

Large Contributions of Negative-Energy States to Forbidden Magnetic-Dipole Transition Amplitudes in Alkali-Metal Atoms

I. M. Savukov, A. Derevianko, H. G. Berry, and W. R. Johnson

Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556

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The influence of negative-energy states (NES) on forbidden magnetic-dipole $ns_{1/2} - (n+1)s_{1/2}$ transitions in alkali-metal atoms is investigated. We find that the NES contributions are significant in almost all cases and, for rubidium, reduce the transition rate by a factor of 8. We tabulate magnetic-dipole (M_1) transition amplitudes for the alkalis. Our M_1 value for cesium, where accurate measurements are available, differs from experiment by 16%. We briefly discuss the feasibility of an experimental test of NES effects.

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It is well known (see, for example, the discussion by Brown and Ravenhall [1]) that the Dirac-Coulomb Hamiltonian has no bound state eigenfunctions in the presence of the electron-electron interaction. The *no-pair* Hamiltonian $H_{n,p}$, derived from quantum electrodynamics has been advocated by Sucher [2] for use in relativistic atomic calculations. Although $H_{n,p}$ leads to very accurate energies, the omitted effects of electron-positron pairs can be significant for the forbidden magnetic-dipole (M_1) transition amplitudes. The first discussion of pair corrections to the M_1 decay rate for the 2^3S_1 state in helium was given by Feinberg and Sucher [3] in 1971. Later, when new lifetime measurements of the 2^3S_1 metastable state of heliumlike ions became available, more accurate calculations of M_1 were performed by several theoretical groups. Lindroth and Salomonson [4] numerically demonstrated the detailed cancellation of one-pair diagrams in the case of heliumlike argon. The decay rates of the same transitions for heliumlike ions with $Z = 2-100$ were calculated within a relativistic configuration-interaction approach by Johnson, Plante, and Sapirstein [5]. They treated contributions of negative-energy states (NES) using second-order many-body perturbation theory (MBPT). Indelicato [6] considered the effects of NES for 2^3S_1 M_1 decay in the multiconfiguration Dirac-Fock approach. Recently we studied NES contributions to transition amplitudes in more detail [7]. In the Pauli approximation, we derived an effective one-pair operator that explicitly reveals cancellation between Coulomb and Breit two-body diagrams and, by using it, found a transition without such cancellation: the neutral helium $2^3S_1-3^3S_1$ transition has a very large NES contribution which reduces the M_1 rate by a factor of 2.9.

There have been no other calculations treating NES contributions to M_1 transitions systematically in multi-electron systems, except for He- and Be-like transitions [8]. In this Letter, we report second-order MBPT calculations of magnetic-dipole $ns_{1/2} - (n+1)s_{1/2}$ transitions in the alkalis including the analysis of one-pair effects. We have discovered an unusually large NES contribution,

which reduces the transition rate by a factor of 8 in Rb, and propose measurements to test NES effects. The results of our calculations for Cs are compared to a previous theoretical determination [9] and to experimental values [10,11]. For the other alkalis, no measurements exist.

We can argue that forbidden magnetic-dipole amplitudes are the most sensitive among electromagnetic transition amplitudes to the accuracy of the relativistic description of an atomic system. As we will demonstrate, several factors contribute to the result: correlation effects, spin-orbit interaction, Breit interaction, retardation effects, and, finally, the negative-energy contributions.

The interaction of an electron with external electromagnetic field in transverse gauge is represented as (atomic units are used throughout the Letter)

$$H_1 = c\alpha \cdot \hat{\epsilon} e^{i\mathbf{k}\cdot\mathbf{r}}, \quad (1)$$

where $\hat{\epsilon}$ is the photon polarization and \mathbf{k} is its wave vector. Multipole expansion and angular reduction (see, e.g., Ref. [5]) lead to the following expression for the relativistic retarded magnetic-dipole reduced matrix element:

$$\begin{aligned} \langle m || M_1 || n \rangle &= c \langle -\kappa_m || C_1 || \kappa_n \rangle (\kappa_m + \kappa_n) \\ &\times \int_0^\infty \frac{3}{k} j_1(kr) (G_m F_n + F_m G_n) dr. \end{aligned} \quad (2)$$

Here G and F are the radial parts of large and small components of atomic orbitals, k is the photon wave number, $\kappa = (j + \frac{1}{2})(-1)^{j+l+1/2}$, and C_1 is the normalized spherical harmonic [12]. In the long-wavelength limit and Pauli approximation this relativistic expression reduces to a conventional nonrelativistic operator

$$M_1 = L + 2S. \quad (3)$$

Even if the general angular selection rules for the M_1 operator are satisfied, this matrix element vanishes when the radial wave functions are orthogonal, i.e., if $\kappa_i = \kappa_j$, but $n_i \neq n_j$. The Einstein A coefficient for the M_1 transition $|I\rangle \rightarrow |F\rangle$ is expressed in terms of the reduced

matrix element as

$$A_{M_1} = \frac{k^3}{3c^2} \frac{|\langle F || M_1 || I \rangle|^2}{2J_I + 1}.$$

We start by utilizing second-order MBPT built on the “frozen-core” Dirac-Hartree-Fock (DHF) potential. This approximation includes both leading correlation and NES effects. We consider matrix elements of the magnetic-dipole operator z between two valence states v and w . For the purposes of this paper, the valence state v represents the ground state orbital $ns_{1/2}$ and the state w represents the first excited s state $(n+1)s_{1/2}$. The first-order value is given by the matrix element taken between DHF orbitals z_{wv} . The second-order correction including both Coulomb (g_{ijkl}) and Breit (b_{ijkl}, b_{ij}) interactions, is given by

$$\begin{aligned} Z_{wv}^{(2)} = & \sum_{i \neq v} \frac{z_{wi} b_{iv}}{\epsilon_v - \epsilon_i} + \sum_{i \neq w} \frac{b_{wi} z_{iv}}{\epsilon_w - \epsilon_i} \\ & + \sum_{na} \frac{z_{an} (\tilde{g}_{wnva} + \tilde{b}_{wnva})}{\epsilon_a + \epsilon_v - \epsilon_n - \epsilon_w} \\ & + \sum_{na} \frac{(\tilde{g}_{wavn} + \tilde{b}_{wavn}) z_{na}}{\epsilon_a + \epsilon_w - \epsilon_n - \epsilon_v}. \end{aligned} \quad (4)$$

Here antisymmetrized matrix elements are defined as $\tilde{g}_{ijkl} = g_{ijkl} - g_{ijlk}$. The one-body matrix element of the Breit interaction is $b_{ij} = \sum_a b_{iaja}$. In the above expressions, index a runs over core orbitals and n extends over virtual orbitals. It is important to note that virtual states include both excited positive- and negative-energy orbitals. Corresponding Feynman diagrams are given in Fig. 1. We emphasize that in the no-pair approximation the summation over virtual states would be limited only to the positive-energy states, i.e. include only the diagrams in the upper panel of Fig. 1. The inclusion of the Breit interaction in our analysis is crucial, because its contribution to the matrix element is of the same order as for the Coulomb interaction. We use a static limit of the Breit interaction, since the energy of the transverse photon is determined by the energy difference of the real electrons, not by that of the virtual electron. The numerical calculations were performed with a relativistic B -spline basis set representation [13] obtained in a cavity of radius 40 atomic units and included 40 positive-energy and 40 negative-energy wave functions for each partial wave.

The results of our calculations are presented in Table I. Note that the first-order Dirac-Hartree values dominate for lighter atoms and become less significant for cesium and francium. This is due to larger second-order no-pair contributions. The values of NES contributions (in the third row) appear to be roughly proportional to Z . In the case of cesium they constitute only a small fraction (4%) which is even smaller for francium (0.6%) due to large no-pair second-order contributions. However, the NES fraction reaches 19% in potassium. The rubidium case is the most surprising: there is cancellation of the two no-pair

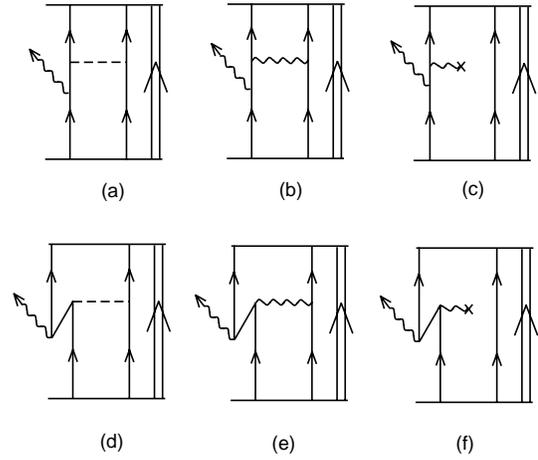


FIG. 1. Principal Feynman-like time-ordered diagrams contributing to M_1 amplitude in the second order. The wavy lines represent photons and the straight lines represent atomic electrons. The double vertical solid lines designate inactive (observing) electrons. Diagrams (a) and (d) are due to Coulomb interaction, (b) and (e) are due to two-body static Breit interaction, and (c) and (f) due to one-body static Breit interaction. The upper panel of diagrams represents no-pair contributions, and the lower panel represents contributions from negative-energy states.

terms and, consequently, a strong dependence of the total value on the negative-energy corrections. Although such cancellation in second order may be coincidental, and more accurate calculations may be necessary, we conclude that a measurement of the M_1 transition in rubidium could provide an excellent test of the NES contributions.

The large relative contribution of NES for forbidden magnetic-dipole transitions is caused by several factors. First, due to unique properties of the M_1 operator (3), the no-pair amplitude is severely suppressed (by a factor of α^2 in the lowest order). Second, the magnetic-dipole operator $(M_1)_{ij}$ in Eq. (2) contains an integral of large and small components of Dirac wave functions. For positive-energy states the small component is significantly weaker than the large component (by a factor of αZ for hydrogenlike ions). For NES, the situation is reversed, and the small component is much larger. In addition, the Pauli approximation expression (3) with its particular δ -function-like properties is no longer valid and one obtains much larger values for M_1 matrix elements between negative and positive states than from positive-positive matrix elements. Finally, the energy denominators of order $2mc^2$ bring the NES contributions to the same level as the contribution from the “regular” positive-energy states. As seen from Table II, NES contributions from the Breit interaction are comparable to those from the Coulomb diagrams because the Breit operator mixes large and small components.

We note that, for Rb, Cs, and Fr, correlation effects are very important, leading to contributions larger than the lowest-order DHF values. The mechanism has been discussed by Dzuba *et al.* [9]. In the Pauli approximation,

TABLE I. Contributions to reduced matrix elements of the M_1 operator in atomic units multiplied by a factor of 10^5 . Row 1, lowest-order DHF value; row 2, second-order no-pair contribution; row 3, negative-energy state contributions in second order; row 4, total value of M_1 matrix element.

Z	Li 3	Na 11	K 19	Rb 37	Cs 55	Fr 87
I	0.91	1.16	1.15	1.38	1.51	2.09
II, no-pair	0.12	0.03	-0.08	-1.86	-10.69	-116
II, NES	0.02	0.13	0.20	0.31	0.40	0.64
Total	1.05	1.06	1.27	-0.17	-8.78	-113

the M_1 matrix element is proportional to the integral of the product of the large components between the states involved. In the first-order forbidden transitions $ns_{1/2} - (n+1)s_{1/2}$ between states with different principal quantum numbers, the radial wave functions are orthogonal and the M_1 rate is zero. Although it is not zero beyond Pauli approximation, it is strongly suppressed. The situation is quite different for $p_{1/2}$ and $p_{3/2}$ matrix elements which are nonzero due to overlapping radial wave functions caused by the spin-orbit interaction. As a result, the second-order contributions dominate due to such matrix elements connecting core and exciting states. This correlation effect becomes overwhelming for heavier elements where spin-orbit coupling is important.

In Table III, we compare our cesium results for the magnetic-dipole reduced matrix element with calculations of Dzuba *et al.* [9] and with measurements from several experimental groups. The transition amplitude used in [9] is related to the reduced matrix element expressed in atomic units as

$$(M_1)_{\text{amp}} = \frac{1}{\sqrt{6}} \langle n_w s_{1/2} || M_1 || n_v s_{1/2} \rangle \times \left| \frac{\mu_B}{c} \right|.$$

The experimental entries for the M_1 matrix element in Table III were obtained from measurements of M_1^{hf}/M_1 and a semiempirical value [11] of the off-diagonal hyperfine mixing amplitude $M_1^{\text{hf}} = 0.8094(20) \times 10^{-5} |\mu_B/c|$. The result of our work, despite approximate treatment of correlation effects, is in reasonable agreement (16%) with the experimental results. Since the negative-energy ef-

TABLE II. Breakdown of negative-energy state contributions to the reduced M_1 matrix element in atomic units, multiplied by a factor of 10^5 . The column "Coulomb" represents contributions from Fig. 1(d), column "Breit two body"—from Fig. 1(e), and column "Breit one body"—from Fig. 1(f).

	Coulomb	Breit		Total
		two body	one body	
Li	-0.015	0.067	-0.029	0.024
Na	-0.020	0.106	0.047	0.133
K	-0.022	0.112	0.106	0.197
Rb	-0.025	0.154	0.174	0.303
Cs	-0.026	0.183	0.239	0.395
Fr	-0.035	0.221	0.450	0.636

fects are marginally smaller than these deviations, it is not possible to draw definitive conclusions about NES effects from available experiments in cesium. The second-order expression (4) is a leading term of the random-phase approximation (RPA). The calculations of Dzuba *et al.* [9] implicitly included the effect of negative-energy states due to the reduction of RPA-like diagrams to the form of a differential equation. However, their analysis did not take into account the Breit interaction. Such an approach misses an important negative-energy contribution. Indeed, we demonstrate in Table II that the NES contribution from the Breit interaction is much larger than that arising from the Coulomb interaction.

The theoretical calculations of the M_1 transition amplitudes in the alkalis clearly demonstrate the important role of negative-energy states. We now discuss experimental possibilities to test these contributions. We compare the NES fractional contributions, defined as the ratio of NES to no-pair contributions, in different alkali-metal atoms in Fig. 2. In the light alkalis (Li, Na, K) the effect is proportional to Z and is maximal for K (19%). For heavy atoms such as Cs and Fr, it is small because of large no-pair contributions. Rubidium, in the middle, has a very large relative effect (65%) and is the most promising. If measurements in the other alkalis reach the precision achieved in Cs, then all alkalis except Fr will be good candidates for testing NES contributions. The accuracy of the calculations, on the other hand, can impose

TABLE III. Comparison of theoretical and experimental values for Cs $6s-7s$ reduced magnetic-dipole matrix element in atomic units, multiplied by a factor of 10^5 . The experimental errors are given in parentheses.

Reference	$\langle 6s M_1 7s \rangle \times 10^5$
Theory	
This work	-8.78
Dzuba <i>et al.</i> [9], 1985	-13.7
Experiment	
Bennett and Wieman [10], 1999	-10.40 (0.03)
Bouchiat and Guéna [11], 1988 ^a	-10.5 (0.1)

^aThe average of previous experimental results corrected for the electric-quadrupole contribution.

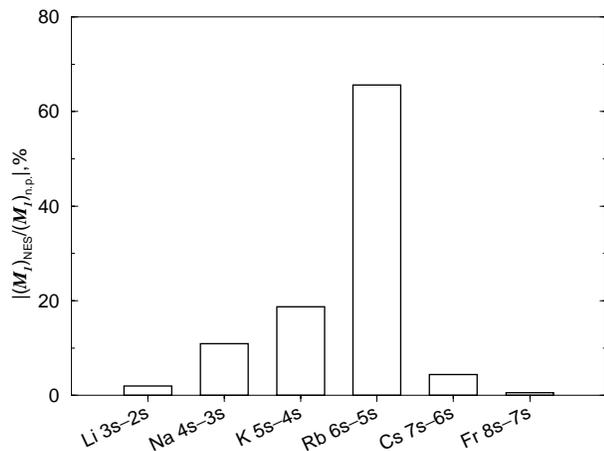


FIG. 2. The relative contributions to the magnetic-dipole (M_1) matrix element $ns - (n+1)s$ in alkali atoms: the ratio of the NES contributions (row 3 of Table I) to the total no-pair contributions (sum of rows 1 and 2 of Table I).

even more severe restrictions than experiment. The accuracy of our calculations, as seen in the deviation from the experiment for Cs, is about 16%; it is expected to be better for lighter elements. More accurate (1%) no-pair calculations are possible, for example, in the relativistic single-double approximation [14]. For Li, precise no-pair configuration-interaction calculations [15] are also possible with an accuracy much better than 1%.

In conclusion, we have presented the results of the second-order MBPT calculations for the forbidden M_1 transitions in the alkalis. Comparisons with experimental and theoretical data for cesium have been made. We have found very large negative-energy state contributions to the M_1 transition amplitudes in the alkalis. The NES amplitude is dominant in the case of rubidium, which could provide the best experimental test of negative-energy contributions in atomic structure.

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