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Hyperfine-structure measurements in ^{183}W and the contact interaction in $5d^N6s$ atoms

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Effective radial parameters of the magnetic dipole hyperfine interaction and g_J values have been determined from atomic beam magnetic-resonance measurements in five metastable states of W. The contact-interaction parameters a_{6s}, important for hyperfine anomaly and Knight shift, are analyzed and tabulated for the $5d^N6s$ configurations.

I. INTRODUCTION

^{183}W, which has a natural abundance of 14%, is the only stable tungsten isotope with nonzero nuclear spin $I=\frac{1}{2}$. Until now several optical measurements of the hyperfine structure (hfs) have been performed,[1-3] but the more precise atomic-beam magnetic-resonance (ABMR) method[4] could not be applied because of the extremely high evaporation temperature of about 3800 K. The development of a universal, crucible-free evaporation technique using a 100-kV electron gun[5] and the improvement of this method during the last years made it now possible to perform ABMR measurements in the atomic states $5d^46s^2\,^5D_{1,2,3,4}$ and $5d^56s\,^7S_3$ with excitation energies up to 6200 cm^{-1}. The ground state $^5D_0$ is not suitable for the ABMR method because of $J=0$. Stimulated by new investigations of the W spectrum,[6,7] intermediate-coupling wave functions have been calculated[8] for the configurations $(5d+6s)^6$. Using these wave functions it is possible to evaluate the effective radial parameters $a_d^{k_sk_l}$ of the $5d^46s^2$ configuration and the contact interaction parameters a_{6s}^{10}. If these parameters are adjusted to the experimental hfs results they include influences of relativistic and configuration-interaction effects on the hfs.[4,9] Our present experiments extend the systematic investigations of these effects in the $4d$ shell[10] to the $5d$ shell. Preliminary results for ^{183}W, assuming pure SL coupling, have already been reported in Ref. 10. Improvement of the values for the radial parameters is important for the extraction of nuclear properties from hfs data. For example, the contact interaction parameter a_s^{10} due to the hyperfine interaction of an unpaired s electron is very sensitive to effects of the finite size of the nucleus. Therefore we give a new compilation of the corresponding parameters $[dP_{6s}(r)/dr]_0^2=4\pi|\Psi_s(0)|^2$ for the $5d^N6s$ series, where $\Psi_s(0)$ is the value of the Schrödinger wave function of the s electron at the nucleus. For the definition of $P(r)$ see, for example, Ref. 11.

II. MEASUREMENTS AND RESULTS

The electronic g_J factors of the five states up to 6200 cm^{-1}, which are needed for checking the quality of the intermediate-coupling (IC) wave functions and for extrapolating the hfs measurements to zero magnetic field, were measured in the 31% isotope ^{184}W. Because of $I=0$ there is no hfs in this isotope. In all cases the double quantum transition $M_J=+1 \leftrightarrow M_J=-1$ has been measured at three settings of the external magnetic field up to 400 Oe, since in second-order perturbation theory this transition is not perturbed by Zeeman interactions with neighboring fine-structure states. The experimental procedure has been described in detail before.[12] The experimental g_J values g_J^{exp} are given in column 2 of Table I. They are well reproduced by the values g_J^{IC} calculated from the intermediate-coupling wave functions (column 3 of Table I).

Using predictions from $\Delta F=0$ resonances measured at magnetic fields up to 300 Oe the $\Delta F=1$ resonances belonging to the hfs intervals of the five states were easily found. Because of $I=\frac{1}{2}$ the hfs splitting of ^{183}W is only caused by magnetic dipole interaction. The corresponding interaction constants A were fitted to the $\Delta F=1$ resonance frequencies measured at magnetic fields of 1 and 2 Oe. The results are given in the last column of Table I. The effects of second-order perturbations due to off-diagonal hfs and Zeeman interactions were calculated using the complete

TABLE I. g_J values and hyperfine interaction constants A of ^{183}W.

State	g_J^{expt}	g_J^{IC}	A (MHz)
$^5D_1$	1.499 26(7)	1.498	29.118(13)
$^5D_2$	1.486 83(8)	1.485	56.261(8)
$^5D_3$	1.478 13(8)	1.475	78.020(6)
$^5D_4$	1.455 14(7)	1.452	88.308(5)
$^7S_3$	1.981 42(5)	1.978	505.592(12)

set of intermediate-coupling wave functions. All fine-structure states up to 23 000 cm^{-1} were regarded as perturbing states. For all investigated atomic states the corrections turned out to be much smaller than the experimental error limits.

Since the intermediate-coupling wave functions for W span the three configurations $(5d+6s)^6$, each A factor depends on $10a^{k_s k_l}$ parameters in the effective operator formalism[9,10] ($3a^{k_s k_l}$ for the open d shell in each of the three configurations and a_{6s}^{10} for the s electron in the $5d^5 6s$ configuration). Therefore several constraints have to be made in order to reduce the degrees of freedom. Other complications arise from the fact that in the SL limit the parameters $a_d^{10}(5d^5 6s)$ and $a_{6s}^{10}(5d^5 6s)$ are linearly dependent, and so also, because $S=L$ for the ^5D multiplet, are the parameters $a_d^{01}(5d^4 6s^2)$ and $a_d^{10}(5d^4 6s^2)$. The existence of such correlations led to irregular values for the effective radial parameters of the $5d^4 6s^2$ configuration, even in an atom so far away from pure SL coupling as W. Because in all other experiments on $5d$- and $4d$-shell atoms the parameter a^{01} was well reproduced by relativistic Hartree-Fock (HF) results of Lindgren and Rosén,[11] $a_d^{01}(5d^4 6s^2)$ was fixed to the value obtained by linear interpolation between the HF values for Ta and Re. $a_d^{01}(5d^5 6s)$, $a_d^{12}(5d^5 6s)$, $a_d^{01}(5d^6)$, and $a_6^{12}(5d^6)$ were related to $a_d^{01}(5d^4 6s^2)$ in nonrelativistic approximation by the ratios of the spin-orbit parameters $\zeta(5d^5 6s)/\zeta(5d^4 6s^2)$ and $\zeta(5d^6)/\zeta(5d^4 6s^2)$, and $a_d^{10}(5d^6)$ was neglected. Errors caused by these assumptions are expected to be small due to the small coefficients of these parameters for all investigated levels. The breakdown of SL coupling reduces the interdependence of $a_d^{10}(5d^5 6s)$ and $a_{6s}^{10}(5d^5 6s)$ only slightly. Therefore $a_d^{10}(5d^5 6s)$ was held equal to $a_d^{10}(5d^4 6s^2)$ in accordance with results in $4d^N 5s^2$ and $4d^{N+1} 5s$ configurations. This assumption influences mainly the value of $a_{6s}^{10}(5d^5 6s)$. But since a_{6s}^{10} and a_d^{10} are of different order of magnitude a change of 100% in a_d^{10} changes the value of a_{6s}^{10} by only 10%, and this situation was felt to be passable. As final results for the remaining free parameters we obtained $a_d^{01}(5d^4 6s^2)=145$ MHz (fixed at the HF value), $a_d^{12}(5d^4 6s^2)=75$ MHz, $a_d^{10}(5d^4 6s^2)=-61$ MHz, $a_{6s}^{10}(5d^5 6s)=3315$ MHz. The differences between the recalculated and the experimental values of the A factors range from 1.4% to 5.8% for the ^5D multiplet. For the $^7S_3$ level the residual is 0.13% Combining the radial parameters with the NMR value of the magnetic dipole moment[13] $\mu_I(^{183}W)=0.116\,205(2)\mu_N$ (uncorrected for diamagnetism) we can evaluate experimental values $\langle r^{-3}\rangle_{k_s k_l}$. The results are $\langle r^{-3}\rangle_{01}=6.54\,a_0^{-3}$, $\langle r^{-3}\rangle_{12}=3.38 a_0^{-3}$, $\langle r^{-3}\rangle_{10}=-2.75 a_0^{-3}$, $[dP_{6s}(r)/dr]_0^2=224.2 a_0^{-3}$. These quantities complete the data of Ref. 10 and are in good agreement with the other results. The value of $\langle r^{-3}\rangle_{10}$, although very sensitive to the intermediate-coupling wave functions, is of the right sign and order of magnitude.

III. CONTACT INTERACTION IN $5d^N 6s$ ATOMS

The s-contact interaction parameter $[dP_{6s}(r)/dr]_0^2$, which is related to a_{6s}^{10} by the equation

$$a_{6s}^{10}=(\mu_0/4\pi)(2\mu_B/h)(\mu_I/I)\tfrac{2}{3}[dP_{6s}(r)/dr]_0^2,$$

plays an important role in $5d$-shell atoms because of its magnitude and the strong configuration mixing in this region. Furthermore, the contact interaction is very sensitive to nuclear effects such as the distribution of nuclear magnetism which gives rise to the Bohr-Weisskopf effect,[14] the dominating part of the hfs anomaly. The second contribution to the hfs anomaly, the Breit-Rosenthal effect,[15] which is due to the deviation of the electrostatic potential inside the nucleus from a Coulomb potential, can reach a considerable amount for large Z, but the isotopic variation is very small. Theoretical HF evaluations of $[dP_{ns}(r)/dr]_0^2$ refer to a point-nucleus model and thus are not isotope dependent. For comparison to such calculations and to recognize systematic

TABLE II. Experimental and theoretical values of the $6s$-contact interaction parameter $[dP_{6s}(r)/dr]_0^2$ (in a_0^{-3}). The experimental point-nucleus values are calculated using the corrections for the Breit-Rosenthal (δ_{BR}) and Bohr-Weisskopf (ϵ_{BW}) effect given in columns 5 and 6.

Z	Isotope	a_{6s}^{10} (MHz)	$[dP_{6s}(r)/dr]_0^2$	$(1-\delta_{BR})$	$(1+\epsilon_{BW})$	$[dP_{6s}(r)/dr]_{0,\text{point}}^2$ Expt.	Theor.
71	^{175}Lu	81 66	203.2	0.941		216	
72	^{179}Hf			0.937			244
74	^{183}W	33 15	224.2	0.930		241	297
75	^{187}Re	198 68	246	0.926		266	355
76	^{189}Os	77 40	281	0.922		305	
77	^{193}Ir	23 69	356.3	0.917	1.071	363	422
78	^{195}Pt	300 00	393	0.911		431	443
79	^{197}Au	30 50	501	0.906	1.078	513	494

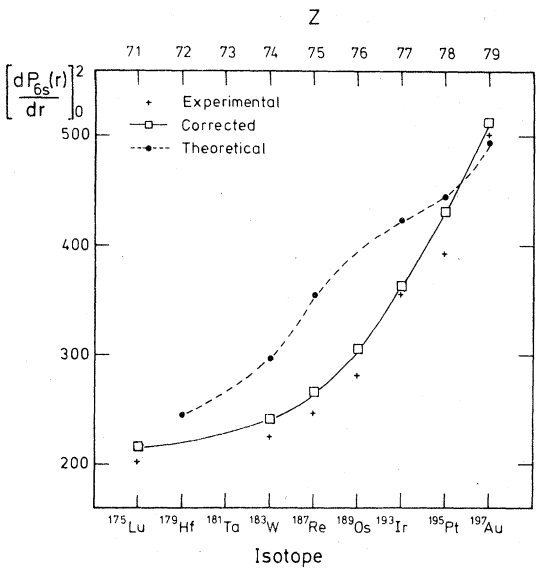

FIG. 1. 6s-contact term in the $5d^N6s$ shell in units of a_0^{-3}.

variations within an atomic shell the experimental parameters have to be corrected according to

$$a_{6s,\,\text{point}} = a_{6s,\,\text{expt}}/[(1+\epsilon_{BW})(1-\delta_{BR})]\,.$$

The Breit-Rosenthal corrections δ_{BR} have been taken from Ref. 16. For the Bohr-Weisskopf correction ϵ_{BW} Moskowitz and Lombardi[17] derived from several mercury isotopes the empirical relation $\epsilon_{BW}[\%] = \alpha/\mu_I$ where for s-electron atomic states $\alpha = \pm 1.13\mu_N (I = l \pm \frac{1}{2})$, l being the orbital angular momentum of the odd neutron. For odd protons opposite signs have to be used.[18] Since calculations of $^1\Delta_s^2$ using this relation yields also perfect agreement with experimental results for Ir (Ref. 19) and Au,[20] this rule was used to calculate the Bohr-Weisskopf correction for Ir and Au for which ϵ_{BW} was expected to be of considerable amount because of the small magnetic moments. In addition to the value for W presented in this paper ABMR measurements have been performed for ^{193}Ir (Ref. 19) and ^{197}Au.[21] For ^{175}Lu a precise value for a_{6s} is given in Ref. 22. Furthermore, we analyzed the optical data of ^{187}Re (Ref. 23) and ^{189}Os (Ref. 24) with respect to the contact term in intermediate coupling using for the d-electrons effective $\langle r^{-3}\rangle_{k_s k_l}$ values extrapolated from measurements in Hf, Ta, W, and Ir.[10] In the same way a rough estimate was obtained for ^{195}Pt (Ref. 25) in pure SL coupling. The experimental values for the contact parameter and the calculated corrections ϵ_{BW} and δ_{BR} are summarized in Table II. The Bohr-Weisskopf correction for the isotopes ^{175}Lu, ^{187}Re, ^{189}Os, and ^{195}Pt was assumed to be negligibly small because of the large magnetic dipole moments. This might be incorrect for ^{183}W. But even in this case the correction ϵ_{BW} was neglected as the complex structure of deformed nuclei leads to very complicated calculations. $[dP_{6s}(r)/dr]_0^2$ is plotted in Fig. 1. In addition theoretical HF values of the 6s-contact interaction parameters[26] are given, which have been calculated because of their importance for the normalization of hyperfine fields obtained by NMR in solids. Although adjusted to the experimental value of Au the theoretical calculations do not reproduce the experimental values very well, and further theoretical efforts are highly desirable.

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