

Hyperfine Interaction and Isomeric Shift in Pt^{195†}

D. AGRESTI, E. KANKELEIT,* AND B. PERSSON
California Institute of Technology, Pasadena, California
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Using the Mössbauer technique the first-excited-state g factor for Pt¹⁹⁵ has been determined to be -0.41 ± 0.03 . The following magnetic fields at the Pt nucleus were found: in an Fe lattice, 1.19 ± 0.04 MG; in a Co lattice, 0.86 ± 0.03 MG; and in a Ni lattice, 0.36 ± 0.04 MG. Isomeric shifts have been detected in a number of compounds and alloys and have been interpreted to imply that the mean square radius for the Pt¹⁹⁵ nucleus in the first excited state is smaller than in the ground state.

INTRODUCTION

IN this work we report on Mössbauer studies of the 99-keV ($\frac{3}{2}^- - \frac{1}{2}^-$) first-excited-to-ground-state transition in Pt¹⁹⁵. This transition has been studied before¹⁻³ to determine the g factor of the $\frac{3}{2}^-$ level by making use of the high internal fields at the Pt nucleus in Pt-Fe alloys. However, accurate values of $g_{3/2}$ and H_{int} could not be determined independently from the Mössbauer-effect measurements, and no isomeric shift has been observed. A shift detected by Atac *et al.*² in a scattering experiment was interpreted as being due to interference between Rayleigh and Mössbauer scattering.

In the present work, Fe, Co, and Ni alloys with a small amount (3 at.%) of Pt were used as absorbers in transmission experiments. Application of magnetic fields parallel and perpendicular to the gamma-ray direction modified the intensities of the transitions between the nuclear magnetic sublevels in the observed spectra, and thereby increased the information obtainable over that from a single measurement using an unpolarized absorber. A simultaneous least-squares analysis of the data obtained for the different field configurations made a determination of both $g_{3/2}$ and H_{int} possible.

EXPERIMENT

The experimental setup is described in another paper.⁴ A source of 2 mCi of Au¹⁹⁵ diffused into a 1-mil platinum foil was used. Most of the alloy absorbers were prepared by high-frequency melting, rolling, and tempering. Some were arc melted and carefully annealed for several days. But no change in the spectra was observed for the two absorber preparations. The measurements were performed at 4.2°K. The perpendic-

ular field was applied with a C-shaped magnet which provided about 2.5 kG in the plane of the disk-shaped absorber, and a superconducting solenoid supplied a longitudinal field of about 40 kG. Absorbers of finely ground alloy dispersed in wax were used both unpolarized and polarized by the longitudinal field. The complete alignment of the absorber was verified in experiments on W¹⁸² in Fe,⁴ in which the same technique was used. The 99-keV line was detected with a NaI(Tl) crystal. About 35% of the pulses within the discriminator window were due to background, originating mainly from x rays. Some typical Mössbauer spectra for the different field configurations and alloys are shown in Figs. 1 and 2. The spectra show little structure, and the individual peaks are not resolved. But an isomeric shift is clearly observed for the alloys.

The spectra were fitted to a sum of Lorentzian line shapes by least-squares computer analysis. The ($\frac{3}{2}^- - \frac{1}{2}^-$) transition is taken to be pure $M1$,^{5,6} thus producing six transitions between the magnetic sublevels. The relative intensity of each transition is constrained in the fit to correspond to that expected for the relevant field configuration. Fits to individual spectra revealed a strong correlation among four parameters, namely, excited-state g factor ($g_{3/2}$), internal field (H_{int}), width (W), and effect, i.e., dip of the absorption spectrum. A unique determination of these parameters was therefore not possible. The situation improved markedly when a simultaneous fit was performed to the spectra for all three field configurations. The parameters $g_{3/2}$, H_{int} , isomeric shift (I.S.), and width corrected for the difference in the absorber thickness were constrained to be the same for our six sets of data. As a consequence, the strong correlations among the four parameters, i.e., the corresponding off-diagonal elements of the error correlation matrix, were reduced up to 50%, and unique and rather precise values can be quoted for these parameters. The results are presented in Table I. The value⁷ $g_{1/2} = 1.21204$ was used.

To check the internal consistency among the individual measurements, drastically changed weights were

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* Present address: Institut für Technische Kernphysik, Darmstadt, Germany.

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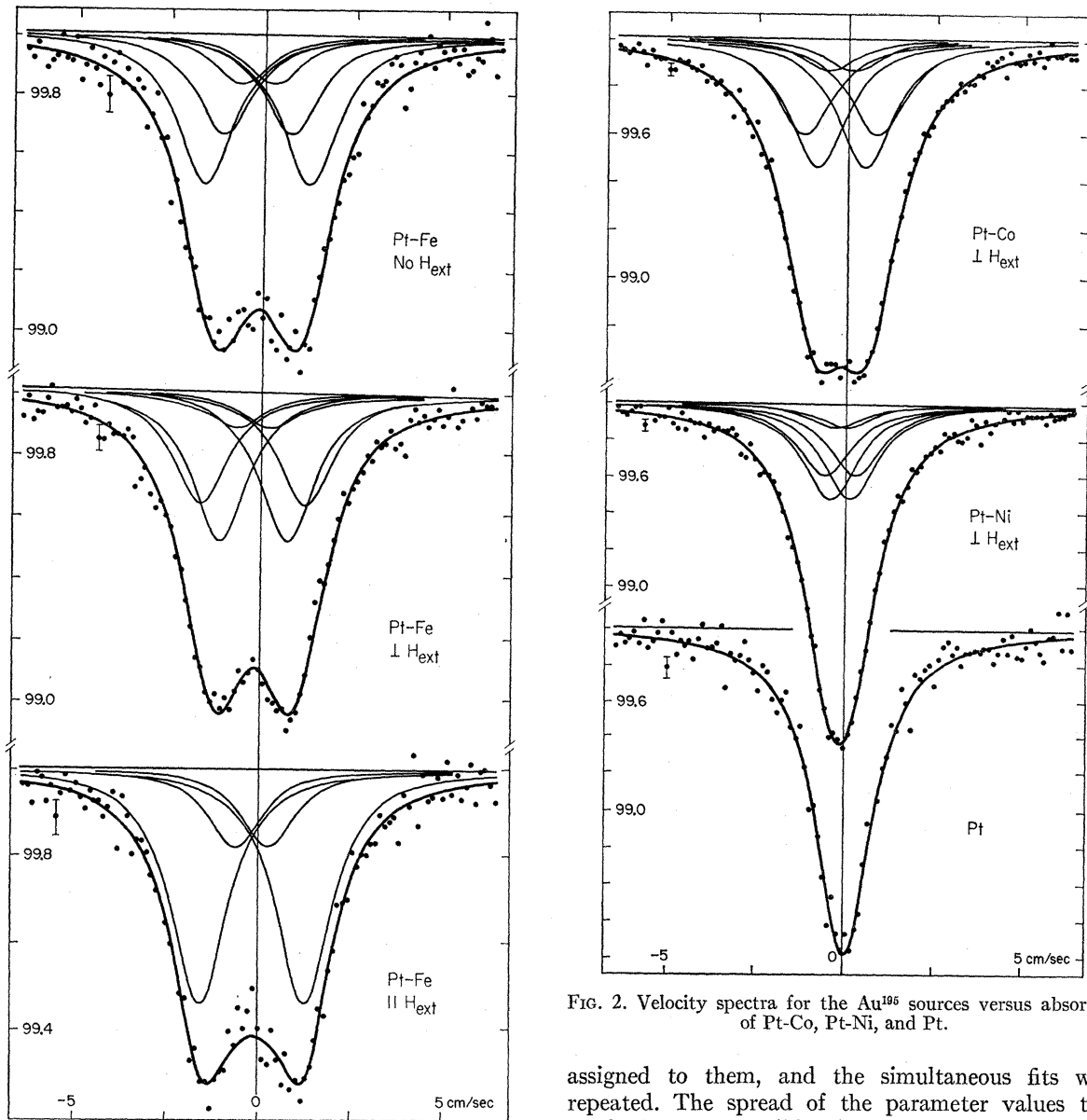


FIG. 1. Velocity spectra for the Au^{196} source versus Pt-Fe absorbers for the different field configurations. The curves result from the simultaneous fit with pure magnetic interaction and pure $M1$ transition.

FIG. 2. Velocity spectra for the Au^{196} sources versus absorbers of Pt-Co, Pt-Ni, and Pt.

assigned to them, and the simultaneous fits were repeated. The spread of the parameter values that resulted was compatible with the standard deviations. In fitting individual spectra, minima for Q^2 could sometimes be obtained for different sets of parameters. With this in mind we repeated the simultaneous fit searching for other minima, but none was found.

TABLE I. Summary of results. The errors given are standard deviations. The widths (W) are extrapolated to zero absorber thickness using $\Theta_D(\text{Pt}) = 230^\circ\text{K}$ and the relation $\Theta_D = \Theta_D(\text{host}) \times [M(\text{host})/M(\text{impurity})]^{1/2}$.

	Pt-Fe	Pt-Co	Pt-Ni	Pt
Thickness (mg Pt/cm ²)	30-60	50	50	50
$g_{3/2}$	-0.41 ± 0.03	-0.41^a	-0.41^a	
H_{int} (MG)	1.19 ± 0.04	0.86 ± 0.03	0.36 ± 0.04	
W (mm/sec)	17.4 ± 0.03	18.6 ± 0.04	17.8 ± 0.06	18.0 ± 0.4
I.S. (mm/sec)	-1.90 ± 0.11	-1.96 ± 0.09	-1.65 ± 0.08	0.06 ± 0.17
$H_{\text{int}}/\mu_{\text{eff}}$ (MG/magneton)	0.54 ± 0.02	0.46 ± 0.02	0.58 ± 0.06	

* Value adopted from the Pt-Fe results.

The velocity spectra observed with the Pt-Fe absorbers seem to be slightly asymmetric (Fig. 1). Quadrupole interaction in the absorber might be the cause of this asymmetry; therefore the simultaneous fit was repeated with a combined magnetic and quadrupole interaction, and a slightly improved fit was obtained. There is also the possibility that the small $E2$ admixture can affect the results. Including this (with $|\delta|=0.12$) further improves the fit. However, the changes in the values of the important parameters ($g_{3/2}$, H_{int} , and I.S.) are much smaller for both these fits than the errors given for them in Table I.

Figure 2 shows spectra from measurements with traverse applied field on Pt-Co and Pt-Ni absorbers. By constraining $g_{3/2}$ to the value obtained from the fit to the Pt-Fe data, H_{int} could be determined for the other alloys. The results for the fits are presented in Table I.

Isomeric shifts for a number of platinum alloys are plotted in Fig. 3 versus the electronegativity⁸ of the host metal. In addition to the alloys mentioned above, shifts were measured for 20 at.% Pt in Au and 0.7 at.% Pt in Al. The alloys are solid solutions except for the Pt-Al alloy. In this case x-ray analysis indicates intermetallic compound formation, which is to be expected since a solid solution at room temperature can be acquired only by extremely rapid quenching.⁹ To obtain further information about isomeric shifts and possible electric quadrupolar interactions, some compounds of Pt have been studied. Isomeric shifts were observed for PtO (-0.34 ± 0.11 mm/sec), PtO₂ (-0.40 ± 0.08 mm/sec), PtCl₂ (-0.1 ± 0.2 mm/sec), and PtCl₄ (-0.3 ± 0.3 mm/sec).

DISCUSSION

The value for the g factor of the $\frac{3}{2}^-$ state determined in this experiment (-0.41 ± 0.03) is in good agreement with that of Atac *et al.*² (-0.43 ± 0.10) and with the recently quoted value (-0.40 ± 0.07) of Buyrn and Grodzins.³ It is just outside the limits ($0.47 > g_{3/2} > -0.60$) given by Benczer-Koller *et al.*¹ who assume $H_{int} = 1.39$ MG for the Pt-Fe alloy. The core-excitation model proposed by de-Shalit¹⁰ and Gal¹¹ should account for the low-energy states in Pt¹⁹⁵. Experimental information is available for the 2⁺ states in Pt¹⁹⁴,^{12,13} so that comparison of the predicted and measured values of the g factor can be made. It turns out that calculations give values very near zero; hence present core-excitation models do not give adequate description of the $\frac{3}{2}^-$ state in Pt¹⁹⁵. As has been pointed out,¹¹ the discrepancy might

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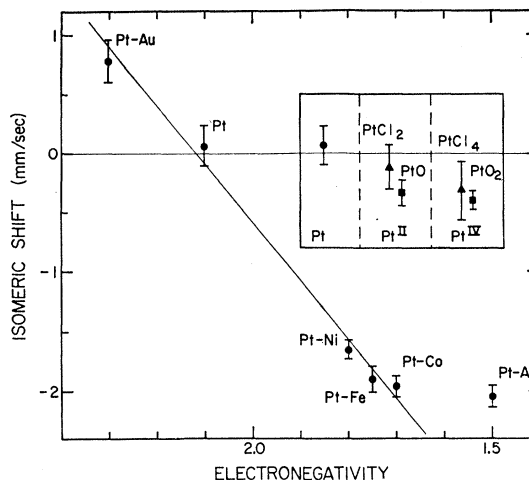


FIG. 3. Plot of the isomeric shift of platinum alloys versus the electronegativity of the host element. The deviation for Pt-Al from the general trend might be because the platinum is not in a solid solution. In the insert isomeric shifts for some platinum compounds are shown related to the valency of platinum.

be removed if configuration mixing is taken into account.

The present result for the magnitude of the internal field at the site of the platinum nucleus in Pt-Fe alloy (1.19 ± 0.04 MG; 3 at.%) differs from what has been obtained by calorimetric determination¹⁴ (1.39 MG; 3.2 at.% Pt) and by neutron-spin-resonance measurements¹⁵ (1.8 MG; 10 at.% Pt). It is shown in Table I that the ratio of the internal field at the platinum nucleus to the effective magnetic moment of the host atom is almost constant, as is to be expected if conduction electron polarization is the cause of the internal fields.¹⁶ The small deviation from constancy has the same trend as is observed for a number of elements dissolved in the ferromagnetic $3d$ transition metals and is particularly pronounced for tungsten.⁴

From Fig. 3 it is seen that the isomeric shift for the alloys decreases with decreasing electronegativity of the host element. This trend is opposite to that observed for Au¹⁹⁷ by Barret *et al.*¹⁷ If we adopt the line of reasoning that decreased electronegativity of the host material results in an increased electron density at the impurity nucleus, it follows that the mean square radius of the Pt¹⁹⁵ nucleus in the first excited state is smaller than that in the ground state.

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