Semiclassical approach to Rydberg-atom intercombination transitions in collisions with electrons

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The intercombination transitions between l-resolved Rydberg levels due to collisions with fast electrons are studied within the Ochkur approximation for principal quantum numbers $n \leq 20$, where l is the angular momentum. The use of the Heisenberg correspondence principle for radial integrals enables one to obtain analytic expressions for any term in the multipole expansion of the cross section for collisions with an arbitrary n and l change. For transitions involving angular momenta that are small compared to the principal quantum numbers, the semiclassical and the exact results are found to be fairly close, within the accuracy consistent with the Born approximation. As a result, the semiclassical approach furnishes a ready estimate for cross sections, and its validity range proves broad enough to cover the region of large momenta transferred to the atom. The numerical results also confirm that dipole transitions have no dominance over the collisions with other Δl values, in accord with the general model of intercombination scattering.

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I. INTRODUCTION

The collision-induced intercombination transitions typically are substantially weaker than those without spin change. Within *LS* coupling, the spin-change transitions are caused exclusively by the exchange interaction between projectile electron and atom. For lower levels intercombination transitions have received comprehensive consideration (see Henry and Kingstone [1], Ochkur [2], and Sobelman, Vainshtein, and Yukov [3]), whereas for highly excited states the experimental information is not available. This is in contrast to spin-conserving transitions, which have lately been a subject of active experimental studies (for recent results see the papers of Sun and MacAdam [4], Rolfes *et al.* [5], and references therein). On the other hand, the account for spin-change effects requires, strictly speaking, partial-wave techniques even for fast projectiles, as opposed to the simple Born approximation for spin-conserving transitions. For these reasons collision-induced spin-change transitions in Rydbergs have not received an adequate theoretical treatment as yet.

Recently Beigman and Matusovsky [6] presented numerical results for intercombination cross sections in *LS* coupling averaged over angular momentum l and in the range of principal quantum numbers $n \leq 10$. Their calculations were performed within the framework of the Ochkur approach [2], which is, in fact, a modification of the Born-Oppenheimer method and is believed to furnish about the same accuracy as the Born approximation does for spin-conserving ($\Delta S = 0$) transitions. Also, the classical binary encounter approximation of Stabler [7] and Webster, Hansen, and Duveneck [8] was shown to give a reasonable estimate for the total cross section. Similar results have been obtained by Borodin, Kazansky, and Ochkur [9] through the integration of a microcanonical distribution function in a phase space.

Further considerations are concerned with the role of angular momentum. First, the l-averaged cross section is, in general, insufficient as its validity is limited by an assumption of statistical equilibrium over orbital quantum numbers. In rarified plasmas or in experiments with selective excitation the latter condition holds only in close proximity to the ionization continuum. For the majority of a discrete spectrum, l-resolved cross sections are needed. Second, as well as being of considerable interest in themselves, l-resolved cross sections are more sensitive to the approximations employed than those integrated over an angular momentum, and therefore present more stringent tests of the major assumptions and computational routines.

The distinctive feature of intercombination transitions is that the dipole transitions are no longer dominant and it is essentially important to have a detailed account of all nondipole $nl \rightarrow n'l'$ transitions. The reason for this is that the typical impact parameters that most contribute to cross sections are smaller than for the $\Delta S = 0$ collisions, and the momentum transferred to the atom is correspondingly larger. As a result, nondipole interactions become dominating contributors to the total cross section.

This situation is in contrast to $\Delta S = 0$ scattering, where multipole transitions have been traditionally considered, at best, as small corrections to leading dipole transitions (although the nontrivial contribution of nondipole interactions has been repeatedly acknowledged: see, for example, the early calculations of Omidvar [10], the systematic studies of Flannery and McCann [11] and also Syrkin [12], which contains an analytical estimate of the nondipole contribution to the cross section).

Direct numerical calculation of $nl \rightarrow n'l'$ amplitudes is feasible only for relatively small n and unsuitable for massive calculations required at large n. The alternative approach is based on the Heisenberg correspondence principle (HCP) for radial matrix elements (RME) (see the paper of Sobeslavsky [13], the reviews of Percival and

Richards [14] and Flannery [15]). So far this method has been used only for $\Delta S=0$ collisions. In particular, Sobeslavsky [13] obtained an analytic RME expression for inelastic Rydberg-electron collisions $\Delta n=1$, $\Delta l=0$. Naccache [16] solved the case $\Delta l=1$, Matsuzawa [17] calculated RME for the quasi-elastic transitions $\Delta n=0$, $\Delta l>0$. The author [12] analyzed the general case of arbitrary $\Delta n>0$, $\Delta l>0$ and suggested a simple analytical formula for Born nondipole cross sections.

The purpose of this paper is to apply the Heisenberg correspondence principle to intercombination transitions. We will first show that the accuracy of the semiclassical approximation in this case is about the same as that for $\Delta S=0$ transitions, and, second, that such an approach enables one to obtain analytic trends for matrix elements. We will primarily consider transitions between levels with angular momenta, small compared to principal quantum numbers. For such l the multipole expansion for the cross sections is employed and then the correspondence principle is applied to the radial matrix elements (large l values require another approach). The exact quantum calculations with hydrogenic wave functions are used for an assessment of the quality of the semiclassical approach. The results apply to transitions in He with some restrictions to s levels because of nonhydrogenic corrections. In heavier atoms the corrections for levels with the lowest angular momenta (s,p) should be considered with respect to specific values of quantum defects.

The material is organized as follows. In Sec. I we outline the Ochkur approach to intercombination transitions and present the multipole expansion and radial integrals for cross sections. Section II reviews the Heisenberg correspondence principle technique for radial matrix elements. Section III contains analytic results for radial integrals and radial factors for arbitrary Δn and multipole parameter κ, supported by systematic comparison of quantum and semiclassical calculations. It also illustrates the data on collisional transition strengths for low l. In conclusion, we present summary of the results.

II. THE OCHKUR APPROXIMATION

Consider an exchange scattering of a fast electron on a Rydberg atom. The singlet-triplet transition $nl\,^1L \rightarrow n'l'\,^3L'$ in a helium configuration may serve as a canonical example for the process of this type (LS coupling is assumed). The extension of the formalism for more complex configurations does not cause a problem. We use the cross section in the momentum representation

$$\sigma=\frac{2\pi}{kk'}\int_{q_{\min}}^{q_{\max}}|f_{\mathrm{BO}}|^2 q\,dq\ , \qquad q_{\min,\max}=|k-k'|,k+k'\ , \tag{1}$$

where exchange effects are accounted for by the scattering amplitude f_{BO} in the Born-Oppenheimer approximation (see, for example, Mott and Massey [18]), k and k' being the projectile electron wave vectors before and after collision. It has been shown by Ochkur [2], that for this approximation to work correctly only a leading term $1/k^2$ of the f_{BO} asymptotic expansion needs to be retained as $k\rightarrow\infty$. This reduces an exchange amplitude to the factorized form, standard for the first-order plane-wave approximation (see Flannery [15]):

$$|f_{\mathrm{BO}}|^2=\left[\frac{\mu}{2\pi\hbar^2}\right]^2\frac{k}{k_0}M|F_{nl\rightarrow n'l'}(q)|^2|T_{\mathrm{el}}|^2\ , \tag{2}$$

$$|F_{nl\rightarrow n'l'}|^2=\frac{1}{2l+1}\sum_{m,m'}|\langle nlm|e^{iqr}|n'l'm'\rangle|^2\ , \tag{3}$$

where $F_{nl-n'l'}$ is a collisional form factor and where $T_{\mathrm{el}}=4\pi e^2/k^2$, in contrast to $T_{\mathrm{el}}=4\pi e^2/q^2$ for $\Delta S=0$ transitions; M is the spin-angular factor and will be specified below. Therefore the exchange cross section in the Ochkur approximation reads (from now on atomic units with Ry for the energy will be used) as

$$\sigma=\frac{8\pi}{k^6}M\int_{q_{\min}}^{q_{\max}}|F_{nl\rightarrow n'l'}(q)|^2 q\,dq\ , \tag{4}$$

which scales as $\sigma\sim k^{-6}$ for fast collisions.

By expanding the plane wave in the form factor over spherical harmonics we arrive at a standard multipole expansion over radial R factors (see, for example, Sobelman, Vainshtein, and Yukov [3]):

$$\begin{aligned}
&\sigma_{nl\rightarrow n'l'}=\frac{8\pi}{k^6}M\frac{1}{2l+1}\Omega_{nl\rightarrow n'l'}\ ,\\
&\Omega_{nl\rightarrow n'l'}=(2l+1)(2l'+1)\\
&\qquad\times\sum_{\kappa}(2\kappa+1)\begin{pmatrix} l & l' & \kappa\\ 0 & 0 & 0\end{pmatrix}^2 R^{\kappa}_{nl\rightarrow n'l'}\ ,\\
&R^{\kappa}_{nl\rightarrow n'l'}=(nn')^{-2}\int_0^{\infty}|\mathcal{R}^{\kappa}_{nl\rightarrow n'l'}(Q)|^2 Q\,dQ\ ,\\
&\mathcal{R}^{\kappa}_{nl\rightarrow n'l'}(Q)=\int_0^{\infty}j_{\kappa}(Qr)P_{nl}(r)P_{n'l'}(r)\,dr\ ,\\
&\kappa=\Delta l,\Delta l+2,\Delta l+4,\ldots,l+l';\Delta l=|l'-1|\ ,
\end{aligned} \tag{5}$$

where $P_{nl}(r)$ is a radial wave function, multiplied by r; $j_{\kappa}(Qr)$ is a spherical Bessel function; $Q=nn'q$; and $M=(2S+1)/2(2S_p+1)$, S and S_p being the spins of the atom and atomic core, respectively, and where we have introduced for convenience the collisional transition strength Ω. As we are interested in fast collisions and because the integral over Q in (5) converges both for small and large momenta, we have also extended the range of integration from $Q=0$ to ∞.

Formulas (5) present closed-form solutions for any l-resolved intercombination cross sections. When performed with exact wave functions, however, the procedure (5) becomes laborious as n increases. In addition, another aspect complicates matters further for intercombination transitions. The integration over momentum in (5) is dominated by the region close to the major maximum of the radial integral, i.e., roughly speaking $Q>\max(\Delta n,\kappa)$. This maximum changes with Δn and κ relatively slow and as a result the same is true for radial factors $R^{\kappa}_{nl\rightarrow n'l'}$. Therefore, all multipoles κ should be included in (5), in contrast to the $\Delta S=0$ transitions where the leading term $\kappa_0=\Delta l$ is already a good approximation. Solving this problem, the Heisenberg correspondence

principle can serve as an efficient alternative, consistent with the spirit of highly excited levels.

III. HEISENBERG CORRESPONDENCE PRINCIPLE FOR RADIAL MATRIX ELEMENTS

The Heisenberg correspondence principle (HCP) evaluates matrix elements at the limit of $n \to \infty$ as classical Fourier components of the dynamical variable along the trajectory of Rydberg electron. It has been used on various occasions, for example, in Refs. [13–15].

As we consider here low orbital numbers, HCP is utilized not directly to the complete matrix elements $\langle nlm|e^{iqr}|n'l'm'\rangle$, but for radial elements $\langle nl|j_\kappa(Qr)|n'l'\rangle$ in accordance with multipole expansion (5). This yields one-dimensional Fourier components of $j_\kappa(Qr)$ over the angular variable θ_n, conjugate to the action $I_n = n\hbar$, i.e.,

$$\langle nl|j_\kappa(Qr)|n'l'\rangle \sim \frac{1}{2\pi}\int_0^{2\pi} j_\kappa(Qr)e^{-i\Delta n\theta_n}d\theta_n \ ,$$
$$r=(1-\varepsilon\cos u) \ , \qquad (6)$$
$$\theta_n = u - \varepsilon\sin u \ ,$$
$$\varepsilon=(1-l_{\max}^2/n^2)^{1/2} \ ,$$

under the conditions that $\max(\Delta n,\Delta l)\ll n,n'$, and $l,l'\ll n,n'$, u and ε being the eccentric anomaly and eccentricity, respectively. It should be emphasized that the correspondence principle in form (6) does not represent, strictly speaking, the exact limit of radial matrix elements as $n\to\infty$ (see [14]), but rather the semiclassical estimate, valid for $\Delta l \to 0$. Indeed, the matrix element $\langle nl|j_\kappa(Qr)|n'l'\rangle$ depends both on l and l', whereas the semiclassical integral (6) does not. An analytical evaluation and numerical results given below show that the semiclassical approximation (6) furnishes the best accuracy in the case of small Δl and $l\ll n$.

IV. RESULTS AND DISCUSSION

Quantum-mechanical calculations for radial factors $R^\kappa_{nl\to n'l'}$ were performed with hydrogenic wave functions. For semiclassical matrix elements we used (6) with $\varepsilon=1$, since for small orbital numbers $l\ll n$ where the present formalism is efficient the deviation of ε from unity has no affect on the results. Consider first the extreme cases, $\kappa=0$ and $\Delta n=0$.

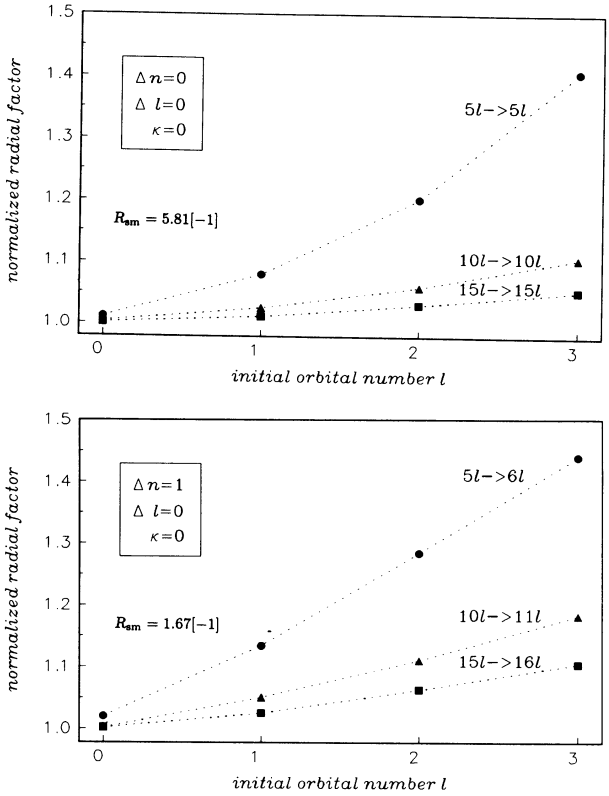


FIG. 1. Quantum radial factors as a function of l for different n, in units of the semiclassical factor for the same Δn and κ. The numbers in brackets denote multiplicative powers of ten. (a) $\Delta n=0$, $\Delta l=0$, $\kappa=0$; and (b) $\Delta n=1$, $\Delta l=0$, $\kappa=0$.

A. $\kappa=0$, arbitrary n change

With the aid of the Poisson representation of Bessel functions (see, for example [19]), (6) immediately integrates to

$$|\mathcal{R}^{(0)}_{\Delta n}(Q)| = \frac{1}{Q}\left|\sin\left[q-\Delta n\tan^{-1}\left[\frac{Q}{\Delta n}\right]\right]\right.$$
$$\left.\times J_{\Delta n}[(\Delta n^2+Q^2)^{1/2}]\right| \ , \qquad (7)$$

TABLE I. Semiclassical radial factors $(nn')^2R^\kappa_{\Delta n}$ in units of α_0^{-4} [Eq. (6)]. Power of 10 notations are used, e.g., $5.81[-1]=5.81\times10^{-1}$.

κ \ Δn	0	1	2	3	4	5	6	8	10
0	5.81[−1]	1.67[−1]	1.11[−1]	8.62[−2]	7.18[−2]	6.22[−2]	5.52[−2]	4.57[−2]	3.94[−2]
1	3.18[−1]	1.08[−1]	7.34[−2]	5.76[−2]	4.82[−2]	4.19[−2]	3.73[−2]	3.09[−2]	2.67[−2]
2	2.25[−1]	8.45[−2]	5.84[−2]	4.62[−2]	3.89[−2]	3.39[−2]	3.02[−2]	2.51[−2]	2.17[−2]
3	1.76[−1]	7.09[−2]	4.98[−2]	4.00[−2]	3.35[−2]	2.93[−2]	2.62[−2]	2.18[−2]	1.89[−2]
4	1.46[−1]	6.17[−2]	4.40[−2]	3.52[−2]	2.99[−2]	2.62[−2]	2.34[−2]	1.96[−2]	1.70[−2]
5	1.28[−1]	5.50[−2]	3.96[−2]	3.19[−2]	2.72[−2]	2.39[−2]	2.14[−2]	1.79[−2]	1.56[−2]
6	1.10[−1]	4.98[−2]	3.63[−2]	2.94[−2]	2.51[−2]	2.21[−2]	1.98[−2]	1.66[−2]	1.44[−2]
8	8.66[−1]	4.35[−2]	3.18[−2]	2.68[−2]	2.28[−2]	1.98[−2]	1.79[−2]	1.55[−2]	1.36[−2]

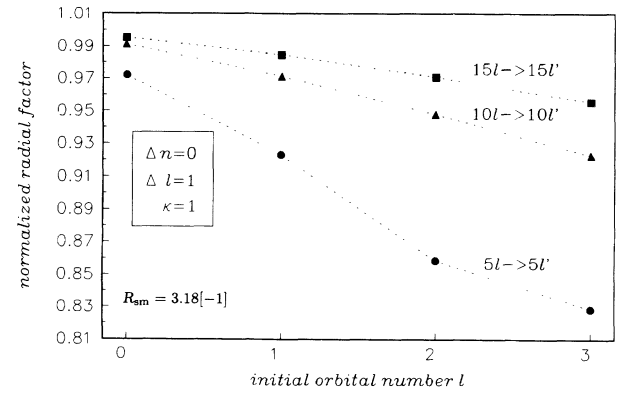
where $J_{\Delta n}$ are integer order Bessel functions. As indicated by Beigman and Syrkin [20], the identical result follows from a quantum treatment of $\langle nl|j_\kappa(Qr)|n'l\rangle$ for arbitrary Δn by means of the Tricomi expansion for hypergeometric functions. Details of calculations are given in the Appendix.

Therefore, when $l \ll n$, the semiclassical and quantum calculations for radial factors R^κ should be in a fairly good agreement as n increases. The comparison is illustrated in Fig. 1 for $\Delta n=0,1$. Throughout all the figures we present the quantum-to-semiclassical ratio of radial factors. The semiclassical radial factors are identified by Δn, κ, and absolute values $R_{\rm sm}$. Also, the numerical data for semiclassical radial factors $R^\kappa_{\Delta n}$ are summarized in Table I. The discrepancy between quantum and semiclassical results rises as l increases, but remains within about 20% for all l up to $l/n \simeq \frac{1}{3}$.

When integrating expression (7) over momentum the major contribution comes from $Q \geq \Delta n$. This yields an analytical approximation for radial factors in the form

$$R^0_{\Delta n}=(nn')^{-2}(\Delta n)^{-2/3}\mathcal{H}_1(\Delta n)\ , \tag{8}$$

where the constant $\mathcal{H}_1(\Delta n)$ varies only slightly with Δn. Comparing (8) with the numerical integration of (7), one has $\mathcal{H}_1=1.67,\ 1.76,\ 1.79,\ 1.81,\ 1.81,\ 1.82\times10^{-1}$ for $\Delta n=1,2,3,4,5,6$, respectively.



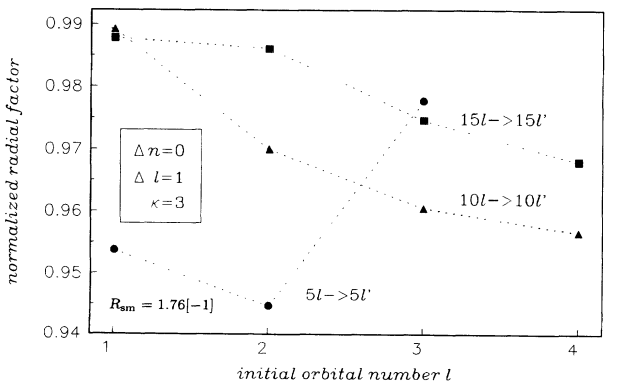


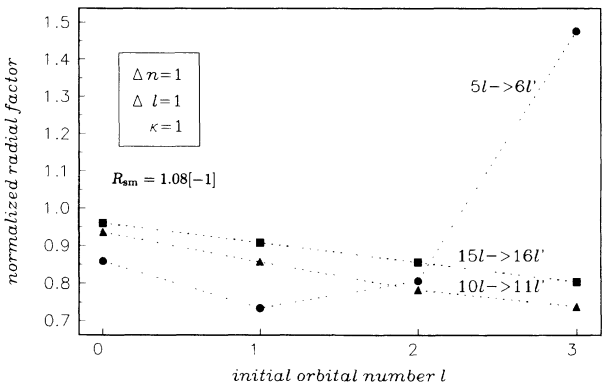
FIG. 2. Quantum radial factors as a function of l for different n, in units of the semiclassical factor for the same Δn and κ. The numbers in brackets denote multiplicative powers of ten. (a) $\Delta n=0$, $\Delta l=1$, $\kappa=1$; and (b) $\Delta n=0$, $\Delta l=1$, $\kappa=3$.

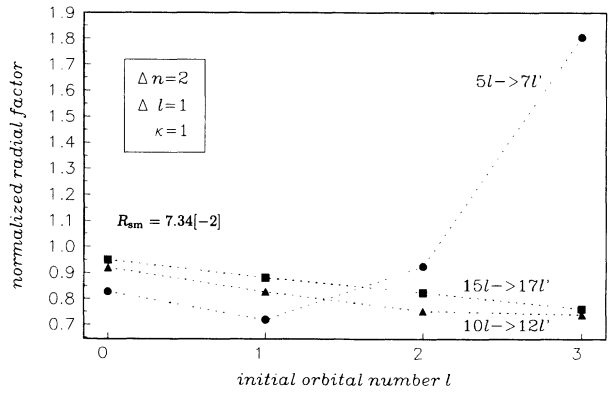
B. $\Delta n=0$, arbitrary κ change

The comparison between semiclassical and quantum calculations is given in Fig. 2. The results show the good quality of the semiclassical approximation especially for $l/n \ll 1$. A difference exceeding $\sim 10\%$ is observed only when l becomes comparable with n.

The semiclassical expressions of RME readily follows from (6) and were first obtained by Matsuzawa [17]:

$$\begin{aligned}\langle nl|j_{2m}(Qr)|n'l'\rangle &= j_m(Q)J_m(Q)\ ,\\ \langle nl|j_{2m+1}(Qr)|n'l'\rangle &= j_m(Q)J_{m+1}(Q)\ ,\end{aligned} \tag{9}$$





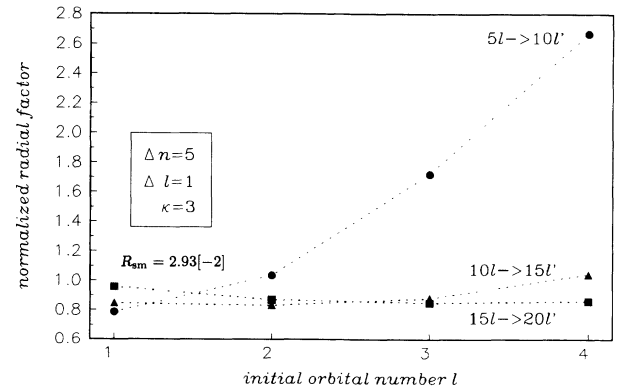


FIG. 3. Quantum radial factors as a function of l for different n, in units of the semiclassical factor for the same Δn and κ. The numbers in brackets denote multiplicative powers of ten. (a) $\Delta n=1$, $\Delta l=1$, $\kappa=1$; and (b) $\Delta n=2$, $\Delta l=1$, $\kappa=1$; (c) $\Delta n=5$, $\Delta l=1$, $\kappa=3$.

[Equation (9) for $m=0$ results from (6) through the Poisson representation and the case $m>0$, in turn, comes from expression for $m=0$ by means of integration over the parameter method].

Using an asymptotic expansion of matrix elements Beigman and Syrkin [20] showed the exact quantum result for $Q<1$ to differ from (9) only by the "eccentricity" type factor, namely,

$$\varepsilon_\kappa=\left\{\prod_{i=1}^{\kappa}\left[1-\frac{(l+1-i)^2}{n^2}\right]\right\}^{1/2}, \tag{10}$$

so that for $\kappa=1$ (10) reduces to "natural" ε from (6) and

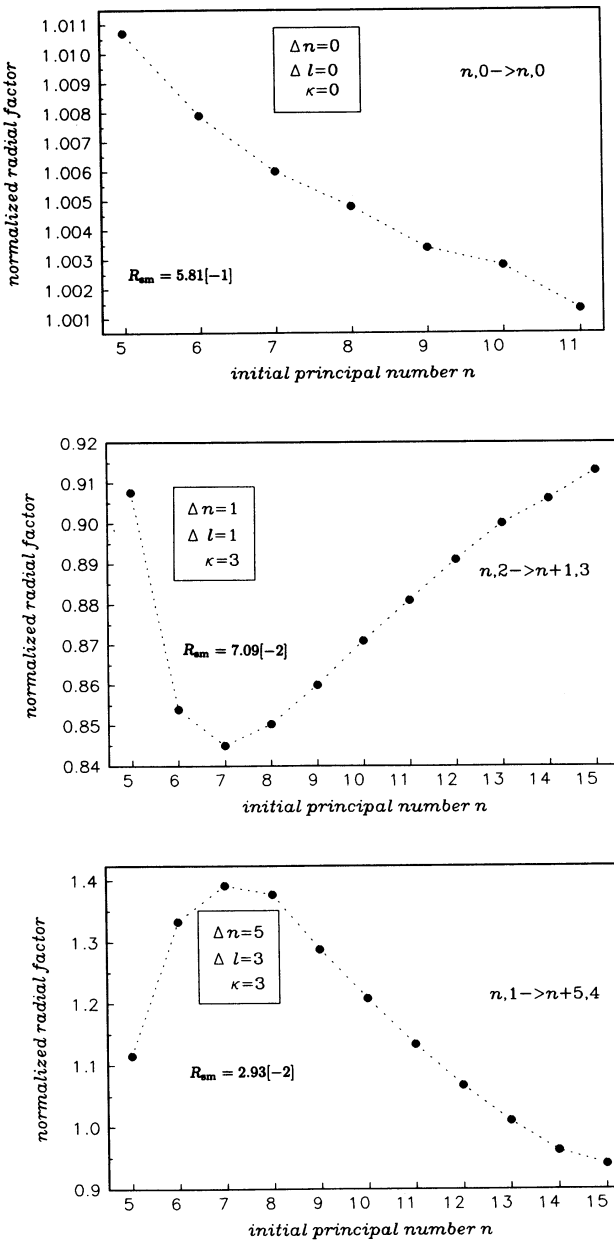


FIG. 4. n dependence of quantum radial factors, in units of the semiclassical factor for the same Δn and κ. (a) $\Delta n=0$, $\Delta l=0$, $\kappa=0$; (b) $\Delta n=1$, $\Delta l=1$, $\kappa=3$; and (c) $\Delta n=5$, $\Delta l=3$, $\kappa=3$.

for $\kappa>1$ we have a product of κ terms (the same expression has been later reproduced by Heim, Trautmann, and Baur [21]).

For intercombination transitions the major contribution comes from $Q>\kappa$ where quantum correction (10) is not valid. An analytic estimate for the radial factor (5), in this case, may be obtained either from (9) or directly from representation (6). The latter proves somewhat more convenient technically and, noticing that J_κ scales as $\kappa^{-1/3}$ at maximum, we have

$$R^{\kappa}_{\Delta n=0}=n^{-4}\kappa^{-2/3}\mathcal{H}_2(\kappa)\ , \tag{11}$$

where the constant $\mathcal{H}_2(\kappa)$ is again a slow function of κ: $\mathcal{H}_2=3.18,\ 3.57,\ 3.66,\ 3.68,\ 3.66,\ 3.64\times10^{-1}$ for $\kappa=1,2,3,4,5,6$, respectively.

C. Arbitrary $\Delta n>0$ and $\kappa>0$ changes

For arbitrary Δn and κ the quality of the semiclassical approach is illustrated in Fig. 3 for $\Delta n=1$, 2, and 5. The typical errors are about 20%. The errors are smallest for minimal possible Δl and gradually increase with Δl and l; the maximal discrepancy of $\sim40\%$ occurs only in the cases when $l\sim n$. In Fig. 4 we also present the accuracy of semiclassical results as a function of n. It is seen, first, that the convergence to the classical expression (6) is not necessarily monotonic and, second, that the ultimate precision depends on the actual parameters of the transition: Δn and Δl, l, and n. Also, these results enable the conclusion that overall, the accuracy of semiclassical matrix elements for exchange transitions is quite similar to that reported for the $\Delta S=0$ transitions (see, for example, [3,12,14,17,20]).

An analytical evaluation of RME in the general case is not readily obtainable. Instead, consider an asymptotic expansion of the radial integral, valid for $Q\gg\Delta n,\kappa$:

$$|\mathcal{R}^{\kappa}_{\Delta n}(Q)|\approx\frac{1}{Q}\left|\sin\left[Q-(\Delta n+\Delta l)\frac{\pi}{2}\right]\times J_{\Delta n}[(\Delta n^2+Q^2)^{1/2}]\right| . \tag{12}$$

The study of this expression along with formulas (8) and (11) suggests the general approximation of radial factors in the form

$$R^{\kappa}_{\Delta n}=(nn')^{-2}\frac{\max(\Delta n^{-1/3},\kappa^{-1/3})}{\Delta n^{1/3}\kappa^{1/3}}\mathcal{H}_3(\Delta n,\kappa)\ . \tag{13}$$

TABLE II. Correction factor $\mathcal{H}_3(\Delta n,\kappa)$, 10^{-1} [Eq. (13)].

κ \ Δn	1	2	3	4	5	6
1	1.08	1.17	1.20	1.21	1.22	1.23
2	1.34	1.17	1.21	1.23	1.25	1.26
3	1.46	1.31	1.20	1.22	1.23	1.25
4	1.55	1.40	1.28	1.20	1.21	1.23
5	1.61	1.46	1.35	1.26	1.20	1.21
6	1.65	1.51	1.40	1.31	1.21	1.20

The behavior of the quasiconstant function $\mathcal{H}_3(\Delta n,\kappa)$ is summarized in Table II. As it follows from (11) and (13), the radial factors R undergo only relatively slow change as functions of κ, namely as $\sim\kappa^{-1/3}$ or $\kappa^{-2/3}$. Therefore, the important conclusion is that all terms in the multipole expansion (5) are practically equally important and as such should be taken into consideration.

D. Cross sections

In the limit of fast collisions the cross sections depend on the projectile velocity as $\sigma\sim k^{-6}$. The dependence on n, l, and Δn and Δl can conveniently be characterized by means of the collisional transition strength Ω. Some of the results for low l are presented in Table III (the data for Ω in this table do not pretend to provide ultimate quantitative accuracy; the data rather illustrate the error, introduced by the semiclassical approximation itself, independent of corrections for nonhydrogenic effects, especially for s and p levels).

Two major conclusions can be drawn from these data. First, for levels with low orbital numbers the semiclassical approach provides reasonable results for radial integrals and cross sections compared to direct quantum calculations, the precision being consistent with that of the Born approximation. Second, the cross sections of dipole transitions demonstrate no dominance over nondipole transitions. Therefore, any calculation of the total cross section should incorporate the contribution from multipole interactions.

TABLE III. Quantum transition strengths $(nn')^2\Omega_{\text{quant}}$ and ratios $A=\Omega_{\text{quant}}/\Omega_{\text{sm}}$.

		Ω	A			Ω	A
	$\Delta n=0$				$\Delta n=1$		
$s\to s$	$5s\to 5s$	0.588	1.010	$s\to s$	$5s\to 6s$	0.171	1.020
	$10s\to 10s$	0.583	1.003		$10s\to 11s$	0.168	1.002
	$15s\to 15s$	0.582	1.001		$15s\to 16s$	0.168	1.002
$s\to p$	$5s\to 5p$	0.927	0.972	$s\to p$	$5s\to 6p$	0.279	0.859
	$10s\to 10p$	0.945	0.991		$10s\to 11p$	0.304	0.936
	$15s\to 15p$	0.948	0.955		$15s\to 16p$	0.310	0.960
$s\to d$	$5s\to 5d$	0.948	0.844	$s\to d$	$5s\to 6d$	0.313	0.748
	$10s\to 10d$	1.05	0.937		$10s\to 11d$	0.344	0.814
	$15s\to 15d$	1.09	0.964		$15s\to 16d$	0.368	0.871
$s\to f$	$5s\to 5f$	0.854	0.693	$s\to f$	$5s\to 6f$	0.613	1.236
	$10s\to 10f$	1.05	0.848		$10s\to 10f$	0.383	0.773
	$15s\to 15f$	1.12	0.908		$15s\to 16f$	0.393	0.793
	$\Delta n=2$				$\Delta n=5$		
$s\to s$	$5s\to 7s$	0.113	1.018	$s\to s$	$5s\to 10s$	0.062	0.998
	$10s\to 12s$	0.111	1.000		$10s\to 15s$	0.061	0.990
	$15s\to 17s$	0.111	1.010		$15s\to 20s$	0.061	0.994
$s\to p$	$5s\to 7p$	0.182	0.828	$s\to p$	$5s\to 10p$	0.097	0.782
	$10s\to 12p$	0.202	0.919		$10s\to 15p$	0.112	0.888
	$15s\to 17p$	0.208	0.949		$15s\to 20p$	0.117	0.979
$s\to d$	$5s\to 7d$	0.226	0.775	$s\to d$	$5s\to 10d$	0.137	0.811
	$10s\to 12d$	0.231	0.791		$10s\to 15d$	0.130	0.769
	$15s\to 17d$	0.246	0.849		$15s\to 20d$	0.137	0.806
$s\to f$	$5s\to 7f$	0.596	1.360	$s\to f$	$5s\to 10f$	0.277	1.330
	$10s\to 12f$	0.372	0.818		$10s\to 15f$	0.185	0.889
	$15s\to 17f$	0.288	0.786		$15s\to 20f$	0.174	0.838
	$\Delta n=0$				$\Delta n=1$		
$p\to p$	$5p\to 5p$	3.309	1.071	$p\to p$	$5p\to 6p$	1.120	1.109
$d\to d$	$5d\to 5d$	7.546	1.182	$d\to d$	$5d\to 6d$	2.768	1.239
	$\Delta n=2$				$\Delta n=5$		
$p\to p$	$5p\to 7p$	0.763	1.115	$p\to p$	$5p\to 10p$	0.427	1.095
$d\to d$	$5d\to 7d$	1.908	1.243	$d\to d$	$5d\to 10d$	1.104	1.241
	$\Delta n=0$				$\Delta n=1$		
$p\to d$	$5p\to 5d$	3.265	0.936	$p\to d$	$5p\to 6d$	1.005	0.781
$p\to f$	$5p\to 5f$	2.838	0.752	$p\to f$	$5p\to 6f$	1.635	1.090
	$\Delta n=2$				$\Delta n=5$		
$p\to d$	$5p\to 7d$	0.683	0.769	$p\to d$	$5p\to 10d$	0.348	0.783
$p\to f$	$5p\to 7f$	1.308	1.242	$p\to f$	$5p\to 10f$	0.872	1.410

This last conclusion looks quite reasonable from the viewpoint of the Ochkur approximation, relating the amplitudes of spin-exchange transitions to the spin-conserving ones. We have already mentioned in the Introduction that the contribution of nondipole interactions is significant for $\Delta S=0$ transitions with large Δn (as it was particularly emphasized in [11] and, also, in [12]). It is natural, then, that they are even more important for intercombination transitions.

V. CONCLUSIONS

We have presented the treatment of intercombination transitions in Rydberg atoms within the framework of the Ochkur approximation, which allows for a transparent closed-form representation of the cross sections by means of multipole expansion. Further, the Heisenberg correspondence principle has been employed to evaluate radial integrals. The results obtained shows that, overall, for orbital numbers l,l' (and Δl) small enough compared to n,n' the semiclassical approximation and exact quantum calculations agree to within 10% to 20%. This is about the same accuracy that is typical for semiclassical methods for spin-conserving transitions as well as for the Born approximation itself.

We have also shown that the validity range of the semiclassical approximation proves broader than that of the quantum asymptotic expansion for matrix elements. The latter applies mostly only for relatively small momenta $Q<1$ transferred to the atom, whereas the correspondence principle provides reasonable accuracy also for large momenta, which is important for intercombination transitions.

The discrepancy between quantum and semiclassical cross sections becomes significant for $l\sim n$. This fact, however, should not be considered as a failure of a semiclassical approach itself, but rather as an insufficiency of the semiclassical approximation for radial matrix elements for large l and Δl. The correct approach for large orbital numbers is to apply the Heisenberg correspondence principle directly to the matrix elements $\langle nlm|e^{iqr}|n'l'm'\rangle$. This will be the subject of a separate work.

For a quality assessment of the above approach as a whole and numerical results in particular, as well as for further progress, it is of primary importance to have experimental results on intercombination cross sections in collisions of electrons with Rydberg atoms.

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APPENDIX

The integral to evaluate is

$$I=\int_0^\infty P_{nl}(r)j_0(Qr)P_{n'l}(r)dr\ . \tag{A1}$$

Since

$$P_{nl}(r)=\frac{2}{n^{l+2}\Gamma(2l+2)}\left[\frac{\Gamma(n+l+1)}{\Gamma(n-l)}\right]^{1/2}(2r)^{l+1}e^{-r/n}F_1\left[-n+l+1,2l+2,\frac{2r}{n}\right], \tag{A2}$$

$j_0(x)=\sin(x)/x$ and $F_1(a,b;x)$ is a confluent hypergeometric function and we have

$$\begin{aligned}I=\frac{nn'}{2Qi}C(n,n',l)\Bigg\{&\left[\frac{1}{n'}+\frac{1}{n}-i\frac{Q}{nn'}\right]^{\alpha+\alpha'-\gamma}\left[\frac{1}{n'}-\frac{1}{n}-i\frac{Q}{nn'}\right]^{-\alpha}\left[\frac{1}{n}-\frac{1}{n'}-i\frac{Q}{nn'}\right]^{-\alpha'}\\&\times F\left[\alpha,\alpha',\gamma,-\frac{4nn'}{[(\Delta n)^2+Q^2]^{1/2}}\right]\\&-\left[\frac{1}{n'}+\frac{1}{n}+i\frac{Q}{nn'}\right]^{\alpha+\alpha'-\gamma}\left[\frac{1}{n'}-\frac{1}{n}+i\frac{Q}{nn'}\right]^{-\alpha}\left[\frac{1}{n}-\frac{1}{n'}+i\frac{Q}{nn'}\right]^{-\alpha'}\\&\times F\left[\alpha,\alpha',\gamma,-\frac{4nn'}{[(\Delta n)^2+Q^2]^{1/2}}\right]\Bigg\},\end{aligned} \tag{A3}$$

where $\alpha=-n+l+1$, $\alpha'=-n'+l+1$, $\gamma=2l+2$, and $F(a,b,c;x)$ is a hypergeometric function and $C(n,n',l)$ is some constant. Using then an asymptotic Tricomi expansion for $F(-n,-n',\gamma;s)$ from (A3) (see Beigman and Syrkin [20]),

$$F(-n,-n',\gamma;s)\approx\frac{\Gamma(\gamma)\Gamma(n'+1)}{\Gamma(n+\gamma)}\left[\frac{(n+\gamma-1)(n+n'+1)}{2}\right]^{-\Delta n/2}e^{-i(\pi/2)n_S(n+n')/2}$$
$$\times J_{\Delta n}\left[\left[\frac{(n+\gamma-1)(n+n'+1)}{2}\right]^{1/2}\right], \tag{A4}$$

valid for $n,n'\to\infty$, $(\Delta n/n)\to 0$, $|s|\sim n^2$. After the series of elementary transformations we arrive at the result

$$|I|=\frac{1}{Q}\left|\sin\left[Q-\Delta n\tan^{-1}\left[\frac{Q}{\Delta n}\right]\right]J_{\Delta n}[(\Delta n^2+Q^2)^{1/2}]\right|. \tag{A5}$$

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