l-resolved intercombination transitions in Rydberg atoms in collisions with electrons

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A systematic study of semiclassical and quantum cross sections of l-resolved intercombination transitions in Rydberg atoms due to collisions with fast electrons is performed in the Ochkur approximation for principal quantum numbers $n \leq 15$ and all possible orbital numbers l and changes Δl. The semiclassical approach provides reasonable estimates for cross sections over the angular-momentum range up to $l \sim n/2$. Intercombination transitions are shown to obey the "Bethe rule": at large l the transitions with n and l changing in the same direction dominate transitions with changes in opposite directions. It is also demonstrated that the more deeply inelastic the collision is, the higher the multipoles Δl that prevail in the total cross section. The semiclassical approach makes it possible to derive simple analytic formulas for cross sections with an explicit dependence on principal and orbital quantum numbers.

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I. INTRODUCTION

The purpose of this paper is to conclude the study of intercombination transitions in Rydberg atoms in collisions with electrons initiated in the previous work [1]. In that work, which hereafter will be referred to as paper I, we have used the Ochkur approximation as a suitable approach to fast collisions with spin change [2]. Radial matrix elements were evaluated by means of the Heisenberg correspondence principle to utilize the quasiclassical properties of highly excited levels [3–6]. That approach enabled one to extract analytic expressions and trends for matrix elements and radial factors and to show that for orbital numbers l,l' (and Δl) small compared to principal quantum numbers n,n', the semiclassical cross sections were in good accord with the exact quantum calculations.

This work extends the treatment to arbitrary angular momenta. Large angular momenta are important, because, first, they furnish the major contribution to the total l-averaged cross section $n \rightarrow n'$ and, second, the corresponding wave functions are clear from nonhydrogenic corrections due to the quantum defect (in contrast to low angular momenta).

We will present a detailed comparison of theoretical quantum and semiclassical cross sections for arbitrary l. The data show that the quality of the semiclassical approximation remains fairly high well beyond the region of its formal validity, i.e., small l values. Especially good accuracy is found for transitions when the principal and orbital quantum numbers change in opposite directions. The results also demonstrate that the intercombination transitions obey the "Bethe rule" [7], much like dipole radiative transitions do: at large l the transitions with n and l changing in the same direction dominate over transitions in opposite directions. Further, we present the evidence of an immediate correlation between the energy transferred to the atom in the course of the collision, i.e., Δn, and the value of Δl for the most intensive l-resolved transitions. In conclusion, we show that, if the orbital numbers l,l' involved are not very small, it is possible, based on analytical trends obtained in paper I, to perform the summation in the multipole expansion and derive simple closed-form approximate formulas for cross sections. These formulas explicitly depend on the initial and final principal and orbital quantum numbers and may serve as guiding estimates.

The paper has the following structure. Section II contains the systematic comparison of quantum and semiclassical calculations along with the discussion of major trends. Section III presents analytic formulas for collision strengths. The summary of the results is given in Sec. IV. Atomic units with Ry for the energy are used throughout the paper.

II. COMPARISON OF QUANTUM AND SEMICLASSICAL RESULTS

Consider the transition $nlL \rightarrow n'l'L'$ with spin change $\Delta S = 1$(LS coupling is assumed) in a Rydberg atom, induced by an exchange collision with a fast electron. According to paper I [1], the Ochkur approximation for the cross section averaged over magnetic quantum numbers gives

$$\sigma_{nl\rightarrow n'l'} = \frac{8\pi}{k^6} M \frac{1}{2l+1} \Omega_{nl\rightarrow n'l'} ,$$
$$\Omega_{nl\rightarrow n'l'} = (2l+1)(2l'+1) \times \sum_{\kappa} (2\kappa+1) \begin{bmatrix} l & l' & \kappa \\ 0 & 0 & 0 \end{bmatrix}^2 \mathcal{R}^{\kappa}_{nl\rightarrow n'l'} , \quad (1)$$

where k is the magnitude of the projectile wave vector, and $M = (2S+1)/2(2S_p+1)$, S and S_p being spins of the atom after collision and the atomic core. Radial factors $\mathcal{R}^{\kappa}_{nl\rightarrow n'l'}$ were obtained with hydrogenic wave functions and also, as an alternative, in the semiclassical approximation

$$\langle nl | j_\kappa(Qr) | n'l' \rangle \sim (2\pi)^{-1} \int j_\kappa(Qr) \exp(-i\Delta n \theta) d\theta$$

where $r = (1-\varepsilon \cos u)$, $\theta = u - \varepsilon \sin u$, u and ε being the eccentric anomaly and eccentricity, respectively. Based on (1) we have systematically compared quantum and

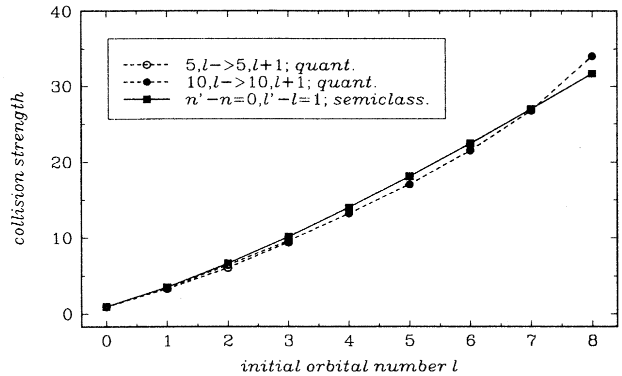

FIG. 1. Quantum and semiclassical collision strengths $(nn')^2\Omega_{nl\to n'l'}$ as functions of l for $\Delta n=0$, $\Delta l=1$.

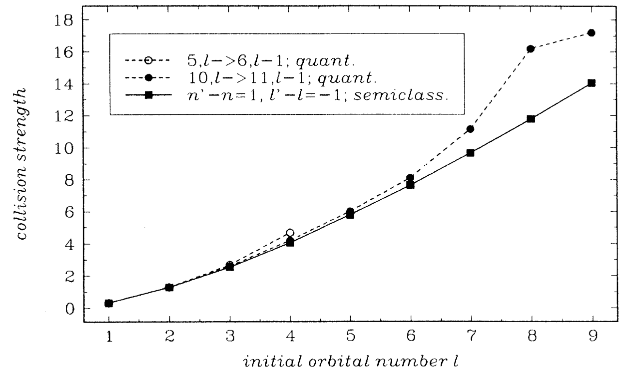

FIG. 3. Quantum and semiclassical collision strengths $(nn')^2\Omega_{nl\to n'l'}$ as functions of l for $\Delta n=1$, $\Delta l=-1$.

semiclassical exchange cross sections for principal quantum numbers $n\le 15$ and all possible orbital numbers l and changes $\Delta l=l'-l$. Below we illustrate the most typical trends for some selected values of n and l changes. All numerical and analytic results are presented in the form of collision strengths Ω as this simplifies the formulas and gives an idea of the relative contributions of transitions with different Δl to the total cross section $n\to n'$.

Elastic transitions $\Delta n=0$ are illustrated in Fig. 1. As expected from paper I, for small l and small changes $\Delta l=l-l'$ the semiclassical results either coincide or are very close to quantum calculations. It is seen, however, that the correspondence principle provides reasonable accuracy even for l that are not actually small. By "reasonable" accuracy we mean an error up to 20–30 %, which is typical for Born-type estimates. In fact, that level of discrepancy is not exceeded approximately up to $l\sim n/2$. We would like also to point out the nonmonotonic relationship between semiclassical and quantum results for elastic transitions. The semiclassical calculations underestimate the cross sections for $\Delta l=0$ and overestimate for $\Delta l>1$. For dipole transitions $\Delta l=1$ both methods seem to be consistently close for all l.

Consider now inelastic transitions $\Delta n>0$. The results are depicted in Figs. 2–5. First of all we observe that the main tendency persists. The quality of the semiclassical approximation remains fairly acceptable within a range of $l\le n/2$. The worst discrepancies over a factor of 2 are found only for maximal $l=n-1$. Further, it is seen that the semiclassical transition strengths always rise smoothly as l increases. Quantum results demonstrate mostly the same trend. Deviations from monotonicity and even quasioscillatory behavior are possible, however, especially for large l and Δl. The reason is that radial factors $\mathcal{R}$ from (1) are not, strictly speaking, smooth functions of l and Δl and the calculations usually show clear evidence of that fact. However, the multipole summation over κ in (1) and the factor of $(2l+1)(2l'+1)$ mitigate this feature, so that traces of nonmonotonicity occasionally show up in the transition strengths, as in Fig. 4.

Interestingly enough, there exists an analogy to the so-called "Bethe rule" for intercombination transitions as well. Bethe and Salpeter indicated [7] that for large l the dipole radiative transitions with n and l changing in the same direction dominate over transitions with opposite n and l change. The comparison of Fig. 2 to Fig. 3 and Fig. 4 to Fig. 5 indicates a similar phenomenon for collisionally induced transitions with spin change. At the same time, as is seen in Figs. 2–5, for inelastic transitions semiclassical strengths are consistently smaller than quantum ones. As a result, the quality of the semiclassical approximation is appreciably higher for transitions with opposite n and l changes, compared to "parallel" change transitions. In fact, in the former case the semiclassical results closely reproduce quantum data practically for all l.

Finally we resort to contributions of different Δl to the

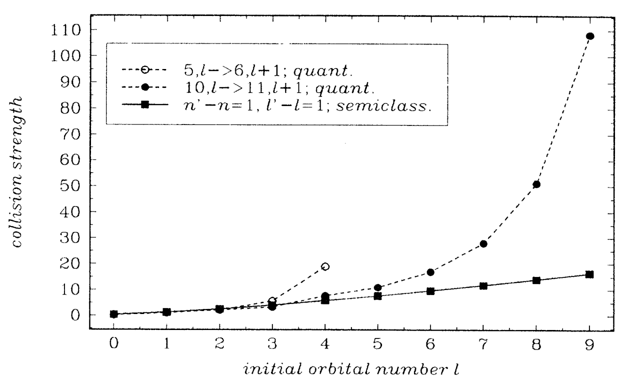

FIG. 2. Quantum and semiclassical collision strengths $(nn')^2\Omega_{nl\to n'l'}$ as functions of l for $\Delta n=1$, $\Delta l=1$.

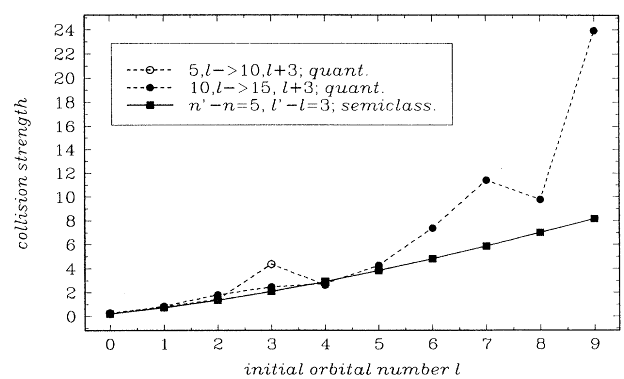

FIG. 4. Quantum and semiclassical collision strengths $(nn')^2\Omega_{nl\to n'l'}$ as functions of l for $\Delta n=5$, $\Delta l=3$.

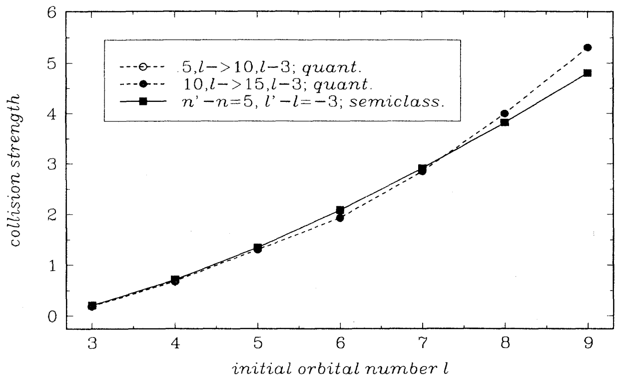

FIG. 5. Quantum and semiclassical collision strengths $(nn')^2\Omega_{nl\to n'l'}$ as functions of l for $\Delta n=5$, $\Delta l=-3$.

total cross section $n\to n'$. In paper I we have already indicated that for spin-change collisions the dipole-allowed transitions had no dominance over dipole-forbidden transitions. Here we would like to elaborate this point further. In fact, there exists a correlation between the Δl value of the transitions that contribute most to the total cross section $n\to n'$ and the extent to which the collision is inelastic, i.e., Δn. In particular, as shown in Fig. 6, for elastic collisions $\Delta n=0$ the collision strength is dominated by monopole and dipole transitions $\Delta l=0,1$. For moderately inelastic collisions $\Delta n=1$ the leading contribution comes from dipole transitions. And for strongly inelastic collisions such as $\Delta n=5$ in Fig. 7 the major role shifts to multipole interactions $\Delta l\gg 1$. This trend is consistent with previous studies by Flannery and McCann [8] and the author [9] and has a natural interpretation. Elastic $\Delta n=0$ and slightly inelastic $\Delta n=1$ collisions are dominated by the long-range projectile-atom interactions, corresponding to small $\Delta l\sim 1$. Large energy transfer to the atom $\Delta n\gg 1$ occurs at relatively small impact parameters, associated with higher multipoles and large angular-momentum transfer $\Delta l\gg 1$. This explains the success of the approach to intercombination transitions based on the binary encounter approximation (Stabler [10], Webster, Hansen, and Duveneck [11], Beigman and Matusovsky [12]).

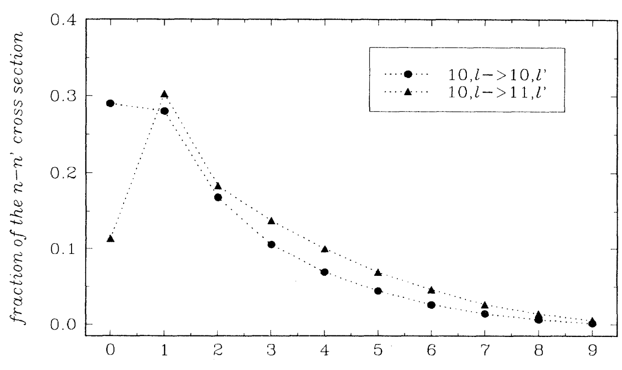

FIG. 6. Relative contribution of all transitions with given $|\Delta l|$ to the total $n\to n'$ strength $\sum_{|\Delta l|}\Omega_{nl\to n'l'}/\Omega_{n\to n'}$ as a function of $|\Delta l|$. $\Delta n=0$ and $\Delta n=1$.

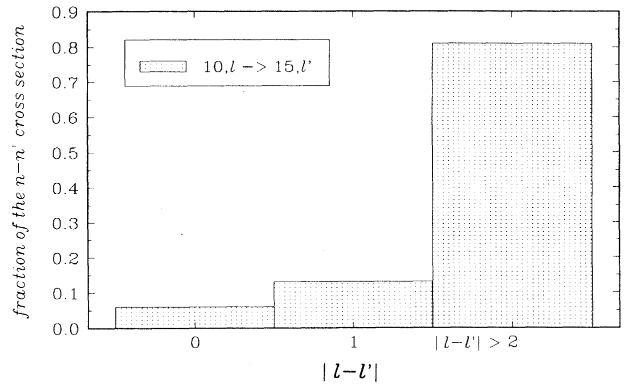

FIG. 7. Relative contribution of all transitions with given $|\Delta l|$ to the total $n\to n'$ strength $\sum_{|\Delta l|}\Omega_{nl\to n'l'}/\Omega_{n\to n'}$ as a function of $|\Delta l|$. $\Delta n=5$.

III. ANALYTIC FORMULAS FOR COLLISION STRENGTHS

Inasmuch as the semiclassical approach furnishes a reasonable accuracy over the fairly broad range of orbital quantum numbers l, it makes it advisable to obtain closed-form analytic formulas based on this approximation.

To that end we use the analytic trends derived in paper I: $\mathcal{R}\sim(\Delta n)^{-2/3}$ if $\kappa=0$; $\mathcal{R}\sim\kappa^{-2/3}$ if $\Delta n=0$; $\mathcal{R}\sim\max(\Delta n^{-1/3},\kappa^{-1/3})\,\Delta n^{-1/3}\kappa^{-1/3}$ if $\Delta n>0$ and $\kappa>0$. Then by means of the completeness conditions for $3j$ symbols along with the asymptotic behavior $\begin{pmatrix} l & l' & \kappa \\ 0 & 0 & 0 \end{pmatrix}^2\sim 1/\kappa$ when $\kappa\gg\Delta l$, multipole expansion (1) reduces to the following formulas:

(1) *For elastic collisions* $\Delta n=0$:

$$\Omega_{nl\to nl'}=n^{-4}(2l+1)(2l'+1)A\frac{(l'+l)^{1/3}-|l'-l|^{1/3}}{l_{\min}}\,, \tag{2}$$

where $A=0.339$ if $\Delta l=0$ and $A=0.513$ otherwise; $l_{\min}=\min(l,l')$.

(2) *For inelastic collisions* $\Delta n>0$:

$$\Omega_{nl\to n'l'}=(nn')^{-2}(2l+1)(2l'+1)\frac{B}{\Delta n^{1/2}}\times\frac{(l'+l)^{2/3}-|l'-l|^{2/3}}{l_{\min}+C}\,, \tag{3}$$

where $B=0.0638$, $C=0$ if $\Delta n=1$, and $C=[l_{\min}/(l_{\min}+5)]^2$ otherwise.

The validity of formulas (2) and (3) assumes that orbital quantum numbers involved are not very small, i.e., $l_{\min}\geq 2$. In that range the error of (2), (3) varies mostly within 5–10%. The stronger deviations over 10% are possible for $\Delta n>0$, although they are not typical. At the same time, whenever (2) and (3) overestimate the semiclassical results, it in fact improves the quality of approximation because the exact quantum calculations for $\Delta n>0$ always exceed the semiclassical data. For $l_{\min}=0$ or 1 it is advisable to use directly (1) which reduces in this case to one or two terms only.

It is instructive to compare the exchange collision strengths (2), (3) with those for nondipole transitions without spin change [9]:

$$\Omega_{nl\rightarrow n'l'} \sim (nn')^2(2l+1)(2l'+1)(2\Delta l+1) \times \begin{bmatrix} l & l' & \kappa \\ 0 & 0 & 0 \end{bmatrix}^2 \frac{\min(\Delta n^{-1}, \Delta l^{-1})}{(\Delta n)^2(\Delta l)^2} . \quad (4)$$

The latter contains pronounced dependence on Δn and Δl, whereas for exchange transitions we observe very slow sensitivity to Δn and Δl.

IV. CONCLUSIONS

We have presented a detailed comparison between semiclassical and quantum results for cross sections of l-resolved intercombination transitions in Rydberg atoms. The data indicate that the quality of the semiclassical approximation is sufficient for reasonable estimates within a fairly broad range of orbital quantum numbers l, i.e., up to $l \sim n/2$. Especially good accuracy is found for transitions when principal and orbital quantum numbers change in opposite directions. It is then shown that the intercombination transitions obey the "Bethe rule": transitions with the same direction of n and l changes dominate over transitions with opposite changes, in direct similarity with dipole radiative transitions. We have also demonstrated that Δl values of the transitions contributing most to the total cross section have a strong correlation with the extent to which the collision is inelastic. And finally, based on the semiclassical approach we have derived simple analytic formulas for the exchange cross sections containing an explicit dependence on the principal and orbital quantum numbers.

In conclusion, we would like to bring attention to the absence of the experimental data on intercombination transitions in Rydberg atoms present calculations could be compared with.

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