

Electric-field gradient at the transition-element impurity ^{193}Ir in the hexagonal transition metals Sc, Y, Lu, Ti, Zr, and Hf

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The electric-field gradient at the transition-element impurity ^{193}Ir in the hexagonal transition metals Sc, Y, Lu, Ti, Zr, and Hf has been determined by Mössbauer measurements. The results are $42.9(11.0) \times 10^{17}$ V/cm² in Sc, $21.5(5.5) \times 10^{17}$ V/cm² in Y, $16.6(4.3) \times 10^{17}$ V/cm² in Lu, $6.5(1.6) \times 10^{17}$ V/cm² in Ti, $6.8(1.7) \times 10^{17}$ V/cm² in Zr, and $11.6(3.0) \times 10^{17}$ V/cm² in Hf. These results suggest that the density of states at the Fermi energy of the host and the electronic structure of the impurity are of major importance for the electric-field gradient in transition-metal alloys. The electric-field gradient of Ir in Sc is the largest value ever observed at a non-rare-earth impurity in a metallic host. In the IVb metals Ti, Zr, and Hf, the field gradient depends sensitively on the concentration of nitrogen and oxygen impurities in the samples.

I. INTRODUCTION

In recent years the electric-field gradient (EFG) at nuclear sites in pure non-cubic metals and dilute alloys has been investigated systematically as a function of host and impurity properties, temperature, and pressure.¹ These studies, which were performed with the aim to gain a better understanding of the electronic contribution to the EFG have shown that in most cases, in particular those involving *sp* metals, the ionic EFG $(1-\gamma_\infty)V_{zz}^{1at}$ and the electronic EFG are strongly correlated²:

$$V_{zz}^{e1} = -k(1-\gamma_\infty)V_{zz}^{1at}. \quad (1)$$

For most pure metals and impurity-host combinations the value of the proportionality factor k is in the range $k=2-5$, depending on the valence of the impurity and the host.^{1,2} A theoretical explanation of this "universal correlation" and in particular for the opposite sign of the ionic EFG and the electronic EFG V_{zz}^{e1} has recently been proposed by Bodendstedt and Perscheid.

There are, however, several cases which do not fit into the concept of a universal correlation between V_{zz}^{e1} and $(1-\gamma_\infty)V_{zz}^{1at}$. In all pure hexagonal group-IIIb and -IVb metals the electronic EFG is proportional to the ionic EFG, but in contrast to Eq. (1) both contributions have the same sign.⁴ The same sign for V_{zz}^{e1} and $(1-\gamma_\infty)V_{zz}^{1at}$ has also been observed for the impurity Hf in Be, Ti, Ru, and Os,⁵ and for 5*d* impurities in Gd.⁶ In the last case the electronic EFG is not even proportional to the ionic contribution, but seems to depend mainly on the number of 5*d* electrons of the impurity.

All these exceptions from the universal correlation involve either transition-element hosts or transition-element impurities. In numerous other transition-element systems, however, the electronic EFG fits well into the universal correlation.¹

At present there is no theoretical concept which could explain these differences, and further experimental data appear necessary in order to recognize the impurity and host properties which mainly determine the electronic EFG in non-cubic transition metals.

In this paper we present an investigation of the EFG and the isomer shift at the transition-element impurity ^{193}Ir in the hexagonal transition metals Sc, Y, Lu, Ti, Zr, and Hf, which was carried out using the Mössbauer effect of the 73-keV transition of ^{193}Ir . Apart from the general interest in the EFG in transition metals, this study has been motivated by the fact that for ^{193}Ir in Gd an EFG of 2×10^{18} V/cm² has been reported,⁷ one of the largest values ever observed for a non-rare-earth impurity in any hexagonal metal host. At the same impurity in Os,⁸ the only other Ir-host combination investigated up to now, the EFG is a factor of 6 smaller. Since the lattice field gradients of Gd and Os differ only slightly, this large difference in the EFG at the same impurity reflects drastic changes in the electronic EFG of these two transition metals. Measurements of the EFG at ^{193}Ir in other transition metals might help to understand the mechanism responsible for these remarkable differences.

II. EXPERIMENTAL DETAILS, DATA ANALYSIS, AND RESULTS

In measurements of the EFG at impurity sites in hexagonal metals a low impurity concentration is of importance. Otherwise, the interpretation of the data might be complicated by possible impurity-impurity interactions. The investigation reported here was therefore carried out as a source experiment.

The 73-keV Mössbauer transition of ^{193}Ir is pop-

ulated by the β decay of ^{193}Os which has a half-life of 30 h. For the source preparation ^{192}Os metal (enrichment 99%) was neutron irradiated and then alloyed with Sc, Y, Lu, Ti, Zr, and Hf by means of an electron gun in a vacuum of 10^{-5} Torr. The purity of the transition metals was 99.95 at.% for Sc, 99.99 at.% for Zr, and 99.9 at.% for Y, Lu, Ti, and Hf. In all cases the Os concentration of the alloys was less than 1 at.%. After melting, the alloys were broken into small pieces in order to reduce the effect of texture and absorption in the source, and in a first series of experiments they were used for the measurements without further heat treatment. In a second series of measurements, the EFG was determined in all metals after the samples had been annealed in a vacuum of 10^{-8} Torr at 1300 K for about 1 h.

The measurements were carried out with a standard double-loudspeaker drive system in transmission geometry, using a $50\text{-}\mu\text{m}$ foil of natural Ir metal as a single line absorber. In most cases both the absorber and the source were kept at 4.2 K. In Sc we also performed a measurement at 77 K in order to estimate the temperature dependence of the EFG. The 73-keV γ rays were detected with an NaI(Tl) detector.

The spectra obtained for the different hosts are shown in Figs. 1 and 2. In the case of Sc, Y, and Lu the spectra were not changed by annealing. Therefore only the spectra measured before annealing are shown (Fig. 1). In the case of Ti, Zr, and Hf the annealing has a considerable effect on

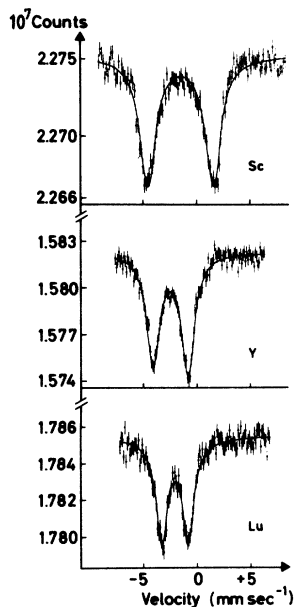


FIG. 1. Mössbauer spectra of ^{193}Ir in Sc, Y, and Lu at 4.2 K, measured with unannealed sources.

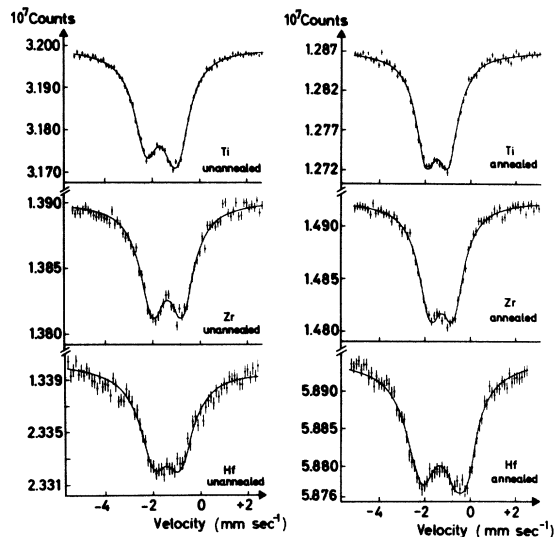


FIG. 2. Mössbauer spectra of ^{193}Ir in Ti, Zr, and Hf at 4.2 K, measured before and after annealing of the sources.

the quadrupole splitting. For these hosts the spectra measured before and after annealing are shown in Fig. 2.

The ground state of ^{193}Ir has spin $\frac{3}{2}$; the spin of the excited state at 73 keV is $\frac{1}{2}$. For a unique quadrupole interaction the Mössbauer spectrum should therefore consist of two lines, which in the case of an axially symmetric EFG are separated by an energy of $eQV_{zz}/2$, the hyperfine splitting of the $\frac{3}{2}$ ground state. V_{zz} is the electric-field gradient, and Q the ground-state quadrupole moment. For a polycrystalline source with an isotropic Debye-Waller factor one expects identical intensities for both lines.

Figure 1 shows that well-split two-line spectra are obtained in Sc, Y, and Lu. In the last two cases the two lines have slightly different intensities. The spectrum measured in Sc at 77 K has the same structure as the one taken at 4.2 K. In the group Ti, Zr, and Hf the ground-state splitting of ^{193}Ir (Fig. 2) is clearly much smaller. Both before and after annealing of the samples the two lines are only poorly separated. In the case of Ti and Zr the splitting decreases by 25%, whereas in the case of Hf it increases by 50% after annealing of the samples.

For the determination of the quadrupole splitting $eQV_{zz}/2$ and the isomer shift (IS) two Lorentzian lines were fitted to the measured spectra. Because of the slight asymmetry of the spectra in some cases, the line intensities were treated as free parameters. The width Γ of the lines was also adjusted. The results of the fits are listed in Table I.

TABLE I. Linewidth Γ , the isomer shift IS, and the quadrupole splitting $eQV_{zz}/2$ of ^{193}Ir in Sc, Y, Lu, Ti, Zr, and Hf. For the IVb metals the results obtained before and after annealing of the samples are given.

Host	T (K)	Γ (mm/sec)	IS (mm/sec)	$eQV_{zz}/2$ (mm/sec)
Sc	4.2	1.24(4)	-1.45(2)	6.16(4)
Sc	77	1.24(11)	-1.36(7)	5.96(14)
Y	4.2	0.98(2)	-1.97(2)	3.08(2)
Lu	4.2	0.88(3)	-2.06(2)	2.39(4)
Ti	4.2	0.87(2)	-1.63(2)	1.27(2)
Ti	4.2	0.76(2)	-1.70(2)	0.94(2)
annealed				
Zr	4.2	0.90(2)	-1.42(2)	1.25(2)
Zr	4.2	0.81(2)	-1.50(2)	0.97(2)
annealed				
Hf	4.2	1.22(4)	-1.22(2)	1.06(5)
Hf	4.2	1.16(4)	-1.34(3)	1.66(3)
annealed				

III. DISCUSSION

A. Annealing effects

The Mössbauer spectra of ^{193}Ir in the group-IVb metals Ti, Zr, and Hf change considerably when the samples are annealed at high temperatures in high vacuum (see Table I and Fig. 2). In the group-IIIb metals Sc, Y, and Lu the same heat treatment leaves the spectra unchanged. In principle a heat treatment in high vacuum may have two effects: removal of lattice defects and outgasing of the sample. The different reactions of the IIIb and the IVb metals to the heat treatment suggest that the changes observed in the IVb metals are probably due to an outgasing of the samples: It is well known that gases such as nitrogen and oxygen are highly soluble in IVb metals, but only moderately soluble in IIIb metals.⁹ Sources of the IVb metals Ti, Zr, and Hf, which were prepared by melting in a vacuum of only 10^{-5} Torr, therefore probably contain a much larger amount of nitrogen and oxygen impurities than samples of the IIIb metals Sc, Y, and Lu. Consequently the properties of the IVb hosts should change much stronger than those of the IIIb hosts, when the samples are cleaned from these impurities by heating in a vacuum of 10^{-8} Torr.

The values listed in Table I show that the outgasing of the IVb hosts affects the line width, the isomer shift, and the quadrupole splitting: As a result of the annealing the line width decreases and the isomer shift increases in all cases. The quadrupole splitting decreases in Ti and Zr, whereas in Hf it increases after annealing.

The reduction of the line width by outgasing is easily understood: Nitrogen and oxygen impurities near the Ir sites perturb the environment of the

probe nuclei, which then experience instead of one unique value a distribution of slightly different electric-field gradients. This leads to a broadening of the Mössbauer lines and consequently the line width should decrease when the gas impurities are removed from the host lattice.

The increase of the isomer shift after outgasing is probably a volume effect: the atomic volume of Ti, Zr, and Hf increases with the nitrogen and oxygen concentration.⁹ Thus the atomic volume decreases when the samples are outgased. This leads to a larger conduction electron density and consequently causes an increase of the isomer shift.

The reasons for the changes of the quadrupole splitting are less clear. The electric-field gradient in hexagonal metals is usually the larger, the stronger the axial ratio c/a of the lattice parameters c and a deviates from its ideal value [$c/a = (8/3)^{1/2}$] for closest packing.¹ In Ti, Zr, and Hf the deviation of c/a from the ideal value decreases with increasing nitrogen and oxygen concentration.⁹ One might therefore expect that the EFG increases when the gas concentration is reduced. Only in Hf, however, the quadrupole splitting increases after outgasing. The decrease of the splitting in Ti and Zr shows that the influence of gas impurities on the EFG in hexagonal metals is more complex. Systematic investigations of the quadrupole splitting as a function of the gas concentration appear necessary for an understanding of these impurity induced changes of the EFG.

In the following discussion of the isomer shift and the EFG we shall only consider results obtained with the outgased samples. These results are listed in Table II.

B. The Isomer shift

Wagner *et al.*¹⁰ have investigated the isomer shift of ^{193}Ir , ^{99}Ru , and ^{197}Au as impurities in numerous metals of the 3d, 4d, and 5d transition series. Their values for ^{193}Ir in Y, Ti, Zr, and Hf are in agreement with our results. The data of Wagner *et al.*¹⁰ reveal some general properties of the isomer shift of transition-element impurities in transition-metal hosts:

(i) The isomer shift of the same impurity in 3d, 4d, and 5d hosts belonging to one column of the periodic table usually decreases from the 3d via the 4d to the 5d metals.

(ii) Within a given transition series the isomer shift decreases with an increasing number of outer d electrons of the hosts.

These two trends are attributed to volume and band-structure effects of the hosts: The atomic volume of the 3d metals is roughly 25% smaller than that of the 4d and 5d metals; therefore the

TABLE II. Measured EFG V_{zz} at 4.2 K, the isomer shift IS, the ionic EFG $(1-\gamma_\infty)V_{zz}^{iat}$, and the electronic EFG V_{zz}^{el} of ^{193}Ir in different IIIb, IVb, and VIIIb transition metals. $\eta(E_F)$ is the density of states at the Fermi energy of these hosts. The electronic EFG V_{zz}^{el} was calculated assuming a positive sign for V_{zz} .

Host	Config.	V_{zz} (10^{17}V/cm^2)	IS (mm/sec)	$(1-\gamma_\infty)V_{zz}^{iat}$ (10^{17}V/cm^2)	V_{zz}^{el} (10^{17}V/cm^2)	$\eta(E_F)$ ($\text{eV}^{-1}\text{spin}^{-1}$)
Sc	$3d^14s^2$	$\pm 42.9(11.0)$	-1.45	0.94 ^c	42.0	2.27 ^l
Y	$4d^15s^2$	$\pm 21.5(5.5)$	-1.96	1.12 ^d	20.4	2.14 ^m
Gd ^a	$4f^75d^16s^2$	$+18.5(4.7)$	-1.99	0.67 ^e	17.8	1.74 ⁿ 2.33 ^o 0.79 ^p
Lu	$4f^{14}5d^16s^2$	$\pm 16.6(4.3)$	-2.06	1.09 ^f	15.5	2.39 ^q 1.44 ⁿ
Ti	$3d^24s^2$	$\pm 6.5(1.6)$	-1.70	2.03 ^g	4.5	0.70 ^r
Zr	$4d^25s^2$	$\pm 6.8(1.7)$	-1.50	1.40 ^h	5.4	0.59 ^r
Hf	$5d^26s^2$	$\pm 11.6(3.0)$	-1.34	1.82 ⁱ	9.8	0.46 ^r
Os ^b	$5d^66s^2$	$\pm 3.3(0.8)$	-0.55	6.17 ^k	-2.9	0.49 ^r 0.20 ^s

^a Reference 7.

^b Reference 8.

^c $T=290\text{ K}$; $Z=3$; c and a from Ref. 21.

^d $T=77\text{ K}$; $Z=3$; c and a from Ref. 22.

^e $T=120\text{ K}$; $Z=3$; c and a from Ref. 23.

^f $T=4.2\text{ K}$; $Z=3$; c and a from Ref. 24.

^g $T=290\text{ K}$; $Z=4$; c and a from Ref. 25.

^h $T=4.2\text{ K}$; $Z=4$; c and a from Ref. 26.

ⁱ $T=4.2\text{ K}$; $Z=4$; c and a from Ref. 27.

^k $T=77\text{ K}$; $Z=8$; c and a from Ref. 28.

^l Reference 29.

^m Reference 30.

ⁿ Reference 31.

^o Reference 32.

^p Reference 33.

^q Reference 34.

^r Reference 35.

^s Reference 36.

conduction electron density and thus the isomer shift in homologous transition metals should be largest in the $3d$ and smallest in the $5d$ hosts. The decrease of the isomer shift in one transition series is related to a decreasing s -electron population and an increasing d -electron population on the Mössbauer atom.

The isomer shifts reported here differ from these general trends in one aspect: In the group Sc, Y, Gd, and Lu, which has the valence-electron configuration $nd^1(n+1)s^2$, $n=3, 4, 5$, and can therefore be considered as one column of the periodic table, the isomer shift is largest in the $5d$ host Lu and smallest in the $3d$ host Sc. This inversion of the general trend cannot be due to a volume effect, as the atomic volume of Sc is 20–25% smaller than that of the $4d$ and $5d$ hosts. The decrease of the isomer shift from Lu to Sc must therefore be due to band-structure changes such as a decrease of the s character and an increase of the d character of the conduction electrons between Lu and Sc. In

this context it is interesting to note the correlation which exists between isomer shift and quadrupole splitting in these IIIb metals (see Table II): As the isomer shift decreases, the quadrupole splitting increases in the order Lu, Gd, Y, Sc. While the isomer shift depends on the s -electron density, the quadrupole interaction is sensitive mainly to the d -electron density at the impurity site. The assumption of an increase of the d character of the conduction electrons from Lu to Sc would therefore satisfy the observed simultaneous increase of the quadrupole interaction and decrease of the isomer shift. To our knowledge, band-structure calculations to support this interpretation are presently not available. Such an interpretation would, however, be in conflict with the pseudopotential calculations of da Silva *et al.*,¹¹ who predict that in a given triad of homologous transition metals the d -electron density at the impurity site increases from the $3d$ to the $5d$ metals.

In the group IVb the data follow this prediction.

Here the isomer shift decreases in the order Ti, Zr, Hf. Part of this decrease is probably a volume effect, since Ti, which has the largest isomer shift, has the smallest atomic volume. Hf and Zr, however, have identical volumes, but different isomer shifts. This suggests an increase of the d character of the conduction electrons from Zr to Hf.

The comparison between group-IIIb and group-IVb metals belonging to the same transition series shows that in the $4d$ and $5d$ hosts the isomer shift decreases with an increasing number of d electrons, in agreement with the general trend observed by Wagner *et al.*¹⁰ In the $3d$ series one generally observes a slightly different behavior. Here the isomer shift first increases, reaches a maximum in the middle in the series, and then decreases. The measured increase of the isomer shift from Sc to Ti fits well into this scheme.

C. Electric-field gradient

In Table II we have listed the values of the electric-field gradient V_{zz} in the different transition-metal hosts at 4.2 K. These values were calculated from the measured quadrupole splitting $eQV_{zz}/2$, using $Q = 0.70(18)b$ for the quadrupole moment of the ground state of ^{193}Ir .¹² The results of the previous measurements in Gd (Ref. 7) and Os (Ref. 8) have been included. The large errors of V_{zz} are exclusively due to the uncertainty of the quadrupole moment. With the exception of Gd, the sign of the EFG could not be determined since the Mössbauer spectrum of a $\frac{1}{2}$ - $\frac{3}{2}$ transition and pure quadrupole interaction in a polycrystalline sample is independent of the sign of $eQV_{zz}/2$. In the case of Gd the positive sign of V_{zz} was deduced from a measurement of the combined magnetic and electric hyperfine interaction in the ferromagnetic phase. In this special case the spectrum is sign sensitive.

Furthermore, Table II contains the values of the ionic-field gradient $(1 - \gamma_\infty)V_{zz}^{1at}$. The lattice EFG V_{zz}^{1at} was calculated, using the analytical expression derived by Das and Pommerantz¹³ for hexagonal metals:

$$V_{zz}^{1at} = \frac{Ze}{4\pi\epsilon_0} \frac{0.0065 - 4.3584(c/a - 1.6333)}{a^3}. \quad (2)$$

Here c and a are the lattice parameters and Z is the charge state of the lattice ions. V_{zz}^{1at} has been calculated for the lowest temperatures, for which the lattice parameters have been measured up to now. These temperatures are given in Table II, which also contains the assumed charge state of the lattice ions and the references from which the lattice parameters were taken. For the Sternheimer correction of Ir we used the value $1 - \gamma_\infty = 43$ for

Ir^{3+} , calculated by Gupta and Sen.¹⁴ Column VI of Table II contains the values of the electronic contribution $V_{zz}^{e1} = V_{zz} - (1 - \gamma_\infty)V_{zz}^{1at}$, which were calculated from the measured value of V_{zz} and the calculated value of the ionic EFG. For an unambiguous calculation of V_{zz}^{e1} the sign of V_{zz} has to be known. This is the case only for Gd.⁷ For the other hosts we assumed the same positive sign.

The values of V_{zz} listed in Table II show several remarkable features:

(i) The EFG of ^{193}Ir in the group-IIIb metals Sc, Y, Gd, and Lu is extremely large: $4.3 \times 10^{18} \text{V/cm}^2$ in Sc and approximately $2 \times 10^{18} \text{V/cm}^2$ in Y, Gd, and Lu. To our knowledge the result for Ir in Sc is the largest EFG value ever observed for a non-rare-earth impurity in a metallic host. Larger values have only been found in some semimetals.¹ A comparison of V_{zz} and $(1 - \gamma_\infty)V_{zz}^{1at}$ in Table II shows that the ionic contribution $(1 - \gamma_\infty)V_{zz}^{1at}$ accounts for only 5% or less of the measured EFG. The ratio

$$k = |V_{zz}^{e1}/(1 - \gamma_\infty)V_{zz}^{1at}|$$

varies from $k \approx 50$ for Sc to $k \approx 15$ for Lu, whereas for most other systems the factor k is of the order of 2–5.¹ Clearly the very large EFG of Ir in Sc, Y, Gd, and Lu is due to an exceptionally high electronic contribution V_{zz}^{e1} in these metals. The large electronic EFG is probably related to the electronic structure of the transition-element impurity Ir $5d^7 6s^2$. It is interesting to note that the EFG of Ir in the IIIb metals is of the same order of magnitude as the EFG of a free Ir ion. Depending on the ionization state, the EFG produced by the unclosed $5d$ shell of a free Ir ion varies between $3 \times 10^{18} \text{V/cm}^2$ and $6 \times 10^{18} \text{V/cm}^2$.¹⁵ Therefore a large electronic contribution to the EFG might be expected, if in the alloy the $5d$ electrons remain strongly localized at the impurity. This picture of a local $5d$ -electron contribution is not in conflict with the EFG values of other transition-element impurities in the IIIb metals. The EFG of Ta $5d^3 6s^2$ and Sc $3d^1 4s^2$ in Y, for example, is $6 \times 10^{17} \text{V/cm}^2$ and $1 \times 10^{17} \text{V/cm}^2$, respectively,^{16,17} i.e., a factor of 4 and 20, respectively, smaller than the EFG of Ir in the same host. For the impurities Ta and Sc, however, the expectation value $\langle r^{-3} \rangle_{nd}$ and thus the EFG produced by an open d shell is a factor of 2–3 and 10–20, respectively, smaller than for Ir.¹⁵

(ii) The electronic structure of the impurity is not the only factor which determines the magnitude of the EFG in transition metals. This is obvious from the difference of the EFG in Sc and Y and also from a comparison of the EFG in the IIIb and the IVb hosts. Between these two groups of metals the EFG decreases strongly by more than a factor of 2, except in the case of Hf, where the decrease

is slightly smaller.

Table II shows that these changes are not due to differences in the ionic EFG $(1 - \gamma_\infty)V_{zz}^{iat}$, which even increases slightly as one goes from the IIIb to the IVb hosts. The pronounced decrease of V_{zz} between these two groups must therefore be related to changes in the electronic structure of the hosts. Watson *et al.*¹⁸ and Piecuch and Janot^{19,20} have theoretically investigated the EFG in pure transition metals and transition-metal alloys. According to these studies the contribution to the EFG from electrons localized in the Wigner-Seitz cell of the nucleus under consideration should be proportional to the density of states at the Fermi energy $\eta(E_F)$. We have calculated the density of states from the electronic specific-heat data available in the literature. The values of $\eta(E_F)$ and the references from which they were calculated are listed in Table II. For the group-IIIb metals the results of different authors differ considerably. Nevertheless, the data indeed suggest a certain proportionality between the EFG and the density of states at the Fermi energy of the different transition metals: Between the groups Y, Gd, Lu and

Ti, Zr, Hf both the EFG and the density of states differ by approximately a factor of 2–3. The very large EFG in Sc, however, cannot be explained on this basis, since the density of states of Sc and the other IIIb metals differ only slightly.

Although the experimental results presented in this paper suggest that in contrast to simple metals the density of states of the host and the electronic structure of the impurity may be of major importance for the EFG in transition-metal alloys, a quantitative interpretation of these results is at present not possible. Considerable theoretical effort will be necessary for a full understanding of the EFG in these systems.

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¹E. N. Kaufmann and R. J. Vianden, *Rev. Mod. Phys.* **51**, 161 (1979).

²R. S. Raghavan, E. N. Kaufmann, and P. Raghavan, *Phys. Rev. Lett.* **34**, 1280 (1975).

³E. Bodendstedt and B. Perscheid, *Hyperfine Interact.* **5**, 291 (1978).

⁴H. Ernst, E. Hagn, E. Zech, and G. Eska, *Phys. Rev. B* **19**, 4460 (1979).

⁵B. Perscheid and G. Kaindl (private communication).

⁶K. Krien, M. Forker, F. Reuschenbach, and R. Trzcinski, *Hyperfine Interact.* **7**, 19 (1979).

⁷B. Perscheid and M. Forker, *Z. Phys.* **B29**, 307 (1978).

⁸F. Wagner and U. Zahn, *Z. Phys.* **233**, 1 (1970).

⁹E. Fromm and G. Gebhardt, *Gase und Kohlenstoff in Metallen* (Springer, Berlin, 1976).

¹⁰F. E. Wagner, G. Wortmann, and G. M. Kalvius, *Phys. Lett.* **42A**, 483 (1973).

¹¹X. A. de Silva, A. A. Gomes, and J. Danon, *Phys. Rev. B* **4**, 1168 (1971).

¹²S. Büttgenbach, M. Herschel, G. Meisel, E. Schrödl, W. White, and W. J. Childs, *Z. Phys.* **263**, 341 (1973).

¹³T. P. Das and M. Pommerantz, *Phys. Rev.* **123**, 2070 (1961).

¹⁴R. P. Gupta and S. K. Sen, *Phys. Rev. A* **8**, 1169 (1973).

¹⁵S. Fraga, J. Karwowski, and K. M. Saxena, *Handbook of Atomic Data* (Elsevier, Amsterdam, 1976).

¹⁶T. Butz and G. M. Kalvius, *J. Phys. F* **4**, 2331 (1974).

¹⁷R. G. Barnes, F. Borsa, S. L. Segel, and D. R. Tongeson, *Phys. Rev.* **137**, A1828 (1965).

¹⁸R. E. Watson, A. C. Gossard, and Y. Yafet, *Phys. Rev.* **140**, A375 (1965).

¹⁹M. Piecuch and Ch. Janot, *J. Phys. (Paris) Colloq.* **C6**, 359 (1976).

²⁰M. Piecuch and Ch. Janot, *Hyperfine Interact.* **5**, 69 (1977).

²¹N. Schmitz-Pranghe and Ph. Dünner, *Z. Metallkd.* **59**, 377 (1968).

²²V. A. Vinkel and V. V. Vorobev, *Sov. Phys. Crystallogr.* **13**, 457 (1968).

²³V. V. Vorobev, Yu. N. Smirnow, and V. A. Finkel, *Zh. Eksp. Teor. Fiz.* **49**, 1774 (1966) [*Sov. Phys.-JETP* **22**, 1212 (1966)].

²⁴J. J. Tonnies, K. A. Gschneidner, and F.H. Spedding, *J. Appl. Phys.* **42**, 3275 (1971).

²⁵R. H. Willens, *Rev. Sci. Instrum.* **33**, 1069 (1962).

²⁶J. Goldak, L. T. Lloyd, and C. S. Barrett, *Phys. Rev.* **144**, 478 (1966).

²⁷M. P. Krug and B. E. Davies, *J. Less-Common Met.* **22**, 363 (1970).

²⁸V. A. Finkel, M. I. Palatnik, and G. P. Kovtien, *Fiz. Met. Metalloved.* **32**, 212 (1971).

²⁹W. W. Weller and K. K. Kelley, U. S., *Bur. Mines, Rep. Invest.* **5984**, 1 (1962).

³⁰L. D. Jennings, R. E. Müller, and F.H. Spedding, *J. Chem. Phys.* **33**, 1849 (1960).

³¹P. Wells, P. C. Lanchester, D. W. Jones, and R. G. Jordan, *J. Phys. F* **6**, 11 (1976).

³²B. Dreyfus, J. C. Michel, and D. Thoulouze, *Phys. Lett.* **A24**, 457 (1967).

³³P. Wells, P. C. Lanchester, D. W. Jones, and R. G. Jordan, *J. Phys. F* **4**, 1729 (1974).

³⁴J. A. Morrison and D. M. T. Newsham, *J. Phys. C* **1**, 370 (1968).

³⁵F. Heininger, E. Bucher, and J. Müller, *Phys. Kondens. Mater.* **5**, 243 (1966).

³⁶J. G. Daunt, *Prog. Low Temp. Phys.* **1**, 202 (1955).