Simple Formula for the Ionization Rate of Rydberg States in Static Electric Fields

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With use of a full three-turning-point WKB analysis, a simple formula is obtained for the ionization rate of hydrogenic atoms. Contrary to current belief, WKB ionization rates are found to be excellent approximations to exact numerical calculations. In addition, the symmetries of the WKB solution are explored and it is shown that the resonance parameters can all be written as simple functions of three objects, $I_\xi^{(j)}$, $I_\eta^{(j)}$, and $\theta^{(j)}$, which can easily be calculated on programmable pocket calculators.

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The advent of highly selective field ionization techniques has reawakened interest in the study of the decay mechanism of Rydberg atoms in the presence of static electric fields. Several experiments in recent years[1] motivated a good deal of theoretical work and important progress in this direction has been achieved. The divergent perturbation series for the energy was proved to be Borel summable.[2] A dispersion relation between the ground-state energy shift and the ionization rate was discovered[3] and generalized.[4] Exact numerical calculation of Stark resonance parameters has also been accomplished.[5] Since the spectrum turns into a continuum no matter how small the electric fields are, these calculations are quite elaborate. The theory of the ionization of hydrogenic species in dc electric fields was reviewed by Yamabe, Tachibana, and Silverstone.[6] The purpose of the present paper is to report a nonperturbative WKB approach to the problem. Although the WKB quantization equations can be analytically expressed in terms of complete elliptic integrals,[7] a fact known to Lanczos,[8] the usual WKB approaches until recently[7] have been to expand the quantization equations in a similar way to the perturbative series. This introduced further approximations into the calculations. Furthermore, when applying the WKB approximation to radial equations, some authors used a factor m^2-1 in the centrifugal term, thereby violating the conditions[9] for the applicability of the WKB quantization. In addition, it is common to see in the literature WKB formulas for the widths of the levels with simple exponential tunneling factors ($e^{-2\sigma}$, etc.). These factors, obtained through different arguments, are accurate only approximately and at energies well below the autoionization limit, a region of little interest. Therefore it should not be surprising to find sometimes in the literature statements that "WKB ionization rates are of little accuracy."

In the present communication we derive a simple formula for the ionization rate of hydrogenic atoms in dc electric fields. This formula is obtained from a full first-order WKB treatment of the three-turning-point scattering problem and avoids the aforementioned problems. By comparison of values obtained from this formula with exact numerical calculations reported in the literature[5] we believe our formula to be reliable around the autoionizing limit. As a byproduct of our calculations we are able to show that the whole three-turning-point problem on hand reduces to the sole calculation of three families of integrals, namely of the objects

$$I_\xi^{(j)}=\int_b^a[(a-\xi)(\xi-b)(\xi-c)]^{-1/2}\xi^j\,d\xi, \tag{1a}$$

$$I_\eta^{(j)}=\int_a^b[(\eta-a)(b-\eta)(c-\eta)]^{-1/2}\eta^j\,d\eta, \tag{1b}$$

and

$$\theta^{(j)}=\int_b^c[(a-\eta)(b-\eta)(c-\eta)]^{-1/2}\eta^j\,d\eta \tag{1c}$$

for $j=-1$, 0, and 1; the other symbols are defined below.

The Hamiltonian for the Stark effect in hydrogen-like atoms is separable in parabolic coordinates ξ and η.[10] With the usual product *Ansatz* for the eigenfunctions[10] the problem reduces to the study of two one-dimensional Schrödinger equations with the potentials $V_\xi=-Z_1/\xi+m^2/4\xi^2+\frac{1}{4}F\xi$ and $V_\eta=-Z_2/\eta+m^2/4\eta^2-\frac{1}{4}F\eta$ where m^2-1 was replaced by m^2 on account of the criterion for applicability of the WKB approximation to radial equations,[9] and where $Z_1 + Z_2 = Z$. F represents the static electric field. The motion along the ξ coordinate is always confined. The η motion is complicated by the fact that the electron can escape from the atom in the direction $\eta \to \infty$. The WKB quantization neglecting tunneling then reads $I_\xi = (n_1 + \frac{1}{2})\pi$ and $I_\eta = (n_2 + \frac{1}{2})\pi$, n_1 and n_2 being the usual parabolic quantum numbers,[10] and I_ξ and I_η the usual phase integrals. These integrals may be very much simplified by the use of the identity

$$I_\xi = \int_b^a (\tfrac{1}{2}E - V_\xi)^{1/2} d\xi$$
$$= \tfrac{1}{2}\int_b^a V_\xi'(\tfrac{1}{2}E - V_\xi)^{-1/2}\xi\, d\xi, \tag{2}$$

obtained after a trivial integration by parts. The first equality sign in Eq. (2) gives us

$$I_\xi = \frac{\sqrt{F}}{2}\left[-I_\xi^{(2)} + \frac{2E}{F}I_\xi^{(1)} + \frac{4Z_1}{F}I_\xi^{(0)} - \frac{m^2}{F}I_\xi^{(-1)}\right]. \tag{3a}$$

From the second equality if follows that

$$I_\xi = \frac{\sqrt{F}}{2}\left[\tfrac{1}{2}I_\xi^{(2)} + \frac{2Z_1}{F}I_\xi^{(0)} - \frac{m^2}{F}I_\xi^{(-1)}\right]. \tag{3b}$$

Combining (3a) and (3b) it is easy to eliminate $I_\xi^{(2)}$ and obtain

$$I_\xi = \tfrac{1}{2}\sqrt{F}\,[\tfrac{1}{3}(a + b + c)I_\xi^{(1)}$$
$$- \tfrac{2}{3}(ab + ac + bc)I_\xi^{(0)} + abcI_\xi^{(-1)}]. \tag{4}$$

The roots $c < 0 \leqslant b < a$ are obtained from

$$-\xi^3 + \frac{2E}{F}\xi^2 + \frac{4Z_1}{F}\xi - \frac{m^2}{F}$$
$$\equiv (a - \xi)(\xi - b)(\xi - c) = 0, \tag{5}$$

where E is the energy. The objects $I_\xi^{(j)}$ can easily be evaluated:

$$I_\xi^{(0)} = gK(k),$$
$$I_\xi^{(-1)} = (g/a)\,\Pi((a - b)/a, k),$$
$$I_\xi^{(1)} = g[cK(k) + (a - c)E(k)], \tag{6}$$

where $g = 2(a - c)^{-1/2}$, $k^2 = (a - b)/(a - c)$, and K, E and Π are standard complete elliptic integrals of the first, second, and third kinds, respectively. For states below the autoionization limit the expression for I_η is obtained from Eq. (4) by replacing $\sqrt{F}$ by $-\sqrt{F}$ and ξ by η. The η roots $0 \leqslant a < b < c$ are now obtained from

$$\eta^3 + \frac{2E}{F}\eta^2 + \frac{4Z_2}{F}\eta - \frac{m^2}{F} \equiv (\eta - a)(b - \eta)(c - \eta) = 0. \tag{7}$$

The required $I_\eta^{(j)}$ are obtained from Eq. (6) by replacing ξ by η. This symmetry reflects the equivalence of V_ξ and V_η under the replacement $Z_1 \to Z_2$ and $F \to -F$.

The critical quantity in the formula for the ionization rate (i.e., for the width of the levels, which is the imaginary part of the complex eigenvalues) is the tunneling integral across the η barrier. The integral θ across the barrier can also be reduced through integration by parts. The final result is

$$\theta = \tfrac{1}{2}\sqrt{F}\,[\tfrac{1}{3}(a + b + c)\theta^{(1)}$$
$$- \tfrac{2}{3}(ab + ac + bc)\theta^{(0)} + abc\theta^{(-1)}] \tag{8}$$

with

$$\theta^{(0)} = gK(k), \quad \theta^{(-1)} = (g/c)\,\Pi((c - b)/c, k),$$
$$\theta^{(1)} = g[aK(k) + (c - a)E(k)], \tag{9}$$

where now $g = 2(c - a)^{-1/2}$ and $k^2 = (c - b)/(c - a)$. In evaluating θ the branches for the integrals were chosen such that $\theta > 0$ for energies below the top of the barrier.

In obtaining the correct WKB ionization rate two points are of special relevance: (i) The contribution from the barrier should be obtained from a detailed analysis of a three-turning-point problem; by so doing one finds instead of the simple exponential factor mentioned before

$$\omega(\theta) = \frac{(1 + e^{-2\theta})^{1/2} - 1}{(1 + e^{-2\theta})^{1/2} + 1}. \tag{10}$$

Of course, in deriving this expression for the present problem (in which $V_\eta \to \infty$ when $\eta \to \infty$) the phase shift is obtained from the comparison of the WKB eigenfunctions with the "core-free" ones, i.e., with the Airy function. This is an important modification of the known results for the three-turning-point problem,[11] which usually assumes $V \to 0$ at infinity. (ii) The separation constant Z_2 depends implicitly on the energy. This was apparently overlooked by Lanczos[8] but was correctly taken into account in the approximate result of Rice and Good.[6,12]

From the usual Breit-Wigner parametrization of the resonances one finds that the width Γ of the levels can be written

$$\Gamma = 2\omega(\theta)(\partial I_\eta/\partial E)^{-1}. \tag{11}$$

It is not difficult to see that

$$\partial I_\eta/\partial E = F^{-1/2}[\tfrac{1}{2}I_\eta^{(1)} + (\partial Z_2/\partial E)I_\eta^{(0)}]. \tag{12}$$

Since at resonance $\partial I_\xi/\partial E = 0$, it follows that

$\partial Z_2 \partial E = -\partial Z_1/\partial E = I_\xi^{(1)}/2I_\xi^{(0)}$. Therefore

$$\Gamma = \frac{4\omega(\theta)F^{1/2}I_\xi^{(0)}}{I_\xi^{(0)}I_\eta^{(1)} + I_\xi^{(1)}I_\eta^{(0)}}. \tag{13}$$

Numerical evaluation of this Γ just requires determination of θ since all $I_\xi^{(j)}$ and $I_\eta^{(j)}$ are automatically obtained during calculation of the eigenvalue E. For energies well below the autoionizing limit $[\omega(\theta) \approx e^{-2\theta}/4]$, Eq. (13) reduces to the approximation obtained by Rice and Good.[12] In this same energy range, by doing the crude approximation $\partial Z_2/\partial E = 0$, we obtain $\Gamma = 8\Gamma_{BS}$, where Γ_{BS} refers to the width as given by Eq. (54.6) of Bethe and Salpeter.[10a] This gives some hint of why the simple WKB formula is usually found to be about 1 order of magnitude too small. At energies near the autoionizing limit it is important to use the full three-turning-point formula to obtain the position of the resonances. This means that $I_\eta = (n_2 + \frac{1}{2})\pi$ should be replaced by $I_\eta + Q(\theta) = (n_2 + \frac{1}{2})\pi$, where $Q(\theta)$ is a "quantum correction" which takes the nonzero tunneling probability into account.[10]

TABLE I. Comparison of present WKB results with exact numerical calculations of Damburg and Kolosov (Ref. 13). Ratio means exact/WKB. All values correspond to $m = 2$. The principal quantum number is given by $n = n_1 + n_2 + m + 1$.

n_1	n_2	Field strength (kV/cm)	Energy ratio	Ioniz. rate ratio
6	3	30	1.0000	1.01
6	3	33	1.0000	1.01
6	3	35	1.0000	1.00
6	3	37	1.0000	0.99
7	2	33	0.9994	1.10
7	2	35	1.0000	1.01
7	2	37	1.0000	1.01
7	2	40	1.0002	0.99
8	1	35	1.0003	1.03
8	1	37	1.0001	1.03
8	1	40	1.0001	1.02
8	1	43	1.0004	1.00
2	8	19	1.0000	1.00
2	8	20	1.0000	1.00
2	8	22	1.0000	0.99
2	8	25	1.0001	0.92
3	7	19	1.0000	1.00
3	7	20	1.0000	1.00
3	7	22	1.0000	1.00
3	7	25	1.0000	0.93
4	6	20	0.9997	1.03
4	6	22	0.9998	1.02
4	6	25	0.9999	0.97
4	6	27	1.0001	0.91
0	11	13.5	1.0000	1.00
0	11	14	1.0000	1.00
0	11	15	1.0000	1.00
0	11	16	1.0000	0.96
0	11	17	1.0000	0.92
1	10	14	1.0004	1.00
1	10	15	1.0000	1.00
1	10	16	1.0000	0.98
1	10	17	1.0000	0.94
1	10	18	1.0000	0.92

In Table I we compare our WKB results with exact numerical work of Damburg and Kolosov.[13] As mentioned by them, certain sublevels in this table cannot be correctly described by perturbation theory. As seen from our table, the WKB resonance parameters, obtained from a full three-turning-point treatment, are excellent approximations to the exact numerical parameters.

In summary, we have shown that the correct WKB treatment of the Stark effect in hydrogenlike atoms is not only able to produce excellent approximations for the real part of the complex eigenvalues but can be used to obtain satisfactory approximations for their imaginary part as well. The whole problem can be reduced to the calculation of three quantities, $I_\xi^{(j)}$, $I_\eta^{(j)}$, and $\theta^{(j)}$. These quantities may be reduced to complete elliptic integrals which, in turn, may easily be evaluated even on programmable pocket calculators.

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Optical Pumping of Dense Charge-Exchange Targets for Polarized Ion Sources

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Optical pumping by a single-frequency cw ring dye laser of thick sodium vapor targets ($NL > 10^{13}$ atoms/cm^2) without buffer gas has been investigated for application in a polarized ion source. The atomic polarization of the sodium was determined from the optical rotation of a second dye laser tuned midway between the D lines. Rate-equation calculations reproduce the experimental data and predict very high polarizations ($P \geq 0.8$) with the use of one or more single-frequency dye lasers in thick alkali-metal vapor targets.

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Recently much interest has been generated over the possibility of producing a polarized H^- ion source by optical pumping.[1] An ion source based on this concept, first suggested by Haeberli[2] and examined more closely by Anderson,[1] has the potential of producing polarized H^- beam currents far in excess of those produced in today's ion sources. In an optically pumped polarized ion source (OPPIS) incident protons capture spin-polarized electrons from an alkali-metal vapor target that is polarized by optical pumping. The atomic hyperfine interaction in hydrogen can then be used to convert the atomic polarization into nuclear polarization.[3] Capture of a second electron in an unpolarized alkali-metal vapor target produces the polarized H^- beam. To achieve efficient polarization transfer during the charge-exchange process, the alkali metal must be optically pumped in a large magnetic field.[4] To achieve efficient neutralization of the proton beam, the optically pumped alkali-metal target must have thickness $NL = (1-5) \times 10^{13}$ atoms/cm^2, which should produce 2.6–13 μA of polarized H^- beam per milliampere of incident H^+ beam in sodium.

Anderson[1] pointed out two major questions which need to be addressed to demonstrate the feasibility of an OPPIS. The first concerns the transfer of atomic polarization in the charge-exchange reaction. We have shown theoretically[4] that at least 75% of the atomic polarization can be transferred. Recent measurements at the Japanese National Laboratory for High Energy Physics (KEK)[5] suggest that the polarization transfer is substantially better than this worst-case estimate. The second question concerns the degree of polarization