Probabilities of radiative transitions between highly excited atomic states

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Approximate expressions are obtained for the Coulomb dipole matrix elements, radiative transition probabilities, and photoionization cross sections for highly excited atomic states. The results are valid for $n-n'\gg 1$ and are different for small $(l\ll n)$ and large $(l\sim n)$ orbital momenta. They are identical in the common region of applicability. The results agree also with the well-known expressions for the case $|n-n'|\ll n$ in the common region of applicability. A quasiclassical substantiation of the empirical Bethe quantum-number selection rules for radiative transitions is presented: upon variation of the principal quantum number n the orbital quantum number l varies with an overwhelming probability in the same direction as n. It is shown that the rule is valid for $l\gg n^{2/3}$ and holds better the larger the difference between the principal quantum numbers, $n-n'$. It is shown that upon photoionization of highly excited atomic states the orbital momentum predominantly increases. A generalization of the quantum-mechanical correspondence principle is proposed for the calculation of the matrix elements between states whose principal quantum numbers differ greatly.

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§1. INTRODUCTION

The interest in spectroscopy of highly excited atomic states is due to many applications: the investigation of interstellar hydrogen, the development of masers and of detectors for long-wave radiation on transitions between highly excited atomic states, and a few others.

In the theoretical analysis of highly excited atomic states, the wave function of the valent atomic electron can be assumed with good approximation to be hydrogenlike and characterized in the discrete spectrum by a principal quantum number n, an orbital number l, and a magnetic number M. This approximation is justified because at $n\gg 1$ the electron is on the average far from the atomic residue, whose influence can be taken into account by replacing n with $n^*=n-\delta_l$, where δ_l is the quantum defect. We shall not distinguish below between n and n^*, although the results remain valid also when the quantum defect is taken into account. The condition $n\gg 1$ means at the same time that the states of the degenerate energy level n can be described by using a quasiclassical approximation.

The probabilities between the radiative transitions between highly excited atomic states are expressed in terms of the reduced dipole-moment matrix element (here and below $e=\hbar=m=1$)

$$R_{nl}^{n'l\pm1}=\int_0^\infty R_{nl}(r)R_{n'l\pm1}(r)r^3dr. \tag{1}$$

If the R_{nl} are Coulomb radial functions, this integral can be calculated analytically in general form. The exact answer contains in this case hypergeometric functions.[1] For transitions between highly excited atomic states, both qualitative estimates and direct calculations using the exact formula are difficult, since generally speaking all three parameters of the hypergeometric functions are large. The available oscillator-strength tables (see, e.g., Ref. 2) make it possible to calculate the probabilities of transitions between relatively low states. However, the quasiclassical character of the highly excited atomic states can yield simpler approximate expressions for the matrix elements.

Thus, it was found in Ref. 3 that for highly excited states that differ little in energy, i.e., when $\Delta n=n-n'\ll n,n'$, the matrix element (1) reduces in accordance with the correspondence principle to a Fourier component of the classical coordinate as a function of the time. In Ref. 4, the Landau method[5] yielded the argument of the exponential in an exponentially small matrix element of a transition with $\Delta n\sim n,n'$ [see (10) below)].

In this paper we obtain, within the framework of the quasiclassical approximation, approximate expressions for the matrix elements and for the transition probabilities, which allow us to describe transitions between states with greatly differing principal quantum number $\Delta n\sim n,\ n'$. The equations obtained differ in form for small $(l\ll n,n')$ and large $(l\sim n,n')$ orbital angular momenta. In the region $n^{2/3}\ll l\ll n$, however, which is common to both cases, they coincide. In addition, at $1\ll\Delta n\ll n$ the obtained formulas coincide with the result of Ref. 3 (a summary of the results of Ref. 3 is contained in the book by Vaĭnshteĭn *et al.*[2]).

The amplitude of a transition from the discrete to the continuous spectrum can be obtained from the amplitude of a transition in the discrete spectrum by means of an analytic continuation, in energy, effected for one of the states by making the substitution $n'\to i/p$, where p is the electron momentum in the continuous spectrum. Quasiclassical states with $n'\gg 1$ go over thus into the states with small momentum $p\ll 1$. On the other hand, the detailed balancing principle make it possible to connect the probabilities of the direct and inverse processes. By the same token, the approximate formulas obtained in the present paper can be used to describe all radiative transitions between states with energies near the boundary of the discrete and continuous spectra, including the description of free-free transitions.

In §2 we present the calculations of the matrix elememts for small and large orbital angular momenta and discuss the approximate Bethe selection rules for them. These results are used in §3 to determine the probabilities of radiative transitions and the photoionization cross sections; their connection with the Kramers formulas are discussed. Section 4 is devoted to a generalization of the correspondence principle of quantum and classical mechanics for matrix elements in the case of states with greatly differing quantum numbers.

§2. QUASICLASSICAL MATRIX ELEMENTS

We deal first with the matrix element (1) in the case of small orbital angular momenta $l \ll n,\ n'$. We recall that no restrictions are imposed on the difference Δn between the principal quantum numbers. For the sake of argument we put $\Delta n > 0$, i.e., $n > n'$. We substitute in (1) the radial functions in the quasiclassical approximation

$$R_{nl}(r)=\frac{a_n}{rp_{nl}^{1/2}(r)}\cos\left(\int\limits_{r_{nl}}^{r}p_{nl}dr-\frac{\pi}{4}\right), \tag{2}$$

where the radial momentum and the normalization constants are equal, respectively, to

$$p_{nl}(r)=(-1/n^2+2/r-(l+{}^1/_2)^2/r^2)^{1/2}, \quad a_n=(2/\pi n^3)^{1/2}. \tag{3}$$

As will be verified below, the significant integration region $r \sim (n')^{4/3}$ is much less than the region of the classical motion $\sim(n')^2$. The radial momentum (3) in the significant region is then weakly dependent on the principal quantum number, and a strong overlap of the wave functions R_{nl} and $R_{n'l\pm1}$ takes place. The phase shifts of the wave functions are in this case of the order of $(n')^{3/2}$, i.e., they are large. To calculate the integral (1) we can therefore replace the cosine products by

$$\frac{1}{2}\cos\left(\int\limits_{r_{nl}}^{r}(p_{nl}-p_{n'l\pm1})dr\right). \tag{4}$$

We have neglected here the difference between the left-hand turning points, which is small at $n, n' \gg 1$. The difference of the momenta in the significant region is of the form

$$p_{nl}-p_{n'l\pm1}=\frac{(n')^{-2}-n^{-2}\pm2lr^{-2}}{2(2r-l^2)^{1/2}}r. \tag{5}$$

It is seen therefore that by equating the phase of the cosine in (4) to unity we obtain the significant integration region indicated above.

At the same time, the significant integration region is located in the classically accessible region; the contribution to the matrix element (1) from the region of the left-hand turning point, where the quasiclassical approximation does not hold, can be easily verified to be smaller than (6) by a factor l/n.

Substituting (2) and (4) in (1) and replacing the upper limit of integration with respect to r by ∞, we obtain after certain transformations

$$R_{nl}^{n'l\pm1}=\frac{2l^2}{3^{1/2}\pi}\frac{(nn')^{1/2}}{(n^2-n'^2)}[K_{2/3}(\xi)\mp K_{1/3}(\xi)]. \tag{6}$$

Here $K_\nu(\xi)$ are MacDonald functions and we have put

$$\xi={}^1/_6l^3(n'^{-2}-n^{-2}).$$

Equation (6) is valid for $l \ll n'$ and $\Delta n \gg 1$. The second inequality ensures the possibility of replacing the upper limit of integration by ∞.

At $n \sim n'$ and $l \lesssim n^{2/3}$ the matrix elements (6) are of the order of $n^{1/3}$, i.e., they are quasiclassically large, as follows from the strong overlap of the wavefunctions at small r. At $l \gg n^{2/3}$ they become exponentially small.

Using various expansions of the MacDonald functions, we can simplify (6) in various limiting cases. Thus, in the case of very small orbital angular momenta $l \ll (n')^{2/3}$, when $\xi \ll 1$, we obtain from (6)

$$R_{nl}^{n'l\pm1}=\frac{12^{2/3}}{3^{1/2}\pi}\Gamma\left(\frac{2}{3}\right)\frac{(nn')^{11/6}}{(n^2-n'^2)^{5/3}}. \tag{7}$$

Here $\Gamma(x)$ is the gamma function. It is seen from (7) that in this limiting case the matrix element does not depend on l at all. Equation (7) is valid not only at $l \gg 1$, but also at small values $l \sim 1$ (s, p, d, and f states). Its importance is due to the fact that highly excited atoms with small orbital angular moments are produced selectively in the experiments.

Even for the lower states of the hydrogen atom, where the quasiclassical approximation is formally not applicable, it leads to results that do not differ greatly from the exact ones. For example, for the $2s \rightarrow 4p$ transition the ratio of the quasiclassical matrix radial dipole element calculated from Eqs. (7) to the exact value [see Eq. (63.4) of Ref. 6] is 0.73.

We note that under the condition $1 \ll \Delta n \ll n$ Eq. (6) coincides with the asymptotic form of the result of Ref. 3, in which we must put $l \ll n$ (for details see, e.g., Ref. 4).

Introducing in (6) the substitution $n' \rightarrow i/p$ in accord with the statements made in the introduction, we obtain the matrix element of the bound-free transition

$$R_{nl}^{pl\pm1}=-\frac{il^2}{3^{1/2}\pi\omega}\left(\frac{p}{n}\right)^{3/2}\left[K_{2/3}\left(\omega\frac{l^3}{3}\right)\pm K_{1/3}\left(\omega\frac{l^3}{3}\right)\right]. \tag{8}$$

Here

$$\omega=p^2/2+1/2n^2$$

is the energy of the absorbed photon. The condition for the applicability of (8) is that the inequalities $l/n \ll 1$ and $lp \ll 1$ be satisfied. We note that in the vicinity of the threshold, namely at $p \ll 1/n$, the matrix dipole element depends on the momentum p like $p^{3/2}$. Next, in the limiting case $l \ll \omega^{-1/3}$, just as for bound-bound transitions, the matrix element (8) does not depend on the orbital angular momentum l.

The matrix element of bound-bound transitions, obtained under the assumption $\Delta n \ll n$, was continued in Ref. 3 [see also Eq. (24.2) of Ref. 2] into the continuous spectrum with the aid of the substitution $\Delta n \rightarrow \omega n^3$. This continuation was not justified, inasmuch as analytic continuation into the continuous spectrum calls for passage through the region $\Delta n \sim n$, where the equation of Ref. 3 do not hold. Thus, the validity of the equations of Ref. 3 for the cross sections for photoionization from the states (n, l) remains unclear. Comparison with (8) shows, however, that the prescription proposed in Ref. 3 for obtaining the matrix element of a bound-free transition gives the correct result at $l \ll n$.

We turn now to the case of large orbital angular momenta, when l/n, $l/n' \sim 1$. The corresponding dipole matrix elements are exponentially small, so that the significant region of integration with respect to r corresponds not to small values of r but to complex r. As noted in the Introduction, the argument of the exponential was determined in Ref. 4 by the Landau quasiclassical method.

To find the pre-exponential factor, we start from the exact expression for the matrix element,[1] although the result could be obtained here, too, with the aid of the generalized correspondence principle (see §4), on the basis of classical Coulomb trajectories (taken only in the complex domain of the coordinate r and of the time t). We present the hypergeometric functions in the exact expression for the matrix element in the form of a contour integral [see Eq. (e.3) of Ref. 5)] and make the following change of the integration variable

$$u = -\frac{(n-n')^2}{4nn'}\left(1+\frac{1}{w}\frac{n+n'}{n-n'}\right),$$

if the conditions

$$\Delta n \gg 1, \quad l \gg (n')^{2/3} \tag{9}$$

are satisfied the resultant integral can be calculated by the saddle-point method. Numerous factorials can be expressed by using the Stirling formula. In the case of the matrix element $R_{nl}^{n'l+1}$, account must be taken of the terms that follow the principal term of the expansion near the saddle point. Leaving out the cumbersome calculations, we present only the final result

$$R_{nl}^{n'l\pm1} = f_\pm \exp\{-l|f(x)-f(x')|\}, \tag{10}$$

where $x = l/n$, $x' = l/n'$ and

$$f(x) = \frac{1}{2x}\ln\frac{1+x}{1-x} + \frac{1}{2}\ln(1-x^2).$$

The pre-exponential factors are (we assume, as before, $n > n'$)

$$\begin{aligned} f_- &= 4\left(\frac{l}{\pi}\right)^{1/2}\left(\frac{nn'}{n^2-n'^2}\right)^{3/2}[(1-x^2)(1-x'^2)]^{-1/4}, \\ f_+ &= \frac{2nn'}{(\pi l^5)^{1/2}}\left(\frac{nn'}{n^2-n'^2}\right)^{5/2}[(1-x^2)(1-x'^2)]^{1/4}. \end{aligned} \tag{11}$$

Thus, f_- corresponds to the case when the principal quantum number n and the orbital momentum l change in the same direction in the course of the transition, while f_+ corresponds to changes in the opposite direction.

At $1 \ll \Delta n \ll n$, as expected, Eq. (10) coincides with the corresponding asymptotic form of Ref. 3.

For the orbital angular momenta $n^{2/3} \ll l \ll n$, Eqs. (10) and (8) yield the same answer with account taken of the pre-exponential factor. This confirms the validity of the calculations of the exponentially small matrix elements by the method described at the beginning of this section.

The matrix elements of bound-free transitions are obtained by analytic continuation of (10). We do not present here the corresponding results, since the method of obtaining them from (10) is obvious. In the same manner we can determine from (8) and (10) the dipole matrix elements of the free-free transitions of a small-momentum electron.

If $x' \to 1$, then the obtained expressions (10) must be slightly modified to take into account the fact that the quantity $(n' = l)!$, contained in the exact formula of Ref. 1, cannot be replaced by using Stirling's formula as was done in the derivation of (10). We then obtain

$$R_{nl}^{n'l-1} = \frac{\pi^{1/4}2^{l+3/2}}{[(n'-l)!]^{1/2}}\frac{l^{5/4}n^2}{(n^2-l^2)^{7/4}}\left(\frac{n'}{e}\right)^{(n'-l)/2} e^{-lf(x)}. \tag{12}$$

In this form, the formula is valid for values of l close to n'. In particular, we obtain from it the well-known exact value of the matrix element R_{nl}^{ll-1} (in which Stirling's formula was used for $n!$ and $l!$). We obtain similarly $R_{nl}^{n'l+1}$ for l close to n'.

Bethe (Ref. 6, p. 420 of Russ. transl.) formulated an empirical selection rule for Coulomb dipole matrix elements: when the principal quantum number n changes, the orbital momentum l changes with overwhelming probability in the same direction as n.

This rule was deduced from an analysis of transitions in which at least one of the principal quantum numbers was small. By the same token, the orbital momenta were small and comparable with the smaller principal quantum number. As to the application of this rule to transitions between highly excited atomic states with n, $n' \gg 1$, it can be seen from (11) that it holds for transitions with large orbital momenta $l \sim n$, n'. Indeed, in this case we have from (11)

$$\frac{f_+}{f_-} \sim \frac{nn'}{l^3} \sim \frac{1}{n}.$$

A similar statement is valid for bound-bound transitions: At $l \gg \omega^{-1/3}$ the transition from the bound to the free state is accompanied by an increase in the orbital momentum.

The Bethe rule, however, is violated at small orbital momenta $l \lesssim n^{2/3}$, and particularly strongly for $l \ll n^{2/3}$, where, as seen from (7), the matrix element does not depend on l at all, and by the same token is the same for transitions with $l' = l \pm 1$.

If we consider the corresponding matrix elements for the case $1 \ll \Delta n \ll n$, we see easily (this was demonstrated in Ref. 4) that in this case their ratio is of the order of $1/\Delta n$. Thus, the larger Δn the better is the Bethe rule satisfied; in this case the orbital momenta should be large enough: $l \gg n^{2/3}$.

As for bound-free transitions, we obtain from (11) at $l \gg \omega^{-1/3}$

$$R_{nl}^{pl-1}/R_{nl}^{pl+1} \sim \frac{1}{l^3\omega} \ll 1,$$

i.e., on going to the continuous spectral the orbital quantum number predominantly increases. Just as in the case of bound-bound transitions, the larger l, the better this rule manifests itself.

§3. PROBABILITIES OF RADIATIVE TRANSITIONS

We obtain first the probability of the spontaneous transitions from the state n with fixed orbital momentum $l \ll n$ into a state with principal quantum number $n' < n$. It is obtained by summing two probabilities with $l' = l \pm 1$. Starting from (6), we obtain

$$w(nl \to n') = \frac{2l^4(n^2 - n'^2)}{9c^3\pi^2 n^5 n'^5}[K_{2/3}^2(\xi) + K_{1/3}^2(\xi)]. \tag{13}$$

Here c is the speed of light ($c = 137$ in atomic units). In particular, it follows from (13) that for very small orbital momenta $l \ll (n')^{2/3}$ the spontaneous-transition probability, as well as the corresponding matrix element (7), does not depend on l. On the contrary, at $(n')^{2/3} \ll l \ll n$ the transition with $l' = l+1$ makes a small contribution to (13) in accord with the Bethe rule.

Averaging (13) over the orbital angular momentum l and replacing summation over l by integration we obtain, as expected, the known Kramers formula [see, e.g., Eq. (23.1) of Ref. 2]. Thus, the main contribution to the average transition probability is made by matrix element with $l \sim n^{2/3}$; the contribution of transitions with $l \ll n^{2/3}$ is small because of the small statistical factor $(2l+1)$ which arises upon averaging because of the degeneracy with respect to the projections of the orbital momentum, and the contribution of transitions with $l \gg n^{2/3}$ is small because of the exponential smallness of the matrix elements.

Expression (13) makes it possible to determine the radiative lifetime of the highly excited state nl. It is determined from the formula

$$\frac{1}{T_{nl}} = \sum_{n'=l}^{n-1} w(nl \to n'). \tag{14}$$

Substituting (13) in (14) and replacing the summation over n' by integration, we obtain for the case $l \gg 1$

$$T_{nl}^{\infty} = \frac{3^{1/2}\pi c^3}{4} n^3 l^2. \tag{15}$$

We note that the relation $T_{nl} \sim n^3$ agrees with the one known from the numerical calculations (see page 422 of the Russian transition of Ref. 6). If, however, l is not too large compared with unity, then expression (15) must be replaced by the more accurate result of the substitution of (13) in (14):

$$T_{nl} = T_{nl}^{\infty}\left[1 - \frac{l}{2\pi\sqrt{3}} K_{1/3}\left(\frac{l}{6}\right) K_{2/3}\left(\frac{l}{6}\right)\right]^{-1}. \tag{16}$$

The probability of the spontaneous radiative transition from a state with large orbital momentum $l \sim n$ is determined, according to the Bethe rule, only by the matrix element f_- [see (11)], and is equal to

$$w(nl \to n') = \frac{4l}{3\pi c^3 n^3 n'^3}[(1-x^2)(1-x'^2)]^{-1/2} \exp\{-2l|f(x) - f(x')|\}. \tag{17}$$

In the case of bound-free transitions, we are dealing with the cross section for the photoionization of the state nl with a transition into the continuous-spectrum state with energy $p^2/2$. If $l \ll n$, then, using the matrix element (8), we obtain

$$\sigma(nl \to p) = \frac{4l^4}{9cn^3\omega}\left[K_{2/3}^2\left(\omega\frac{l^3}{3}\right) + K_{1/3}^2\left(\omega\frac{l^3}{3}\right)\right], \tag{18}$$

where ω is the frequency of the absorbed photon. The photorecombination cross section can be easily obtained from (18) with the aid of the detailed balancing principle, and is not presented here. We note once more [see the discussion following Eq. (8)] that the photoionization cross section (18) was correctly intuited in Ref. 3 [see Eq. (24.1) of Ref. 2].

It is easy to verify that when the cross section (18) is averaged over the orbital momentum l we obtain the known Kramers formula [see Eq. (24.4) of Ref. 2], as should be the case.

We now turn to transitions with $l \sim n$. The photoionization cross section is determined then by analytical continuation of the matrix element (10), and in accord with the Bethe rule only the transition with $l' = l+1$ makes a contribution. We obtain

$$\sigma(nl \to p) = \frac{4\pi l}{c\omega^2 n^3}\left[\left(1 - \frac{l^2}{n^2}\right)(1 + l^2p^2)\right]^{-1/2} \exp(-2lF), \tag{19}$$

where F is given by

$$F = \frac{1}{p}\operatorname{arctg}(lp) + \frac{l}{2}\ln(1 + l^2p^2) - n\operatorname{arth}\left(\frac{l}{n}\right) - \frac{l}{2}\ln\left(1 - \frac{l^2}{n^2}\right). \tag{20}$$

Of course, Eqs. (19) and (20) cannot be obtained by any analytic continuation of the equations of Ref. 3.

We note that Eqs. (19), (20), and (18) coincide in the common region of validity of $n^{2/3} \ll l \ll n$.

§4. GENERALIZED CORRESPONDENCE PRINCIPAL

By tracing the procedure used in the beginning of §2, we can see that the calculation method used for small angular momenta can be formulated as a certain extension of the correspondence principal for the calculation of the matrix elements with large change of the principal quantum number. We note for this purpose that in the essential integration region, corresponding to small values of R, the momenta p_{nl} and $p_{n'l\pm1}$ are practically equal. This means that the classical trajectories are almost the same for the initial and final states at small r. We designate this classical coordinate of the electron by $r_l(t)$. From an analysis of the first part of §2 it is seen then that

$$R_{nl}^{n'l\pm1} = \frac{1}{T_n^{1/2}T_{n'}^{1/2}} \int r_l(t) \cos\left(\int_0^t \omega(t')\,dt'\right) dt. \tag{21}$$

Here T_n and $T_{n'}$ are the classical periods of motion for the initial and final states, and depend on the corresponding quantum numbers n and n', while the quantity

$$\omega(t) = \frac{1}{2n'^2} - \frac{1}{2n^2} \pm \frac{l}{r_l^2(t)} \tag{22}$$

takes into account not only the change of the energy in the transition, but also the change of the centrifugal energy $(l+\frac{1}{2})^2/2r^2$ with increasing (+ sign) or decreasing (− sign) orbital angular momentum l. The region of the integration with respect to the time t in (21) corresponds formally to one of the periods of motion of the particle, but the actual integration in (21) takes place only over the region where the initial and final trajectories are close to each other. For example, for small orbital angular momenta $l \ll n$, a contribution is made only by the region of small r, and the upper limit of the integration in (21) can be extended to infinity, since the integral converges soon enough.

Actually, the result (21) is valid not only for the case of small orbital momenta $l \ll n$, but also in the general case. At $l \sim n$, by calculating the integral in (21) by the saddle-point method, we verify that the essential integration region is in the complex plane of the time t, and the initial point t_0 is obtained from the condition $\omega(t_0) = 0$. In the vicinity of this point, obviously, the initial and final trajectories are the same, despite the strong difference between n and n'. It is the corresponding coordinate that should be substituted in (21) (it is, of course, complex in this case).

In the case of close values of n and n', Eq. (21) goes over, as it should, into the well-known formula of the correspondence principle for quantum-mechanical matrix elements.

Of course, all the foregoing is closely connected with Landau's[5] general theory of calculating quasiclassical matrix elements. In the case of small orbital angular momenta it is important to take into account the point where the potential energy becomes infinite in the considered problem, i.e., the point $r = 0$. In the case of large orbital momenta l, it is important to take into account the point where the difference $E_n - E_{n'}$ between the effective potential energies $U_l - U_{l'}$, which take into account the centrifugal barrier [see Eq. (51.7) of Ref. 5]. Those sections of the classical trajectories which differ greatly in the initial and final states make no substantial contribution to the sought quasiclassical matrix element: the only important region is where these trajectories are close to one another, and it does not matter whether this takes place in the region of the real motion of the particle or in the region of the analytical continuation of the real trajectories into the complex space-time plane.

We note in conclusion that the equations derived, together with known results (see Refs. 3 and 7), provide a comprehensive description of the transitions between quasiclassical states in either a discrete or continuous spectrum characterized by definite orbital momenta, for the Coulomb problem.

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