

Relativistic calculation of the $2^3S_1 - 1^1S_0$ magnetic dipole transition rate and transition energy for heliumlike argon

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A calculation of the magnetic dipole transition $2^3S_1 - 1^1S_0$ in heliumlike argon is presented. The calculation is performed in a relativistic framework and includes the Coulomb interaction, in the no-virtual-pair approximation, to all orders. Contributions from the exchange of virtual photons, i.e., from the Breit interaction, are of the same formal order of magnitude as contributions from the Coulomb interaction and are treated on the same footing. The combination of a relativistic treatment and the inclusion of correlation in the calculation results in an improved accuracy compared with previous calculations. The lifetime of the 2^3S_1 state is predicted to be 209.4 ± 0.4 nsec, which includes an estimate of neglected effects, ± 0.2 nsec, and numerical errors, ± 0.2 nsec. This result agrees with experiment, 202 ± 12 nsec, within present error bars. A detailed breakdown of the contributions to the transition energy is also presented. The contribution to the ground-state energy of Ar^{16+} from the mixture of the Breit and the Coulomb interaction is calculated to all orders. This contribution is 9% smaller than the result in the Pauli approximation.

I. INTRODUCTION

The transition from the $1s2s^3S$ state to the ground state of helium and heliumlike ions is dominated by the magnetic dipole decay. This was demonstrated by Griem¹ following the work by Gabriel and Jordan,² who observed the decay of the $1s2s^3S$ state in solar plasma and it was in contradiction with the earlier suggestion by Breit and Teller³ that the decay should be dominated by multiphoton processes.

The first measurement of the decay of a 2^3S state was done on heliumlike argon by Marrus and co-workers⁴⁻⁶ using beam-foil spectroscopy. The measured lifetime was 172 ± 30 nsec while most theoretical predictions available at the time^{7,8} or produced later⁹⁻¹³ range from 205 to 215 nsec. These theoretical predictions are, of course, of varying accuracy as will be discussed below. In a later experiment¹⁴ efforts were made to avoid the spurious dependence on the distance from the foil which had been seen by Bednar *et al.*¹⁵ in experiments with chlorinelike and sulfurlike helium. This second measurement gave a lifetime of 202 ± 20 nsec for Ar^{16+} . Lin and Armstrong¹⁶ have tried to explain the position-dependent results with the presence of Li-like ions in the ion beam. Certain configurations of the Li-like ions can undergo a $M1$ transition of a very similar transition energy as the heliumlike ion. Since these configurations of the Li-like ion can decay also by autoionization, the lifetime will be shorter than for the heliumlike ion. A measurement which cannot distinguish between the two ions will then measure a too short lifetime for heliumlike argon.

Recently, Hubricht and Träbert¹⁷ measured the lifetime of 2^3S of heliumlike argon using slow recoil ions produced by heavy-ion-atom collisions. The x-ray radiation is measured in coincidence with the recoil ions. Their result is 202 ± 12 nsec and there is hope for new experiments with an accuracy of 1–2% in the near fu-

ture.¹⁷ This makes new theoretical efforts interesting. The aim of the present work is to obtain an accuracy well below 1%.

The magnetic dipole transition from 2^3S to 1^1S is a purely relativistic effect and the leading contribution to the transition matrix element is of order $\alpha^3 Z^2$ a.u. Earlier theoretical works have been either relativistic, but then inside a one-particle description of the ion,^{10,11,13} or made in the Pauli approximation using nonrelativistic but correlated wave functions as in the work by Drake⁷ and by Anderson and Weinhold.¹² The most important of the missing terms in Refs. 7 and 12, i.e., higher-order relativistic terms which contribute in relative order $\alpha^2 Z^2$, has been explicitly calculated by Lin.^{18,19} This correction can be added to the very accurate nonrelativistic results^{7,12} and will then give a prediction of the lifetime with an accuracy of approximately 1%, as will be discussed further in Sec. VI. In recent years a development has taken place which has made possible relativistic calculations of much better accuracy.²⁰⁻²² Relativistic correlation can now be treated to all orders as demonstrated on neutral helium by Blundell *et al.*²³ and Salomonson and Öster²² and on lithium and beryllium⁺ by Blundell *et al.*²⁴ In the present work, which uses the method developed by Salomonson and Öster, the magnetic dipole transition rate from 2^3S to 1^1S in heliumlike argon is calculated relativistically using completely correlated wave functions. This applies to correlation due to the Coulomb as well as to the Breit interaction. Due to the purely relativistic nature of this transition the effect of the Breit interaction enters in the same formal order as the Coulomb interaction, i.e., it gives contributions of order $\alpha^3 Z$ a.u. to the magnetic dipole matrix element. This will be discussed in Sec. IV. The mixture of the Breit interaction and other perturbations is an interesting subject which has not been studied in any detail in the literature. In a recent calculation Lindroth *et al.*²⁵ studied the effect of the inclusion of

the Breit interaction in the orbitals on the same footing as the Coulomb interaction when evaluating the enhancement of a possible electric dipole moment of the electron. The expectation value of various powers of r were also studied. The enhancement factor prediction for thallium decreased with 2% and the expectation value of, e.g., r^{-2} with as much as 5%. Even in cases when the Breit interaction enters in relative order α^2 Ry it may thus have a significant effect. For the transition rate $2^3S_1-1^1S_0$ in heliumlike argon the result, when hydrogenlike wave functions are used to calculate the $M1$ -matrix element, is overestimated with slightly more than 15%. The effect due to the Breit interaction is about a seventh this and opposite in sign. A neglect of the Breit interaction would then lead to an underestimate of the transition rate with 2%, i.e., an overestimate of the lifetime with the same fraction.

The effect of Breit interaction on the $M1$ -matrix element was included by means of the Pauli approximation in Refs. 7 and 12. In Ref. 13 the effect of the Breit interaction in the wave functions was not considered.

In Sec. II a review of the basic theory is given. The calculation method is discussed in Sec. III and finally the results are given in Secs. IV and V.

II. MAGNETIC DIPOLE TRANSITION

Formulas for electric and magnetic multipole transitions have been given in some detail by Akhiezer and Berestetsky²⁶ and by Grant²⁷ and only a brief introduction will be given here. We consider an atom interacting with the photon vector potential

$$H = \sum_i c \boldsymbol{\alpha}_i \cdot (\mathbf{p}_i + e \mathbf{A}_i) + \beta m_i c^2 - \frac{e}{4\pi\epsilon_0} \frac{e\mathbf{Z}}{r_i} + \sum_{i < j} V_{ij}, \quad (2.1)$$

where the transverse vector potential \mathbf{A} , when expressed in plane waves, is given by

$$\begin{aligned} \mathbf{A}(i) &= \sum_{k,q} \left[\frac{\hbar}{2\omega\epsilon_0 V} \right]^{1/2} [a_{k,q}^\dagger \boldsymbol{\epsilon}_q e^{-i(\mathbf{k}\cdot\mathbf{r}_i - \omega t)} \\ &\quad + a_{k,q} \boldsymbol{\epsilon}_q e^{i(\mathbf{k}\cdot\mathbf{r}_i - \omega t)}] \\ &= \sum_{k,q} \left[\frac{\hbar}{2\omega\epsilon_0 V} \right]^{1/2} \mathbf{A}(i)_{k,q}. \end{aligned} \quad (2.2)$$

(Formulas and equations are given in SI units. However,

er, in the subsequent sections numbers are usually given in atomic units, a.u., where e , π , $4\pi\epsilon_0$, and m_e have the value unity and the value of c , the speed of light, is $1/\alpha \approx 137$.) The dimensionless vector $\mathbf{A}_{k,q}$ has been introduced for later convenience. V_{ij} is the two-particle potential including the Coulomb and the Breit interaction between the electrons. The choice of V_{ij} as well as the related problem with projection operators will be discussed in Sec. III.

The lifetime for a certain state is the inverse of the transition rate summed over all possible final states. Expressions for transition rates between states n and n' for a photon emitted into a solid angle element $d\Omega$ can be found, e.g., in Ref. 28,

$$A_{d\Omega}^{k,q} = \frac{3}{8\pi} \left[\frac{4}{3\hbar} \frac{1}{4\pi\epsilon_0} \frac{\omega^3}{c^3} \right] \left| \langle n' | \frac{ec}{\omega} \boldsymbol{\alpha} \cdot \mathbf{A}_{k,q} | n \rangle \right|^2 d\Omega, \quad (2.3)$$

where

$$\omega = c|\mathbf{k}| = \frac{E_n - E_{n'}}{\hbar}.$$

A plane-wave description of \mathbf{A} , as in (2.2), is not suitable for our present problem and an expansion into spherical waves can be obtained by using the relation

$$\boldsymbol{\epsilon}_q e^{-i(\mathbf{k}\cdot\mathbf{r})} = \boldsymbol{\epsilon}_q \sum_{l=0}^{\infty} (-1)^l (2l+1) j_l(\omega r/c) P_l(\cos\gamma), \quad (2.4)$$

which can be found, e.g., in Ref. 29. In expression (2.4) j_l are spherical Bessel functions and P_l Legendre polynomials with γ the angle between \mathbf{k} and \mathbf{r} . For convenience we may consider the plane wave moving in the z direction with polarization vectors $\boldsymbol{\epsilon}_q$ where $q = \pm 1$ corresponds to left and right circular polarization. Note that this z axis is independent of the direction of quantization of the atomic states. The relation (2.4) can then be written

$$\begin{aligned} \boldsymbol{\epsilon}_q e^{-i(kz)} &= \sum_{l=0}^{\infty} (-1)^l (2l+1) j_l(\omega r/c) \\ &\quad \times \left[\frac{4\pi}{(2l+1)} \right]^{1/2} \boldsymbol{\epsilon}_q Y_{l0}(\theta, \varphi), \end{aligned} \quad (2.5)$$

where θ is the angle between \mathbf{r} and the z axis. The matrix element in (2.3) can now be written

$$\left\langle n' \left| \frac{ec}{\omega} \boldsymbol{\alpha} \cdot \mathbf{A}_{k,q} \right| n \right\rangle = \left\langle n' \left| \sum_{l=0}^{\infty} \frac{ec}{\omega} (-1)^l (2l+1) j_l(\omega r/c) \boldsymbol{\alpha} \cdot \boldsymbol{\epsilon}_q C_0^l \right| n \right\rangle, \quad (2.6)$$

where the components of the C tensor is related to the spherical harmonics as

$$C_q^l = \left[\frac{4\pi}{2l+1} \right]^{1/2} Y_{lq}.$$

The scalar product $\boldsymbol{\alpha} \cdot \boldsymbol{\epsilon}_q$ gives the q component of $\boldsymbol{\alpha}$. We can express $\alpha_q C_0^l$ in terms of coupled operators of well-defined rank as

$$\alpha_q C_0^l = \sum_L \{ \boldsymbol{\alpha} C^l \}_q^L \langle 1q10 | Lq \rangle.$$

This can be used to rewrite (2.6) as follows:

$$\left\langle n' \left| \sum_{l=0}^{\infty} \sum_L \frac{ec}{\omega} (-i)^l (2l+1) j_l(\omega r/c) \{ \alpha C^l \}_q^L \langle 1q|0|Lq \rangle \right| n \right\rangle. \quad (2.7)$$

The study of the transition $2^3S_1-1^1S_0$ requires an interaction which preserves parity and, in order to allow a transition from $J=1$ to 0, is in the form of a tensor operator of rank one. Only the term with $l=L=1$ in the expansion (2.7), i.e. the magnetic dipole term, has this property.

The aim is now to get an expression for the transition rate due to the magnetic dipole interaction. For this special case, where the interaction is in the form of a tensor operator of rank one, we may proceed as follows. With $L=l=1$ the expression (2.3) can now be written

$$\frac{3}{8\pi} \left[\frac{4}{3\hbar} \frac{1}{4\pi\epsilon_0} \frac{\omega^2}{c^3} \right] \left| \left\langle n' \left| \frac{-i3}{\sqrt{2}} \frac{ec}{\omega} j_1(\omega r/c) \{ \alpha C^1 \}^1 \right| n \right\rangle \cdot \epsilon_q \right|^2 d\Omega, \quad (2.8)$$

where ϵ_q , with $q=\pm 1$, is either of the two circular polarization vectors of the transverse vector field \mathbf{A} . The q component, corresponding to the photon coordinate system, of the spherical vector $\langle n' | \{ \alpha C^1 \}^1 | n \rangle$ is here written as a scalar product of this vector and the photon polarization vector ϵ_q . The summation over the two polarizations ϵ_q for each direction of \mathbf{k} , and a following integration over the directions of \mathbf{k} , can now be performed noting that the integration is equivalent to an integration over all angles between the two vectors in (2.8). This is shown in detail in Ref. 28. The transition rate between n and n' can now be written in the usual form given, e.g., by Bethe and Salpeter,³⁰

$$A_n^{n'} = \frac{4}{3\hbar} \frac{1}{4\pi\epsilon_0} \frac{\omega^3}{c^3} |\mathbf{M}_n^{n'}|^2, \quad (2.9)$$

where the magnetic dipole operator \mathbf{M}^1 is given by

$$\mathbf{M}^1 = \frac{-i3ec}{\sqrt{2}\omega} j_1(\omega r/c) \{ \alpha C^1 \}^1. \quad (2.10)$$

The lifetime for a certain magnetic substate of 2^3S_1 is obtained as the inverse of the transition rate (2.9) averaged over initial states and summed over final states, i.e., as the inverse of

$$\bar{A}_{nLJ}^{n'L'J'} = \frac{1}{2J+1} \frac{4}{3\hbar} \frac{1}{4\pi\epsilon_0} \frac{\omega^3}{c^3} \sum_{M,M'} |\mathbf{M}_{nLJM}^{n'L'J'M'}|^2. \quad (2.11)$$

It may be noted that the transition rate from a particular state $nLJM$ to all the magnetic substates M' of the state $n'L'J'$, without regard to the polarization of the emitted radiation, is independent of M .

It is illustrating to examine the $M1$ operator in the nonrelativistic limit as well as in the limit of small energy transfer. For small values of $\omega r/c$ a series expansion of the Bessel function would be valid and in lowest order it gives

$$\mathbf{M}^1 = \frac{-ier}{\sqrt{2}} \{ \alpha C^1 \}^1 = \frac{e}{2} \boldsymbol{\alpha} \times \mathbf{r}, \quad (2.12)$$

which is the result for homogeneous magnetic field over the region of the atom. For the $2^3S_1-1^1S_0$ transition,

which lies in the x-ray region, such an approximation is not valid and we will use the full expression (2.10). In the vicinity of the involved energies, around 3 keV, the $M1$ -matrix element scale approximately linearly with the energy transfer and the use of (2.12) would lead to an overestimate of the $M1$ -matrix element with around 40%.

In the nonrelativistic limit, i.e., after a lowest-order Foldy-Wouthuysen transformation has been performed, (2.12) reduces to

$$\mathbf{M}^1 = \frac{-e}{2mc} (l+2s) = \frac{-\mu_B}{c\hbar} (l+2s). \quad (2.13)$$

For the transition in question the operator (2.13) gives zero. Since it has no radial dependence, it cannot combine states with orthogonal radial function as the $1s$ and $2s$ one-particle states. Even for the exact 1^1S_0 and 2^3S_1 states (2.13) will give zero due to the symmetric respective antisymmetric nature of the radial functions. Drake⁷ has carried the Foldy-Wouthuysen transformation one step further and obtained the first nonvanishing $M1$ operators to be used in the nonrelativistic limit. We will instead employ the original operator (2.10). This operator is notationally of order α , but since the first nonrelativistic limit does not contribute to the transition in question, the size of the contributions from the operator will instead be of order $\alpha^3 Z^2$ a.u. The cancellation of the α term has to be obtained by numerical means. Some care must be taken to assure numerical stability. An advantage of the method used by Drake is, of course, that this cancellation then is taken care of explicitly. However, an accuracy of more than 2% cannot be obtained in helium-like argon without the inclusion of relativistic effects beyond the lowest nonvanishing order. Since the $M1$ operator contributes in order $\alpha^3 Z^2$ a.u. the leading contribution to the transition rate (2.9) will be of order $\alpha^9 Z^{10}$ a.u. and it is thus rapidly growing with Z as is well known.⁷

It may be noted, as clearly seen in (2.13), that the $M1$ operator is defined in units of the magnetic dipole moment divided by c . This is a convenient definition which gives the familiar form of the transition rate (2.9), i.e., the same form of the transition rate as for an electric dipole transition, albeit with the electric dipole operator replaced by \mathbf{M}^1 .

III. RELATIVISTIC PAIR FUNCTION

Recently, Salomonson and Öster have shown how relativistic all-order pair functions can be obtained by summation over a complete set of eigenvectors to a discretized single particle Dirac Hamiltonian,²² e.g., the Hamiltonian for a hydrogenlike system,

$$h_i^D = c\alpha_i \cdot \mathbf{p}_i + \beta m_i c^2 - \frac{e}{4\pi\epsilon_0} \frac{eZ}{r_i}. \quad (3.1)$$

The pair functions should describe the interactions among the electrons. It is well known that the extension of the Dirac equation to the many-electron atom requires some special considerations.^{31,32} The direct addition of the Coulomb

$$\sum_{i < j} \frac{e^2}{4\pi\epsilon_0} \frac{1}{r_{ij}} \quad (3.2)$$

and/or the Breit interaction

$$-\frac{1}{2} \frac{e^2}{4\pi\epsilon_0} \sum_{i < j} \left[\frac{\alpha_i \cdot \alpha_j}{r_{ij}} + \frac{(\alpha_i \cdot \mathbf{r}_{ij})(\alpha_j \cdot \mathbf{r}_{ij})}{r_{ij}^3} \right] \quad (3.3)$$

(given above in the limit of small energy transfer between the electrons) to the Dirac Hamiltonian is obviously wrong since such a Hamiltonian would allow bound states to autoionize, emitting one electron into the positive-energy continuum and another electron into the negative-energy continuum. In order to respect that the negative-energy states are not available for the electrons, unless a hole has been created by a preceding production of an electron-positron pair, the positive- and negative-energy continua have to be treated differently. Formally this is done by surrounding the two-particle operators with projection operators. When working with a discrete basis set, as in the present work, such projection operators are easily obtained as restrictions in the summation over excited states. If the two-particle operators are surrounded by projection operators onto positive-energy states, that implies a neglect of the effects caused by creation and subsequently annihilation of virtual electron-positron pairs. Such effects contribute beyond $\alpha^2 Ry$ to the energy. If this no-virtual-pair approximation is used, the Hamiltonian for a many-electron system in the absence of the photon vector potential can be written

$$H = \sum_i c\alpha_i \cdot \mathbf{p}_i + \beta m_i c^2 - \frac{e}{4\pi\epsilon_0} \frac{eZ}{r_i} + \sum_{i < j} \Lambda_{ij}^{++} V_{ij} \Lambda_{ij}^{++}, \quad (3.4)$$

where V_{ij} , when working in the Coulomb gauge and neglecting terms in the Hamiltonian which contributes beyond order $\alpha^2 Ry$ to the eigenvalue, is given by the sum of (3.2) and (3.3) above. The projection operators Λ^{++} give unity when working on products of positive-energy solutions to (3.1) and zero when working on products involving at least one negative-energy solution.

The eigenstates to H for a heliumlike system can be written as

$$\Psi = \{ \Phi_a \Phi_b + \rho_{ab}(1,2) \}, \quad (3.5)$$

where the curly brackets denote antisymmetrization and Φ is an eigensolution to the one-particle Hamiltonian h^D . If we choose to work with nonantisymmetrized pair functions, as was done in Ref. 22, the pair function ρ can be written

$$\rho_{ab} = \sum_{rs \neq ab, ba} \frac{|rs\rangle \langle rs | \Lambda_{ij}^{++} V_{ij} \Lambda_{ij}^{++} | ab + \rho_{ab} \rangle}{\epsilon_a + \epsilon_b - \epsilon_r - \epsilon_s} - \sum_{cd = ab, ba} \frac{|\rho_{cd}\rangle \langle cd | \Lambda_{ij}^{++} V_{ij} \Lambda_{ij}^{++} | ab + \rho_{ab} \rangle}{\epsilon_a + \epsilon_b - \epsilon_r - \epsilon_s}. \quad (3.6)$$

A first approximation of ρ is obtained if the pair function is neglected on the right-hand side of (3.6). An iterative scheme can then be set up to obtain the full pair function as described in Ref. 22.

In order to calculate the transition rate (2.11) it is necessary both to know the transition energy to the desired accuracy and to calculate the transition matrix element. The contributions to the energy which are calculated in this work are treated within the no-virtual-pair approximation. This is discussed in Sec. IV B. For the transition matrix element, however, a certain class of virtual-pair contributions have been included. The $M1$ operator is a one-particle operator and it is thus possible to include it in the single-particle Hamiltonian. This corresponds to a redefinition of the meaning of positive- and negative-energy states. Positive-energy eigenstates to the Hamiltonian

$$h_i = h_i^D + ec\alpha_i \cdot \mathbf{A}_i \quad (3.7)$$

include effects of the $M1$ operator, which corresponds to the creation and annihilation of electron-positron pairs when described from the point of view of Hamiltonian (3.1). Since the $M1$ -matrix element is off-diagonal, i.e., it mixes small and large components in the wave function, these effects will contribute in order $\alpha^3 Z$, i.e., in leading order in the fine-structure constant α . The calculation of the transition matrix element is discussed in Sec. IV A.

IV. CALCULATION OF THE MAGNETIC DIPOLE TRANSITION RATE

A. Magnetic dipole matrix element

The lowest-order contribution to the magnetic dipole matrix element (2.10) is given in Fig. 1 and is of order $\alpha^3 Z^2$ a.u. as mentioned in Sec. I. In Table I the contribution from this diagram is given using hydrogenlike wave functions; the result is 2.296198×10^{-5} a.u. Through the spherical Bessel function $j_1(\omega r/c)$ in (2.10), the matrix element depends on the energy for the transition in question. We have here used $\omega = 114.072$ as given in Table III and discussed further in Sec. IV B. For comparison, the value obtained with hydrogenlike energies is 2.160926×10^{-5} a.u.

In Fig. 2 diagrams containing correlation in the initial, (a)–(d), or final, (e)–(h), states are shown. The two-

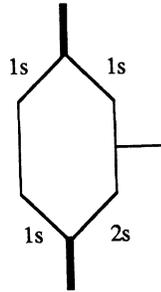


FIG. 1. Lowest-order contribution to the $M1$ -matrix element. There is also a corresponding diagram with the $M1$ interaction in index 1, where particle one and two in the initial state have been exchanged.

particle interaction can be either the Coulomb or the Breit interaction. The diagrams in Fig. 2 that contain positive-energy solutions to h_D only contribute in order $\alpha^3 Z$ for the Coulomb interaction, (a) and (e) but for the

Breit interaction, (c) and (g), there will be an additional factor of $\alpha^2 Z^2$ since the Breit interaction mixes large and small components of the wave function. For comparison, these diagrams are anyway included in Table I. For the diagrams in Fig. 2 which contain a sum over negative energy states, i.e., (b), (d), (f), and (h), in one index the situation is quite different. The $M1$ operator is approximately proportional to the matrix element of $\alpha \mathbf{x} \mathbf{r}$, as can be seen in (2.12). Since the Dirac operator α gives unity between positive- and negative-energy states the matrix element will be governed by the integral over \mathbf{r} which is of the order $1/Z$. The Breit interaction gives a factor of αZ in each index when evaluated between positive-energy states, but is of the order unity when evaluated between one positive- and one negative-energy state. The opposite is true for the Coulomb interaction. In addition, the radial integral over $1/r_{12}$ gives a factor of Z in either case. Finally, the creation of a virtual electron-positron pair gives an energy denominator of order $2mc^2$. In both

TABLE I. Contributions to the $M1$ -matrix element from Figs. 1, 2, 3, and 4.

		$M1$ -matrix element (a.u.)	Transition rate (sec^{-1})
Hydrogenlike wave functions, ^a energies ^b from Table III	Fig. 1	$+2.296\ 198 \times 10^{-5}$	$5.587\ 888 \times 10^6$
“Second order” (pure second order is given in parentheses) ^c			
Coulomb	Fig. 2(a), $ i_+\rangle$ Fig. 2(e), $ i_+\rangle$ Fig. 2(b), $ i_-\rangle$ Fig. 2(f), $ i_-\rangle$	$-0.1213 (-0.1227) \times 10^{-5}$ $-0.0475 (-0.0448) \times 10^{-5}$ $-0.0112 (-0.0118) \times 10^{-5}$ $-0.0126 (-0.0123) \times 10^{-5}$	
Breit	Fig. 2(c), $ i_+\rangle$, $l_i=0$ Fig. 2(c), $ i_+\rangle$, $l_i=2$ Fig. 2(g), $ i_+\rangle$ Fig. 2(d), $ i_-\rangle$, $l_i=0$ Fig. 2(d), $ i_-\rangle$, $l_i=2$ Fig. 2(h), $ i_-\rangle$	$< 1.10^{-11}$ $< 1.10^{-9}$ -0.0005×10^{-5} -0.0078×10^{-5} $+0.0078 \times 10^{-5}$ $+0.0238 (+0.0238) \times 10^{-5}$	
“Third order”			
Coulomb	Fig. 3(a) Fig. 3(b)	-0.0018×10^{-5} $< 1 \times 10^{-9}$	
Breit	Fig. 3(c) Fig. 4(b) Fig. 4(f)	$+0.0007 \times 10^{-5}$ $+0.0004 \times 10^{-5}$ -0.0012×10^{-5}	
Normalization contribution ^d		-0.0025×10^{-5}	
Sum (numerical error is given in parentheses)		$2.123(1) \times 10^{-5}$	$4.775(5) \times 10^6$
Neglected effects ^e		(1)	(5)

^aThis value is calculated using analytical wave functions obtained in a point nucleus potential. The use of the potential from an extended nucleus would reduce the value with less than 0.001%.

^bThe $M1$ -matrix element itself depends on the transfer energy and the use of hydrogenlike (Dirac) energies, $\omega = 121.9845$ a.u., gives a $M1$ -matrix element of $2.160\ 926 \times 10^{-5}$ a.u. and a transition rate of $6.051\ 890 \times 10^6$ sec^{-1} .

^c $|i\rangle$ denotes the intermediate line in Fig. 2. In Fig. 2 lines going up represent positive-energy states $|i_+\rangle$ and lines going down negative-energy states $|i_-\rangle$.

^dIt is often convenient in perturbation theory to use intermediate normalization, i.e., the initial one-particle description of the two-particle wave function is normalized, leaving the total wave function unnormalized. The total result has then to be divided with the norm, in this case $\langle \Psi_{\text{initial}} | \Psi_{\text{initial}} \rangle^{1/2} \langle \Psi_{\text{final}} | \Psi_{\text{final}} \rangle^{1/2}$.

^eThe most important of the neglected effects are probably due to retardation beyond the Breit interaction and to radiative effects as discussed in Sec. IV A 1.

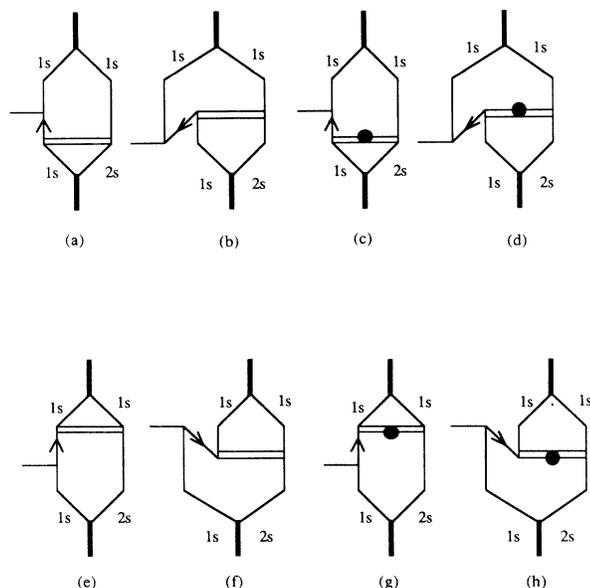


FIG. 2. These contributions to the $M1$ -matrix element enter in second and higher orders. A line with a dot represents the Breit interaction. The double lines indicate that the Coulomb interaction has been iterated to all orders. The Breit interaction has been kept to lowest order, but it has been allowed to mix with the Coulomb interaction to all orders. It is, however, only when the Breit interaction is evaluated between one negative- and one positive-energy state that it is contributing in leading order as discussed in Sec. IV A. Lines going down represent negative-energy states. The addition of exchange diagrams as well as diagrams with the $M1$ interaction in index 2 is assumed.

cases we thus get a contribution $\alpha^3 Z$ a.u. In Table I it can be seen that these entries (e.g., the third, fourth, and tenth lines) are indeed of the same order of magnitude and, in addition, there is a detailed cancellation between these terms. The contribution for the Breit interaction including positive-energy intermediate states only (e.g., the fifth, sixth, and seventh lines) is one order of $\alpha^2 Z^2$ smaller. The largest contributions from Fig. 2 comes naturally from the Coulomb interaction including positive-energy solutions (a) and (e), since these diagrams account for the screening of the nuclear charge by each electron as seen by the other.

One may question the use of the frequency independent form of the Breit interaction (3.3) between positive- and negative-energy solutions to h_D (3.1), as in Figs. 2(d) and 2(h), since the approximation (3.3) is valid only in the limit of small energy transfer between the electronic states. However, the intermediate states in Figs. 2(d) and 2(h), although negative-energy solutions to the Hamiltonian h_D , are actually part of a positive-energy solution to Hamiltonian (3.7).

The double lines in Fig. 2 indicate that the Coulomb interaction has been iterated to all orders where the contributions in each iteration decrease as $1/Z$. The Breit interaction has been kept to lowest order, but it has been allowed to mix with the Coulomb interaction to all or-

ders. In Table I the results from Fig. 2 are shown. The contributions obtained in pure second order, i.e., with the two-particle interaction in Fig. 2 representing one interaction only, has been separated out for comparison. For the Coulomb interaction only intermediate s states contribute in pure second order. For the Breit interaction, however, even $d_{3/2}$ states will contribute to the correlation in the 2^3S_1 state. However, to order $\alpha^3 Z$ a.u. the Breit contribution in the 2^3S_1 state vanishes. This is expected since for S states in the nonrelativistic limit the Breit interaction is proportional to a δ function $\delta(r_{12})$ when evaluating between product functions. The triplet state has no charge density at $r_1=r_2$ and a matrix element of $\delta(r_{12})$ will then vanish.

The diagrams in Figs. 3 and 4 contribute in third and higher orders. The diagram 3(a), which includes positive-energy states only, contributes in order α^3 a.u. for the Coulomb interaction. The corresponding diagrams for the Breit interaction 4(a) and 4(d) is one factor of $\alpha^2 Z^2$ smaller and has not been considered here. Even the diagrams which include one negative-energy state, 3(b) and 3(c), contribute in order α^3 a.u. for the Coulomb interaction. The corresponding Breit interaction diagrams contribute also in order α^3 a.u. when the Breit interaction is evaluated between one positive- and one negative-energy state, as in diagrams 4(b) and 4(f). This can be understood in the same way as for the corresponding diagrams in Fig. 2. Diagrams 4(c) and 4(e) are a factor of $\alpha^2 Z^2$ smaller and are thus neglected.

The result for the lowest-order diagram, Fig. 1, is obtained with analytical wave functions. The contributions from Figs. 2, 3, and 4 are obtained with pair functions in two grid sizes, 91 and 121 points in each direction. The results have then been extrapolated. The partial wave expansions includes up to d excitations. As a test a calculation including up to h excitations has also been performed for the smaller number of grid points. The difference cannot be seen in the number of figures quoted here.

Neglected effects

Table I includes all contributions to the $M1$ -matrix element of order α^3 a.u. The combination of radiative effects and the $M1$ interaction will give contributions of order $\alpha^4(\ln\alpha)Z^2$ a.u.,³³ but all such terms can be shown

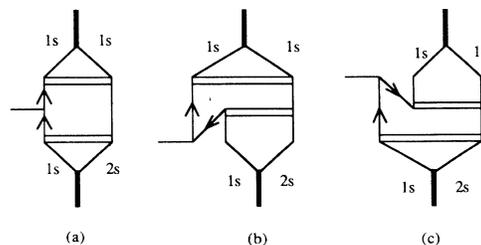


FIG. 3. Contributions to the $M1$ -matrix element which enter in third and higher orders. Lines going down represent negative-energy states. The addition of exchange diagrams as well as diagrams with the $M1$ interaction in index 2 is assumed.

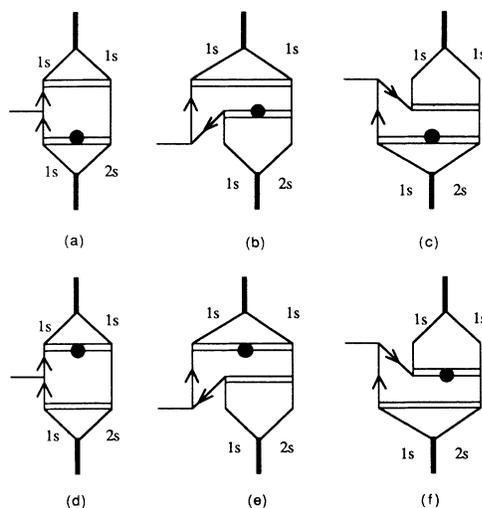


FIG. 4. Contributions to the $M1$ -matrix element, involving the Breit interaction, which enter in third and higher orders. The Breit interaction is represented by the line with the dot. Lines going down represent negative-energy states. The addition of exchange diagrams as well as diagrams with the $M1$ interaction in index 2 is assumed.

to cancel.^{34,35} Radiative effects also give rise to contributions of order $\alpha^4 Z^2$ a.u. as do contributions from retardation effects beyond what is included in the Breit interaction, such terms are illustrated in Fig. 5(a) and 5(b), respectively. As reviewed by Sucher,¹⁹ even all terms of this order cancel. Remaining neglected terms, which arise from radiative effects and from virtual-pair corrections to the electron-electron interaction, are of order $\alpha^4 Z$ a.u. Since this is α/Z relative to the leading contributions, we estimate the neglected contributions to the $M1$ -matrix element to 0.001×10^{-5} a.u.

B. Transition energy $1^1S_0-2^3S_1$

The result for the transition energy is summarized in Tables II and III. The one-particle energies, which are given in Table II, are described by the Dirac energy, the reduced mass correction, and the Lamb shift as given by Johnson and Soff.³⁶ In Table III the two-particle contributions to the energy in heliumlike argon are added. The correlated Coulomb and Breit interactions are obtained by using pair functions to all orders as discussed in Sec.

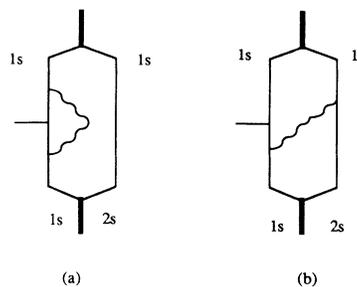


FIG. 5. Examples of contributions to the $M1$ -matrix element which are not included in the calculation. (a) illustrates the contributions from the self-energy; (b) shows a contribution from retardation beyond what is concluded in the Breit interaction. As in Figs. 2–4 there are also corresponding diagrams with intermediate lines going down, representing negative-energy states.

III. The Breit interaction itself is treated in lowest order, but it is permitted to mix in all possible ways with the Coulomb interaction, Fig. 6. The Breit interaction is very small in the 2^3S_1 state. This is since the nonrelativistic limit of the Breit interaction vanishes for a triplet S state when it is described by product functions as discussed in Sec. IV A. Both the Coulomb and the Breit interactions are treated in the no-virtual-pair approximation. Contributions that include one or several virtual electron-positron pairs will enter in order $\alpha^3 Z^3$ Ry. Compared with the corresponding no-virtual-pair contributions, given in Table III on the fourth and fifth lines, this is in relative order $\alpha^3 Z^3$ Ry for the Coulomb and αZ Ry for the Breit interaction. Higher-order retardation is also contributing in order $\alpha^3 Z^3$ Ry. The errors given in Table III for the higher-order two-particle contributions are mainly due to these two sources. The main total error in Table III comes, however, from the neglect of the screening of the self-energy. This effect was considered by Indelicato, Gorceix, and Desclaux³⁷ in a calculation of the transfer energy of several of the allowed transitions among the $n = 1$ and 2 shells in heliumlike systems. They included the screening of the self-energy with an approximate method and a comparison with their results indicates that 10% of the one-particle self-energy is a likely contribution for heliumlike argon.

The transition rate (2.11) depends on the cube of the transition energy. In order to assure accuracy of 0.1%,

TABLE II. Energies for the 1s and 2s states of hydrogenlike argon ($Z = 18$) given in atomic units. The error in the last digit is given in parentheses.

	1s	2s	ΔE
Dirac energy	-162.7049	-40.7204	121.9845
Reduced mass correction	0.0022	0.0006	
Lamb shift ^a	0.0419(0.2)	0.0057(0.03)	
Sum	-162.6607(0.2)	-40.7142(0.03)	121.9465(0.2)

^aJohnson and Soff (Ref. 36) include self-energy (0.0447 a.u. for the 1s state and 0.0060 a.u. for the 2s state) and other effects such as vacuum polarization, field shift, etc.

TABLE III. 1^1S_0 and 2^3S_1 energies of heliumlike argon, given in atomic units. The error in the last digit is given in parentheses.

	1^1S_0	2^3S_1	ΔE
One-particle energies (See Table II)	-325.3214(0.4)	-203.3749(0.2)	121.9465(0.2)
First-order Coulomb interaction	11.3223	3.4068	-7.9155
First-order Breit interaction	0.0780	$< 1 \times 10^{-5}$	-0.0780
Higher-order Coulomb interaction ^a	-0.1584(3)	-0.0484(1)	0.1100(4)
Higher-order Breit interaction ^b	-0.0085(10)	$< 1 \times 10^{-4}$	0.0085(10)
Screening of the self-energy ^c	(40)	(5)	(45)
Mass polarization ^d	(2)	(1)	(1)
Sum ^c	-314.0880(60)	-200.0165(7)	114.072(7)

^aThe errors given are mainly due to the neglected effects of virtual electron-positron pairs. Such effects enters in relative order $\alpha^3 Z^3$ Ry. The numerical errors are estimated to one unit in the last digit for 1^1S_0 and less than that for 2^3S_1 . These errors may also easily be reduced by a more detailed study of the effect of the numerical grid size as was done in Ref. 22.

^bThe calculation giving higher-order Breit interaction, i.e., the mixture of one order of the Breit interaction itself and all orders in the Coulomb interaction has been performed inside the no-virtual-pair approximation and the error given is a rough estimate of the possible contributions from one or several intermediate virtual pairs. The numerical accuracy for the no-virtual-pair calculation is discussed in Sec. V.

^cThe main error given in the final numbers are due to the screening of the self-energy which is supposed to contribute with approximately 10% of the one-particle self-energy (Ref. 37).

^dFor s states the mass polarization does not contribute in lowest order. Therefore it is rather small and contributes approximately with 10% of the reduced mass contribution (see Table II).

the transition energy has then to be known within three units of the fifth figure. The results in Table III are well below that.

The mixture of the Breit and Coulomb interactions has been treated to lowest order in helium and lithium by Johnson and co-workers.^{20,38} The magnetic part of the Breit interaction was calculated for a few heliumlike systems to that order also in Ref. 21. In that work s excitations were also treated to all orders. Gorceix, Indelicato, and Desclaux,³⁹ have performed a multiconfigurational Dirac-Fock (MCDF) calculation, including some effects beyond lowest order, for the mixture of the magnetic and the electrostatic correlation. However, no complete relativistic all-order calculation has been presented in the literature for the Breit interaction. Since the Breit interaction is interesting in itself, we present in Sec. V a somewhat more detailed calculation of this interaction in the ground state than what is actually needed to predict the transition energy for the magnetic dipole transition.

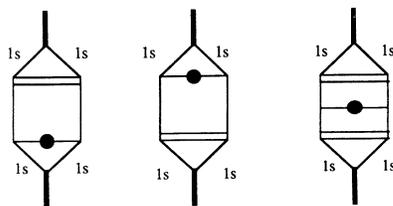


FIG. 6. Contributions to the energy of the ground state of helium-like systems caused by the Breit interaction in the no-virtual-pair approximation. The double line indicates that the Coulomb interaction has been iterated to all others.

V. BREIT INTERACTION

The effect of the exchange of one transverse virtual photon between the electrons can, when treated in Coulomb gauge and in the limit of small energy transfer, be shown to lead to the Breit interaction as given in (3.3). A convenient formula for evaluating (3.3) has been given, e.g., by Grant and Pyper.⁴⁰ When hydrogenlike wave functions are used, the expectation value of this operator gives a contribution of order $\alpha^2 Z^3$ Ry. In second and higher order (3.3) can mix with the Coulomb interaction and, provided it is evaluated between positive states only, it then gives correctly all contributions to order α^2 Ry. If effects beyond that, as effects corresponding to creation and annihilation of virtual electron-positron pairs, are to be considered, it is no longer sufficient to treat the exchange of virtual photons in the limit of small energy transfer. The interaction to use in that case has been given by Brown⁴¹ and Mittlemann.⁴²

An alternative form of the Breit interaction is obtained if Feynman gauge is used. The two alternative forms can be shown to give identical results when the expectation value is evaluated between single-particle states obtained in a local potential. However, when treated together with the Coulomb interaction, the agreement between the two forms is not as easily obtained.^{43,44} As shown by Ref. 45, crossed photons have to be considered in Feynman gauge in order to take all effects of order α^2 Ry into account. In Table IV the contributions to the Breit interaction in the ground state of heliumlike argon, when treated in Coulomb gauge and in the no-virtual-pair approximation, are given. The final result includes all effects to order α^2 Ry. In addition, many effects of order $\alpha^3 Z^3$ and higher are included. The result then show a deviation from the value by Pekeris,⁴⁶ which includes α^2 effects

TABLE IV. All-order Breit interaction obtained in the no-virtual-pair approximation (Fig. 6) for the ground state of heliumlike argon given in a.u. Errors are given in parentheses.

$l s^2 \rightarrow$	Breit interaction (a.u.)
s^2	-0.005 076
p^2	-0.002 415
d^2	-0.000 498
f^2	-0.000 202
g^2	-0.000 104
h^2	-0.000 060
i^2	-0.000 037
j^2	-0.000 026
k^2	-0.000 018
l^2	-0.000 013
m^2	-0.000 010
Sum ^a	-0.008 46(2)
Extrapolated	-0.008 51(3)
Nonrelativistic limit ^b	-0.009 33

^aThe errors given are numerical and from the extrapolation of the partial wave expansion.

^bPekeris (Ref. 46) has calculated the Breit interaction in the Pauli approximation for $Z=1-10$. These results can be fitted to a Z expansion formula from which the nonrelativistic result can be obtained for any Z . The difference between the relativistic and nonrelativistic results are of relative order $\alpha Z Ry$ as expected.

only. (Recently, even more accurate results have been published by Drake;⁴⁷ however, the Breit interaction alone is not deducible from the values given.)

In Table IV the contributions to the Breit interaction for each term in the partial-wave expansion is shown. In

their accurate study of the ground state of helium Salomonson and Öster²² found that the relativistic shifts in the Coulomb interaction converged as $(l_{\max} + 0.5)^{-2}$. This is considerably slower than the nonrelativistic convergence $(l_{\max} + 0.5)^{-4}$.⁴⁸ The relativistic convergence rate has also been considered by Kutzelnigg.⁴⁹ However, the pure $(l_{\max} + 0.5)^{-2}$ behavior is only obtained in the nonrelativistic limit. Already for rather modest nuclear charges, as $Z=18$ as in this work, the convergence rate is significantly faster. This is true both for the Coulomb and the Breit interactions. The tail of the partial-wave expansion is, however, much more affected by relativistic effects than the total energy. This seems to be true not only for the relativistic shift in the nonrelativistic limit, as seen by the much slower convergence for the relativistic than the nonrelativistic contributions, but for higher relativistic corrections as well. The origin of this behavior is likely to be the singular behavior of the relativistic contributions, which is seen as a δ function, $\delta(r_{12})$, in the nonrelativistic limit.

The numbers in Table IV are obtained in one single grid size and the numerical errors given are estimated effects of a grid extrapolation. Complete calculations in a few grid sizes and a following grid extrapolation would reduce the errors significantly, see Ref. 22. The uncertainty in the extrapolation in the partial-wave expansion is harder to overcome before a more complete understanding of the convergence pattern is obtained.

VI. CONCLUSIONS AND COMPARISON WITH OTHER CALCULATIONS

The $M1$ transition $2^3S_1-1^1S_0$ in heliumlike argon has been calculated with an estimated accuracy better than 1%. The 2^3S state can also decay by two-photon emis-

TABLE V. Comparison of different theoretical estimates of the lifetime of the 2^3S_1 state of heliumlike argon.

Reference	Lifetime (nsec)	Numerical error (if given)	Estimation of neglected effects	Transition energy (a.u.) (if given)
Drake (Ref. 7) ^a	212.4			113.7422
Drake with corrections from Lin (Refs. 18 and 19) ^b	207.5			114.223
Beigman and Safronova (Ref. 8) ^c	209			114.3
Johnson and co-workers (Refs. 10 and 11) ^d	208			
Anderson and Weinhold (Ref. 12) ^a	212.4	± 0.5		
Krause (Ref. 13) ^e	209.3			
This work	209.4	± 0.2	± 0.2	114.072(7)

^aObtained in the nonrelativistic limit, the nonrelativistic transition energy is used.

^bThe most important terms which are missing in Ref. 7 are higher-order relativistic effects of relative order $\alpha^2 Z^2$. These terms have been calculated explicitly by Lin (Refs. 18 and 19) and are found to be $1.07 \alpha^2 Z^2$ relative to the *hydrogenlike* result. Of this correction $0.69 \alpha^2 Z^2$ is due to the leading relativistic corrections, i.e., $\frac{11}{128} \alpha^2 Z^4$ a.u., to the hydrogenlike transition energy. The transition energy given in the last column is obtained by adding this correction to the nonrelativistic transition energy given by Drake (Ref. 7).

^cObtained in the nonrelativistic limit, albeit with a transition energy of 114.3 a.u. This transition energy is not explicitly given in Ref. 8, but can be deduced from given results. Note, however, that the screening corrections to the transition matrix element are added with the wrong sign in Ref. 8, which has been corrected here. Screening is included to lowest order only.

^dIncludes screening but not correlation.

^eIncludes screening but not correlation. The effect of the Breit interaction on the $M1$ -matrix element is not considered.

sion. The transition rate for that process has been studied by Drake, Victor, and Dalgarno⁵⁰ and it is expected to be three to four orders of magnitude less important than the $M1$ transition. Therefore the inverse of the $M1$ -transition rate should give the lifetime of the 2^3S state, i.e., 209.4 ± 0.4 nsec. Half of the error is due to effects which have been neglected in the calculation as discussed in Sec. IV A 1. It may be noted that these effects have not been considered in other works⁷⁻¹³ either and when comparing the present result with previous calculations, Table V, the numerical error (0.2) should be considered.

When comparing different theoretical results it is convenient to consider the following summary of the sizes of different contributions to the $M1$ -matrix element discussed in Sec. IV:

$$M1 \propto \alpha^3 Z^2 \left[1 + \alpha^2 Z^2 + \dots + \frac{1}{Z} + \alpha^2 Z + \dots + \frac{1}{Z^2} + \frac{\alpha}{Z} + \dots + \frac{\alpha}{Z} + \dots \right]. \quad (6.1)$$

The first groups of terms give the contributions from Figs. 1 and 2, and 3 and 4, respectively. The first term in every group is the leading contribution from each diagram as discussed in Sec. IV A. With the method used by Drake⁷ and by Anderson and Weinhold¹² only this first term in the three first groups of terms is included. The following terms are higher-order relativistic corrections from the same diagrams, which are all included in this calculation due to the use of relativistic wave functions. The second term in the first group of terms has been calculated explicitly by Lin^{18,19} and can be added to the results in the nonrelativistic limit^{7,12} as is shown in Table V. In the works by Johnson and co-workers^{10,11} correlation is omitted. Thus many contributions from the second and third groups are missing. Some terms of this size are, however, included by means of screening. The same applies to the work by Krause.¹³ In Ref. 13 all terms which arise from the Breit interaction are missing. Such terms would enter in group two and onwards. The term on the fourth line indicates the order of magnitude of the neglected contributions from Fig. 5 as discussed in Sec. IV A 1. Such contributions are neglected in all the calculations presented in Table V.

It may be noted that formula (6.1) applies strictly only if the nonrelativistic hydrogenlike transition energy is used to evaluate the Bessel function in the $M1$ operator (2.10). When the screened energy is used additional factors of decreasing powers of Z will be introduced.

The lifetime 209.4 nsec obtained in the present work is 1.4% shorter than the result 212.4 nsec given by Drake⁷ and by Anderson and Weinhold.¹² The main difference is that the calculations in Refs. 7 and 12 are obtained in the nonrelativistic limit. Approximately half of the difference can be attributed to the transition energy. Drake used nonrelativistic energies while the present cal-

ulation is made with the transition energy including all relativistic effects to order α^2 Ry and the dominating effects of order α^3 Ry. The remaining difference is probably due to relativistic contributions beyond lowest order which are included when relativistic wave functions are used to evaluate the matrix element, see (6.1). When Drake's results are corrected with the term given by Lin,^{18,19} i.e., $1.07 \alpha^2 Z^2$ relative to the hydrogenlike result in the nonrelativistic limit, the result 207.5 nsec is obtained. This result is nearly 1% shorter than the present prediction. Half of this difference may again be directly attributed to the energy. The addition of the contributions of relative order $\alpha^2 Z^2$ to the transition energy gives a result 114.223 a.u., which is slightly larger than the present value 114.072(7) a.u. The difference, which is mainly due to the inclusion of the Breit interaction and the Lamb shift in the present work, will give rise to an increase in the lifetime of around 0.5%. It is not unlikely that the differences in the transition matrix element is of the same size. That the present calculation really agrees with the calculation of Lin concerning the terms of relative order $\alpha^2 Z^2$ can be seen by comparing the relativistic hydrogenlike result for the transition matrix element, 2.1609×10^{-5} a.u. given in Table I, with the result in the nonrelativistic limit which can be deduced from Drake, Eq. (14) in Ref. 7, 2.1538×10^{-5} a.u. This gives a relative correction to the *transition rate*, see Eq. (2.11), of $0.38 \alpha^2 Z^2$, which then should be added to the correction of the same relative order arising from the transition energy $0.69 \alpha^2 Z^2$; see also Table V. The total correction of this order obtained here is then $1.07 \alpha^2 Z^2$, i.e., in excellent agreement with Lin. The difference compared to the work by Johnson *et al.*,^{10,11} 208 nsec, is less than 1%. Correlation is neglected in Refs. 10 and 11, but it is harder to compare these studies with this work since only the final results are given in Refs. 10 and 11. The close agreement between the work by Krause,¹³ 209.3, and this work is probably a coincidence since the calculation¹³ neglects the effect on the $M1$ -matrix element of the Breit interaction which, as seen in Table I, change the result with 2%. Correlation is also neglected in Ref. 13. The calculation by Biegman and Safronova⁸ is performed in the nonrelativistic limit. The transition energy includes, however, some relativistic corrections. This, and the fact that the screening is included to lowest order only in Ref. 8, explains the deviation from the results by Drake.⁷ Note that the result quoted in Ref. 8 is obtained when the screening corrections to the transition matrix element is added with the wrong sign. This has been corrected in Table V.

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