

Correlation and Negative Continuum Effects for the Relativistic $M1$ Transition in Two-Electron Ions using the Multiconfiguration Dirac-Fock Method

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The role of the $E < -mc^2$ continuum and of correlation in the multiconfiguration Dirac-Fock calculation of nondiagonal one-electron operators is described. Because of its importance and symmetry which emphasize both effects, we study the $1s2s^3S_1 \rightarrow 1s^2^1S_0$ relativistic $M1$ transition in two-electron ions. It is shown that both contributions have the same magnitude, and must be included even at low Z . These contributions improve significantly the agreement between theory and experiment at all Z , reducing the disagreement for Nb from 2.7 to -1.8 times the experimental error. [S0031-9007(96)01383-X]

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In the past few years much effort has been placed on the understanding of the relativistic many-body problem in atoms. Both few-electron heavy ions and inner shells of heavy atoms have been intensely studied. Because the direct use of bound state QED (as was recently done in [1,2]) is very often impractical for the evaluation of level energies in systems with more than one electron, approximate methods, based on an effective relativistic Hamiltonian, have been developed, in which correlation energy can be evaluated in the same way as in the non-relativistic case. Multiconfiguration Dirac-Fock (MCDF) and relativistic many-body perturbation theory (RMBPT) are the most widely used methods for that purpose. Following Brown and Ravenhall [3], Sucher [4] pointed out in 1980 that the existence of the $E < -mc^2$ continuum did not easily allow one to generalize to the relativistic case the nonrelativistic methods for energy calculations: A proper form of the electron-electron interaction with projection operators onto the $E > mc^2$ continuum must be used, leading to the so called no-pair Hamiltonian, $\mathcal{H}^{\text{np}} = \sum_{i=1}^m \mathcal{H}_D(r_i) + \sum_{i<j} \mathcal{V}(|\mathbf{r}_i - \mathbf{r}_j|)$, where \mathcal{H}_D is a one-electron Dirac operator and \mathcal{V} is an operator representing the electron-electron interaction $\mathcal{V}_{ij} = \Lambda_{ij}^{++} V_{ij} \Lambda_{ij}^{++}$, with

$$V_{ij} = \frac{1}{r_{ij}} - \frac{\boldsymbol{\alpha}_i \cdot \boldsymbol{\alpha}_j}{r_{ij}} - \frac{\boldsymbol{\alpha}_i \cdot \boldsymbol{\alpha}_j}{r_{ij}} [\cos(\omega_{ij} r_{ij}) - 1] + (\boldsymbol{\alpha}_i \cdot \nabla_i)(\boldsymbol{\alpha}_j \cdot \nabla_j) \frac{\cos(\omega_{ij} r_{ij}) - 1}{\omega_{ij}^2 r_{ij}}. \quad (1)$$

Here $\Lambda_{ij}^{++} = \Lambda_i^+ \Lambda_j^+$ is an operator projecting onto the positive energy states to avoid introducing unwanted pair creation effects, $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the interelectronic distance, ω_{ij} the energy of the photon exchanged between the electrons, and $\boldsymbol{\alpha}_i$ are Dirac matrices. The first term in Eq. (1) represents the regular Coulomb interaction, the second one is the magnetic (Gaunt) interaction, and the rest is the retardation (in Coulomb gauge) [5,6].

Whether the use of projection operators is necessary or not in the MCDF case has been controversial [6-8],

and it was not at all clear on how to include them in practice. This has been settled recently as a method has been proposed which enables one to identify and to project out the spurious contributions from the $E < -mc^2$ continuum in the MCDF case [9]. Yet, little attention has been paid, so far, to the role of the $E < -mc^2$ continuum in the evaluation of quantity other than energy, such as transition probabilities.

The purpose of this Letter is to clarify this last point, taking as an example the $1s2s^3S_1 \rightarrow 1s^2^1S_0$ relativistic $M1$ transition. I report here a complete multiconfiguration Dirac-Fock calculation, in which correlation contribution to the energy and the radial matrix element are taken into account, I discuss the effect of the $E < -mc^2$ continuum, and provide accurate transition rates for high Z ions.

The relativistic $M1$ is of particular interest, because precise experiments are available for $2 \leq Z \leq 54$, and because the effects I want to discuss are enhanced; the transition being completely of relativistic origin. Atomic numbers down to $Z = 2$ are studied to show the interplay between correlation and the $E < -mc^2$ continuum.

There is no explicit expression for Λ^{++} , except at the Pauli approximation [10]. In the MCDF method, the wave function is usually obtained numerically through, e.g., finite difference methods, from an inhomogeneous integro-differential equation in which the inhomogeneous term comes from exchange. There is thus no simple way to construct a projection operator. In Ref. [9] an approximation to such an operator is constructed by developing the solution of the *inhomogeneous* integrodifferential equation on the basis set representing all the solutions of the *homogeneous* equation. The expansion over the complete set of the homogeneous solutions will explicitly include the contribution of the negative continuum. Restricting the expansion to the solutions whose eigenvalues are greater than $-mc^2$ removes this negative continuum contribution in the same way as in RMBPT calculations.

The use of the MCDF wave functions for the evaluation of nondiagonal operators such as the transition rates

is not straightforward. The two available MCDF codes [11,12] are designed with the assumption that all spin orbitals of identical symmetry in all configurations are orthogonal. This does not lead to any problem when evaluating energies but makes it more difficult to use fully relaxed wave functions when computing matrix elements between two different states. The standard procedure is to use wave functions for the initial and final states, in which all spectator-electron orbitals are frozen. Thus the orthonormality of spin orbitals is preserved. The relativistic $M1$ is a very interesting case in that respect, because it involves two orbitals of identical symmetry ($1s$ and $2s$). Using wave functions that are Slater determinants, the matrix element $\langle 1s'2s'|(M1(r_1) + M1(r_2))|1s^2\rangle$ gives $\langle 2s'|M1|1s\rangle\langle 1s'|1s\rangle - \langle 1s'|M1|1s\rangle\langle 2s'|1s\rangle$ (primes denote initial state orbitals). If spin orbitals in the initial and final states are orthogonal, then this reduces to the usual matrix element. This happens, for example, if the $2s'$ orbital has been calculated using the frozen $1s$ orbital in lieu of $1s'$. Otherwise, the overlap $\langle 2s'|1s\rangle$ is $\approx 10^{-3}$. But the matrix element $\langle 1s'|M1|1s\rangle$ is much larger than $\langle 2s'|M1|1s\rangle$, and the final value is $\approx 40\%$ off if this term is neglected. It agrees with the frozen core calculation to four significant figures otherwise.

For correlated wave functions there is no such alternative: One must use full relaxation in both the initial and final states. Correlation orbitals (orbitals with effective occupations small compared to one) are very different from occupied orbitals, because the exchange potential, not the direct potential, dominates their behavior (particularly when the Gaunt interaction is made self-consistent [9]). In that case, overlaps cannot be neglected, and any MCDF calculation of an off-diagonal operator is meaningless if one does not properly include nonorthogonality contributions. I have written a code that can generate all possible matrix elements between two arbitrary MCDF wave functions following the formalism established by Löwdin (1955) [13]. Because it leads to very large calculations for many-electron atoms, this formalism is seldom used [14,15]. The two-electron system, however, can be evaluated easily. The initial wave function is taken as $1s2s^3S_1 + 3s4s^3S_1 + \dots + 4f5f^3S_1$ and the ground state wave function as $1s^2^1S_0 + 2s^2^1S_0 + \dots + 5g^2^1S_0$ [9,16,17].

The results obtained with this procedure are in better agreement with experiment at $Z = 41$ than a simple Dirac-Fock value (using the same transition energy). Yet, they are still very inaccurate at lower Z , recovering only half of the difference between the single configuration Dirac-Fock values and experiment. A closer analysis shows that this is due to the use of the no-pair Hamiltonian, i.e., the exclusion of the $E < -mc^2$ continuum from the correlated wave functions used for the evaluation of the radial $M1$ matrix element. The MCDF method must be viewed as a way to do an all-order resummation of standard, i.e., non-QED perturbation theory. In that sense, $E < -mc^2$ states, which are reinter-

preted in QED as positive energy positrons, are not properly taken into account, and lead to infinite perturbation contributions to the energy [Fig. 1(a)]. The use of \mathcal{H}^{np} in the determination of the energy with the MCDF method avoids spurious contributions due to diagrams such as the one in Fig. 1(a). The situation is very different for perturbation correction to transition rates (or other one-electron operators), as was noted already in a RMBPT calculation [18]. This can easily be understood from the diagram in Fig. 1(b). In that case, the $E < -mc^2$ state is part of the first order correction to the wave function (or, in other words, a piece of the *reduced* Green's function). As the $M1$ operator is a one-electron operator, the energy denominator in that case is just $E_- - E_{2s}$, which never cancels out. Thus one should use \mathcal{V}_{ij} to evaluate correlated energies, and V_{ij} to evaluate wave functions used in the $M1$ matrix element. In Ref. [18], the total contribution of diagrams such as those in Fig. 1(b) is adding up to 0. This is due to the specific potential used in this reference for building unperturbed states. The present calculation proves that the effect of the $E < -mc^2$ continuum on the $M1$ rate is much larger with a fully self-consistent MCDF potential than in the RMBPT case.

Since the Gaunt interaction is treated self-consistently, I could not reach convergence for very large sets of configurations [9], and could not do calculations for $Z \geq 54$, although techniques have been developed recently to improve those problems when the Gaunt interaction is not self-consistent [19]. However, the $M1$ matrix element converges very fast when summing over angular momentum, and adding configurations involving only s and p electrons was enough to reach convergence again as described in Ref. [18]. This fact can be checked on the calculation with projection operators, for which all configurations up to $n = 5$ and $l = 4$ are included. At

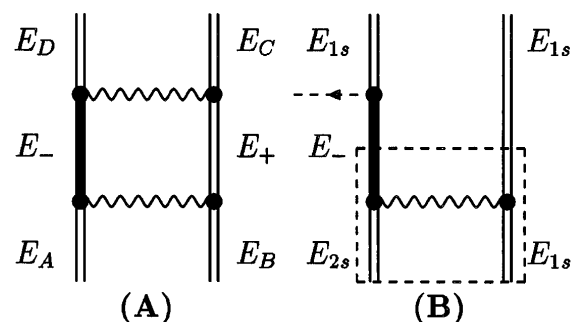


FIG. 1. (a) Second order perturbation contribution to the energy to be excluded by the projection operator. The energy denominator is $E_+ + E_- - E_A - E_B$, with $E_- < -mc^2$ (negative energy continuum), $E_+ > mc^2$ (positive energy continuum), E_A, E_B bound state energies. This denominator can always be canceled out. Including a projection operator suppresses the matrix element, avoiding the problem. The suppressed terms can only be evaluated using QED [1,2]. (b) Contribution to the $M1$ matrix element with intermediate $E < -mc^2$ state. The area inside the dashed box corresponds to the first order correction to the wave function (reduced Green's function).

TABLE I. Relativistic $M1$ transition rate(s). DF: single configuration Dirac-Fock lifetime. MCDF_{np} : MCDF lifetime evaluated with a no-pair Hamiltonian. $\%/DF$: variation relative to DF lifetime. $\text{MCDF}_{e^+e^-}$: MCDF lifetime evaluated without projection operators in the wave function. $\%/np$: variation relative to MCDF_{np} lifetime. The same high precision energy is used for all three calculations.

Z	DF	MCDF_{np}	$\%/DF$	$\text{MCDF}_{e^+e^-}$	$\%/np$
2	1.288×10^4	0.875×10^4	-47.2%	0.782×10^4	-11.8%
3	6.637×10^1	5.603×10^1	-18.5%	5.058×10^1	-10.8%
6	2.367×10^{-2}	2.220×10^{-2}	-6.6%	2.103×10^{-2}	-5.5%
7	4.439×10^{-3}	4.205×10^{-3}	-5.6%	4.012×10^{-3}	-4.8%
10	9.935×10^{-5}	9.584×10^{-5}	-3.7%	9.270×10^{-5}	-3.4%
12	1.467×10^{-5}	1.424×10^{-5}	-3.0%	1.385×10^{-5}	-2.8%
16	7.372×10^{-7}	7.214×10^{-7}	-2.2%	7.059×10^{-7}	-2.2%
17	3.939×10^{-7}	3.860×10^{-7}	-2.1%	3.781×10^{-7}	-2.1%
18	2.183×10^{-7}	2.142×10^{-7}	-1.9%	2.100×10^{-7}	-2.0%
22	2.763×10^{-8}	2.720×10^{-8}	-1.6%	2.676×10^{-8}	-1.6%
23	1.749×10^{-8}	1.723×10^{-8}	-1.5%	1.697×10^{-8}	-1.6%
26	4.964×10^{-9}	4.899×10^{-9}	-1.3%	4.833×10^{-9}	-1.4%
35	2.342×10^{-10}	2.319×10^{-10}	-1.0%	2.298×10^{-10}	-0.9%
36	1.753×10^{-10}	1.736×10^{-10}	-1.0%	1.721×10^{-10}	-0.9%
41	4.589×10^{-11}	4.549×10^{-11}	-0.9%	4.516×10^{-11}	-0.7%
47	1.117×10^{-11}	1.108×10^{-11}	-0.8%	1.102×10^{-11}	-0.6%
54	2.631×10^{-12}	2.613×10^{-12}	-0.7%		
92	8.285×10^{-15}	8.245×10^{-15}	-0.5%		

$Z = 6$, including higher l and levels up to $n = 5$ gives only a 0.06% change. For low Z the maximum n used in the set of configurations was 4. For $Z \geq 18$ the calculation had to be limited to $n \leq 3$. However, in this case, correlation plays a relatively smaller role.

The results of these calculations are presented in Table I. The comparison between both calculations and experiment is presented in Fig. 2, together with other calculations, and in Table II. The effect of correlation on the radial part of the matrix element ranges from 47% at $Z = 2$ to 0.5% at $Z = 92$. As can be seen in Table II and Fig. 2, the $0.96\% \pm 0.35\%$ disagreement between the Dirac-Fock calculation and the experiment for Nb becomes 0.1% when correlation to the matrix element is included using MCDF wave functions evaluated with projection operators. For $Z = 6$ the difference decreases from $15 \pm 0.2\%$ to 7.8% but is not canceled. In the same way, at $Z = 18$, the difference between the RMBPT value of [18] and the MCDF value decreases from $4.1 \pm 0.2\%$ to 2.3%. Most of the remaining difference is recovered when removing the projection operators from the wave function used to evaluate the $M1$ matrix element (obviously, the energies must still be computed with projection operators). For the same elements, the latter calculation gives $0.64 \pm 0.35\%$ (Nb), $2.4 \pm 0.2\%$ (C), and $0.3 \pm 0.2\%$ (Ar), respectively, which reduces significantly the difference between calculation and the most precise experiments to date ($Z = 6, 10, 41, 47$). At that stage, it should be noted that, if the magnetic part of the interaction (1) is not included in the evaluation of the wave function, the transition range changes by almost 5% at $Z = 6$, thus destroying the agreement between theory and experiment. This has, however, little effect at high Z .

In this paper, I have shown that the MCDF method can reproduce very accurately the relativistic $M1$ transition rate, provided that correlation is properly accounted for, using fully relaxed wave functions, evaluated with both the Coulomb interaction and the magnetic interaction. One is then obliged to properly take into account the non-orthogonality between spin orbitals. I have also shown that the $E < -mc^2$ continuum plays a very special role

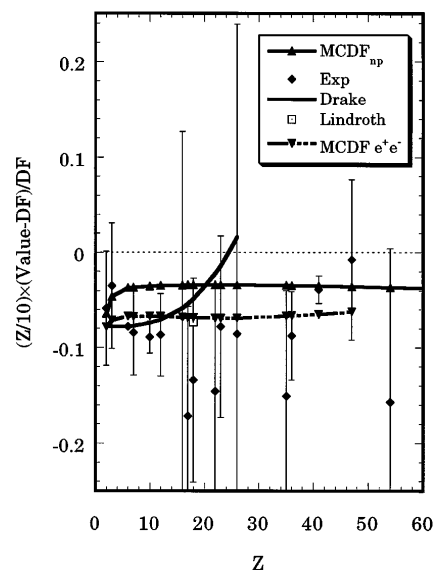


FIG. 2. Comparison between theory and experiment. All rates are normalized to the DF value. DF: single configuration Dirac-Fock used as a reference. MCDF: highly correlated Dirac-Fock calculation using a no-pair Hamiltonian. $\text{MCDF}_{e^+e^-}$: same without the projection operator. Other calculations from Refs. [18,33-36].

TABLE II. Comparison between the different DF approximations used in this work and experiment. Exp: experimental value. Prec.: experimental precision. Relative differences are calculated using experimental values as a reference.

Z	Exp.	Ref.	Prec.	DF	MCDF _{np}	MCDF _{e⁺e⁻}
2	0.909×10^4	[20]	30.0%	41.7%	-3.8%	-13.9%
3	5.860×10^1	[21]	22.0%	13.3%	-4.4%	-13.7%
6	2.509×10^{-2}	[22]	0.2%	15.0%	7.8%	2.1%
7	3.905×10^{-3}	[22]	6.4%	13.7%	7.7%	2.8%
10	9.050×10^{-5}	[23]	1.7%	9.8%	5.9%	2.4%
12	1.361×10^{-5}	[24]	3.6%	7.8%	4.7%	1.8%
16	7.060×10^{-7}	[25]	12.2%	4.4%	2.2%	0.0%
17	3.540×10^{-7}	[25]	6.8%	11.3%	9.0%	6.8%
18	2.020×10^{-7}	[26]	5.9%	8.1%	6.0%	4.0%
22	2.580×10^{-8}	[26]	5.0%	7.1%	5.4%	3.7%
23	1.690×10^{-8}	[27]	4.1%	3.5%	2.0%	0.4%
26	4.800×10^{-9}	[27]	12.5%	3.4%	2.1%	0.7%
35	2.241×10^{-10}	[28]	3.2%	4.5%	3.5%	2.5%
36	1.710×10^{-10}	[29]	1.3%	2.5%	1.5%	0.6%
41	4.545×10^{-11}	[30]	0.35%	0.95%	0.1%	-0.64%
47	1.115×10^{-11}	[31]	1.8%	0.2%	-0.6%	-1.2%
54	2.554×10^{-12}	[32]	3.0%	3.0%	2.3%	

in the calculation, particularly at low Z where it is the least expected (this is also true of the magnetic interaction). The discrepancy between theory and the most precise experiments is significantly reduced. Residual discrepancy remains at low Z due to the relatively limited size of the configuration set. At $Z = 41$, the precision of the experiment suggests that radiative corrections to the transition rate, beyond the one included in the transition energy, should be investigated to account for the remaining discrepancy. The formalism and program used for this calculation can be readily used to study forbidden transitions, or transitions for which correlation is very important such as two-electron one-photon transitions, in more complex ions or atoms. It must also be used for other nondiagonal one-electron operators as those in hyperfine quenching or parity violation.

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