PHYSICAL REVIEW A

GENERAL PHYSICS

THIRD SERIES, VOLUME 30, NUMBER 2 AUGUST 1984

Semiempirical Sternheimer shielding factors for the atomic 4*f* and 5*d* shells

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(Received 19 March 1984)

Sternheimer shielding factors are often obtained empirically by comparing nuclear electric quadrupole moment values derived from the hyperfine structure (hfs) of many-electron atoms with those determined by Coulomb-excitation or mesic-atom measurements. A large scatter in the resulting shielding factors arises from the lack of consistency in the approaches used in the analyses of the hfs. In the present work the radial hfs integrals are evaluated with a consistent configuration-average Dirac-Fock method. The scatter in the shielding factors obtained is reduced substantially and the results are in reasonable agreement with Sternheimer's original estimates.

I. INTRODUCTION

In addition to its intrinsic interest, the hyperfine structure (hfs) of atomic levels has been used for many years as a means of evaluating the multipole moments of the nuclei of the atoms involved. The availability of nuclear magnetic-dipole moments obtained by direct measurements has led to a rather good understanding of the procedures to be followed in extracting such moment values from measurements of magnetic-dipole hfs. Direct measurements of nuclear electric quadrupole moments are much less extensive and precise, and have become possible only recently.

In extracting nuclear moment values from hfs measurements a wide variety of procedures is available. All involve, in effect, direct evaluation of the angular factors in the hfs by the rules of angular momentum theory. The radial factors are then obtained either empirically or by *ab initio* calculations. Configuration-interaction and correlation effects are taken into account in the analysis to the extent feasible. In obtaining the quadrupole moment from hyperfine structure, Sternheimer[1] has shown that the moment itself causes a shielding or polarization of the electron shells.[2] The true nuclear quadrupole moment Q is obtained from the apparent or hfs value Q', evaluated as above without considering the shielding, from the relation

$$Q=[1/(1-R_{nl})]Q' , \tag{1}$$

where R_{nl} is the Sternheimer shielding (or antishielding) factor for electrons in the shell *nl*. Unfortunately, Eq. (1) is of limited use for obtaining the true moment from the hfs value because our knowledge of the shielding factors is very limited. It is clear that a reliable table of shielding factors would be of great value both for determining true quadrupole moments where hfs data exist and for achieving a more fundamental understanding of the shielding itself.

In addition to the analysis of quadrupole hfs in atomic fine-structure levels, two other methods are commonly used to obtain nuclear quadrupole moments: x-ray spectroscopy of mesic atoms and Coulomb-excitation studies of the relevant nuclei. Both methods avoid the complications introduced by the many-electron nature of the hfs. In the first method, the quadrupole hfs of the (μ or π) mesic x rays gives the desired information since the electric-field gradient at the nucleus is due almost entirely to the meson in such an atom. In the second method, nuclear Coulomb-excitation data can give accurate quadrupole moments for both ground and excited nuclear states if the states are part of a normal rotational band.

The field of nuclear quadrupole moments has been reviewed a number of times. Summaries of moments derived from atomic hfs studies have recently been given by Olssen and Rosén[3] for the 3*d* shell, and by Büttgenbach[4] and Olssen and Rosén[5] for the 4*d* and 5*d* shells. A comprehensive review of ground-state quadrupole moments derived by all methods has been given by Löbner *et al.*[6]

The present study was stimulated by the recent work of Tanaka *et al.*,[7] who made rather precise quadrupole moment measurements for a number of μ-mesic atoms and then derived empirical Sternheimer shielding factors R_{nl} for these atoms by comparing their moment values with those published earlier based on hfs studies. Examination of the Sternheimer factors they obtained reveals a large scatter in the values for different atoms with the same unfilled electron shell.

It appeared to the present authors that this scatter might well arise from (a) the different procedures used by the many authors for extracting the quadrupole hfs radial parameters

$$b_{02}(nl)=\frac{e^2}{h}Q'\langle r^{-3}\rangle_{nl}^{02} \quad (2)$$

from their measured hfs splittings, and (b) the wide range of approaches used by the same authors for evaluating the quantity $\langle r^{-3}\rangle_{nl}^{02}$ in order to obtain the hfs value of Q' from the $b_{02}(nl)$ value. Our approach is to treat (b) by using a configuration-average Dirac-Fock method consistently in evaluating the radial integrals. Although there is no way to avoid the scatter due to (a) short of a complete and consistent reanalysis of all the hfs data, it is possible by examining the original references to assess their reliability and to comment on the most likely causes of inconsistencies.

It is worthwhile to review some of the ways in which inconsistencies can arise in reducing measured hfs intervals to a final value of $b_{02}(nl)$.[4,8] Since the effective operator theory[9] usually used in the interpretation of hfs depends on three radial parameters for the dipole and three more for the quadrupole hfs (one of which is b_{02}, as discussed above), it is necessary to obtain the dipole (A) and quadrupole (B) hfs constants for at least three states of a given electron configuration (more than three are needed if there is more than one open shell). Eigenvectors are obtained for the levels of interest from least-squares fits to all the known fine-structure levels of the configuration (such fits frequently span several competing configurations explicitly and may take account of far-configuration mixing with additional parameters). A wide choice of procedures and approximations is available in (a) obtaining the eigenvectors and (b) in fitting the resulting parametrized intermediate-coupling (and often multiconfiguration) hfs expressions to a limited set of measured hfs A and B values. Corrections for second-order hyperfine structure (hfs interactions with other atomic states) are often necessary and can also be made in various ways. The hfs papers cited in this work approach these problems differently in detail, and these differences can introduce considerable inconsistency.

II. PRESENT APPROACH

In this study, we calculate the radial parameters $\langle r^{-3}\rangle_{nl}^{02}$ with a configuration-average Dirac-Fock method. Essentially, these are multiconfiguration Dirac-Fock calculations which include the average contributions from all jj-coupled states that arise from a given electron configuration. In particular, instead of adjusting the configuration weights until self-consistency is achieved, they are chosen to be the statistical weights of the corresponding jj configurations and are kept fixed throughout the

TABLE I. Nuclear quadrupole moment information for $4f$-shell atoms. Column 1 gives the nucleus and columns 2 and 3 give the true nuclear quadrupole moment and method of measurement—mesic atom (μ) or Coulomb excitation (CE). Columns 4 and 5 give the electron configuration and the measured hfs quadrupole parameter $b_{02}(4f)$. Column 6 gives the present configuration-average Dirac-Fock value for $\langle r^{-3}\rangle_{4f}^{02}$. Column 7 gives the hfs value of the quadrupole moment obtained by using $b_{02}(4f)$ and $\langle r^{-3}\rangle_{4f}^{02}$ in Eq. (2). The final column gives the semiempirical Sternheimer shielding factor R_{4f} calculated from the true and hfs Q values using Eq. (1).

	Q (true)		hfs				
Isotope	Value (b)	Method	Configuration	$b_{02}(4f)$ (MHz)	Calculation $\langle r^{-3}\rangle$ (a.u.)	Calculation Q' (hfs) (b)	R_{4f}
$^{159}_{65}$Tb	1.432[a]	μ	$4f^85d6s^2$	2337[e,f]	8.9220	1.115	0.221
$^{159}_{65}$Tb	1.47[b,c]	CE	$4f^85d6s^2$	2337[e,f]	8.9220	1.115	0.241
$^{159}_{65}$Tb	1.432[a]	μ	$4f^96s^2$	2173[e]	8.3440	1.108	0.226
$^{159}_{65}$Tb	1.47[b,c]	CE	$4f^96s^2$	2173[e]	8.3440	1.108	0.246
$^{161}_{66}$Dy	2.64[c,d]	CE	$4f^{10}6s^2$	4335[g]	9.0926	2.029	0.231
$^{163}_{66}$Dy	2.648[a]	μ	$4f^{10}6s^2$	4578[g]	9.0926	2.143	0.191
$^{163}_{66}$Dy	2.62[c,d]	CE	$4f^{10}6s^2$	4578[g]	9.0926	2.143	0.182
$^{165}_{67}$Ho	3.57[b,c]	CE	$4f^{11}6s^2$	6131[h]	9.8720	2.643	0.260
$^{165}_{67}$Ho	3.57[b,c]	CE	$4f^{11}6s^2$	6054[i]	9.8720	2.610	0.269
$^{167}_{68}$Er	3.565[a]	μ	$4f^{12}6s^2$	6959[j]	10.6833	2.772	0.222
$^{167}_{68}$Er	3.62[b,c]	CE	$4f^{12}6s^2$	6959[j]	10.6833	2.772	0.234

[a]See Ref. 7.
[b]See Ref. 15.
[c]See Ref. 16.
[d]See Ref. 17.
[e]See Ref. 18.
[f]See Ref. 13.
[g]See Ref. 19.
[h]See Ref. 20.
[i]See Ref. 21.
[j]See Ref. 22.

calculation. The new values of $\langle r^{-3}\rangle_{nl}^{02}$ are used in Eq. (2) to obtain the hfs quadrupole moment values Q' from the published $b_{02}(nl)$ values. Semiempirical Sternheimer shielding factors R_{nl} are then deduced from the hfs and "true" moment values using Eq. (1).

III. RESULTS AND DISCUSSION

The Sternheimer shielding factors obtained in this study are given for the $4f$ shell in Table I and for the $5d$ shell in Table II. In each table the nucleus is specified in

TABLE II. Nuclear quadrupole moment information for $5d$-shell atoms. Column 1 gives the nucleus and columns 2 and 3 give the true nuclear quadrupole moment and method of measurement—mesic atom (μ) or Coulomb excitation (CE). Columns 4 and 5 give the electron configuration and the measured hfs quadrupole parameter $b_{02}(5d)$. Column 6 gives the present configuration-average Dirac-Fock value for $\langle r^{-3}\rangle_{5d}^{02}$. Column 7 gives the hfs value of the quadrupole moment obtained using $b_{02}(5d)$ and $\langle r^{-3}\rangle_{5d}^{02}$ in Eq. (2). The final column gives the semiempirical Sternheimer shielding factor R_{5d} calculated from the true and hfs Q values using Eq. (1). Numbers are given only for neutral atoms except as noted for ^{157}Gd II.

	Q (true)		hfs				
	Value				Calculation	Calculation	
Isotope	(b)	Method	Configuration	$b_{02}(5d)$ (MHz)	$\langle r^{-3}\rangle$ (a.u.)	Q (hfs) (b)	R_{5d}
$^{151}_{63}$Eu	0.903[a]	μ	$4f^7 5d 6s$	513[n]	1.3741	1.589	−0.760
$^{153}_{63}$Eu	2.412[a]	μ	$4f^7 5d 6s$	1312[n]	1.3741	4.064	−0.685
$^{153}_{63}$Eu	2.50[b,c,d]	CE	$4f^7 5d 6s$	1312[n]	1.3741	4.064	−0.626
$^{157}_{64}$Gd II	1.36[a]	μ	$4f^7 5d 6s$	1194[o]	3.1612	1.608	−0.182
$^{157}_{64}$Gd II	1.32[d,e]	CE	$4f^7 5d 6s$	1194[o]	3.1612	1.608	−0.215
$^{157}_{64}$Gd II	1.36[a]	μ	$4f^7 5d 6p$	1218[o]	3.3213	1.561	−0.148
$^{157}_{64}$Gd II	1.32[d,e]	CE	$4f^7 5d 6p$	1218[o]	3.3213	1.561	−0.179
$^{155}_{64}$Gd	1.30[a]	μ	$4f^7 5d 6s^2$	1066[p]	2.8353	1.600	−0.231
$^{155}_{64}$Gd	1.31[d,e]	CE	$4f^7 5d 6s^2$	1066[p]	2.8353	1.600	−0.225
$^{159}_{65}$Tb	1.432[a]	μ	$4f^8 5d 6s^2$	1256[q,r]	2.8968	1.845	−0.289
$^{159}_{65}$Tb	1.47[c,d]	CE	$4f^8 5d 6s^2$	1256[q,r]	2.8968	1.845	−0.255
$^{175}_{71}$Lu	3.46[f]	μ	$5d 6s^2$	3511[s]	3.1110	4.803	−0.388
$^{175}_{71}$Lu	3.51[d,g]	CE	$5d 6s^2$	3511[s]	3.1110	4.803	−0.368
$^{175}_{71}$Lu	3.46[f]	μ	$5d^2 6s$	4017[t]	2.5354	6.743	−0.949
$^{175}_{71}$Lu	3.51[d,g]	CE	$5d^2 6s$	4017[t]	2.5354	6.743	−0.921
$^{177}_{72}$Hf	3.365[a]	μ	$5d^2 6s^2$	4663[u]	4.3563	4.556	−0.354
$^{177}_{72}$Hf	3.15[d,h]	CE	$5d^2 6s^2$	4663[u]	4.3563	4.556	−0.449
$^{179}_{72}$Hf	3.793[a]	μ	$5d^2 6s^2$	5269[u]	4.3563	5.148	−0.357
$^{179}_{72}$Hf	3.74[d,h]	CE	$5d^2 6s^2$	5269[u]	4.3563	5.148	−0.376
$^{181}_{73}$Ta	3.28[i]	μ	$5d^3 6s^2$	4839[s]	5.5757	3.694	−0.126
$^{181}_{73}$Ta	3.28[d,j,k]	CE	$5d^3 6s^2$	4839[s]	5.5757	3.694	−0.126
$^{181}_{73}$Ta	3.28[i]	μ	$5d^4 6s$	4895[s]	4.9571	4.203	−0.281
$^{181}_{73}$Ta	3.28[d,j,k]	CE	$5d^4 6s$	4895[s]	4.9571	4.203	−0.281
$^{185}_{75}$Re	2.19[i]	μ	$5d^5 6s^2$	4844[s]	8.1323	2.535	−0.158
$^{185}_{75}$Re	1.96[l]	CE	$5d^5 6s^2$	4844[s]	8.1323	2.535	−0.293
$^{187}_{75}$Re	2.08[i]	μ	$5d^5 6s^2$	4585[s]	8.1323	2.400	−0.154
$^{187}_{75}$Re	2.00[j,l]	CE	$5d^5 6s^2$	4585[s]	8.1323	2.400	−0.200
$^{185}_{75}$Re	2.19[i]	μ	$5d^6 6s$	4225[s]	7.4625	2.410	−0.100
$^{185}_{75}$Re	1.96[l]	CE	$5d^6 6s$	4225[s]	7.4625	2.410	−0.230
$^{187}_{75}$Re	2.08[i]	μ	$5d^6 6s$	3999[s]	7.4625	2.281	−0.097
$^{187}_{75}$Re	2.00[j,l]	CE	$5d^6 6s$	3999[s]	7.4625	2.281	−0.141
$^{191}_{77}$Ir	0.816[a]	μ	$5d^7 6s^2$	2327[v]	10.9431	0.905	−0.109
$^{193}_{77}$Ir	0.751[a]	μ	$5d^7 6s^2$	2105[v]	10.9431	0.819	−0.090
$^{191}_{77}$Ir	0.816[a]	μ	$5d^8 6s$	1897[v]	10.2152	0.790	0.031
$^{193}_{77}$Ir	0.751[a]	μ	$5d^8 6s$	1716[v]	10.2152	0.715	0.048
$^{197}_{79}$Au	0.547[m]	μ	$5d^9 6s^2$	1985[s]	14.0600	0.601	−0.098

[a]See Ref. 7.
[b]See Ref. 23.
[c]See Ref. 15.
[d]See Ref. 16.
[e]See Ref. 24.
[f]See Ref. 25.
[g]See Ref. 26.
[h]See Ref. 27.
[i]See Ref. 28.
[j]See Ref. 29.
[k]See Ref. 30.
[l]See Ref. 31.
[m]See Ref. 32.
[n]See Ref. 11.
[o]See Ref. 33.
[p]See Ref. 34.
[q]See Ref. 18.
[r]See Ref. 13.
[s]See Ref. 4.
[t]See Ref. 12.
[u]See Ref. 35.
[v]See Ref. 14.

column 1, and the value and method of measurement for the true quadrupole moment in columns 2 and 3 (μ for mesic atom, and CE for Coulomb excitation). The electron configuration in which the hfs is measured and the b_{02} value appear in columns 4 and 5. Column 6 gives the present configuration-average Dirac-Fock $\langle r^{-3}\rangle_{nl}^{02}$ value and column 7 gives the hfs Q' value from Eq. (2). The final column gives the semiempirical shielding factor R_{nl} computed from Eq. (1). The choice of nuclei included is determined by the requirement that dependable values for the true nuclear quadrupole moment and the quadrupole atomic hfs both be available. It may be noted that the shielding factor is extremely sensitive to the moment values used in calculating it. Table II shows, for example, that the 10% difference between the Q values measured for ^{185}Re by the mesic-atom and Coulomb-excitation methods leads to a factor of 2 difference in the computed shielding factors. Despite this sensitivity, the overall consistency of the R_{nl} values in Tables I and II is noteworthy.

The $4f$-shell shielding factors of Table I are plotted in Fig. 1 against the number of $4f$ electrons in the shell. Sternheimer factors derived from Coulomb-excitation moment values are indicated by circles and those from mesic-atom moment values by crosses. The consistency between the two methods is very good, and indicates a shielding factor for the $4f$ shell of about $+0.23(2)$. This is to be compared with an estimate some years ago by Sternheimer[1,10] of $+0.1$. Although good values of the hfs quadrupole moment Q' are available for several atoms in the first half of the $4f$ shell, there are as yet no precise mesic-atom or Coulomb-excitation measurements of the true moments. The values given for R_{4f} in Table I and Fig. 1 are somewhat larger than those of Tanaka *et al.*[7] and, in addition, show considerably less scatter.

The $5d$-shell shielding factors of Table II are plotted in Fig. 2; they are based about equally on moment values from mesic-atom measurements (crosses) and Coulomb-excitation measurements (circles). The right-hand section of the figure shows the dependence of the shielding factors on the number of $5d$ electrons in the shell. The left-hand section shows data for configurations containing a single $5d$ electron in conjunction with one or more additional open shells. The R_{5d} values based on hfs in the $4f^7 5d6s$ configuration of ^{151,153}Eu (Ref. 11), and on the $5d^2 6s$ configuration of ^{175}Lu (Ref. 12) in Table II are omitted from Fig. 2 because they are in sharp disagreement with all the other values and are consequently of questionable reliabilty. Since there is relatively good agreement between the mesic-atom and the Coulomb-excitation moment values for ^{153}Eu and ^{175}Lu, the problem probably arises in deriving proper b_{02} values from the measured hfs. Several possible sources of such difficulties in the analysis of the Eu I hfs may be noted: (1) the $4f^7 5d6s$ levels studied are rather highly excited (13 000–16 000 cm^{-1}) and may contain more configuration-interaction effects than could be taken into account; (2) the reduction of the number of free hfs parameters (though unavoidable) may have been too drastic, especially in the use of Casimir factors instead of through *ab initio* calculations of the radial parameter ratios; and perhaps most important, (3) the eigenvectors used are based on a severe truncation of the $4f^7$ core. Bauche-Arnoult *et al.*[13] have shown that taking the entire $4f^8$ core into account explicitly is necessary for a satisfactory treatment of hfs in the $4f^8 5d6s^2$ configuration of Tb I. The interpretation of the hfs of such complex configurations is an extremely difficult problem and one cannot say with certainty exactly where the problem arises.

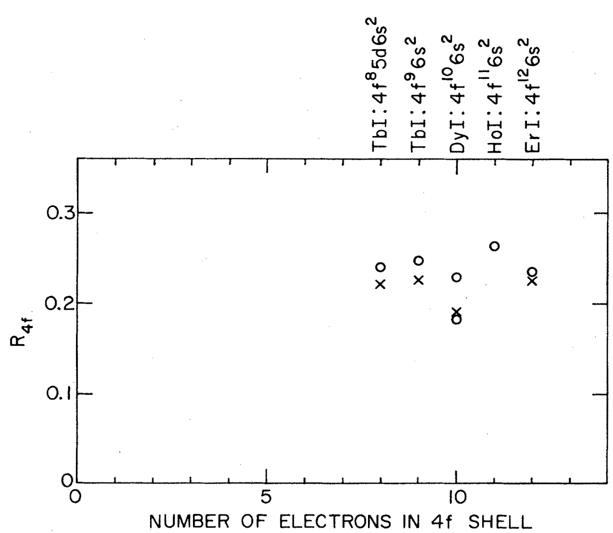

FIG. 1. Semiempirical Sternheimer shielding factors for the $4f$ shell. The values of R_{4f} are plotted against the number of electrons in the $4f$ shell for the $4f^N 6s^2$ configurations of the neutral rare-earth atoms. R_{4f} is obtained by comparing the quadrupole moments determined from hfs studies with the true moments, obtained from mesic-atom measurements (for the crosses) and from Coulomb-excitation studies (for the circles).

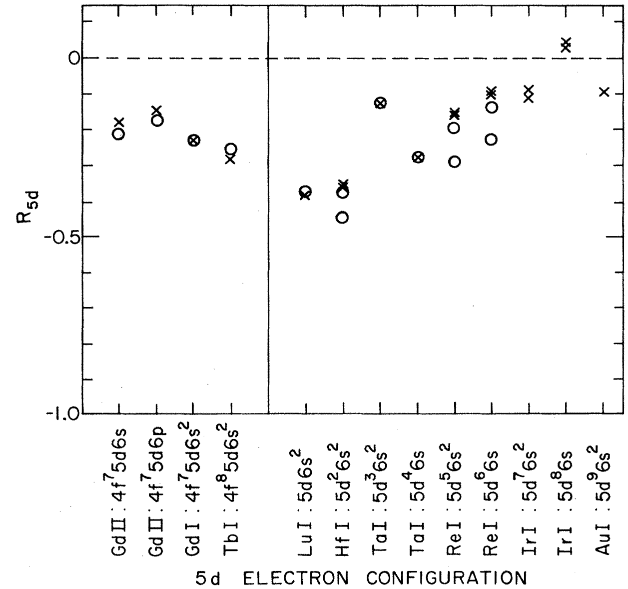

FIG. 2. Semiempirical Sternheimer shielding factors for the $5d$ shell. R_{5d} is shown in the right-hand section for $5d^N 6s$ and $5d^{N-1}6s^2$ configurations, and at the left-hand section for more complex configurations. R_{5d} is obtained by comparing the quadrupole moments derived from hfs studies with the true moments, obtained from mesic-atom measurements (for the crosses) and from Coulomb-excitation studies (for the circles).

In the case of ^{175}Lu, the hfs has been measured[12] for many levels of both odd and even parity configurations. In examining the multiconfiguration least-squares fits to the fine-structure levels, one notes that several levels are strongly mixed and the residuals are rather large for some levels; still higher-lying configurations, which were ignored in the fits, appear to be a problem. In the hfs analysis, the large number of hfs parameters was reduced to a tractable number by setting some equal to zero and assuming reasonable relationships between others. Poor fits to the quadrupole hfs constants for several of the $5d^26s$ states occur, and may play a role in the unexpectedly large value found for b_{02}. It is very difficult to achieve a reliable analysis of hfs if several configurations are strongly interacting.

The $b_{02}(5d)$ values quoted for the $5d^86s$ configuration of Ir I are from Büttgenbach *et al.*[14] Büttgenbach,[4] on reanalyzing the hfs data, has required the ratio of the b_{02} parameters for the configurations $5d^86s$ and $5d^76s^2$ to be the same as that of the corresponding spin-orbit constants. Under this constraint, the b_{02} value for the $5d^86s$ configuration in ^{193}Ir increases to 1992 MHz, and the corresponding shielding factor R_{5d} changes from $+0.048$ to -0.105. Although this R_{5d} value is more consistent with those on either side of it in Fig. 2, there is some arbitrariness in the method used to obtain it.

For the $5d$ shell, Sternheimer[1] has calculated (with the omission of exchange terms) $R_{5d}=-0.384$ for Pr I and -0.443 for Tm I. On including the exchange terms crudely, he has estimated[10] that $R_{5d}=-0.3$ for the $5d$ shell. If one interprets the data of Fig. 2 to represent scatter about a constant value, we find the semiempirical value to be about -0.27 ± 0.10. One might, however, be justified in noting a linear trend in the $5d$-shell data when the $4f$ shell is closed (right-hand section of Fig. 2), with R_{5d} changing from about -0.4 for $5d^1$ to 0 or -0.1 for $5d^9$. The values in the left-hand section of Fig. 2 are perhaps less reliable because of the presence of one or more additional open shells and the consequently increased difficulty of achieving a clear interpretation of the hfs.

IV. CONCLUSIONS

In conclusion, our knowledge of Sternheimer shielding factors, though much improved from that of ten years ago, is still in a very elementary state. Semiempirical values are strongly dependent on the many procedures used in interpreting the hfs as has been briefly outlined above, and the evaluation of the shielding effects by *ab initio* calculations has so far had only limited quantitative success. It is hoped that the present semiempirical treatment will be useful both as a listing of known values for the $4f$ and $5d$ shells and in stimulating more sophisticated *ab initio* efforts at understanding the interaction between the nuclear quadrupole moment and the electronic shells.

ACKNOWLEDGMENT

This research was supported by the U.S. Department of Energy (Office of Basic Energy Sciences), under Contract No. W-31-109-Eng-38.

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