The nature of the strongly forbidden $M1$ transitions and the g-factor anomalies in heavy atoms

O. P. Sushkov, V. V. Flambaum, and I. B. Khriplovich

Institute of Nuclear Physics, Siberian Division of the USSR Academy of Sciences

A new mechanism is proposed for the appearance of strongly forbidden $M1$-transition amplitudes and corrections to the g factor in a heavy atom with a single outer electron. Expressions are obtained for these quantities in the case of an outer s or p electron. Numerical computations are carried out for cesium and thallium.

1. INTRODUCTION

The question of the causes of the deviation of the g factor of an atom with one outer electron from the g factor of the free electron was actively discussed many years ago at the time of the first measurements of the anomalous magneitc moment of the electron.[1-3] The strongly forbidden $M1$ transitions in the heavy atoms have of late attracted attention in connection with the ongoing search for parity nonconservation in these atoms as a result of the weak interaction of the electron with the nucleus.[4-10]

The g-factor anomalies of the alkali atoms have been measured in a number of experiments.[11-13] If in the light atoms the corrections to the g factor is almost constant, in the heavy atoms it changes its sign and increases rapidly with Z. Hence we can conclude that the corrections to the g factor in the heavy and light atoms are due to different mechanisms. It is well known that, to explain this anomaly in the light atoms, it is sufficient to take only the relativistic effects into account.[14,15] As regards the heavy atoms, however, it was proposed long ago that here a large contribution is made by the mixing of the configurations jointly with the spin-orbit interaction.[16] It was natural to assume that the latter effect is important for the strongly forbidden $M1$ transitions in the heavy atoms.[5,6] Specifically, in Refs. 5, 6, and 16, the topic of discussion is the corrections arising in fourth-order perturbation theory: second order in the Coulomb mixing of the configurations and second order in the spin-orbit interaction. The corresponding quantitative computations were, however, not carried out. Therefore, the decisive argument in favor of this mechanism was the fact that all the remaining attempts to explain the observed effects, say in cesium, turned out to be untenable. Yet when we carried out the corresponding computation for cesium, we surprisingly found its result to be roughly 100 times smaller than the experimental result. This discrepancy for cesium is also noted in Neuffer and Commins's paper,[17] with which we became acquainted only after the completion of the present investigation.

In the present paper we propose a new mechanism for the phenomena under discussion. They can arise not in fourth-order, but in third-order, perturbation theory: first-order in the Coulomb mixing of the configurations and second-order in the spin-orbit interaction. We compute the g factors of the $6s_{1/2}$ state of cesium and the $6p_{1/2}$ and $6p_{3/2}$ states of thallium, as well as the amplitudes of the $M1$ transitions $6s_{1/2} \rightarrow 7s_{1/2}$ in cesium and $6p_{1/2} \rightarrow 7p_{1/2}, 8p_{1/2}, 9p_{1/2}$ in thallium, taking into account, along with the configuration mixing, the normal relativistic corrections. We do not consider the corrections due to the hyperfine interaction, since their computation is trivial and, besides, they can be experimentally separated out.

2. THE g FACTOR OF CESIUM

To elucidate the cause of the appearance of the g-factor anomaly of cesium, it is convenient to represent the correction due to configuration mixing in the following form:

$$\frac{1}{2}\delta g = \langle J, J_z = \frac{1}{2} | L_z + 2S_z - 2J_z | J, J_z = \frac{1}{2} \rangle = -\langle J, J_z = \frac{1}{2} | L_z | J, J_z = \frac{1}{2} \rangle. \quad (1)$$

In the LS-coupling approximation there is in the cesium atom an outer $6s$ electron above the filled shells, i.e., the ground state is $^2S_{1/2}$. In order for the matrix element $\langle L_z \rangle$ to be different from zero, it is necessary for the admixture into the $^2S_{1/2}$ state of states with $L \neq 0$ to be taken into account simultaneously in the bra $\langle J, J_z = \frac{1}{2} |$ and in the ket $| J, J_z = \frac{1}{2} \rangle$. It is clear that for this purpose it is necessary, firstly, to open the closed shells and, secondly, to take into account the spin-orbit interaction, which changes L. In Refs. 16, 5, and 6 it is assumed that the opening of the shells occurs only as a result of the residual Coulomb interaction. It is from this assumption that the fourth-order perturbation theory arose.

The main point of the present paper is the observation that to the opening of the shells leads the off-diagonal spin-orbit interaction alone, i.e., after the switching on of the spin-orbit interaction, the wave function of a closed shell is no longer an eigenfunction of the operator $\hat{L}$ with $L_z = 0$.[1)] But, of course, the average value $\langle 5p^6 | \hat{L} | 5p^6 \rangle = 0$. In order for the matrix element of L_z in the formula (1) to be nonzero, it is necessary that the angular momentum of the excitations of the closed shells be correlated with the spin of the outer electron into account once. Thus, the effect arises even in third-order perturbation theory: second-order in the spin-orbit interaction and first-order in the Coulomb interaction.

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Thus far we have carried out the discussion in terms of the LS scheme, treating the spin-orbit interaction as a perturbation. However, the computation looks significantly simpler in the jj scheme with the use of relativistic radial wave functions, where the effect formally arises in first-order perturbation theory. In this case in the matrix element (1) it is sufficient to take into account in the ket or bra the admixture by the exchange Coulomb interaction of the states with an excited inner-shell electron. The corresponding correction to the g factor is equal to

$$\delta g_2 = -4 \sum_{nn'ljj'_z} \left\langle 6s, nljj_z \left| \frac{e^2}{r} \right| n'lj'j_z, 6s \right\rangle \frac{\langle n'lj'j_z | L_z + 2S_z | nljj_z \rangle}{E_{nlj} - E_{n'lj'}}, \quad (2)$$

where n, l, j, and j_z respectively denote the principle quantum number, the orbital quantum number, the total angular momentum of the electron. The states $|nljj_z\rangle$ lie beneath the $6s$ state, while the states $|n'lj'j_z\rangle$ lie above it (naturally, the continuous spectrum should be included here).

The matrix element $\langle n'lj'j_z | L_z + 2S_z | nljj_z \rangle$ for $n \neq n'$ is equal to zero if $j = j'$, but is nonzero in first order in the spin-orbit interaction for $j' = \bar{j} \equiv 2l - j$[18] (the fact that it is not too much suppressed in heavy atoms was earlier discovered by means of a numerical computation by Neuffer and Commins[19]):

$$\langle n'l\bar{j}j_z | L_z + 2S_z | nljj_z \rangle = -\frac{[(l+\frac{1}{2})^2 - j_z^2]^{\frac{1}{2}}}{2l+1} \alpha(n'l\bar{j}; nlj), \quad (3)$$

where $\alpha(n'l\bar{j}; nlj)$ is the overlap integral of the corresponding radial wave functions. In first order in the spin-orbit interaction $\alpha(n'l\bar{j}; nlj) \approx -\alpha(n'lj; nl\bar{j})$. From this it is very easy to verify that (2) is, in fact, a quantity that is of second order in the spin-orbit interaction. The thus found correction to the g factor of the ground state of cesium is equal to

$$\delta g_3 = -\frac{8}{3} \sum_{nn'lj} \frac{l(l+1)}{(2l+1)^2} \frac{G^l(6s, nlj; 6s, n'l\bar{j})}{E_{nlj} - E_{n'l\bar{j}}} \alpha(nlj; n'l\bar{j}). \quad (4)$$

Here G^l is the exchange Coulomb integral. It should be noted that in the formula (4) the dependence of the G^l on j and $\bar{j}$ (i.e., on the spin-orbit interaction) is not of less importance than the dependence on their energy denominator. A numerical computation in the effective-potential approximation (see the Appendix) on the basis of the formula (4) yields $\delta g = 3.66 \times 10^{-4}$.

Let us consider the other mechanisms that contribute to the correction to the g factor. These are, first, the relativistic corrections to the magnetic-moment operator of the outer electron; second, the magnetic interaction of the outer electron with the inner-shell electrons; and, finally, the above-mentioned contribution, connected with configuration mixing, of fourth-order perturbation theory. The relativistic wave function of the outer electron has the form

$$u = \frac{1}{r} \begin{pmatrix} f\Omega_{jlj_z} \\ ig\Omega_{jl'j_z} \end{pmatrix}. \quad (5)$$

With the aid of the Dirac equation, the exact relativistic expression for the magnetic-moment operator of the outer electron can be reduced to a form in which the small corrections ($\sim\alpha^2$) are explicitly separated

out (we set $\hbar = c = 1$):

$$M_{rel} = -|\mu_B|(1+2s)\left\{1 - \int \left[g^2(r) + \frac{\varepsilon - V(r)}{\varkappa - \frac{1}{2}} rf(r)g(r)\right]dr\right\}. \quad (6)$$

Here ε is the energy, $V(r)$ is the effective potential, $\varkappa = (-1)^{j+1/2-l}(j+\frac{1}{2})$. A numerical computation for cesium ($\varkappa = -1$) on the basis of this formula yields $\delta g_{rel} = -0.38 \times 10^{-4}$.

A correction to the g factor, due to the magnetic interaction of the outer electron with the inner-shell electrons, arises in the Coulomb gauge ($\mathrm{div} A = 0$) from the diamagnetic term $e^2 A^2/2m^2c^2$ in the Hamiltonian. In the present case $A = A_e + A_H$, where A_H, A_e are the vector potentials of the external field and the outer electron. The corresponding correction to the magnetic moment can be reduced to the form

$$M_{dia} = -\frac{1}{3}|\mu_B|\alpha^2\left\langle (1+2s)\left[\frac{2(\varkappa+1)}{2\varkappa-1}W_1+W_2\right]\right\rangle,$$
$$W_1 = \frac{a_B}{r^3}\int_0^r n(r_1)r_1^2 4\pi r_1^2\,dr_1, \quad W_2 = a_B\int_r^\infty \frac{n(r_1)}{r_1}4\pi r_1^2\,dr_1. \quad (7)$$

Here $n(r_1)$ is the core-electron density and a_B is the Bohr radius. The matrix element in (7) is evaluated, using the wave functions of the outer electron. In cesium ($\varkappa = -1$) the numerical computation based on this formula yields $\delta g_{dia} = -0.12 \times 10^{-4}$. The same result can be obtained on the basis of the Breit Hamiltonian for the interaction of two electrons[20] after making the substitution $p \to p - eA/c$ in it.

Besides the direct magnetic interaction, the exchange magnetic interreaction, as well as the exchange spin-orbit interaction, contributes to the correction to the g factor. The computation of their contribution for cesium yields ($\mathrm{Ry} = me^4/2\hbar^2$):

$$\delta g_{exch} = \frac{\alpha^2}{2\,\mathrm{Ry}}\sum_{nl} G^l(6s, nl; 6s, nl) \lesssim 1 \cdot 10^{-6}. \quad (8)$$

Here the summation is performed over the closed subshells. Thus, as was to be expected, the exchange magnetic interaction is significantly weaker than the direct interaction (7).

The contribution to the g factor of the relativistic corrections and the direct magnetic interaction has been computed before.[5,6,17] Our numerical results agree with the results given in these papers.

The estimation of the above-mentioned contribution of fourth-order perturbation theory in the case when the closed shells are opened by the Coulomb interaction with the outer electron shows that $\delta g_4 \lesssim 10^{-6}$. Let us note that there arises in the formal computation of the fourth-order multipole order (it is precisely such a correction, with the difference between the Coulomb integrals for the $p_{1/2}$- and $p_{3/2}$-electrons neglected, that has been considered before[16,5,6 2]). It is clear, however, that this term need not be taken into account, since the F^0 integrals they are matrix elements of the centrally symmetric self-consistent field, which has already been taken into account in the wave functions in the solution of the Dirac equation. It is not difficult to verify by means of a direct calculation that the indicated correction is already contained in the formula

(4).

Adding all the calculated contributions, we obtain

$$\delta g = \delta g_3 + \delta g_{rel} + \delta g_{dia} = 3.2 \cdot 10^{-4}. \tag{8'}$$

Allowing for the fact that our computation of the Cou-lomb integrals in one and the same—for all the elec-trons—effective potential constitutes a very rough approximation, the agreement with the experimental value of $\delta g = 2.18 \times 10^{-4}$ (Ref. 11) is quite satisfactory.

3. THE AMPLITUDE OF THE STRONGLY FORBIDDEN $6s_{1/2} \to 7s_{1/2}$

The amplitude of the $M1$ transition $6s_{1/2} \to 7s_{1/2}$,

$$M_z = \langle J, J_z = \tfrac{1}{2} | \hat{M}_z | J, J_z = \tfrac{1}{2} \rangle \,,$$

can be computed in exactly the same way as the cor-rection to the g factor.[3] The contribution due to the mixing of the configurations is equal to (cf. (4)):

$$M_3 = \frac{2}{3} | \mu_B | \sum_{nn'lj} \frac{l(l+1)}{(2l+1)^2} \frac{G^l(6s, nlj; 7s, n'lj) + G^l(7s, nlj; 6s, n'lj)}{E_{nlj} - E_{n'lj}}$$
$$\times \alpha(nlj; n'lj) = -0.76 \cdot 10^{-4} | \mu_B | . \, . \tag{9}$$

The relativistic correction to the amplitude of the $1(nlj) \to 2(n'lj)$ $M1$ transition of the outer electron has the form (cf. (6)):

$$M_{rel} = | \mu_B | (1+2s) \left\{ \int_0^\infty \left[g_1 g_2 + \frac{1}{2\varkappa-1}(\varepsilon_2 - V(r))f_1 g_2 r + (\varepsilon_1 - V(r))f_2 g_1 r \right] dr \right.$$
$$\left. + \frac{m}{5(2\varkappa-1)} \int_0^\infty (f_1 g_2 + f_2 g_1) k^2 r^3 \, dr \right\}, \tag{10}$$

where $k = \varepsilon_2 - \varepsilon_1$ ($\hbar = c = 1$). The second term in the curly brackets arose from the expansion of the expo-nential function in the photon wave function. For the $6s_{1/2} - 7s_{1/2}$ transition in cesium ($\varkappa = -1$), $M_{rel} = 0.010 \times 10^{-4} | \mu_B |$. The contribution of the magnetic correc-tions is computed with the aid of the formula (7), and is equal to $M_{dia} = 0.03 \times 10^{-4} | \mu_B |$.

The M_{rel} and M_{dia} estimates are in accord with the results of Refs. 5, 6, 17, 21, and 22. All the remain-ing contributions to the $M1$ amplitude, as well as to the g-factor anomaly, are small. The total amplitude is equal to

$$\langle 6s, j_z = \tfrac{1}{2} | \hat{M}_z | 7s, j_z = \tfrac{1}{2} \rangle = M_3 + M_{rel} + M_{dia} = -0.63 \cdot 10^{-4} | \mu_B |; \tag{11}$$

the experimental value (see Ref. 7) of $M_z = -(0.424 \pm 0.034) \times 10^{-4} | \mu_B |$.

Let us note that our calculation reproduces the ratio $M_z / \delta g$ virtually exactly.

4. THE g FACTORS OF THE $6p_{1/2}$ and $6p_{3/2}$ STATES AND THE AMPLITUDES OF THE STRONGLY FORBIDDEN $M1$ TRANSITIONS $6p_{1/2} \to 7p_{1/2}$, $8p_{1/2}$, $9p_{1/2}$ IN THALLIUM

In thallium, for the corrections to the g factor of the ground state and for the matrix elements of the $M1$ transitions $6p_{1/2} \to np_{1/2}$, the third-order mechanism, which was dominant in cesium, does not "work," since, as is easy to verify, the Coulomb interaction does not lead to the transition $p_{1/2}, lj \to p_{1/2}, l\bar{j}$.

The contribution to δg of the relativistic corrections to the magnetic-moment operator of the $6p_{1/2}$ electron

is computed from the formula (6) with $\varkappa = 1$ (the effec-tive potential used by us is described in the Appendix): $\delta g_{rel} = -1.07 \times 10^{-4}$. This number coincides with the number obtained in Ref. 19. The computation from the formula (7) of the magnetic interaction of the outer electron with the inner-shell electrons yields $\delta g_{dia} = -1.23 \times 10^{-4}$. Notice that the formula (7) does not agree with the analogous expression obtained in Ref. 19. However, numerically, this discrepancy does not turn out to be very important. The calculation of the contribution of the exchange magnetic and spin-orbit interactions in thallium turns out to be significantly more tedious than the calculation in cesium. Its numerical value is equal to $\delta g_{exch} = 0.19 \times 10^{-4}$. As for the correction of fourth-order perturbation theory, in this case it turns out to be less than 2×10^{-6}. The total correction to the g factor of the $6p_{1/2}$ state is

$$\delta g = \delta g_{rel} + \delta g_{dia} + \delta g_{exch} = -2.1 \cdot 10^{-4}. \tag{12}$$

The quantity δg computed by us is in good agreement with the experimental value $\delta g = -2.012(18) \times 10^{-4}$ (Ref. 23), and numerically close to the result of the calcula-tion in Ref. 19.

The amplitude of the $M1$ transition $6p_{1/2} \to 7p_{1/2}$ is also due largely to the relativistic and diamagnetic corrections, the contribution of which is computed from the formulas (6) and (7): $M_{rel} = 0.172 \times 10^{-4} | \mu_B |$; $M_{dia} = 0.170 \times 10^{-4} | \mu_B |$; $M_{exch} = -0.18 \times 10^{-5} | \mu_B |$. How-ever, here an appreciable role is played by the con-figuration-mixing effects arising in fourth-order per-turbation theory. In this case the contribution of the $6s6p_{1/2} \to nsmp$ excitations apparently predominates. With allowance for the above-indicated uncertainty in the magnitude of the Coulomb integrals, we obtained for it the estimate $M_4 = -(0.1 - 0.5) \times 10^{-5} | \mu_B |$. Thus, the calculated value of the transition amplitude is close to

$$M_z = M_{rel} + M_{dia} + M_{exch} + M_4 = 2.9 \cdot 10^{-5} | \mu_B |. \tag{13}$$

This value of M_z differs in sign from both the experi-mental value, $M_z = (-2.11 \pm 0.30) \times 10^{-5} | \mu_B |$,[8] and the earlier computed value, $M_z = (-3.2 \pm 1) \times 10^{-5} | \mu_B |$.[19] It is possible that the difference in sign is connected with the inconsistency of the definitions of the magnetic moment (for our definition of M_z, see the footnote, 3, to the formula (9)). Unfortunately, we were not able to establish unequivocally the sign with which M_z is de-fined in Refs. 8 and 19.

A similar computation for the amplitudes of the $M1$ transitions $6p_{1/2} \to 8p_{1/2}$, $9p_{1/2}$ yields

$$M_z(6p_{1/2} \to 8p_{1/2}) = 1.8 \cdot 10^{-5} | \mu_B |, \quad M_z(6p_{1/2} \to 9p_{1/2}) = 1.2 \cdot 10^{-5} | \mu_B |. \tag{14}$$

As for the correction to the g factor of the $6p_{3/2}$ state, the contribution made to it by the third-order mechan-ism considered in Sec. 2 is equal to

$$\delta g_3 = -\frac{4}{5} \sum_{nn'lj} \left\{ \frac{(l-1)l(l+1)}{(2l-1)(2l+1)^2} G^{l-1} + \frac{l(l+1)(l+2)}{(2l+1)^2(2l+3)} G^{l+1} \right\}$$
$$\times \frac{\alpha(nlj; n'lj)}{E_{nlj} - E_{n'lj}}, \tag{15}$$

$$G^k \equiv G^k(6p_{1/2}, nlj; 6p_{1/2}, n'lj).$$

According to the numerical computations, $\delta g_3 = 2.6 \times 10^{-4}$. The relativistic and magnetic corrections are computed from the formulas (6) and (7) with $\varkappa = -2$:

$$\delta g_{\text{rel}} + \delta g_{\text{dia}} = -0.63 \cdot 10^{-4} - 0.38 \cdot 10^{-4} = -1.01 \cdot 10^{-4}.$$

Generally, our calculations overestimate the Coulomb integrals for the electron excitations from a closed subshell by a factor of $1.5-2.5$ (see the calculation of δg in cesium in the present paper and the calculation of the hyperfine structure of thallium, lead, and bismuth in Ref. 24). Therefore, the various contributions to δg substantially cancel each other out. In this situation we can only give the estimate

$$\delta g \lesssim 1 \cdot 10^{-4}. \qquad (16)$$

We do not know the experimental value of this quantity.

APPENDIX

For all the computations in cesium we used the effective potential

$$V(r) = -\frac{(Z-1)}{r(1+\eta r)^2}\frac{e^{-\mu/d}+1}{e^{(r-\mu)/d}+1} - \frac{1}{r}, \qquad (A.1)$$

$$Z=55, \quad \mu = 3.1a_B, \quad d = 0.4a_B, \quad \eta = 2.235a_B^{-1}.$$

Actually, this is the Tietz potential[25] truncated at large distances. For $r < \mu$, this potential is well approximated by the Thomas-Fermi potential. The potential (A.1) reproduces the experimental energies and the fine structure of all the states (including the inner-shell ones) with an error not worse than a few percent.

In thallium we used the potential proposed in Ref. 19:

$$V(r) = -\frac{(Z-1)}{r(1+\eta r)^2}e^{-r/d} - \frac{1}{r}, \qquad (A.2)$$

$$Z=81, \quad d = 3.8775a_B, \quad \eta = 2.5937a_B^{-1}.$$

The error in the energy and fine-structure fits here is also not worse than a few percent.

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[1] V. G. Zelevinskii has drawn our attention to the fact that this circumstance leads to the appearance of a paramagnetic correction, $\sim (Z\alpha)$,[4] to the diamagnetic susceptibility of an inert gas. The magnitude of this correction can easily be computed with the aid of the formula (3).

[2] One of the present authors (I. B. Khriplovich) takes the opportunity to note that an error in the preprint of his work published in Ref. 6 was corrected after he had become acquainted with M. and C. Bouchiat's paper.[5]

[3] We define $\hat{M}_z$ in such a way that $\hat{M}_z \to -|\mu_B|(\hat{L}_z + 2\hat{S}_z)$ in the nonrelativistic limit. All the radial wave functions used by us are positive in the $r \to 0$ limit.

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