RECEIVED BY TIC AUG 2 1974

CONF-740711--2

STRUCTURE INFORMATION FROM ATOMIC-BEAM MAGNETIC-RESONANCE
STUDIES OF MANY-ELECTRON ATOMS

by

William J. Childs

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Presented at:

4th International Conference on Atomic Physics,
Ruprecht-Karl Universitat, Heidelberg, Germany, July 22-26, 1974

U of C-AUA-USAEC

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

MASTER

U. S. GOVERNMENT

10-11-1974

STRUCTURE INFORMATION FROM ATOMIC-BEAM MAGNETIC-RESONANCE STUDIES
OF MANY-ELECTRON ATOMS*

William J. Childs

Argonne National Laboratory

Argonne, Illinois

INTRODUCTION

In this talk there will be time only for a very brief review of the role that atomic-beam magnetic-resonance (ABMR) has played in the recent development of our understanding of atomic structure. I have chosen to give a few examples of the different types of contributions such research can make, rather than to present extensive tabulation of results.

We cannot here touch on the enormous contribution of ABMR work to nuclear physics, since the pioneering work of Rabi, Zacharias, Millman, and Kusch¹ back in 1938-1940. In the realm of atomic physics, studies using such techniques make possible direct and unambiguous measurement of the electronic angular momentum J , g factor g_J , and hyperfine interaction constants A , B , C , ... of an atomic state. In addition, many other details of atomic structure, such as electron-coupling schemes, admixtures, $\langle r^{-3} \rangle$ values, tests for configuration interaction including core polarization, the presence of relativistic effects and more can be deduced from the quantities measured directly.

THEORETICAL BACKGROUND

Before we consider particular atomic-beam experiments and their results, it is desirable to review the theoretical background briefly.

* Work performed under the auspices of the U.S. Atomic Energy Commission.

Nearly all of the transitions observed in APMR are magnetic-dipole, and occur between different sublevels of a single atomic state $|\alpha SLJ\rangle$. The transition energy is therefore simply the difference between appropriate eigenvalues of the matrix containing the hyperfine and Zeeman Hamiltonians. Even when the theories are put in their simplest form, such a treatment normally allows quantitative description of a large number of precision observations in terms of a very small number of hyperfine constants and a g factor. In taking account of the dipole hyperfine structure, if one distinguishes between the orbital and spin-dipole contributions of orbiting ($l > 0$) electrons to the field at the nucleus, one can understand crudely the J -dependence of the dipole A -factors observed for the different members of a multiplet. Bauche and Judd² in their study of Pu have shown that allowing for the effect of core polarization (polarization of inner, closed s -shells by the outer partially filled electron shell) can markedly improve our understanding of the J -dependence. In 1965, however, Harvey,³ in examining the hfs of O and F, showed that the two parameter approach ($\langle r^{-3} \rangle$ for electrons in the partially filled shell and a second parameter for core polarization) was not consistent with his experimental results. He found that for consistency, one had to allow different values of $\langle r^{-3} \rangle$ for the orbital and spin-dipole contributions to the A factors, thus requiring a total of three parameters.

In order to make the inclusion of relativistic effects in many-electron hyperfine calculations more convenient, Sandars and Beck⁴ in 1965 introduced an effective operator that, when interacting between nonrelativistic LS states produced the same result as the true hyperfine operator between real states. They started with the general form of the hyperfine Hamiltonian

$$\mathcal{H}_{\text{hfs}} = \sum_{k \geq 1} \underline{T}_n^{(k)} \cdot \underline{T}_e^{(k)}, \quad (1)$$

in which $\underline{T}_n^{(k)}$ and $\underline{T}_e^{(k)}$ refer respectively to operators in the nuclear and electronic spaces, and chose for the electronic part the effective operator

$$\underline{T}_e^{(k)}(\text{eff}) = \sum_{k_s, k_l} P^{(k_s, k_l)k} \underline{U}^{(k_s, k_l)k} \quad (2)$$

in which the P 's are constants. The $\underline{U}^{(k_s, k_l)k}$ are double tensor operators of rank k_s in spin space, k_l in orbital space, and k in combined or J space. They showed quite generally that there are at most three such operators $\underline{U}$ for each order k of the hyperfine interaction, and they showed precisely which three are required. Of the various ways of expressing their result for the magnetic-dipole interaction, the most commonly used for an electron configuration of the type $n\ell^N$ is

$$\mathcal{H}_{\text{hfs}}^{(M-1)} = \underline{\underline{I}} \cdot \sum_i [a_{\underline{\underline{1}}}^{01} \underline{\underline{L}}_i - \sqrt{10} a_{\underline{\underline{1}}}^{12} \left\{ \underline{\underline{S}} \cdot \underline{\underline{C}} \right\}_i^{(2)}]^{(1)} + a_{\underline{\underline{1}}}^{10} \underline{\underline{S}}_i] , \quad (3)$$

in which the superscripts on the a's (which are basically the nuclear dipole moment μ times the P's of Eq. (2)) give the values of k_s and k_ℓ for the associated operator. It should be noted that there are three operators, as required by Sandars and Beck, and three a's. The presence of the contact term a^{10} is important, and it does not necessarily vanish for $\ell \neq 0$ electrons.

The corresponding expression for the electric-quadrupole hyperfine interaction may be written, for ℓ^N configurations ($\ell > 0$), as

$$\mathcal{H}_{\text{hfs}}^{(E-2)} = \underline{\underline{T}}_n^{(2)} \cdot \sum_i \left[P_{\underline{\underline{U}}_i}^{(02)^2 (02)^2} + P_{\underline{\underline{U}}_i}^{(13)^2 (13)^2} + P_{\underline{\underline{U}}_i}^{(11)^2 (11)^2} \right] , \quad (4)$$

in which the first term, of rank two in the (electronic) orbital space, is equivalent to the second-order spherical harmonic of the nonrelativistic quadrupole Hamiltonian. The other two terms are entirely relativistic in origin, are normally much smaller than the first term, and are spin-dependent. In writing the quadrupole Hamiltonian, the P's are sometimes replaced by parameters $b^{(k_s, k_\ell)}$ which are basically the same but with the nuclear quadrupole moment Q included. They are thus analogous to the $a^{(k_s, k_\ell)}$ of the dipole interaction.

Sandars and Beck also showed, for a pure ℓ^N configuration, exactly what values are required for the a's, b's, or P's for the effective operator theory to give the correct relativistic result. They showed that each radial quantity $a^{(k_s, k_\ell)}$ or $b^{(k_s, k_\ell)}$, for a given value of the appropriate nuclear moment μ or Q , is given as a definite linear combination of three relativistic radial integrals, each of which has $\langle r^{-3} \rangle$ as its nonrelativistic limit. For the dipole quantities, for example, we may indicate the equivalence (not one-to-one) as

$$\begin{pmatrix} a^{01} \\ a^{12} \\ a^{10} \end{pmatrix} \leftrightarrow \mu \begin{pmatrix} F_{++} \\ F_{--} \\ F_{+-} \end{pmatrix} , \quad (5)$$

in which the dipole integrals $F_{jj'}$ are

$$F_{jj'} \propto \int_0^\infty \frac{P_j Q_{j'} + Q_j P_{j'}}{r^2} dr \rightarrow \langle r^{-3} \rangle_{jj'} \quad (6)$$

and in which P_j and Q_j are the large and small components of the relativistic radial wave function. As indicated in Eq. (6), the nonrelativistic limit of $F_{jj'}$ is $\langle r^{-3} \rangle$ for all j, j' . Because of the way we chose to write the dipole hyperfine Hamiltonian (Eq. (3)), it is convenient to define an effective $\langle r^{-3} \rangle$ value for each operator in the expression by the relation

$$a^{k_s, k_\ell} = \frac{2\mu_B \mu_N}{h} \frac{\mu_I}{I} \langle r^{-3} \rangle^{k_s, k_\ell} \quad (7)$$

in which all symbols have the conventional meaning.

In applying the theory to a real atom, it is clear that we must know in detail the composition of the atomic states studied in order to be able to evaluate the expectation values of the hyperfine Hamiltonians. If the eigenvector of a state is accurately known, and if the nuclear dipole moment μ is known, the A factor for the state may be expressed as a linear combination of the three quantities

$$\langle r^{-3} \rangle_{01}, \langle r^{-3} \rangle_{12}, \langle r^{-3} \rangle_{10}, \quad (8)$$

and if configuration interaction is ignored, these three quantities can be calculated explicitly. Differences between the "observed" and calculated $\langle r^{-3} \rangle$ values can then be regarded as measures of other structural effects. The situation for the quadrupole structure is similar except for the lack of directly measured values of Q . For the magnetic-octupole and electric-hexadecapole hyperfine interactions seen by atomic-beam workers, the $\langle r^{-3} \rangle$ values are replaced by $\langle r^{-5} \rangle$ values.

We also see that since the theory is essentially a three-parameter description, it is highly desirable to make measurements in as many states of a configuration as possible so that the parameters will be over-determined.

APPLICATIONS OF THEORY TO OBSERVED HYPERFINE STRUCTURE

An interesting atom to which to apply the theory is the rare-earth Sm, for which Woodgate⁵, Robertson et al.⁶, and others⁷ have made extensive ABMR measurements of the hyperfine structure in six atomic states of two stable isotopes.

The eigenvectors of the $4f^6 6s^2 {}^7F_{0,1,2,3,4,5,6}$ ground multiplet were determined by Judd and Lindgren⁸ and by Conway and Wybourne⁹ by fitting the parameterized fine-structure eigenvalues to the observed level energies. Although the purity they found is above 34% for the states of interest, it is essential to keep careful account of all the small admixtures when analyzing the hyperfine structure theoretically.

The g_J values measured by ABMR¹⁰ are so precise that relativistic and diamagnetic effects had to be taken into account⁷⁻⁹ before comparison of the predicted values with experiment. It is found⁷ that the differences between theoretical and experimental g values occur only in the sixth figure for each of the six $J \neq 0$ states. This degree of agreement is very unusual, however; differences of up to several percent often occur even for atoms for which multi-configuration eigenvectors are available.

From the Sandars-Beck effective-operator theory, using the best available eigenvectors and the known dipole moment, one may express the A factors for the six 7F states ($J \neq 0$) as linear combinations of the three $\langle r^{-3} \rangle$ values for the $4f$ -electron shell. For example, the result for the 7F_3 state of ${}^{147}\text{Sm}$ is

$$\begin{aligned} A({}^7F_3) &= -11.1346 \langle r^{-3} \rangle^{01} + 2.7979 \langle r^{-3} \rangle^{12} - 11.0206 \langle r^{-3} \rangle^{10} \\ &= -50.2396 \text{ MHz} , \end{aligned} \quad (9)$$

where the experimental result, corrected for second-order hyperfine structure, is given on the right. Since we have one of these equations for each of the six states of 7F , the three $\langle r^{-3} \rangle$ values are overdetermined, and we can obtain the results of Table I by a least-squares fit¹. In the second column, we notice first that a nonzero value is obtained for $\langle r^{-3} \rangle^{10}$ (often an indication of core polarization), and second, that there is a slight difference between the values required for the orbital and spin-dipole $\langle r^{-3} \rangle$. Rosén¹¹ has used an optimized relativistic Hartree-Fock-Slater technique to calculate these integrals ab initio, and his results are seen to be in very good agreement with experiment. The difference between the calculated and experimental values of the ratio $\langle r^{-3} \rangle^{01} / \langle r^{-3} \rangle^{12}$ was found to be relatively insensitive to the method of calculation, and is interpreted as evidence of configuration interaction. The excellent agreement between theory and experiment for $\langle r^{-3} \rangle^{10}$ strongly argues that the contact-like term arises almost entirely from relativistic effects rather than from core polarization or other form of configuration interaction. This is in sharp contrast to the situation in the $3d$ -shell, as we shall see. Table I also shows the striking success modern self-consistent relativistic calculations can have in explaining the quadrupole hyperfine structure. It

should be noted that the two entirely relativistic contributions to the quadrupole hyperfine structure, though small, are understandable at least crudely.

Quantity	Experimental Value (MHz)	Calculated Value (MHz)	Difference (%)
$\langle r^{-3} \rangle^{01}$	6.339(5)	6.225	1.8
$\langle r^{-3} \rangle^{12}$	6.420(5)	6.709	-4.5
$\langle r^{-3} \rangle^{01} / \langle r^{-3} \rangle^{12}$	0.987	0.928	6.4
$\langle r^{-3} \rangle^{10}$	-0.215(5)	-0.230	-7.0
$\langle r^{-3} \rangle^{13} / \langle r^{-3} \rangle^{02}$	0.024	0.030	
$\langle r^{-3} \rangle^{11} / \langle r^{-3} \rangle^{02}$	-0.010	-0.014	

Table I. Comparison of $\langle r^{-3} \rangle$ values deduced from ABMR hfs studies of ^{147}Sm and those calculated by Rosen with a relativistic, optimized Hartree-Fock-Slater technique.

Not all atoms are so well behaved (or understood) as Sm, however. The near earth Ce has 47 levels below 7000 cm^{-1} , of which 33 have been seen¹² by ABMR. In Tb, nearly 20 levels lie below 5000 cm^{-1} , and the ABMR technique has been used¹³ to study the hfs of the lowest 17. In Tb, the configurations $4f^9 6s^2$ and $4f^8 5d 6s^2$ both lie low, and in fact the atomic ground state was only identified by Klinkenberg and van Kleef¹⁴ in 1970. Interpretation of the hfs results for the even-parity states has been difficult because of the extreme complexity of obtaining good eigenvectors from the many thousands of levels expected from $4f^8 5d 6s^2$ and $4f^8 5d^2 6s$. Saying that the problem is only a practical one rather than a fundamental one isn't entirely satisfying, and doesn't lead to improved understanding. In spite of such difficulties, Arnoult and Gersenkorn's¹⁵ severely truncated eigenvectors have had surprising success in accounting for the dipole hyperfine constants, and theoretical work has continued.¹⁶

One of the most interesting features of the Tb analysis is that the value of the ^{159}Tb nuclear quadrupole moment could be extracted¹⁵ from the hyperfine structure of the $4f$ and $5d$ electron shells independently, and the results differed by more than 30%. The intrinsic quadrupole moment has been measured¹⁷ by Coulomb

excitation studies, however, and if the usual projection factor for rotational states is accepted, the resulting "experimental" shielding factors¹⁸ are in good agreement with those calculated by Sternheimer¹⁹.

CONFIGURATION INTERACTION AND HYPERFINE STRUCTURE

Wybourne²⁰ and others have shown that if it is not too strong, the effect of configuration interaction on hyperfine structure is simply to distort the values of $\langle r^{-3} \rangle$ deduced from the data. The importance of configuration interaction in the hyperfine structure of the 3d-shell atoms has long been apparent. A large amount of experimental ABMR data has been obtained²¹⁻²⁷ in a number of laboratories, and the results have been summarized by Winkler²⁸, Childs²⁹, and Armstrong³⁰. Two effects in particular stand out. The first is that the value of the ratio

$$\alpha = \frac{\langle r^{-3} \rangle_{01}}{\langle r^{-3} \rangle_{12}}$$

is observed to be much further from 1 than could be explained¹⁸ by relativistic effects alone, and undergoes substantial changes as one goes through the 3d-shell. Bauche-Arnoult³¹ has studied the problem with some success by perturbation theory; it is clearly associated with configuration interaction.

The second effect of interest is the surprisingly large value found for the core-polarization constant $\chi \propto a^{10}/(u/I)$. The experimental results are summarized in Fig. 1, in which the core-polarization constant is plotted. The uncertainties in the points are difficult to assign because they arise principally from uncertainties in the intermediate-coupling eigenvectors used and are not experimental in origin. The magnitudes of χ are all much too large¹⁸ to be due to the contact-like relativistic term of the Sandars-Beck effective operator, but are due to core polarization. Freeman and Watson³², Bagus, Liu, and Schaefer³³, and others have had some success in accounting for the general trend through the 3d-shell with spin-unrestricted Hartree-Fock calculations, but the magnitudes of the calculated values are only 35-75% of the observed values for most 3d-shell atoms. A many-body calculation by Kelly³⁴ for Fe shows rather good quantitative agreement with experiment.

Some studies of core-polarization effects in the 3p-shell have been made. Although experimental ABMR results for some 4d- and 5d-shell atoms are becoming available, principally from Bonn, the theoretical situation here is more difficult. It may well be that core polarization and other configuration-interaction effects cannot be adequately taken into account without making the calculations relativistically.

Figure 1. Observed variation of the core-polarization constant χ in the ground term of 3d-shell atoms.

ABMR measurements by Ting³⁵ of the hyperfine constants in the $5d6s^2\ ^2D_{3/2,5/2}$ ground term of ^{139}La show that the effects of configuration interaction are very strong. In the absence of configuration interaction, the value calculated from the theory for the ratio

$$A(^2D_{5/2})/A(^2D_{3/2})$$

is 0.429, while the experimental value is 1.291. Although it is simple to analyze this result on the assumption that the discrepancy is due to core polarization, the result of so doing leads to very unphysical values of the radial parameters. It is certainly not due to relativistic effects. Stein³⁶, Wilson³⁷, and more recently Ben Ahmed et al.³⁸ have made 3-configuration least-squares fits to the levels of $5d6s^2$, $5d^26s$, and $5d^3$, and find explicitly strong configuration-interaction admixtures ($\sim 15\%$) in the ground multiplet. With such eigenvectors it has been possible to fit the hyperfine structure observed³⁹ by ABMR for 13 low levels of ^{139}La reasonably well, although the $\langle r^{-3} \rangle$ values deduced are only in fair agreement with Hartree-Fock calculations³⁷. An interesting effect in La is the apparent variation of $\langle r^{-3} \rangle_{5d}$ from state to state, seen both in the experimental hfs³⁹ and in Hartree-Fock calculations by Wilson³⁷. The agreement is only qualitative and the effect should be investigated further. Off-diagonal hyperfine structure (that between two different atomic states) has been used³⁹ with ABMR in La to see how radial parameters vary from state to state, and further work of this kind would be very interesting.

RELATIVISTIC EFFECTS IN HYPERFINE STRUCTURE

Making relativistic calculations of atomic hyperfine structure can be very difficult, especially if configuration-interaction effects are to be included, and experimental measurements of purely relativistic effects are very valuable as a guide for the further development of the theory and of calculational procedures. Perhaps the best place to look for relativistic effects in hyperfine structure is where the nonrelativistic limit predicts no structure. The inherently high precision of the ABMR technique is well suited for this purpose.

Although it is easy to find states of ℓ^N configurations for which the nonrelativistic theory predicts a zero A factor, a non-zero result can always be due to configuration interaction rather than to relativistic effects. For example, a nonzero A value is observed for a number of atomic ground states that arise from half-filled shells, like $d^5\ ^6S_{5/2}$. Nonzero A factors have also been seen in the 3P_1 state of the configuration p^2 , for which the orbital and spin-dipole contributions are predicted to cancel exactly non-relativistically.

Evans, Sanders, and Woodgate⁴⁰ recognized that a significant part of the very small quadrupole hyperfine constant B for the ground S state of half-filled shells could be relativistic in origin, and they made accurate ABMR measurements for 55Mn , $3d^5 4s^2 6s_{5/2}$, and $151,153\text{Eu}$, $4f^7 6s^2 8s_{7/2}$. Their calculations showed that the purely relativistic contribution due to the term $P(11)_{21}(11)_2$ of eq. (4) was much larger than the ordinary contribution of intermediate coupling. Explanation of the exact value observed for the B factor (the quadrupole moments are known from other experiments) has been the subject of several theoretical investigations^{40,41}, and depends on accurate evaluation of the relevant quadrupole relativistic radial integrals. The same problem arises in the $4d^5 5s 7s_3$ ground state of Mo, and has been investigated⁴² in several ABMR experiments.

Relativistic effects can sometimes be seen even when the non-relativistic theory does not predict a zero result. Thus in the p^2 configuration of Sn and Pb, the two states with $J = 2$ can be regarded in first approximation as linear combinations of the two LS basis states 3P_2 and 1D_2

$$\begin{aligned} |^1D_2'\rangle &= \sqrt{1 - \alpha^2} |^1D_2^{LS}\rangle + \alpha |^3P_2^{LS}\rangle \\ |^3P_2'\rangle &= -\alpha |^1D_2^{LS}\rangle + \sqrt{1 - \alpha^2} |^3P_2^{LS}\rangle, \end{aligned} \quad (10)$$

where α is the mixing coefficient. If one sets up the matrix of the electrostatic and spin-orbit interactions for these states in Sn, for example, one finds⁴³ that to fit the observed energies one needs

$$\alpha (\text{energy}) = 0.320,$$

as shown in Table II. The ABMR measurements^{43,44} of the g factors of the $J = 2$ states show equal and opposite perturbations from the LS-limit values, and this perturbation leads directly to the independent result⁴³

$$\alpha (\text{Zeeman}) = 0.323,$$

in nearly perfect agreement. A nonrelativistic analysis of the observed dipole hfs however leads to the somewhat different result

$$\alpha (\text{hfs, nonrelativistic}) = 0.472.$$

When the hyperfine structure is analyzed using the correct relativistic radial integrals, the value found⁴⁵ for α is

$$\alpha (\text{hfs, relativistic}) = 0.366,$$

which is very much closer to the value found from the other two observables. The effects are much more pronounced⁴⁵ in the heavier Pb atom. The situation has been analyzed in detail for both atoms by Lindgren and Rosen⁴⁶.

Observable Used	Value of α Obtained
Energy separation	0.320
Zeeman effect	0.323
Dipole hfs	
(nonrelativistic analysis)	0.472
(relativistic analysis)	0.366

Table II. Dependence of the admixture coefficient α for the $J = 2$ states of Sn on the observable used.

HYPERFINE STRUCTURE OF ACTINIDE ATOMS

Nearly all of the problems faced by theoreticians attempting to calculate atomic hyperfine structure become much more severe for the heaviest elements, and for this reason experimental results are badly needed. Not only does the multiplicity of levels become very large with a resulting complexity of all eigenvectors, but relativistic effects become much more important. Still worse is the fact that it may be difficult or impossible to separate the calculation of mixing effects from relativistic effects.

Considerable ABMR work has been done by Marrus, Armstrong and others at Berkeley⁴⁶ on Pa, Np, Pu, and Am, and such work has been important for recognition² of core polarization in the actinides. More recently, Goodman, Diamond, Stanton, and Fred⁴⁷ at Argonne have measured the g factor of the atomic ground state of ²⁵⁴Fm, and in this way were able to identify it as $5f^{12}7s^2 \ ^3H_6$. The same authors have now measured⁴⁸ the hyperfine structure of ²⁵³Es and shown that the measured dipole hfs constant A is consistent with the independently measured dipole moment μ and Lewis's⁴⁹ relativistic Dirac-Slater radial hyperfine integrals. The eigenvector used for analysis of the hfs was obtained by fitting the experimental g_J value.

SUMMARY AND CONCLUSIONS

I have tried in this brief review to show by examples some of the various ways ABMR experiments can yield information about details of the structure of many-electron atoms. I have only hinted at the important interplay and mutual stimulation of the experimental results and the theoretical development. All contributions to nuclear physics have been omitted entirely.

Apart from the quantities measured directly, the experiments have been of great value for the insight they give for calculation of $\langle r^{-3} \rangle$ and $|\psi_s(0)|^2$ values by ab initio methods, and in recent years with the inclusion of relativistic effects in such calculations. Hyperfine structure is very sensitive to impurities in a state, including those due to core polarization and other forms of configuration interaction, and is therefore well-suited for such investigations. The development of the detailed theory of the Zeeman effect, including corrections^{8,9} for diamagnetic and relativistic effects, was closely correlated with high-precision ABMR measurements. The value of atomic-beam measurements of quadrupole hyperfine structure has been important in understanding shielding effects.

In the years just ahead many areas of investigation are open for atomic-beam research. Comprehensive studies of 4d- and 5d-shell atoms are needed in order to sort out the effects of configuration interaction from those of relativity. Only with such information is it possible to know which simplifications in calculation can safely be made. Much work remains to be done in the region of the actinides, where all aspects of atomic structure become so complex. For lighter atoms, the use of laser excitation to populate metastable states opens up a wide field of research. If hyperfine studies were made of enough states in a single element, it might well be possible to achieve a much more comprehensive understanding of the structure.

Finally, the further development of self-consistent field and many-body techniques for calculations can be expected to suggest and to profit from specific ABMR measurements throughout the periodic table.

REFERENCES

1. I. I. Rabi, J. R. Zacharias, S. Millman, and P. Kusch, Phys. Rev. 53, 318 (1938); P. Kusch, S. Millman, and I. I. Rabi, Phys. Rev. 57, 765 (1940).
2. J. Bauche and B. R. Judd, Proc. Phys. Soc. (London) 83, 145 (1964).
3. J. S. M. Harvey, Proc. Roy. Soc. (London) A285, 581 (1965).
4. F. G. H. Sandars and J. Beck, Proc. Roy. Soc. (London) A289, 97 (1965).
5. G. K. Woodgate, Proc. Roy. Soc. (London) A293, 117 (1966).
6. R. G. H. Robertson, J. C. Waddington, and R. G. Summers-Gill, Canad. J. Phys. 46, 2499 (1968).
7. W. J. Childs and L. S. Goodman, Phys. Rev. A6, 2011 (1972). It may be noted that in making the least-squares fit to the dipole A factors for Table I, the additional constraint $\langle r^{-3} \rangle_{01} - \langle r^{-3} \rangle_{10} = 6.554(23) \text{ a}^{-3}$ (obtained experimentally at strong field in ref. 5) was used.
8. B. R. Judd and I. Lindgren, Phys. Rev. 122, 1802 (1961).
9. J. G. Conway and B. G. Wybourne, Phys. Rev. 130, 2325 (1963).
10. F. M. J. Pichanick and G. K. Woodgate, Proc. Roy. Soc. (London) A263, 89 (1961); see also ref. 7.
11. A. Rosén, J. Phys. B (Atom. Molec. Phys.) 2, 1257 (1969).
12. W. J. Childs and L. S. Goodman, Phys. Rev. A1, 1290 (1970).
13. W. J. Childs, Phys. Rev. A2, 316 (1970); unpublished ABMR work on Tb has also been done by K. H. Chan and P. J. Unsworth.
14. P. F. A. Klinkenberg and Th. A. M. van Kleef, Physica 50, 625 (1970).
15. C. Arnoult and S. Gerstenkorn, J. Opt. Soc. Am. 56, 177 (1966).
16. C. Bauche-Arnoult and J. Bauche, to be published.
17. M. C. Olesen and B. Elbek, Nucl. Phys. 15, 134 (1960); B. Elbek, Thesis, Univ. of Copenhagen, 1963; see also Nuclear Data Tables A7, 527 (1970).
18. I. Lindgren and A. Rosén, Case Stud. Atom. Phys., to be published.
19. R. M. Sternheimer, Phys. Rev. 146, 140 (1966); private communication, (1970).
20. B. G. Wybourne, Spectroscopic Properties of Rare Earths (Interscience, New York, 1965), pp. 148-151.

21. W. J. Childs, Phys. Rev. A4, 1767 (1971); 156, 71 (1967).
22. W. J. Childs and L. S. Goodman, Phys. Rev. 156, 64 (1967); 148, 74 (1966); 170, 50 (1968); 170, 136 (1968).
23. W. J. Childs and B. Greenebaum, Phys. Rev. A6, 105 (1972).
24. H. Gebauer, private communication, (1973).
25. K. H. Channappa and J. M. Pendlebury, Proc. Phys. Soc. (London) 86, 1145 (1965).
26. G. K. Woodgate and J. S. Martin, Proc. Phys. Soc. (London) 70A, 485 (1957).
27. W. Fischer, Z. Physik 161, 89 (1961).
28. R. Winkler, Phys. Letters 23, 301 (1966).
29. W. J. Childs, Phys. Rev. 160, 9 (1967).
30. L. Armstrong, Jr., Theory of the Hyperfine Structure of Free Atoms (Wiley-Interscience, New York, 1971).
31. C. Bauche-Arnoult, Proc. Roy. Soc. (London) A322, 361 (1971).
32. A. J. Freeman and R. E. Watson, Phys. Rev. 131, 2566 (1963); 123, 2027 (1961).
33. P. S. Bagus, B. Liu, and H. F. Schaefer, III, Phys. Rev. A2, 555 (1970).
34. H. P. Kelly, Phys. Rev. A2, 1261 (1970).
35. Y. Ting, Phys. Rev. 108, 295 (1957).
36. J. Stein, J. Opt. Soc. Am. 57, 333 (1967).
37. M. Wilson, Phys. Rev. A3, 46 (1971).
38. Z. Ben-Ahmed, C. Bauche-Arnoult, and J. F. Wyart, private communication, (1974).
39. W. J. Childs and L. S. Goodman, Phys. Rev. A3, 25 (1971).
40. L. Evans, P. G. H. Sandars, and G. K. Woodgate, Proc. Roy. Soc. (London) A289, 108 (1965); A289, 114 (1965).
41. M. A. Coulthard, Proc. Phys. Soc. (London) 90, 615 (1967); J. P. Desclaux, private communication, (1971).
42. J. M. Pendlebury, and D. B. Ring, J. Phys. B5, 386 (1972); S. Büttgenbach, M. Herschel, G. Meisel, E. Schrödl, W. Witte, and W. J. Childs, Z. Physik 266, 271 (1974).
43. W. J. Childs, Phys. Rev. 4, 439 (1971).
44. W. J. Childs and L. S. Goodman, Phys. Rev. 134, A66 (1964).
45. A. Lurio and D. A. Landman, J. Opt. Soc. Am. 60, 759 (1970).

46. See, for example, J. Faust, R. Marrus, and W. A. Nierenberg, Phys. Letters 16, 71 (1965); L. Armstrong, Jr. and R. Marrus, Phys. Rev. 144, 994 (1966).
47. L. S. Goodman, H. Diamond, H. Stanton, and M. S. Fred, Phys. Rev. A4, 473 (1971).
48. L. S. Goodman, H. Diamond, and H. Stanton, private communication, (1974).
49. W. B. Lewis, private communication (1972).