

## Multilevel effects on the balance of dipole-allowed to dipole-forbidden transitions in Rydberg atoms induced by a dipole interaction with slow charged projectiles

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(Received 26 June 1995)

In collisions of Rydberg atoms with charged projectiles at velocities approximately matching the speed of the Rydberg electron  $v_n$  (matching velocity),  $n$  being the principal quantum number of the Rydberg level, the dipole-forbidden transitions with large angular-momentum transfer  $\Delta l \gg 1$  substantially dominate over dipole-allowed transitions  $\Delta l = 1$ , although both are induced by the dipole interaction. Here it is shown that as the projectile velocity decreases the adiabatic character of the depopulation depends on the energy distribution of states in the vicinity of the initial level. If the spectrum is close to degeneracy (as for high- $l$  levels) the dipole-forbidden depopulation prevails practically over the entire low-velocity region, down to velocities  $\sim n^3[\Delta E/Ry]v_n$ , where  $\Delta E$  is the energy spacing adjoining to the level due to either a quantum defect or the relevant level width or splitting, whichever is greater. If the energy gaps are substantial (as for strongly nonhydrogenic  $s$  and  $p$  levels in alkali-metal atoms), then the fraction of dipole transitions in the total depopulation reaches a flat minimum just below the matching velocity and then grows again, making the progressively increasing contribution to the low-velocity depopulation. The analytic models based on the first-order Born amplitudes (rather than the two-level adiabatic approximation) furnish reasonable estimates of the fractional dipole-allowed and dipole-forbidden depopulations.

PACS number(s): 34.60.+z, 34.80.Dp

### I. INTRODUCTION

The dominant status of dipole transitions in charged projectile scattering on atoms is traditionally associated with fast collisions. At large velocities the dipole potential causes dipole  $\Delta l = 1$  transitions only, whereas nondipole transitions with large-angular-momentum change  $\Delta l \gg 1$  are induced solely by multipole interactions. It is for this reason that dipole cross sections dominate over nondipole, although this dominance is not as strong as for radiative transitions and becomes even less pronounced in transitions with large energy change  $\Delta n > 1$ . This standard picture is typical for the Born approximation and is valid, strictly speaking, for fast collisions only. For decades it has been a common practice to rely on the Born approximation in atomic collisions, including even relatively slow collisions, primarily because of its computational simplicity.

In slow collisions, however, due to alternative channels available for excitation, the dipole interaction acquires a different role, thus radically changing the scheme of transitions [1,2]. At intermediate velocities the atom-projectile interaction is not weak and is no longer a mere perturbation. Instead it couples together a broad variety of levels forcing an electron to visit many states in one single act of collision. Since the major term in the atom-projectile interaction is a dipole coupling, the whole process can be viewed as a chain of virtual dipole transitions. At low velocities the probability of this transition chain results in large  $\Delta l$  change and considerably surpasses the probability of the pure dipole  $\Delta l = 1$  transition. It is in this sense that one says the dipole interaction causes dipole-forbidden transitions and the total depopulation cross section is therefore dominated by nondipole transitions  $\Delta l \gg 1$  in contrast to fast collisions.

This structure of transitions in slow collisions (which re-

lates readily to earlier works of Beigman, Vainshtein, and Sobel'man [3] on the action-angle approach to the quasiclassical scattering, Percival and Richards [4] on the strong correspondence principle, Presnyakov and Urnov [5] on equally spaced systems, and Flannery [6] on the equivalent oscillator theorem) was acknowledged in the work of Beigman and Syркин [1] and was tested in detail experimentally in several works of MacAdam and co-workers (see [2] and references cited therein). Since the early 1980s this group has been performing extensive studies on Rydberg atom collisions with charged projectiles over a broad range of velocities. To handle collisions in the domain below the matching velocity the experimental technique was greatly improved, especially in the elimination of stray electric-field effects. Eventually measurements were extended from matching velocities  $v = v_n$  down to  $v \sim 0.2v_n$ , where  $v_n$  is the orbital velocity of a Rydberg electron with the principal quantum number  $n$ . The accord finally attained between the theory and experiment for the depopulation of  $d$  levels in Na (whose quantum defect is 0.0155) confirms the validity of the dipole-forbidden mechanism of transitions in slow collisions. In addition, recently Irby and co-workers [7,8] performed the measurements for the depopulation of strongly nonhydrogenic  $s$  levels in Na and again observed that the fraction of dipole transitions in total depopulation falls from nearly 80% at  $v \sim 10v_n$  to about 20% at  $v = 2v_n$ , similar to results for  $d$  levels, although shifted to higher velocity. Due to quantum defects, however, these two cases differ significantly and this is a major point of the present paper. In what follows we shall show that overall the velocity dependence of the dipole fractional depopulation is the curve with a minimum and that the evolution from direct transitions in fast collisions to a chain of virtual transitions at intermediate velocities (which is common for any multilevel atomic system) represents only a downhill part of the curve preceding the minimum. As the

projectile velocity drops below the matching velocity the actual balance between dipole-allowed and dipole-forbidden transitions truly depends on the type of the spectrum or, more precisely, on the energy-level splitting. Namely, for the substantial energy spacing in the manifold adjacent to the initial level (as for low- $l$  levels with large quantum defects) the minimum is reached readily below the matching velocity, beyond which point the role of dipole transitions increases again. In contrast, if the levels separation is small (as for large- $l$  levels), then the minimum shifts into the deep adiabatic region, so that the depopulation remains dipole-forbidden practically over the entire low-velocity region. We shall also see that the analytical models based on the first-order Born amplitudes rather than two-level adiabatic approximation furnish reasonable estimates of the fractional dipole-allowed and dipole-forbidden depopulation.

The material is organized as follows. Section II reviews the semiclassical close-coupling method. In Sec. III we first discuss a transparent and exactly solvable model of an infinite number of equally spaced levels illustrating the main idea and compare it to the calculations and experimental data for the depopulation of  $ns$  levels in Na, symmetric with respect to two main channels  $ns \rightarrow np \rightarrow (n-1)d \rightarrow \dots \rightarrow (n-1)l$  and  $ns \rightarrow (n-1)p \rightarrow (n-2)d \rightarrow \dots \rightarrow (n-2)l$ . Section IV contains the semi-infinite models suitable for  $nd$  levels in Na, depopulating primarily via one channel  $n, l > 2$  and appropriate generalizations, which allow for unequal spacing (for both one- and two-channel depopulation) and considerably improve the correspondence to the data on realistic spectra. The summary of the results is presented in Sec. V. Atomic units with Ry for the energy are used throughout the paper.

## II. CLOSE-COUPPING METHOD

A full quantum-mechanical treatment of collisionally induced transitions in Rydberg atoms at intermediate and low velocities reduces to highly impractical numerical integration of a large number of atom plus projectile radial Schrödinger equations that describe transitions to numerous accessible states. Two major simplifications can be made. First is the impact parameter approximation [9]. Since the scattering of charged particles on atoms is induced predominantly by the long-range dipole interaction, then within a broad velocity range of practical interest the projectiles (ions and, to a limited extent, electrons) can be considered to move along classical trajectories. In addition, for collisions with neutral atoms accompanied by relatively small energy change  $\Delta n \sim 1$ , the projectile motion may be assumed (to a reasonable accuracy) to be rectilinear and uniform. For further details on the impact parameter method, see [6,10,11]. A second simplification takes into account that our major concern here is the depopulation dependence on the orbital quantum number change  $\Delta l$ . In this context the role of the magnetic quantum numbers  $m$  is minor and by considering transitions between  $m$ -averaged levels based on a suitable averaging procedure (see below) one preserves the major features of the process while significantly reducing the number of equations. Therefore in the semiclassical close-coupling approach the cross section is given as an integral of the transition probability over impact parameter

$$\sigma_{n_0 l_0 - n l} = \int |a_{nl}(\rho, \infty)|^2 2\pi\rho d\rho, \quad (1)$$

where transitions amplitudes  $a_{nl}$  are obtained from the close-coupling system

$$i\dot{a}_{nl}(\rho, t) = \sum_{n'} \sum_{l'=\pm 1} V_{nl-n'l'}(\rho, t) \times \exp(-i\omega_{nl-n'l'}t) a_{n'l'}(\rho, t), \quad (2)$$

with the initial condition  $|a_{nl}(\rho, -\infty)| = \delta_{n, n_0} \delta_{l, l_0}$ , where  $\omega_{nl-n'l'}$ , and  $V_{nl-n'l'}$ , are the energy spacing and interaction matrix elements between levels  $nl$  and  $n'l'$ , respectively. As the leading interaction is dipole (i.e., only  $\Delta l = \pm 1$  terms are retained) the solution of system (2) describes chains of virtual dipole transitions. These chains are contributed mostly by the coupling of closest neighbors  $nl \leftrightarrow n'l \pm 1$ , so that for practical purposes it suffices to keep only two terms  $V_{nl-n'l \pm 1}$  in each equation (2). For the  $m$ -averaged dipole potential we choose

$$V(\rho, t) = \bar{d}_{nl-n'l'} \frac{\rho' + vt}{(\rho'^2 + v^2 t^2)^{3/2}}, \quad \rho' = \rho + R_0, \quad (3)$$

$$\bar{d}_{nl-n'l'} = \left( \frac{1}{2l+1} \sum_m | \langle nlm | z | n'l \pm 1m \rangle |^2 \right)^{1/2},$$

where  $\bar{d}$  is a properly averaged dipole moment and  $R_0$  is a regularization parameter  $\sim 0.5n^2$ , which eliminates the potential singularity as  $\rho \rightarrow 0$ . For fast collisions the averaged potential (3) exactly reproduces the first-order Born approximation. For low velocities  $v < v_n$  the quality of  $m$  averaging has been previously checked by direct comparison with the nonaveraged results and found to provide reasonable accuracy for cross sections [1]. In what follows dipole matrix elements are obtained in the Coulomb approximation with the aid of nonhydrogenic corrections according to [12].

Consider now the collisional depopulation of low- $l$  Rydberg states in Na. Quantum defects of  $s$ ,  $p$ ,  $d$ , and  $f$  levels are 1.348, 0.855, 0.0155, and 0.00145, respectively (see, for example, [13] and references cited therein). For the slightly nonhydrogenic  $d$  level the ratio of  $nd$ - $nf$  splitting to  $np$ - $nd$  splitting is 0.0167 and therefore the  $nd$  level depopulates overwhelmingly to the almost degenerate manifold of  $n, l > 2$  states, whereas the depopulation of the strongly nonhydrogenic  $ns$  level is dominated by two practically equally likely channels:  $ns \rightarrow np \rightarrow (n-1)d \rightarrow (n-1)f \rightarrow \dots \rightarrow (n-1)l$  and  $ns \rightarrow (n-1)p \rightarrow (n-2)d \rightarrow (n-2)f \rightarrow \dots \rightarrow (n-2)l$ . Even with all above assumptions the system (2) still requires numerical solution, which involves, in particular, for  $30s$  depopulation, up to  $\sim 4n$  differential equations and is quite laborious for the low-velocity region. In this regard, before we resort to a numerical approach it is advisable to consider a model that allows for analytic solution and provides an immediate insight into the structure of transitions.

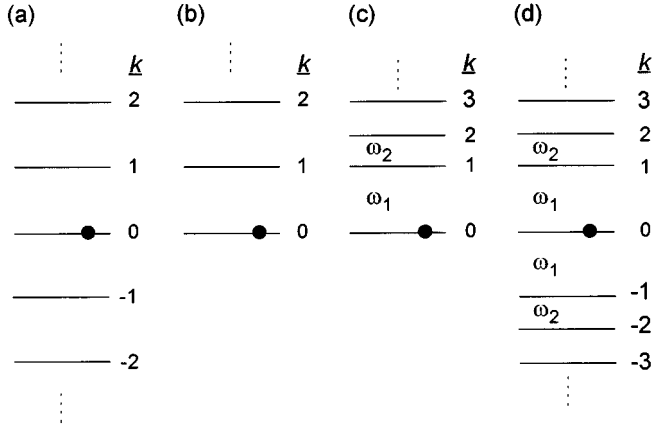


FIG. 1. Schemes of levels: (a) infinite equally spaced levels system; (b) semi-infinite equally spaced levels system; (c) semi-infinite unequally spaced levels system; (d) infinite unequally spaced levels system.

### III. INFINITE EQUALLY SPACED LEVELS SYSTEM AND DATA FOR REALISTIC SPECTRA

If we assume level splitting  $\omega$  and matrix elements  $V_{nl-n'l'}$  within a depopulation channel independent of quantum numbers  $n$  and  $l$ , then one comes to so-called infinite equally spaced levels system (IESLS); thereby Eq. (2) reduces to

$$i\dot{a}_k(\rho, t) = V(\rho, t) \exp(i\omega t) a_{k-1}(\rho, t) + V(\rho, t) \times \exp(-i\omega t) a_{k+1}(\rho, t), \quad (4)$$

with  $-\infty < k < \infty$  and initial condition  $|a_k(\rho, -\infty)| = \delta_{k,0}$ . The scheme of levels is given in Fig. 1(a) and below we always assume for convenience the initial depopulated level corresponding to  $k=0$ . The assumption of equal level spacing is especially restrictive, but we shall see that the model still preserves the most of the important features. The solution of (4) first found by Presnyakov and Urnov [5] is

$$|a_k(\rho, \infty)| = |J_k(2P_1)|, \quad (5)$$

where  $J_k(x)$  is an ordinary Bessel function of the order  $k$  and  $P_1$  is the modulus of the first-order (Born) amplitude, so that (see Seaton [10])

$$P_1^2(\rho, v) = \left| \int V(\rho, t) e^{i\omega t} dt \right|^2 = \left( \frac{2d}{\rho v} \right)^2 \chi(\beta), \quad (6)$$

$$\chi(\beta) = \beta^2 [K_0^2(\beta) + K_1^2(\beta)],$$

where  $\beta = \omega\rho/v$  and  $K_0, K_1$  are modified Bessel functions. If we identify  $k$  with an angular momentum change  $\Delta l$ , then the probability of  $\Delta l$  transition is

$$W_{\Delta l} = J_{\Delta l}^2(2P_1). \quad (7)$$

The total probability of the depopulation is equal to  $W_i = 1 - J_0^2(2P_1)$ . It is convenient to present the contribution of dipole  $\Delta l = \pm 1$  transitions as the ratio

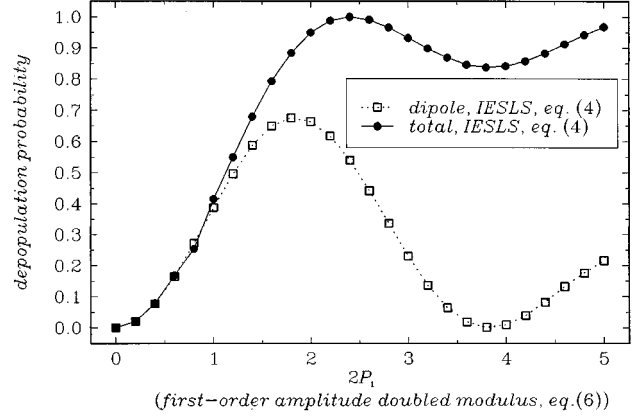


FIG. 2. Total and dipole depopulation probabilities in infinite, equally spaced levels system.  $P_1$  is a modulus of the first-order (Born) amplitude (6).

$$R = (\sigma_1 + \sigma_{-1}) / \sigma_{\text{tot}}$$

$$= 2 \left( \int J_1^2(2P_1) \rho d\rho \right) / \left( \int [1 - J_0^2(2P_1)] \rho d\rho \right). \quad (8)$$

When analyzing (8) we first note that  $P_1$  is unconditionally small in the high-velocity (Born) limit as  $1/v$  and therefore the total cross section is dominated by dipole transitions, i.e.,  $R \rightarrow 1$ . In the opposite limit of low velocities the situation strongly depends on the energy splitting  $\omega$ .

If  $\omega \neq 0$  then  $P_1$  is again small, now exponentially as  $\sim \exp(-\omega\rho/v)$  and again  $R \rightarrow 1$  as  $v \rightarrow 0$ . In other words, in very slow collisions due to the adiabaticity condition (Massey parameter  $\Delta E a_0 n^2 / \hbar v \gg 1$ ) the dipole transitions tend to prevail again and because of the presence of the manifold of adjacent levels the corresponding probability is given exactly by the Born formula (6). Based on the behavior of  $J_1^2(x)$  and  $1 - J_0^2(x)$  (see Fig. 2), we readily conclude that in between two extremes  $v \rightarrow 0$  and  $\infty$ ,  $R$  reaches a minimum whose location is determined by the maximum of  $P_1$  over  $\rho$  and  $v$ . From (6) it immediately follows that the minimum sought is attained at the reduced projectile velocity  $\tilde{v} = v^* \approx 2\delta/3$  [where the reduced velocity is the velocity in units of the Rydberg electron velocity  $v_n$  and  $\delta$  is the difference between quantum defects related to levels spacing  $\Delta E$  by  $\delta \approx 0.5n^3(\Delta E/Ry)$ ]. The value of that minimum is approximately  $R^* \approx 0.36$ . The calculation of  $R$  according to the IESLS model (4)–(8) for 30s depopulation with  $\delta = \delta_s - \delta_p = 0.493$  is given in Fig. 3 and demonstrates good accord with estimates of  $v^*$  and  $R^*$ . In summary, the IESLS model predicts the dipole fractional depopulation to behave as a curve with a minimum, whose value is about 40% and independent of level spacing. The location of the minimum  $v^*$ , however, is immediately related to  $\delta$ , essentially as  $v^* \sim \delta$ . Accordingly, the dipole depopulation of strongly nonhydrogenic levels  $\delta \sim 1$  reaches the minimum in the close submatching velocity domain and then rises again. On the contrary, for very small quantum defects  $\delta < 0.01$  typical for the levels with large orbital quantum numbers  $l$ , the minimum moves into the deep adiabatic region so that the entire

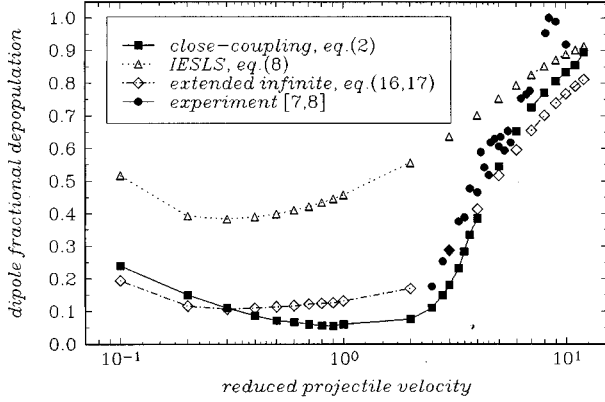


FIG. 3. Fractional dipole depopulation of 30s level in Na.

low-velocity area of practical interest is dominated by the progressively increasing contribution of the dipole-forbidden transitions.

The extreme case of degenerate levels with  $\omega=0$  is of no interest because of the well-known logarithmic divergence of dipole and total depopulation cross sections at large impact parameters. This divergency is regularized by accounting for various small level splittings due to inelastic and radiative level broadening or internal plasma interactions, thereby reducing the problem to the case of small nonzero energy spacing.

Compare now this picture to the direct numerical close-coupling calculations of the 30s depopulation according to (2) and (3) for the real spectrum (see Fig. 3) along with available experimental data [7,8]. We observe the IESLS results converging to the close-coupling data for velocities higher than the matching velocity at about  $\tilde{v}>5$ . The model is also consistent with numerical results (2) in predicting a flat minimum of  $R$  in submatching region and slow  $R$  increase as  $v\rightarrow 0$ . On the other hand, the actual minimum is much deeper, approximately 7–10 %, meaning that the model is quantitatively invalid in this range, overestimating  $R$  up to a factor of 5. It is quite obvious that the cause is equal spacing. Indeed, the actual  $30p-29d$  splitting is about three times less than the one for  $30s-30p$ . This increases both 30s and 30p depopulations, but apparently the 30p level is affected directly and therefore much stronger, which considerably reduces the dipole fraction in the total depopulation. To a considerable extent this drawback is eliminated by semi-infinite models originally proposed in [1] for the depopulation of higher- $l$  levels that allow for unequal spacing.

#### IV. SEMI-INFINITE MODELS AND ITS EXTENSIONS

We resort now to the depopulation of  $d$  levels in Na. As mentioned above, this process is highly asymmetric towards the manifold of closely spaced levels with large  $l$  and therefore, the previous model, infinite in both directions does not apply. Instead, consider an equally spaced system bound from one end [Fig. 1(b)]

$$i\dot{a}_k(\rho, t) = V(\rho, t)\exp(i\omega t)a_{k-1}(\rho, t) + V(\rho, t) \times \exp(-i\omega t)a_{k+1}(\rho, t), \quad (9)$$

$$i\dot{a}_0(\rho, t) = V(\rho, t)\exp(-i\omega t)a_1(\rho, t),$$

with  $1 \leq k < \infty$  and initial condition  $|a_k(\rho, -\infty)| = \delta_{k,0}$ . According to [1], the solution for such a semi-infinite system is given by

$$\begin{aligned} |a_k(\rho, \infty)| &= J_k(2P_1) + J_{k+2}(2P_1) \\ &= [(k+1)J_{k+1}(2P_1)/P_1], \quad 0 \leq k < \infty. \end{aligned} \quad (10)$$

More realistic is a system where the gap  $\omega_1$  between levels 0 and 1 is greater than the constant spacing  $\omega_2$  between the other levels [Fig. 1(c)]. Using the technique of a Volterra integral equation for the generating function, the approximate solution can be written as [1]

$$W_k = |a_k(\rho, \infty)|^2 = f_k W_t,$$

$$f_k = [kJ_k(2P_1(\omega_2))/P_1(\omega_2)]^2, \quad k > 0, \quad (11)$$

$$W_0 = 1 - W_t,$$

where  $W_t(\omega_1, \omega_2)$  is the total depopulation probability of the initial level  $k=0$ , depending, strictly speaking, on both  $\omega_1$  and  $\omega_2$ , and factors  $f_k$  give the fractional contribution of transitions with various  $\Delta l=k$ . The first important feature of this solution is that it automatically satisfies the normalization condition  $\sum_k f_k = 1$  regardless of the approach used for the total probability  $W_t$  and is flexible towards various approximations for  $W_t$  [such as a single gap  $\omega_1$  normalized Born approximation  $W_t(\omega_1) = P_1/(1+P_1)$  used in [1]]. Second, the structure of (11) clearly demonstrates that it is the second gap  $\omega_2$  (and not  $\omega_1$ ) that determines the role of dipole depopulation. If  $\omega_2$  is large enough, then dipole transitions dominate, as only  $f_1 \rightarrow 1$  when  $v \rightarrow 0$ , whereas for all other  $k > 1$  we have  $f_k \rightarrow 0$ . In the opposite case of  $\omega_2 \approx 0$  the dipole contribution becomes negligible for small  $v$  because  $f_1/(1-f_1) \sim 1/N^3$ , where  $N$  is the number of levels in the depopulation channel (in our case  $N \sim n-1$ ). The latter situation is just the case for the  $d$  level depopulation and, overall, the solution (11) works reasonably well, except that it predicts a too fast drop in the dipole fractional depopulations as we move deeper into the submatching region. This happens because the major assumption behind the solution (11) is that the coupling within a closely spaced manifold  $k > 0$  is tighter than between an initial level  $k=0$  and the adjacent manifold  $k > 0$ . Accordingly, the transitions in the manifold occur equally likely in both directions  $k \leftrightarrow k+1$ , whereas at the bottom of the spectrum the transitions take place primarily in the direction  $k=0 \Rightarrow k=1$ . This holds, however, only for fairly fast collisions when the first-order amplitude  $P_1$  is small enough. In passing the vicinity of the matching velocity,  $P_1$  rises substantially, which tightens the coupling between the bottom level 0 and the manifold. As a result, the solution (11) progressively underestimates the probability right below the matching velocity (up to a factor of  $\sim 2$ ) and then becomes completely invalid for lower velocities. The situation can be improved as follows.

First, we choose  $W_t$  such that at the limit of equal spacing  $W_t$  becomes identical to the result for the total probability in the semi-infinite model (10). This reduces  $W_t$  to the form

$$W_t = \{J_1(2P_1(\omega_1))/P_1(\omega_1)\}^2. \quad (12)$$

In fact, as we shall see below, somewhat better results follow if  $W_t$  is given as in the IESLS model (7):

$$W_t = 1 - J_0^2(2P_1(\omega_1)) \quad (13)$$

[although (13) is inconsistent with (10) in the Born limit, which is insignificant for slow collisions].

Second, we account for the fact that despite the limitations related to the equal spacing, the semi-infinite equally spaced model (9) is more preferable at very low velocities as it makes no assumptions on the coupling strength. Therefore, one can, at the qualitative level, combine both models with equal and unequal spacing. If we introduce the impact parameter  $\rho^*$  such that  $P_1(\rho^*)=1$ , then, when integrating the probability over the impact parameter we use the unequally spaced model (11) for  $\rho > \rho^*$  and the equally spaced one (10) otherwise. Thus the approximation for the one-directional depopulation, which hereafter will be referred to as an extended semi-infinite model, can be summarized as follows: if  $\rho > \rho^*$ ,

$$W_k = [kJ_k(2P_1(\omega_2))/P_1(\omega_2)]^2 W_t, \quad k > 0, \quad (14)$$

$$W_0 = 1 - W_t,$$

and if  $\rho < \rho^*$ ,

$$W_k = [(k+1)J_{k+1}(2P_1(\omega_2))/P_1(\omega_2)]^2, \quad k \geq 0,$$

where

$$W_t = \{J_1(2P_1(\omega_1))/P_1(\omega_1)\}^2 \quad (15a)$$

or

$$W_t = 1 - J_0^2(2P_1(\omega_1)). \quad (15b)$$

Based on these formulas and taking into account small differences of quantum defects corresponding to the  $\omega_1(d-f)$  and  $\omega_2(f-g)$  gaps (0.014 and 0.0015, respectively), one can expect the velocity  $v^*$  minimizing the contribution of  $\Delta l=1$  transitions to the total depopulation to occur in the ultra-adiabatic region and therefore the entire submatching velocity domain to exhibit increasing dipole-forbidden fractional depopulation. The numerical results for  $28d$  depopulation following from (14) and (15) and supporting that conclusion are given in Fig. 4 in comparison with exact close-coupling calculations [1] and the experiment [2]. One can see indeed quite reasonable accuracy, especially for low velocities, regarding both the profile of the fractional depopulation and its absolute values.

Return now to the  $s$  depopulation. For the unequally spaced scheme of levels given in Fig. 1(d) we can treat the symmetric  $s$  depopulation in essentially the same manner, combining the semi-infinite unequally spaced model (12) for large impact parameters with an infinite equally spaced

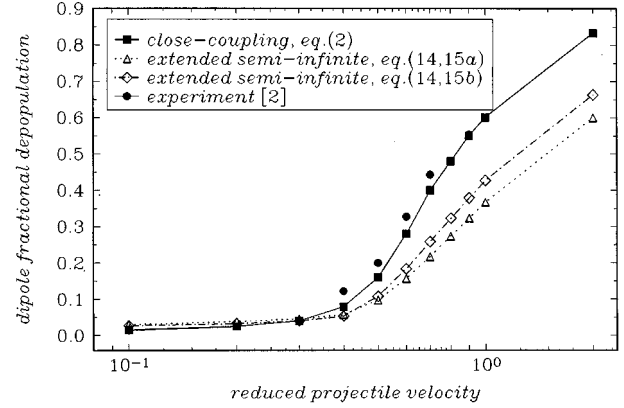


FIG. 4. Fractional dipole depopulation of  $28d$  level in Na.

model for small  $\rho$ . Omitting the elementary transformations, the symmetric extended model may be written as follows: if  $\rho > \rho^*$ ,

$$W_k = [kJ_k(2P_1(\omega_2))/P_1(\omega_2)]^2 (W_t/2), \quad |k| > 0, \quad (16)$$

$$W_0 = 1 - W_t$$

and if  $\rho < \rho^*$ ,

$$W_k = J_k^2(2P_1)^2,$$

where

$$W_t = 1 - J_0^2(2P_1(\omega_1)) \quad (17)$$

When applying this model to the  $30s$  depopulation (see Fig. 3) we find significant improvement over the simple IESLS model in the submatching domain. The remaining discrepancies, such as the location of the depopulation minimum, are difficult to eliminate in a simple framework of an extended model and need a more elaborated approach. The same also applies to  $p$  levels, which depopulate via two asymmetric but quite competing channels  $np \rightarrow ns$  and  $np \rightarrow (n-1)d$ .

The fact that it is the first-order (Born) amplitude  $P_1$  that appears as an effective argument throughout the above-considered analytical solutions (and not a two-level adiabatic approximation, as this might have seemed natural at the first glance) has an immediate implication for the transitions in very slow collisions. It was shown (see [14]) that in the limit  $v \rightarrow 0$  the adiabatic probability obtained from the asymptotic solution of the two-level system drops exponentially faster than the Born probability, roughly speaking, as  $W_{ad} \sim W_{Born} \exp(-d/\rho v)$ , where  $d$  is an  $m$ -averaged dipole moment from (3). The two-level approximation, however, holds only if the coupling between two given levels is stronger than their coupling with the rest of the spectrum, i.e.,  $\omega_2 > \omega_1$ . It is readily clear this is not the case for Rydberg levels where any two neighboring levels are to no extent isolated from the adjacent manifold, the presence of which plays an essential role in the increase of the two-level probability. In particular, in the IESLS model the probability of dipole transitions reduces exactly to the Born formula in the adiabatic limit. In turn, in the real system the energy spacing converges to zero and therefore the probability and the cor-

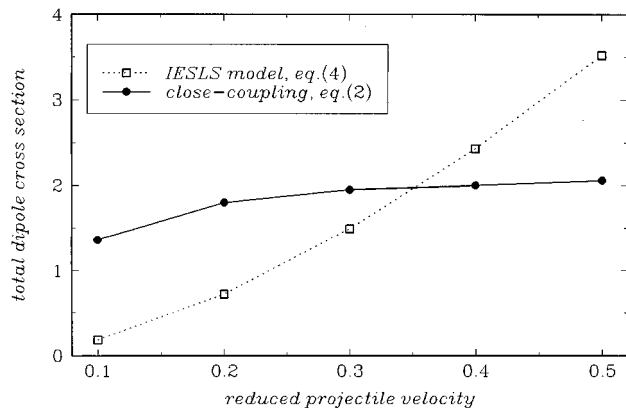


FIG. 5. Absolute dipole depopulation of  $30s$  level in Na, in units of  $\pi a_0^2 n^4$ .

responding cross section decrease at low velocities noticeably slower than in Born approximation (see Fig. 5).

It should be emphasized, in conclusion, that for the ultra-adiabatic region  $\tilde{v} \ll 0.1$  all models considered predict the dipole depopulation to become again a dominant case. However, as seen in Fig. 5, at  $\tilde{v} \sim 0.1$  the dipole cross section is comparable to an area of the Rydberg electron orbit, which means that the semiclassical impact parameter approximation is no longer valid below that velocity. This seems to be the most serious problem in studying an adiabatic depopulation and the relevant method is yet to be developed.

## V. CONCLUSION

The numerical simulation of the close-coupled transitions in Rydberg atoms induced by collisions with slow charged

projectiles reveals the nonmonotonic behavior of the total depopulation character in terms of the balance between dipole-allowed and chain dipole-forbidden transitions. In particular, the dipole fractional depopulation of strongly nonhydrogenic levels reaches a minimum near the matching velocity in contrast to the monotonic decrease with the decreasing velocity for hydrogenic levels with large orbital momenta. Although the exact depth and the location of that minimum result from the interplay between transition probabilities and level splittings in the depopulation channel, the major features are explained by means of the simple multi-level models, attributing the behavior of the dipole depopulation to the energy spacing in the vicinity of the initial level. For large energy gaps the minimum occurs at the beginning of the submatching area, then giving way to the increasing dipole contribution. For closely spaced levels the minimum shifts into the deep adiabatic region and the dipole-forbidden transitions dominate, for the most part, in slow collisions. Quantitatively the models are in a reasonable correspondence with the numerical calculations. The results also indicate that even for slow collisions the problem remains essentially multilevel and the two-level adiabatic approximation is not applicable.

## ACKNOWLEDGMENTS

The author is greatly indebted to K. MacAdam for collaboration, stimulating discussions, and useful comments on the manuscript; V. Irby and K. MacAdam for information on the experiments prior to publication and clarification of the experimental problems; and M. Flannery for a critical reading of the manuscript. This work was supported in part by Maritime College Joint Fund.

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