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Radiative Decay Rates of Metastable One-Electron Atoms*

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Decay rates of the $2s_{1/2}$ metastable states of the hydrogen isoelectronic sequence are determined to lowest order in the fine-structure constant α, and to all orders of αZ, for elements with atomic numbers Z ranging from 1 to 92. The $M1$ decay rate is given in closed analytical form, whereas the $E1$ rate is determined numerically. The two-photon $M1$ decay rate is considered and found to be of negligible importance throughout the periodic table.

There has been renewed interest during the past few years in accurate theoretical calculations of the decay rates of metastable states of the hydrogen and helium isoelectronic sequences, especially for large values of the atomic number Z.[1] Such interest arises partly because of the possibility of measuring the decay rates accurately,[2] and partly because of the importance of direct decay as competition to induced decay in the metastable quenching measurements of the Lamb shift.[3]

The $2s_{1/2}$ metastable state in the hydrogen sequence decays by two competing processes; the emission of two $E1$ photons, and the emission of a single $M1$ photon. Both of these rates were estimated by Breit and Teller,[4] who showed that the two-$E1$ decay mode is highly favored for hydrogen. Subsequently, Spitzer and Greenstein[5] and Shapiro and Breit[6] evaluated the two-$E1$ rate nonrelativistically. Precise closed-form calculations of the two-$E1$ rate have been done in later years[7] which confirm the results of the older papers. The $M1$ rate vanishes in the nonrelativistic limit as pointed out by Breit and Teller[4]; however, it is still possible to extract a lowest nonvanishing approximation in αZ from the relativistic $M1$ matrix element, and to give a simple nonrelativistic expression for the $M1$ rate. Such a formula is quoted by Marrus and Schmieder.[2] For heavy elements we expect corrections of order $(\alpha Z)^2$ to these nonrelativistic results.

The theory of both one- and two-photon decays to lowest order in α is well understood, and is summarized in texts on quantum electrodynamics.[8] Because of the complexity of the general expressions, especially for two-photon decay, there has been no attempt to evaluate the corresponding matrix elements beyond the lowest order in αZ. It is the purpose of the present Letter to give detailed numerical results for one- and two-photon decays, which avoid any expansion in αZ, and thus are correct to lowest order in α for all $Z < 137$.

M1 decay.—The transition rate for $M1$ decay is[9]

$$w(M1) = \tfrac{4}{9}\alpha\omega^3|M(\omega)|^2, \qquad (1)$$

where ω is the photon energy, and where the matrix element for $M1$ decay is given by

$$M(\omega) = (3/\omega)\int_0^\infty dr\, j_1(\omega r)(G_2F_1 + G_1F_2). \qquad (2)$$

In Eq. (2) $j_1(\omega r)$ is a spherical Bessel function of order 1, and $G(r)$ and $F(r)$ are the large- and small-component radial Dirac functions. The subscripts 1 and 2 refer to the $2s_{1/2}$ and $1s_{1/2}$ states, respectively. The $M1$ matrix element can be evaluated in closed analytical form as

$$M(\omega)=-\frac{(\alpha Z)^2}{m}\left(\frac{2(N-1)}{N+2}\right)^{1/2}\left(\frac{2}{N+1}\right)^{2\gamma+1}N^{\gamma-1}[{}_2F_1(\gamma+\tfrac{1}{2},\gamma+1,\tfrac{5}{2},-\eta^2)+\tfrac{2}{5}N\eta^2\,{}_2F_1(\gamma+\tfrac{3}{2},\gamma+2;\tfrac{7}{2},-\eta^2)], \quad (3)$$

with

$$\eta=N\omega/m\alpha Z(N+1), \quad N=[2(1+\gamma)]^{1/2}, \quad \gamma=[1-(\alpha Z)^2]^{1/2}.$$

In the nonrelativistic limit, $N\sim 2$, $\gamma\sim 1$, and $\eta\sim 0$, so that the $M1$ decay rate reduces (in terms of the Rydberg of frequency Ry) to

$$w_{\rm NR}(M1)=\tfrac{1}{35}\pi\alpha^9 Z^{10}\,{\rm Ry}=2.496\times 10^{-6}Z^{10}\ {\rm sec}^{-1}, \quad (4)$$

which is the nonrelativistic rate quoted in Ref. 2. Evaluating the decay rate using Eq. (3), we obtain the results presented in column 2 of Table I, where we list $10^6 Z^{-10} w(M1)$. It can be seen that the fully relativistic calculation increases the rate by a factor of about 2 for $Z=92$ over the rate determined by Eq. (4), while for $Z=1$ the lowest-order calculation is completely satisfactory.

TABLE I. Decay rates in $\sec^{-1}$ for the elements.

Z	$10^6 \times Z^{-10}$ w(M1)	Z^{-6} w(2E1)	$w_{\rm Tot}$ ($\sec^{-1}$)[a]
1	2.4959	8.2290	8.2245
2	2.4964	8.2278	5.2651 (2)
3	2.4971	8.2260	5.9963 (3)
4	2.4981	8.2234	3.3684 (4)
5	2.4993	8.2200	1.2845 (5)
6	2.5009	8.2159	3.8345 (5)
7	2.5028	8.2111	9.6670 (5)
8	2.5049	8.2055	2.1536 (6)
9	2.5073	8.1992	4.3660 (6)
10	2.5100	8.1922	8.2171 (6)
12	2.5164	8.1759	2.4568 (7)
14	2.5239	8.1568	6.2146 (7)
16	2.5326	8.1348	1.3926 (8)
18	2.5425	8.1100	2.8492 (8)
20	2.5537	8.0825	5.4342 (8)
22	2.5661	8.0522	9.8111 (8)
24	2.5798	8.0192	1.6961 (9)
26	2.5949	7.9837	2.8326 (9)
28	2.6114	7.9455	4.6024 (9)
30	2.6292	7.9049	7.3151 (9)
34	2.6694	7.8162	1.7586 (10)
38	2.7158	7.7181	4.0289 (10)
42	2.7690	7.6110	8.9071 (10)
46	2.8296	7.4953	1.9104 (11)
50	2.8983	7.3713	3.9821 (11)
54	2.9759	7.2395	8.0691 (11)
58	3.0635	7.1000	1.5901 (12)
62	3.1623	6.9531	3.0490 (12)
66	3.2738	6.7991	5.6965 (12)
70	3.3999	6.6381	1.0385 (13)
74	3.5425	6.4702	1.8505 (13)
78	3.7046	6.2955	3.2299 (13)
82	3.8894	6.1139	5.5318 (13)
86	4.1012	5.9253	9.3157 (13)
90	4.3454	5.7295	1.5455 (14)
92	4.4817	5.6287	1.9809 (14)

[a]Numbers in parentheses represent the power of 10 by which the rate is to be multiplied. A reduced-mass correction has been included in the total decay rate.

Two-E1 decay.—The calculation of the two-$E1$ rate is somewhat more involved. We follow the classic treatment of Brown, Peierls, and Woodward[10] to reduce the two-photon matrix element to tractable form. The results of the calculation can be summarized as follows:

$$dw/d\omega_1 = (8/27\pi)\alpha^2\omega_1^3\omega_2^3\{\tfrac{1}{3}[E_1^2(\omega_1,\omega_2)+E_1^2(\omega_2,\omega_1)] + \tfrac{2}{3}[E_{-2}^2(\omega_1,\omega_2)+E_{-2}^2(\omega_2,\omega_1)] - \tfrac{2}{9}E_1(\omega_1,\omega_2)E_1(\omega_2,\omega_1) + \tfrac{4}{9}E_{-2}(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1) + \tfrac{8}{9}[E_{-2}(\omega_1,\omega_2)E_1(\omega_2,\omega_1)+E_1(\omega_1,\omega_2)E_{-2}(\omega_2,\omega_1)]\}, \quad (5)$$

where

$$E_\kappa(\omega_1,\omega_2) = (3/\omega)\int_0^\infty dr\, j_1(\omega_2 r)[G_{1s_{1/2}}(r)S_\kappa(r,\omega_1) + F_{1s_{1/2}}(r)T_\kappa(r,\omega_1)]. \quad (6)$$

The symbols S_κ and T_κ represent the large and small components of the $2s_{1/2}\to p_{1/2}$ and $2s_{1/2}\to p_{3/2}$ perturbations of the initial-state wave function induced by photon 1 for $\kappa=+1$ and -2, respectively. These perturbations satisfy inhomogeneous Dirac equations

$$(m-\epsilon_1+\omega_1+V)S_\kappa(r,\omega_1) + (d/dr-\kappa/r)T_\kappa(r,\omega_1) = (3/\omega_1)j_1(\omega_1 r)G_{2s_{1/2}}(r),$$
$$-(d/dr+\kappa/r)S_\kappa(r,\omega_1) - (m+\epsilon_1-\omega_1-V)T_\kappa(r,\omega_1) = (3/\omega_1)j_1(\omega_1 r)F_{2s_{1/2}}(r). \quad (7)$$

In the above equations ω_1 and ω_2 are the energies of the two photons ($\omega_1+\omega_2=\omega$), and ϵ_1 is the energy of the $2s_{1/2}$ bound state. The relativistic result goes over to the familiar nonrelativistic form when retardation is neglected, and when Eqs. (7) are simplified in the Pauli approximation, and expanded in terms of a complete set of nonrelativistic p-state wave functions.

In the relativistic case it is a straightforward task to solve Eqs. (7) and to evaluate the matrix elements (6) numerically. It is convenient to express the results in the form suggested by Spitzer and Greenstein,[5]

$$dw/dy = Z^6(9\alpha^6/2^{10})\psi(y,Z)\ \mathrm{Ry}, \quad (8)$$

where y is the fraction of the photon energy carried off by one of the photons.

Typical spectra are given for $y=0$ to $\frac{1}{2}$ in Table II. The total decay is obtained by integrating Eq. (8) from 0 to $\frac{1}{2}$. In column 3 of Table I we give relativistic values of $Z^{-6}w(2E1)$. For comparison it should be noted that the highly accurate nonrelativistic calculation of Klarsfeld[7] corrected for the currently accepted value of the fine-structure constant is

$$w_{\mathrm{NR}}(2E1) = (8.2292 \pm 0.0001)Z^6\ \mathrm{sec}^{-1}, \quad (9)$$

which is only slightly larger than the relativistic result for $Z=1$.

In column 4 of Table I we give the total decay rate w_{tot} in sec^{-1}. The numbers in column 4 have been corrected for the reduced mass of each element. The values Ry $= 3.289\,842\,3\times10^{15}$ sec^{-1} and $\alpha^{-1} = 137.036\,02$ have been used in preparing the tables. The largest source of error in the numerical calculations is in the final integration of the function $\psi(y,Z)$, and that error is at most two parts in the last quoted figure.

Two-M1 decay and other modes.—It is conceivable that for the heavier elements the two-$M1$ decay mode could give some competition to the principal decay modes. The two-$M1$ rate can be calculated in much the same way as the two-$E1$ rate. Neglecting retardation and going to the nonrelativistic limit, one finds

$$w_{\mathrm{NR}}(2M1) = \frac{\omega^7|\langle 2|r^2|1\rangle|^2}{27\cdot 35\cdot m^3}\ \mathrm{Ry} = 1.38\times10^{-11}Z^{10}\ \mathrm{sec}^{-1}, \quad (10)$$

so that for all values of Z the rate is a negligible fraction of the $M1$ rate.

The decay modes two-$E2$ and $M1+E2$ have

TABLE II. Frequency distribution for two-$E1$ decay. The spectral function $\psi(y,Z)$ is defined by Eq. (8) of the text.

y	$\psi(y,1)$	$\psi(y,20)$	$\psi(y,40)$	$\psi(y,92)$
0.0	0.0	0.0	0.0	0.0
0.0625	2.032	1.938	1.688	0.833
0.1250	3.158	3.073	2.832	1.725
0.1875	3.844	3.770	3.553	2.433
0.2500	4.284	4.219	4.025	2.964
0.3125	4.570	4.511	4.337	3.349
0.3750	4.748	4.695	4.536	3.610
0.4375	4.847	4.797	4.646	3.761
0.5000	4.879	4.830	4.682	3.811

not been investigated, but the corresponding decay rates are presumably of the same order of magnitude as the two-$M1$ rate and therefore negligible.

The principal corrections to the present results are thus the radiative corrections, which are expected to be of order $\alpha/2\pi \sim 0.1\%$, and the corrections for nuclear finite size which are expected to be negligible for two-$E1$ decays, but which may be as important as radiative corrections for the $M1$ decays of heavy elements.

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[9]Natural units $\hbar = c = 1$, unit of length = 1 cm, and m = electron rest mass are used except where otherwise noted.

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Radiative Decay of the 2P State of Atomic Hydrogen: A Test of the Exponential Decay Law*

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In Jaynes's semiclassical radiation theory, the decay of an excited atomic state depends on its initial population. For the radiative decay of the 2P level in atomic hydrogen, produced by pulsed excitation from the 2S level, the theory predicts a decay rate $\Gamma = (1-f)\Gamma_0 + \beta P$, where $\beta = f\Gamma_0$. Here f is the initial 2S population, Γ_0 is the natural 2P decay rate, and P is the 2P population relative to f. For $f = 0.086 \pm 0.016$, and P values in the range 0.15 to 0.64, we have measured $\beta = -(2.8 \pm 2.8)\times 10^6$ sec^{-1}. The corresponding value from the semiclassical theory is $\beta = +(53.8 \pm 10.0)\times 10^6$ sec^{-1}.

One of the earliest predictions of quantum radiation theory was that an excited quantum state should decay exponentially in time, in a manner independent of the way it was produced.[1] Recently, Jaynes and collaborators have proposed a semiclassical theory in which the decay of an excited state depends on the initial population σ of that state.[2] We test the dependence of the characteristic decay rate Γ on σ as predicted by the semiclassical theory applied to a three-level atom. This is done by measuring the radiative decay of the 2P state of atomic hydrogen for different values of σ.

As a three-level atom, we consider the 1S ground state, 2S metastable state, and 2P state of atomic hydrogen. The natural decay rate of the 2P state is $\Gamma_0 = 6.25\times 10^8$ sec^{-1}.[3] An atomic hydrogen beam predominantly in the 1S and 2S levels is subjected to a short electric field pulse. The $2P_{1/2}$ level is excited by Stark coupling to the nearly degenerate $2S_{1/2}$ level, while other levels are not appreciably excited because of their relatively large energy separations.[4] Using Jaynes's theory to calculate the radiation rate for the subsequent 2P to 1S decay,[5] we find for small values of the initial 2S population that the radiation evolves as $\exp(-\Gamma t)$, where t is the time after termination of the pulse, and

$$\Gamma = (1-f)\Gamma_0 + \beta P. \tag{1}$$

Here, f is the fractional 2S population entering the pulse region, P is the relative 2S to 2P con-