using the implantation technique.[15]

The Mössbauer absorption spectrum for sample 5 is also shown in Fig. 1. Allowing for line broadening in the thick sample, the data could be analyzed on the basis of a single field and natural linewidth. The internal field value obtained with this assumption is ~1.0 mOe. No error is assigned, since the spectrum is in fact somewhat better fit by a multivalued internal field. In any case, the width of the curve demands a large average magnetic field. A quadrupole interaction could be expected for this tetragonal[16] alloy. However, even a $qQ\sim5\times10^{-17}$ V/cm^2 would broaden each line by less than 3.0% and produce practically no assymetry. The spectra are therefore quite insensitive to usual quadrupole strengths.

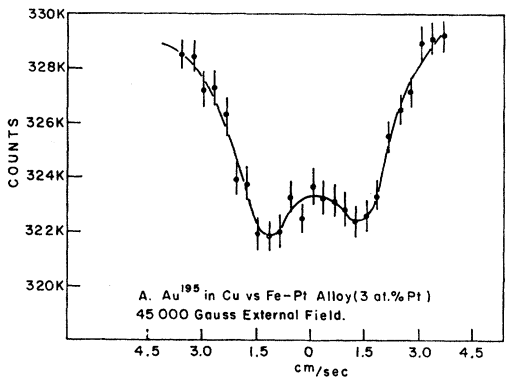

FIG. 4. The Mössbauer absorption spectrum taken at 4.2°K for the Au^{195} in Cu source and the 3 at.% Pt Fe-Pt alloy absorber. A 45 000 G external field was applied parallel to the direction of propagation of the γ rays.

The carrier-free Au^{195} source gave a much lower value for H_{int} than the 3% alloy; reannealling at 720°C for 50 h did not change the result. We believe the anomaly is due to source preparation. Au diffused in Fe tends to concentrate in grain boundaries, and H_{int} of run 6 may well represent such positions.

DISCUSSION

Internal Field

The magnitude and sign of $H_{\text{int}}(\text{Pt})$ observed in these alloys (approximately independent of composition) are very close to the values reported for dilute concentrations of neighboring transition metals alloyed in iron.[1,2] Accurate theoretical calculations for the transferred hyperfine fields in transition metal alloys are not yet available; however, the nature of the mechanisms responsible for their existence have been investigated by several authors.[17] The dominant contributions to $H_{\text{int}}(\text{Pt})$ in such alloys arise from conduction electron polarization due to *s-d* exchange coupling and from core polarization due to a localized ($5d$) moment on the solute atoms. However, neutron scattering experiments on Pt-Fe alloys[18] have shown that there is no localized moment on the Pt atom; therefore, the entire $H_{\text{int}}(\text{Pt})$ must come from conduction electron polarization. For a negative conduction electron spin density at the Pt sites, the hyperfine field is -34×10^3 kOe per spin[19]; thus 0.07 unpaired conduction electrons are sufficient to account for the observed $H_{\text{int}}(\text{Pt})$, 1260 kOe. This value of 0.07 unpaired conduction electrons is in reasonable agreement with theoretical estimates based on exchange polarizing the calculated energy bands of pure platinum.[20]

Magnetic Moment

The magnetic moment of the $\frac{3}{2}^-$ state in $_{78}\text{Pt}^{195}$ is far below the Schmidt value of -1.91, though it is close to that of the $\frac{3}{2}^-$ ground state of $_{80}\text{Hg}^{201}$ ($\mu=-0.5567$). Freed and Kisslinger[21] were able to account for the

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latter result by assuming a reasonable pairing interaction strength admixing seniority-one states and considering seniority-three states as perturbations. In their survey work they varied parameters smoothly and did not consider the details of the actual single particle states, so that their result is equally applicable to the excited state in Pt^{195}. Arima and Horie[22] were also able to account for the value of $\mu(Hg^{201})$ by configuration mixing, but their oscillator potential is unrealistically deep. Finally, one may consider a particle-core coupling model[23] which could account for the $\frac{3}{2}^-$, $\frac{5}{2}^-$ doublet at 99 and 129 keV. In the simplest model the core would be the first 2+ state of Pt^{194}; and one obtains, using its moment (see above) together with that of the ground state of Pt^{195}, the prediction of $\mu(\frac{3}{2})\simeq+0.10$, which is far from the measured value. This lack of agreement is not, however, a fair test of the particle-core coupling picture since there is another close-lying $\frac{3}{2}$, $\frac{5}{2}$ doublet at 210 and 240 keV, and none of the states involved is expected to be particularly pure.

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Range-Energy Relation and Energy Loss of Fission Fragments in Solids*

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The energies of fission fragments transmitted through Al, Ni, Ag, Au, and U-Pd have been measured with surface-barrier detectors. The results, converted to the Lindhard-Scharff-Schiott (LSS) dimensionless parameters $k\rho$ (range) and ϵ (energy), were found to fit the empirical equation

$$k\rho=1.158\epsilon^{1/2}[1-\exp(0.0987\epsilon)]+0.01939\epsilon$$

in the region $12\leq\epsilon\leq348$. These results are 10–20% lower than those predicted by the LSS theory. A description is given of the effect of relatively simple modifications to this theory, and suggestions are presented for a more rigorous theoretical approach.

I. INTRODUCTION

A KNOWLEDGE of the range of fission fragments and their energy loss in solid and gaseous media is important to practical applications of fissio-chemistry and to direct energy conversion via the fission electric cell. Such knowledge is also directly applicable to radiation-induced ionization for space-charge neutralization in thermionics and to nonthermal ionization in magnetohydrodynamics.

Neither the Bohr theory[1] nor the Lindhard-Scharff-Schiott (LSS)[2] theory fits the available data; furthermore, empirical equations derived from different sets of experimental data cannot be compared. One of the complications in trying to unify the available data is the obvious difference between the slowing-down mechanisms in solids and gases; i.e., the density effect associated with gases at low pressures.[3] While we have performed differential energy-loss experiments in both media, this paper gives the results only for the solid absorbers; the gaseous absorber work will be reported at a later date.

We have derived an empirical range-energy expression from our data for fission fragments in Al, Ni, Ag, Au, and U-Pd absorbers that gives fair agreement ($\pm5\%$) with other experimental results for heavy ions in the moderate-energy range. We have compared this expression to the LSS theory and present suggestions that can be used as a basis for a more detailed approach to this problem.

The LSS theory, based on a Thomas-Fermi model of the interacting atoms, considers energy losses by electron stripping (ionization) and by nuclear collisions (nonionization) as uncorrelated and continuous processes. Recent publications[4] present experimental results in terms of this theory in which the energy E in MeV and the range R in mg/cm^2 are reduced to dimensionless

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