Sternheimer free determination of the ^{59}Co nuclear quadrupole moment from hyperfine-structure measurements

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Taking into account results of earlier hyperfine-structure (hfs) measurements, the hfs of altogether 20 fine-structure (fs) levels has been analyzed by the simultaneous parametrization of the one- and two-body interactions in the atomic hfs for the model space $(3d+4s)^{N+2}$ $(N=7)$. The values of the one- and two-body hfs parameters have been determined. The evaluation of the nuclear quadrupole moment of ^{59}Co including Sternheimer corrections up to second order yielded the value of 0.41(1) b. Moreover, the values of the magnetic-dipole constants A and the electric-quadrupole constants B for all levels of the model space $(3d+4s)^9$ have been predicted.

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INTRODUCTION

In a previous paper [1] interpreting the hyperfine structure (hfs) of high-lying metastable levels of the cobalt atom, Guthöhrlein and Keller stated that the obtained hfs constants could not be explained satisfactorily within the framework of the hfs theory given by Sandars and Beck [2]. The existing differences between measured A and B constants and values estimated according to the theory of Sandars and Beck may originate from far configuration interactions, which their theory neglects.

However, recently an alternative parametrization, of hfs interactions has been developed by Dembczyński *et al.* [3], which takes into account all one- and two-body contributions to the hfs splitting of electronic levels separately and explicitly. With this parametrization the Sandars and Beck radial parameters $a^{\kappa k}$ and $b^{\kappa k}$ are also determined more accurately. This is important, especially in connection with the parameters b^{13} and b^{11}, which represent the relativistic effects only. Hence a test of relativistic effects influencing the hfs is also possible. As major progress this parametrization method offers the evaluation of a nuclear quadrupole moment free of Sternheimer corrections [4] up to second order. This possibility has been tested so far for the ^{47}Ti atom only [5].

In our hfs analysis we use the highly accurate results first obtained by von Ehrenstein, Kopfermann, and Penselin [6] and later extended to further levels by Childs and Goodman [7] using the atomic-beam magnetic-resonance method. Even higher-lying metastable levels were investigated applying Doppler-free and Doppler-limited laser spectroscopy by Guthöhrlein and coworkers [1,9], Wenzel [8], Ibrahim-Rûd [10], and Baier [11].

Section I gives some remarks about the necessary fine-structure analysis. The evaluation of the radial parameters of the hfs interaction is described in Sec. II. In Sec. III we compare the experimental results with an *ab initio* theoretical calculation. Section IV gives a general discussion and determination of the nuclear quadrupole moment of ^{59}Co.

I. REMARKS ON FINE-STRUCTURE ANALYSIS AND INTERMEDIATE-COUPLING WAVE FUNCTIONS

As we have shown in an earlier paper [12], the accuracy of the eigenvector amplitudes has a particularly strong influence on the calculation of the effective one-electron hfs parameters starting from the experimental A and B constants. Therefore we have tried to take into account first- and second-order effects on the fine structure (fs) of the cobalt atom as comprehensively as possible with the experimental data available at present.

It is known for the $3d$ elements that the interaction with distant configurations via Coulomb interaction affects the hyperfine structure very strongly [13] and it has also an influence on the fs-term positions as well as on the spin-orbit splittings of the terms. Therefore the fine-structure analysis together with calculations of the eigenvector amplitudes required for hfs interpretation should take these effects into account.

In previous work concerning $3d$ elements [5,12–15] the fine-structure analysis was limited to the model space $(3d+4s)^{N+2}$. In the cobalt atom, just as in the other $3d$ elements, the model space $(3d+4s)^9$ is not well isolated from the other electronic system configurations because some of the levels of configurations ($3d^8 5s$, $3d^7 5s 4s$, and

$3d^7 4d 4s$) are situated below the levels $^2_1D_{3/2,5/2}$ belonging to the model-space configuration $3d^7 4s^2$. Therefore in this work we perform fine-structure analysis using an extended model space, i.e., the analysis of the following system:

$$3d^7 4s^2 + 3d^8 4s + 3d^9 + 3d^7 5s 4s + 3d^8 5s + 3d^8 4d + 3d^7 4d 4s + 3d^8 6s + 3d^7 6s 4s + 3d^6 4d 4s^2 + 3d^7 4p^2 \ . \quad (1)$$

The configuration system $(3d+4s)^9$ forms a subspace of extended model space and will be referred to below as "model space," contrary to the whole system (1), called "extended model space." The analysis of the extended model space allows one to determine quantitatively the interaction between states of the model space and states of the remaining configurations belonging to the extended system. One- and two-electron excitation effects on the term structure of the subspace $(3d+4s)^9$ have been taken into account by the method described in [16].

In this work the effects of the one-electron excitation on the spin-orbit splittings of the terms [electrostatically correlated spin-orbit interaction (ELSO)] have been completely taken into account. These effects have exactly the same origin as those which affect the hyperfine splittings of the levels. Therefore it is reasonable to parametrize the above effects by means similar to hyperfine-structure parametrization [3]. The fine structure of the terms is affected only by the $3d \rightarrow n'''d \rightarrow 3d$ electron excitations ($n'''d$ labeling empty d shells); also, the effect of one-electron excitation $4s \rightarrow n'''d \rightarrow 3d$ gives nonzero off-diagonal matrix elements of spin-orbit interaction between the states belonging to component configurations of the model space $(3d+4s)^9$ [17], hence only equivalents of hfs parameters a_i ($i=1,2,3,4,5,7,8$; a_i were defined in [3] and their physical meaning will be explained in Sec. II) are necessary to consider ELSO perturbations of the fine structure. Parameters of ELSO interaction have been designated as P_i and their definitions can be obtained by substitution of $[2\mu_I\mu_B/I]\ \langle 3d|r^{-3}|n'''d\rangle$ by $\zeta(n'''d,3d)$ in the definitions of a_i given in [3]. Thus the fs parameters P_i introduced here are defined as follows:

For the electron excitation $3d \rightarrow n'''d \rightarrow 3d$,

$$P_1 = \sum_{n'''} (3d||C^{(0)}||3d)(3d||C^{(0)}||n'''d) \times R^0(3d3d,3dn'''d)\zeta(n'''d,3d)/\Delta E \ , \quad (2)$$

$$P_2 = \sum_{n'''} (3d||C^{(2)}||3d)(3d||C^{(2)}||n'''d) \times R^2(3d3d,3dn'''d)\zeta(n'''d,3d)/\Delta E \ , \quad (3)$$

$$P_3 = \sum_{n'''} (3d||C^{(4)}||3d)(3d||C^{(4)}||n'''d) \times R^4(3d3d,3dn'''d)\zeta(n'''d,3d)/\Delta E \ , \quad (4)$$

$$P_4 = \sum_{n'''} (3d||C^{(0)}||n'''d)(4s||C^{(0)}||4s) \times R^0(3d4s,n'''d4s)\zeta(n'''d,3d)/\Delta E \ , \quad (5)$$

$$P_5 = \sum_{n'''} (3d||C^{(2)}||4s)(4s||C^{(2)}||n'''d) \times R^2(3d4s,4sn'''d)\zeta(n'''d,3d)/\Delta E \ . \quad (6)$$

For the electron excitation $4s \rightarrow n'''d \rightarrow 3d$,

$$P_7 = \sum_{n'''} (3d||C^{(2)}||3d)(4s||C^{(2)}||n'''d) \times R^2(3d4s,3dn'''d)\zeta(n'''d,3d)/\Delta E \ , \quad (7)$$

$$P_8 = \sum_{n'''} (3d||C^{(2)}||n'''d)(4s||C^{(2)}||3d) \times R^2(3d4s,n'''d3d)\zeta(n'''d,3d)/\Delta E \ . \quad (8)$$

The parameters P_1, P_2, and P_3 connected with ELSO effect within a $3d^N$ core are similar to those defined by Judd, Crosswhite, and Crosswhite [18] for a $4f$ core. Introduction of the P_i parameters improves not only the fs fit and the accuracy of eigenvector amplitudes but also allows the possibility to compare effects arising from the same kind of electron excitations and observed independently in fine and hyperfine structure of the atom.

We would like to point out that an attribute of the method we use is the assumption that the orbital $3d$ and also the open-$n'l'$ and the inner-$n''l''$ shell orbitals are common to all states within the extended model space. As a consequence we can define, e.g., one spin-orbit parameter $\zeta(3d,3d)$ for all configurations of the extended model space, and we express the strength of spin-orbit interaction in particular configurations using P_i parameters. For example, the relations between one-configuration spin-orbit parameters ζ_{nl} usually used for configurations that are members of the model space $(3d+4s)^{N+2}$ are

$$\zeta_{3d}(3d^{N+M}4s^{2-M}) = \zeta(3d,3d) + [2/(2l+1)][1-(N+M)]P_1 - [2(2-M)/\sqrt{2l+1}]P_4 + [2/(2l+1)]P_6\delta(M,0) \ , \quad (9)$$

where $M=0,1,2$.

The level-fitting calculation (fs fit) for 132 energy levels attributed to the extended model space have been carried out. With 232 parameters, 55 of which were treated as free, a very good fit with a mean-square deviation of 4 cm^{-1} has been achieved. The method of energy-matrix construction for the extended model space and details of the fine-structure analysis for ^{59}Co will be presented separately [17,19]. Below we give only the values of spin-orbit and P_i parameters, which are necessary for discussion of configuration-interaction effects (see Sec. III). In Table I are given the experimental level values, calculated eigenvalues, percentages of first and second components, and total percentage contribution from the states belonging to model space $(3d+4s)^9$. The hfs parametrization method [3] we use in Sec. II refers explicitly to states of

TABLE I. Extract from the fs fit for the extended model space relating to the levels of the model space $(3d+4s)^9$.

Obs. level (cm^{-1}) [30]	Calc. eigenvalue (cm^{-1})	ΔE	Largest eigenvalue component (%)	Next-lgst. eigenvalue component (%)	Total from model space (%)	Calc. g_J	Obs. g_J	Δg_J
			$J=\frac{1}{2}$					
14 399.28	14 395.11	4.17	97.3 a ^4_3P	1.3 a ^2_3P	98.8	2.642	2.651 0	0.009
16 195.68	16 194.24	1.44	98.2 b $(^3_2P)^4P$	0.4 g $(^2_3D,^3D)^4P$	98.4	2.668	2.682	0.014
18 775.01	18 774.96	0.05	62.7 b $(^3_2P)^2P$	34.8 a ^2_3P	98.4	0.686	0.695	0.009
21 215.90	21 218.74	−2.84	63.4 a ^2_3P	34.8 b $(^3_2P)^2P$	98.8	0.677	0.680	0.003
	46 394.30		96.8 b $(^1_0S)^2S$	2.2 g $(^2_3D,^3D)^2S$	97.0	2.002		
			$J=\frac{3}{2}$					
1 809.33	1 812.24	−2.91	98.9 a ^4_3F	0.3 j $(^3_4F)^4F$	99.2	0.399 7	0.399 4	−0.000 3
5 075.83	5 082.61	−6.78	98.5 b $(^3_2F)^4F$	0.4 b $(^1_2D)^2D$	98.9	0.400	0.404	0.004
14 036.28	14 036.27	0.01	94.9 a (^4_3P)	3.5 a ^2_3P	98.8	1.719	1.722	0.003
15 774.04	15 775.57	−1.53	69.3 b $(^3_2P)^4P$	25.7 b $(^1_2D)^2D$	98.6	1.475	1.476	0.001
16 470.60	16 458.48	12.12	63.8 b $(^1_2D)^2D$	28.9 b $(^3_2P)^4$	98.9	1.099	1.101	0.002
18 389.57	18 390.51	−0.94	50.7 b $(^3_2P)^2P$	36.4 a ^2_3P	98.6	1.295	1.300	0.005
20 500.71	20 498.23	2.48	46.9 a ^2_3P	39.5 b $(^3_2P)^2P$	98.8	1.277	1.284	0.007
23 152.57	23 147.14	5.43	61.7 a ^2_3D	17.4 a ^2_1D	99.4	0.868	0.790	−0.078
28 470.51	28 462.76	7.75	88.9 c ^2D	8.2 c ^2_3D	97.3	0.800	0.907	0.107
	53 918.18		66.9 a ^2_1D	15.9 a ^2_3D	84.0	0.801		
			$J=\frac{5}{2}$					
1 406.84	1 408.22	−1.38	99.0 a ^4_3F	0.3 j $(^3_4F)^4F$	99.1	1.028 84	1.028 26	−0.000 58
4 690.18	4 692.59	−2.41	98.1 b $(^3_2F)^4F$	0.7 b $(^3_2F)^2F$	98.9	1.028	1.027	−0.001
8 460.81	8 464.94	−4.13	97.2 b $(^3_2F)^2F$	0.7 b $(^3_2F)^4F$	98.4	0.860	0.802	−0.058
13 795.52	13 800.24	−4.72	98.2 a ^4_3P	0.3 j $(^5_4D)^4P$	98.7	1.600	1.604	0.004
15 184.04	15 186.18	−2.14	76.1 b $(^3_2P)^4P$	21.8 b $(^1_2D)^2D$	98.6	1.511	1.515	0.004
16 778.16	16 786.47	−8.31	74.7 b $(^1_2D)^2D)$	22.1 b $(^3_2P)^4P$	99.0	1.290	1.296	0.006
21 920.09	21 924.20	−4.11	68.0 a ^2_3D	21.9 a ^2_1D	99.4	1.201	1.240	0.039
27 497.06	27 504.66	−7.60	88.3 c ^2_1D	8.8 c ^2_3D	97.3	1.200	1.200	0.000
	34 639.33		98.9 a ^2_3F	0.1 j $(^1_4G)^2F$	99.2	0.857		
	54 274.86		66.6 a ^2_1D	18.4 a ^2_3D	86.5	1.174		
			$J=\frac{7}{2}$					
816.00	814.84	1.156	99.1 a ^4F	0.3 j $(^3_4F)^4F$	99.1	1.238 43	1.237 78	−0.000 65
4 142.66	4 140.11	2.551	97.4 b $(^3_2F)^4F$	1.5 b $(^3_2F)^2F$	98.9	1.237 13	1.237 61	−0.000 53
7 442.41	7 438.48	3.927	96.8 b $(^3_2F)^2F$	1.5 b $(^3_2F)^4F$	98.4	1.144	1.147	0.003
17 233.68	17 232.11	1.570	98.7 a ^3_2G	0.7 b $(^1_2G)^2G$	99.5	0.889	0.883	−0.006
23 207.76	23 208.23	−0.475	98.4 b $(^1_2G)^2G$	0.7 a ^2_3G	99.1	0.889	0.883	−0.006
	34 933.20		99.1 a ^2_3F	0.1 j $(^1_4G)^2F$	99.2	1.143		
			$J=\frac{9}{2}$					
0.00	−3.22	3.22	98.9 a ^4_3F	0.3 j $(^3_4F)^4F$	99.2	1.333 57	1.332 89	−0.000 68
3 482.82	3 475.97	6.85	98.8 b $(^3_2F)^4F$	0.3 g $(^2_3H,^3D)^4F$	98.9	1.333 98	1.333 43	−0.000 55
16 467.90	16 470.97	−3.07	96.9 a ^2_3G	1.9 a ^2_3H	99.5	1.108	1.109	0.001
22 475.36	22 468.52	6.84	97.1 a ^2_3H	1.7 a ^2_3G	99.5	0.914	0.921	0.007
23 184.23	23 184.81	−0.58	98.0 b $(^1_2G)^2G$	0.6 a ^2_3G	99.1	1.110	1.098	−0.012
			$J=\frac{11}{2}$					
21 780.47	21 786.02	−5.55	99.5 a ^2_3H	0.2 j $(^3_4F)^2H$	99.5	1.091	1.100	0.009

[a] $3d^74s^2$.
[b] $3d^84s$.
[c] $3d^9$.
[d] $3d^75s4s$.
[e] $3d^85s$.
[f] $3d^84d$.
[g] $3d^74d4s$.
[h] $3d^86s$.
[i] $3d^76s4s$.
[j] $3d^64d4s^2$.

the model space $(3d+4s)^{N+2}$. For these reasons, we decided to truncate (and, of course, to renormalize) the eigenvectors to states of the model space. Another method for achieving such truncated eigenvectors is simply to carry out the fs fit with the levels of only the model space by varying the parameters associated with that system. However, the number of known levels is not sufficient to consider all predicted fs interactions. Neglecting one of these interactions and reducing the number of free parameters in the fs fit can cause a systematic error in the calculated eigenvector amplitudes. In order to avoid such a situation we used the fact that the positions of the missing levels $3d^74s^2\,{}^2F_{5/2,7/2}$ and $3d^84s\,{}^2S_{1/2}$, which are practically "pure" model-space levels (see Table I), are very well predicted by fs fits performed for the extended model space. Hence we can take the predicted positions of these levels as "known" into the model-space fs-fit calculations. This cannot be done for the other missing levels $3d^74s^2\,{}^2_1D_{3/2,5/2}$, which are only 84% and 86.5%, model-space levels, respectively. It should be mentioned that the predicted positions of these levels, obtained in the model-space fs fit, are placed 625 and 549 cm^{-1} below the positions calculated in the case of extended model space (see Table I). Some results of the extended-model-space calculation are also given in Table II. In this way, two sets of the eigenvectors for the model space have been obtained. Comparison of these two sets: (a) truncated and renormalized, and (b) from repeated fs fits for the model space $(3d+4s)^9$ with assumed positions of the levels $3d^74s^2\,{}^2F_{5/2,7/2}$ and $3d^84s\,{}^2S_{1/2}$, shows that no essential differences appear. Likewise, the values of hfs parameters evaluated independently using these two eigenvector sets were approximately the same. Thus in the remainder of this work we discuss only the results obtained in model space, as only in this case is it possible to compare electron-excitation effects observed on fine and hyperfine structure simultaneously, because of their suitable definitions. This will be discussed in more detail in the following paragraph.

TABLE II. Values of spin-dependent parameters obtained in fs-fitting procedures (in cm^{-1}).

	fs-fitting procedure		
	Extended model space	Model space	Hartree-Fock calculations [19]
$\xi(3d,3d)$	878(6)	927(8)	
$\xi(3d^74s^2)$	500(4)	504(11)	534.58
$\xi(3d^84s)$	447(3)	448(9)	480.13
$\xi(3d^9)$	386(3)	379(8)	428.98
$M^0(3d^74s^2)$	1.77(8)	1.63(32)	1.93
$M^0(3d^84s)$	1.47	1.37	1.65
$M^0(3d^9)$	1.16	1.09	1.37
$M^2(3d^74s^2)$	0.86(9)	0.96(22)	1.05
$M^2(3d^84s)$	0.70	0.80	0.89
$M^2(3d^9)$	0.54	0.65	0.74
P_1	154(3)	171(6)	
P_2	74(6)	62(7)	
P_3	50	41	
P_4	=0 assumed	=0 assumed	
P_5	−22(2)	−30(8)	
P_7	70(28)	14(58)	
P_8	136(19)	94(37)	

Table II shows that the two sets of the values of parameters ζ and P_1 calculated in model space and in extended model space differ from each other. These discrepancies are due to different definitions in both cases. For example, in the definition of P_i the sum running over n''' starts with $n'''=5$ for the extended model space and with $n'''=4$ in the case of the model space. It is worth pointing out that the values of the single-configuration parameters $\zeta(3d^{N+M}4s^{2-M})$ usually used can be recalculated by means of Eq. (9) from the above two sets of parameters, and the values of $\zeta(3d^{N+M}4s^{2-M})$ are almost the same. We assumed that this agreement is a test of quality of our approach and that it indicates a good accuracy of eigenvectors obtained in this work.

In spite of the fact that the orbit-orbit, spin–other orbit, and spin-spin interactions are very weak, the values of the Marvin integrals M^0 and M^2 fitted by us are close to the theoretical ones [20].

II. EVALUATION OF hfs PARAMETERS

In order to evaluate hfs radial parameters the parametrization method for the hfs analysis of $3d$-shell atoms, presented previously [3], has been used. The parametrization method considers each of the one- and two-body hfs interactions as a product of an angular and a radial part. The angular part can be calculated exactly. If precise wave functions in the intermediate coupling scheme are available, the radial part is determined in a semiempirical manner as described below.

For each contribution the radial part is common for all states of the model space whereas the corresponding angular parts depend on the individual fine structure state [3]. The hfs constants A and B for each fs state can be expressed as a sum of products called "theoretical expressions" and are defined as [3]:

$$A(\psi)=\sum_{\substack{\kappa k,nl\\ \kappa k=01,12,10\\ nl=3d,4s}}\alpha_{nl}^{\kappa k}(\psi)a_{nl}^{\kappa k}+\sum_{i=1}^{11}\alpha_i(\psi)a_i\ , \tag{10}$$

$$B(\psi)=\sum_{\substack{\kappa k,nl\\ \kappa k=02,13,11\\ nl=3d}}\beta_{nl}^{\kappa k}(\psi)b_{nl}^{\kappa k}+\sum_{i}^{8}\beta_i(\psi)b_i\ , \tag{11}$$

where $a_{nl}^{\kappa k}$ and $b_{nl}^{\kappa k}$ are hfs one-body radial parameters and $\alpha_{nl}^{\kappa k}(\psi)$ and $\beta_{nl}^{\kappa k}(\psi)$ are their corresponding angular coefficients in the intermediate coupling (IC) scheme, the "real" fine-structure state ψ are written in a SL base. The terms $\alpha_i(\psi)a_i$ and $\beta_i(\psi)b_i$ represent the contributions of the two-body hfs interactions and an index i marks the radial parameters and angular coefficients corresponding to the following excitations:

Index i	Excitation
1, . . . , 5	open $3d$ shell$\rightarrow$empty $n'''d$ shell$\rightarrow$open $3d$ shell
6	closed $n''s$ shell$\rightarrow$open $3d$ shell$\rightarrow$closed $n''s$ shell
7,8	$4s$ shell$\rightarrow$empty $n'''d$ shell$\rightarrow$open $3d$ shell
9	open $4s$ shell$\rightarrow$empty $n'''s$ shell$\rightarrow$open $4s$ shell
10	closed $n''s$ shell$\rightarrow$open $4s$ shell$\rightarrow$closed $n''s$ shell
11	closed $4s$ shell$\rightarrow$empty $n'''s$ shell$\rightarrow$open $3d$ shell, and two-electron excitation: closed $n''s$ shell$\rightarrow$open $3d$ shell and closed $4s$ shell$\rightarrow$open $n''s$ shell

The angular coefficients $\alpha_{nl}^{\kappa k}(\psi),\alpha_i(\psi)$ and $\beta_{nl}^{\kappa k}(\psi),\beta_i(\psi)$ are calculated with a computer program that is based on the formulas given in Ref. [3] and they are available from the authors. The radial parameters $a_l^{\kappa k}$, a_i, $b_{nl}^{\kappa k}$, and b_i are fitted to the experimentally determined hfs constants A and B using the theoretical expressions (10) and (11). For such linear-equation systems all parameters can be deduced, if there is a sufficient number of linearly independent equations. Moreover, the two-body hfs contributions are N- and SL-dependent and hence only hfs measurements including different SL terms allow one to estimate these contributions. The desirable condition to evaluate all one- and two-body radial parameters is only fulfilled if experimental A and B constants are known for the levels belonging to at least ten different SL terms of all three configurations of the model space $(3d+4s)^{N+2}$. The above requirement can be weakened for an atomic system with strong spin-orbit and interconfiguration mixing of fine-structure states, as in that case the admixtures from other states may be significant. In our hfs analysis we use the very accurate results first obtained by von Ehrenstein, Kopfermann, and Penselin [6] and later extended to further levels by Childs and Goodmann [7] using the atomic-beam magnetic-resonance method. Even higher-lying metastable levels were investigated applying Doppler-free and Doppler-limited laser spectroscopy by Guthöhrlein and co-workers Wenzel [8], Ibrahim-Rûd [10], and Baier [11]. The total number of measured magnetic-dipole hfs constants A and electric-quadrupole hfs constants B are 20 and 17, respectively. Moreover, the accuracies of the hfs constants measured using the method mentioned above are very different (see Table III).

It should also be pointed out that the accuracies of the angular coefficients appearing in theoretical expressions (10) and (11) are strongly dependent on the precision of the eigenvector amplitudes. The states $3d^7 4s^2\,{}^4F_J$ are quite close to pure SL coupling; hence in this case the inaccuracies of relevant eigenvectors obtained from fs fits have negligible influence on the quality of corresponding theoretical expressions [3]. Taking the above into account, we have weighted the theoretical expressions in the hfs fit in the following way: $A(3d^7 4s^2\,{}^4F_J)=A(a^{\kappa k},a_i)$ was weighted by a factor of 1000, $A(3d^8 4s\,{}^4F_{9/2,7/2,5/2})=A(a^{\kappa k},a_i)$ by a factor of 100, and the other expressions by a factor of 1. The same weighing procedure was repeated with corresponding B expressions.

As one can see from Table III, there are 20 and 17 equations for the magnetic-dipole and electric-quadrupole hfs constants, respectively. These constants were measured for levels belonging to eight terms only. Thus, according to criteria mentioned above, this was not enough to avoid all linear dependencies in both A- and B-equation systems. For this reason additional assumptions, which are taken from Hartree-Fock calculations [21] or from fine-structure analysis (see Sec. I), had to be included in our hfs-fitting procedure.

The two-body hfs parameters a_1 and a_4 or b_1 and b_4 describe the same electron excitation: $3d\rightarrow n'''d\rightarrow 3d$, first in the core $3d^N$ involving only $3d$ electrons and second in the mixed configuration $3d^N 4s$ involving $3d$ and $4s$ electrons. The radial integrals arising from these parameters are of rank 0; thus the relevant angular coefficients α_1,α_4 and β_1,β_4 are SL independent. The coefficients mentioned above show dependence only on N and M, where N is the number of $3d$ electrons and $2-M$ defines the number of $4s$ electrons. It causes a linear dependence between the parameters a_{3d}^{01}, a_{3d}^{12}, a_1, and a_4 or between b_{3d}^{02}, b_1, and b_4, so that they cannot be simultaneously evaluated without additional assumptions. Sometimes the assumption can be deduced from Hartree-Fock calculations but, as in the case of ^{59}Co, the relevant data are not available; thus the values of radial parameters a_4 and b_4 have been set equal to 0. As a consequence the parameters $a_{3d}^{01'},a_{3d}^{12'},a_1'$ and $b_{3d}^{02'},b_1'$ obtained from fitting the procedure to the experimental data should be interpreted as

$$a_{3d}^{01'}=a_{3d}^{01}-2(N+1)(2l+1)^{-1/2}a_4\ , \tag{12}$$

$$a_{3d}^{12'}=a_{3d}^{12}-2(N+1)(2l+1)^{-1/2}a_4\ , \tag{13}$$

$$b_{3d}^{02'}=b_{3d}^{02}-2(N+1)(2l+1)^{-1/2}b_4\ , \tag{14}$$

$$a_1'=a_1-(2l+1)^{1/2}a_4\ , \tag{15}$$

$$b_1'=b_1-(2l+1)^{1/2}b_4\ , \tag{16}$$

The above relations can be derived from [3].

The angular coefficients α_9 and α_{10} occurring in terms of expressions (10), in which electron excitations $4s\rightarrow n'''s\rightarrow 4s$ and $n''s\rightarrow 4s\rightarrow n''s$ are described, differ by sign only. Therefore, if the relation estimated theoretically from Hartree-Fock calculations would not have been used, only a mixed parameter $a_{9,10}\equiv a_9-a_{10}$ could be determined from the experimental data.

The results of hfs fits performed with the above assumptions are given in Table III. This table shows also the predicted values of hfs constants A and B for all not-yet-investigated levels of the model space $(3d+4s)^9$. The radial integrals have been obtained from the hfs parameters using the following relations [3,22]:

$$a_{3d}^{\kappa k}=95.4128 g_I \langle r^{-3}\rangle^{\kappa k}, \quad \kappa k=01,12,10 , \tag{17}$$

$$a^{12}(\mathrm{IC})=95.4128 g_1 \langle r^{-3}\rangle_{\mathrm{IC}}^{12} , \tag{18}$$

$$a_{4s}^{10}=95.4128\frac{2}{3}\left[\frac{dP}{dr}\right]_{r=0}^{2} F^{10} , \tag{19}$$

$$a_1=95.4128 g_I t_i, \quad i=1,\ldots,8 , \tag{20}$$

$$a_i=\tfrac{2}{3}95.4128 g_I t_i, \quad i=9,10,11 , \tag{21}$$

$$b^{\kappa k}=234.9624 Q\langle r^{-3}\rangle^{\kappa k}, \quad \kappa k=02,13,11 , \tag{22}$$

$$b^{02}(\mathrm{IC})=234.9624 Q\langle r^{-3}\rangle_{\mathrm{IC}}^{02} , \tag{23}$$

$$b_i=234.9624 Q t_i, \quad i=1,\ldots,8 , \tag{24}$$

where $g_I=1.317$ [23], F^{10} is a relativistic correction factor [22], Q is the nuclear quadrupole moment (here 0.41 b; see Sec. IV), all parameters are in MHz, and all radial integrals ($\langle r^{-3}\rangle$ and t_i) are in atomic units.

III. COMPARISON OF THE EXPERIMENTAL RESULTS WITH THEORETICAL CALCULATIONS

Usually the hfs parameters are discussed for each configuration separately. Using the relations given in [3] the corresponding parameters for each configuration (hereafter called configuration parameters) can be obtained from model-space parameters (Table IV) as follows:

TABLE III. Comparison of the experimental hfs constants $A_{\mathrm{expt}}, B_{\mathrm{expt}}$ and the values $A_{\mathrm{calc}}, B_{\mathrm{calc}}$ recalculated from the model-space parameters. The constants are given in MHz. Numbers in brackets denote reference numbers.

State		A_{expt}	A_{calc}	$\frac{\Delta A}{A_{\mathrm{expt}}}\times 100\%$	B_{expt}	B_{calc}	$\frac{\Delta B}{B_{\mathrm{expt}}}\times 100\%$
$3d^74s^2$	$^4_3F_{3/2}$	1042.981(1) [7]	1042.980	0.0001	67.618(20) [7]	67.629	−0.017
$3d^74s^2$	$^4_3F_{5/2}$	613.349(3) [7]	613.356	−0.0012	67.541(50) [7]	67.49	0.08
$3d^74s^2$	$^4_3F_{7/2}$	490.567(2) [7]	490.556	0.0022	94.501(36) [7]	94.54	−0.04
$3d^74s^2$	$^4_3F_{9/2}$	450.283(1) [7]	450.288	−0.0011	139.230(30) [7]	139.224	0.004
$3d^8(^3_2F)4s$	$^4F_{3/2}$	303(2) [9]	304	0.4	−82(40) [9]	−56	32
$3d^8(^3_2F)4s$	$^4F_{5/2}$	562.183(3) [7]	562.167	0.0029	−54.806(250) [7]	−54.64	0.31
$3d^8(^3_2F)4s$	$^4F_{7/2}$	668.919(3) [7]	668.936	−0.0026	−79.221(200) [7]	−78.836	0.5
$3d^8(^3_2F)4s$	$^4F_{9/2}$	828.799(4) [7]	828.806	−0.0009	−118.751(300) [7]	−119.10	−0.3
$3d^74s^2$	$^4_3P_{1/2}$	−710.8(9) [8]	720.0	1.3			
$3d^74s^2$	$^4_3P_{3/2}$		323			146	
$3d^74s^2$	$^4_3P_{5/2}$	178.9(3) [1]	167.1	6.6	−170(14) [1]	−187	−10
$3d^8(^3_2F)4s$	$^2F_{5/2}$	1108.8(1.3) [10]	1117.7	−0.8	−93(20) [10]	−96	−3.2
$3d^8(^3_2F)4s$	$^2F_{7/2}$	391.5(1.4) [10]	393.8	−0.6	−106(27) [10]	−114	−7
$3d^8(^3_2P)4s$	$^4P_{1/2}$	1727.1(1.0) [11]	1707	1.2			
$3d^8(^3_2P)4s$	$^4P_{3/2}$	457.9(3) [1]	430.3	6.0	−90(4) [1]	−80	11
$3d^8(^3_2P)4s$	$^4P_{5/2}$	1124.8(7) [1]	1110.2	1.3	144(9) [1]	159	−11
$3d^8(^1_2D)4s$	$^2D_{3/2}$		341			18	
$3d^8(^1_2D)4s$	$^2D_{5/2}$	1387.5(1.0) [10]	1364.7	1.7	127(15) [10]	127	0
$3d^8(^3_2P)4s$	$^2P_{1/2}$	592.7(1.9) [10]	586.2	1.1			
$3d^8(^3_2P)4s$	$^2P_{3/2}$	332.0(1.5) [1]	331.5	0.2	101(10) [1]	75	26
$3d^74s^2$	$^2_3G_{7/2}$	839.4(4) [1]	831.5	0.9	−97(3) [1]	−85	13
$3d^74s^2$	$^2_3G_{9/2}$	611.9(1.3) [10]	595.8	2.6	−84(15) [10]	−89.0	−6
$3d^74s^2$	$^2_3P_{1/2}$		444				
$3d^74s^2$	$^2_3P_{3/2}$		162			77	
$3d^74s^2$	$^2_3H_{9/2}$		771			−404	
$3d^74s^2$	$^2_3H_{11/2}$		670			−431	
$3d^74s^2$	$^2_3D_{3/2}$		913			169	
$3d^74s^2$	$^2_3D_{5/2}$		552			148	
$3d^8(^1_2G)4s$	$^2G_{7/2}$		238			−468	
$3d^8(^1_2G)4s$	$^2G_{9/2}$		1077			−509	
$3d^9$	$^2_1D_{3/2}$		1020			−147	
$3d^9$	$^2_1D_{5/2}$		315			−204	
$3d^74s^2$	$^2_3F_{5/2}$		721			−101	
$3d^74s^2$	$^2_3F_{7/2}$		666			−142	
$3d^8(^1_0S)4s$	$^2S_{1/2}$		4353				
$3d^74s^2$	$^2_1D_{3/2}$		1117			−242	
$3d^74s^2$	$^2_1D_{5/2}$		497			−365	

TABLE IV. The hfs parameters of the model space and the value of their corresponding radial integrals. The uncertainties given in parentheses are the standard deviations.

	Parameter (MHz)	Radial integral (a.u.)		Parameter (MHz)	Radial integral (a.u.)
a_{3d}^{01}	1152(57)	9.2(5)	b_{3d}^{02}	870.3(4.7)	9.03(5)
a_{3d}^{12}	1180(56)	9.4(5)	b_{3d}^{13}	22.1(1.7)	0.23(2)
a_{3d}^{10}	−86.0(4)	−0.684(4)	b_{3d}^{11}	−6.8(3)	−0.07(3)
a_{4s}^{10}	3077(30)	36.7(4)			
a_{IC}^{12}	188(209)	2(2)	b_{IC}^{02}	31(33)	0.32(34)
a_1'	195(22)	1.55(18)	b_1	151.1(1.8)	1.57(2)
a_2	69(21)	0.55(18)	b_2	72.5	0.75
a_3	46(14)	0.37(11)	b_3	48.3	0.50
a_5	−20(20)	−0.16(16)	b_5	−21.1	−0.22
a_6	−28(9)	−0.223(72)	b_6	−42.3	−0.44
a_7	90(50)	0.72(40)	b_7	68.0	0.71
a_8	274(151)	2.2(1.2)	b_8	128.4	1.33
a_9'	799(25)	6.4(2)			
a_{11}	−2664(1030)	−21.2(8.2)			

$$a^{\kappa k}(d^{N+M}s^{2-M}) = a^{\kappa k} + [2(1-N-M)/5]a_1 - [2(2-M)/\sqrt{5}]a_4 + \tfrac{2}{5}\delta(M,0)a_5 ,$$

$$\kappa k = 01, 12 \text{ (and in corresponding form for } b^{\kappa k},\ \kappa k = 02,\ M = 0,1,2,\ N = 7 , \tag{25}$$

$$a_{3d}^{10}(3d^{N}4s^{2}) = a_{3d}^{10},\quad a_{3d}^{10}(3d^{N+1}4s) = a_{3d}^{10} - \tfrac{2}{5}a_9 , \tag{26}$$

$$a_{3d}^{10}(3d^{N+2}) = a_{3d}^{10} + \tfrac{2}{5}(a_{10} - a_9),\quad a_{4s}^{10}(3d^{N+1}s) = a_{4s}^{10} . \tag{27}$$

The configuration radial integrals are given in Table V together with the results of relativistic Hartree-Fock (RHF) and relativistic optimized-Hartree-Fock-Slater (ROHFS) calculations by Lindgren and Rosèn [22] and Olsson and Rosèn [24]. Table V also shows experimental results given by other authors [1,7] who neglected two-body hfs effects. Generally, we can say that careful fs calculations with precise eigenvectors and taking into account two-body hfs interaction improved the agreement between experimental results and theoretical calculations. As one can see from this table, the ratio $\langle r^{-3}\rangle^{01}/\langle r^{-3}\rangle^{12}$ is quite close to the theoretical prediction. The strong disagreement between theoretical and experimental values, $\langle r^{-3}\rangle_{4s}^{M}$, observed earlier has been removed by taking into account the s-electron excitations: $4s \rightarrow n'''s \rightarrow 4s$ and $n''s \rightarrow 4s \rightarrow n''s$ (parameters a_9, a_{10}) [3].

A similar situation has already been observed in the neighboring iron atom [14]. The value of the "core-polarization" parameter a_{3d}^{10} for the component configuration $3d^{8}4s$ cannot be determined within the framework of its exact definition. The reason is the linear dependence of one- and two-body operators acting on s electrons [25]. Therefore only the following parameters can be determined from hfs-splitting measurements: the core polarization parameter a_{3d}^{10} for the component configurations with closed ns shells, the parameter a_{4s}^{10}, and the mixed parameter $a_{9,10} = a_9 - a_{10}$. The use of the hfs parametrization method [3] allows the extraction of configuration interaction effects and therefore the values of one-body hfs parameters obtained in this way are suited for studying relativistic effects. The values of radial integrals ($\langle r^{-3}\rangle^{01}$, $\langle r^{-3}\rangle^{12}$, $\langle r^{-3}\rangle^{11}$, and $\langle r^{-3}\rangle^{13}$) and especially their ratios (see Table V) confirm theoretical prediction [22,24].

In order to analyze configuration-interaction effects the following ratios of corresponding two- and one-body fs and hfs parameters have been taken:

$$\frac{a_1'}{a_{3d}^{01'}} = 0.17(2),\quad \frac{b_1'}{b_{3s}^{02'}} = 0.174(3) ,$$
$$\frac{P_1}{\zeta(3d,3d)} = 0.18(1) , \tag{28}$$

$$\frac{a_2}{a_{3d}^{01'}} = 0.06(3),\quad \frac{b_2}{b_{3d}^{02'}} = 0.084(15) ,$$
$$\frac{P_2}{\zeta(3d,3d)} = 0.067(7) , \tag{29}$$

$$\frac{a_5}{a_{3d}^{01'}} = -0.02(2),\quad \frac{b_5}{b_{3d}^{02'}} = -0.025(5) ,$$
$$\frac{P_5}{\zeta(3d,3d)} = -0.03(1) . \tag{30}$$

According to the definitions of these parameters all ratios given in (28) should be equal. This is also valid for the

proportions (29) and (30). The above requirements are fulfilled within experimental accuracy. Hence we can conclude that electron excitations $3d \rightarrow n'''d \rightarrow 3d$ and $4s \rightarrow n'''d \rightarrow 3d$ affect spin-orbit splitting and magnetic-dipole and electric quadrupole hyperfine structure in the same way. Moreover, consistency of ratios (28) and agreement of experimental results with theoretical calculations permit one to determine the nuclear quadrupole moment of ^{59}Co with high accuracy. It should be also mentioned that the SL-dependent as well as the N-dependent contributions to the Sternheimer shielding or antishielding factors R [26] can be expressed as a function of the ratios of the b_i/b_{nl}^{02} parameters.

IV. DETERMINATION OF THE NUCLEAR QUADRUPOLE MOMENT OF ^{59}Co

As the one-body parameters $a_{nl}^{\kappa k}, b_{nl}^{\kappa k}$ are now free of the above-mentioned N- and SL-dependent contributions, separated by the use of an independent set of two-body parameters, the determination of the nuclear quadrupole moment from an extended set of one- and two-body hfs

TABLE V. Experimental hfs radial integrals and corresponding theoretical values for the configuration of the model space $(3d+4s)^9$. All values are given in atomic units.

		Calc. values [24]				This
		RHF	ROHFS	Expt.		work
$\langle r^{-3}\rangle^{01}$	$3d^74s^2$	5.852[a]		5.51[b]	5.59(7)[c]	5.379
	$3d^84s$	5.301	5.513	5.1[b]	4.92(9)[c]	4.822
	$3d^9$	4.765	4.984			4.201
$\langle r^{-3}\rangle^{12}$	$3d^74s^2$	5.994[a]		5.96[b]	5.77(72)[c]	5.602
	$3d^84s$	5.457	5.678	5.1[b]	6.82(43)[c]	5.045
	$3d^9$	4.962	5.183			4.424
$\langle r^{-3}\rangle^{01}/\langle r^{-3}\rangle^{12}$	$3d^74s^2$	0.976		0.924[b]	0.969[c]	0.960
	$3d^84s$	0.971	0.971	1.000[b]	0.721[c]	0.956
	$3d^9$	0.960	0.962			0.950
$\langle r^{-3}\rangle^{10}_{3d}$	$3d^74s^2$	−0.0655[a,d]		−0.684[b]	−0.55(11)[c]	−0.651
	$3d^84s$	−0.0730[d]	−0.0769	−1.39(14)[b]	−1.67(27)[c]	
	$3d^9$	−0.0940[d]	−0.0942			−3.231
χ_{contact}	$3d^74s^2$	−0.67[a]		−0.93[b]		−0.88
	$3d^84s$					
	$3d^9$					−4.70
$\langle r^{-3}\rangle^{M}_{4s}$	$3d^74s^2$					
	$3d^84s$	35.379	48.958	50(1)[e]	53(1)[c]	36.7(4)
	$3d^9$					
$\langle r^{-3}\rangle^{02}$	$3d^74s^2$	5.859[a]				5.182
	$3d^84s$	5.305	5.518			4.642
	$3d^9$	4.765	4.986			4.015
$\langle r^{-4}\rangle^{13}$	$3d^74s^2$	0.274[a]				0.23
	$3d^84s$	0.281	0.299			0.23
	$3d^9$	0.322	0.331			0.23
$\langle r^{-3}\rangle^{11}$	$3d^74s^2$	−0.109[a]				−0.07
	$3d^84s$	−0.120	−0.127			−0.07
	$3d^9$	−0.150	−0.152			−0.07
$\langle r^{-3}\rangle^{11}/\langle r^{-3}\rangle^{13}$	$3d^74s^2$	−0.398		−0.292[b]	−0.41[c]	−0.31(6)
	$3d^84s$	−0.427	−0.425	+0.762[b]	−1.5[c]	−0.31(6)
	$3d^9$	−0.466	−0.459			−0.31(6)

[a]From Lindgren and Rosén [22].
[b]Calculated by Lindgren and Rosén [22] from the experimental data of Childs and Goodmann [7].
[c]Calculated from experimental data [1].
[d]Relativistic part only.
[e]From Olsson and Rosén [24].

parameters [3] obtained from experiment is possible:

$$Q_1=\frac{2\mu_I g_i}{e^2}\frac{b_{nl}^{\kappa k}}{a_{nl}^{\kappa k}}\frac{(1+\Delta_{ll'}^{(\kappa k)1})F^{\kappa k}}{(1+\Delta_{ll'}^{(\kappa k)2})R^{\kappa k}}\ , \tag{31}$$

$F^{\kappa k}$ and $R^{\kappa k}$ are relativistic correction factors [22], $\Delta_{ll'}^{\kappa k}$ are the contributions of one-body excitations of closed shells to empty shells [26], and

$$Q_2=\frac{2\mu_I g_I}{e^2}\frac{b_i}{a_i},\quad i=1,\ldots,8\ , \tag{32}$$

where Q_2 is the nuclear quadrupole moment including Sternheimer corrections up to second order. Using the most accurate parameters obtained in this work we can evaluate from a_{3d}^{01} and b_{3d}^{02} [Eq. (31)]

$$Q_1=0.404(15)\ \text{b}$$

and alternatively from a_1 and b_1 [Eq. (32)]

$$Q_2=0.414(15)\ \text{b}\ .$$

The very small discrepancy between Q_1 and Q_2, below the experimental limits of error, indicates that one-body screening effects represented by Δ_{ll} in (31) play no important role in cobalt, as has also been observed for titanium [5]. Thus we can take the mean $Q=0.41(1)$ b as the value of the nuclear quadrupole moment of ^{59}Co, including Sternheimer corrections up to second order. The values of Q evaluated in this work are greater than the values obtained previously [1,7]. Q was determined for the first time by Ehrenstein, Kopfermann, and Penselin [6] on the basis of very simple evaluations: $Q(^{59}\text{Co},3d^7 4s^2;{}^4F_{9/2})=0.404(40)$ b. Childs and Goodman [7] using very precise measurements, gave two different values for the terms $3d^7 4s\ {}^{24}F$ and $3d^8 4s\ {}^4F$:

$$Q(^{59}\text{Co},3d^7 4s^2;{}^4F)=0.380\ \text{b}\ ,$$

$$Q(^{59}\text{Co},3d^8 4s;{}^4F)=0.345\ \text{b}\ .$$

The average values for particular configurations obtained recently by Guthöhrlein and Keller [1] are as follows:

$$Q(^{59}\text{Co},3d^7 4s^2)=0.343(13)\ \text{b}\ ,$$

$$Q(^{59}\text{Co},3d^8 4s)=0.351(16)\ \text{b}\ .$$

Differences between the values mentioned above can be explained by the fact that two-body effects on hfs depend on the SL term and on the type of configuration. Hence the Q values determined previously from hfs measurements for one term only or those averaged for the particular configuration, and neglecting two-body hfs effects, were not quite correct. Therefore the values mentioned above cannot be directly compared with these evaluated in this work, where screening effects have been taken into account.

The parametrization method used in this work allows one to estimate quantitatively almost all contributions to the hfs constants A and B [see (10) and (11)]. Using the radial parameters given in Table IV and calculated angular coefficients $\alpha_{nl}^{\kappa k}(\psi),\alpha_i(\psi)$ and $\beta_{nl}^{\kappa k}(\psi),\beta_i(\psi)$ we can estimate one-body contributions and also in detail two-body contributions representing particular kinds of electron excitations. In this way it is possible to interpret the hfs splitting of the level $3d^8 4s\ {}^2P_{3/2}$ which was discussed in [1]. Table I shows strong interconfiguration mixing for this level: the leading percentage amounts to 50.7% for the state $3d^8 4s\ {}^2P_{3/2}$, with an admixture of 36.8% from the state $3d^7 4s^2\ {}^2P_{3/2}$. In such cases off-diagonal effects (excitation: closed $4s$ shell$\rightarrow$empty $n'''s$ shell$\rightarrow$open $3d$ shell) are very significant and their contribution to this hfs A constant amounts to $\alpha_{11}a_{11}=95$ MHz. It is likely that the contribution of such an effect to the other $^2P_{3/2}$ state should be also important, and we have predicted its value to be -100 MHz. A similar strong two-body core-polarization effect was observed for the first time in titanium and vanadium by Johann, Dembczyński, and Ertmer [13].

Recently, Beck [27] published results of a new approach to the theoretical calculations of hfs obtained for the elements with open-subshell d configurations, which removed large discrepancies between theory and experiment [28,29]. Thus a comparison between results of this new theoretical approach and our predictions would be very interesting. We hope that predicted values of the A and B constants given in Table III can be a stimulus for further experimental and theoretical hfs investigations in the cobalt atomic spectrum.

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