Effect of extra resonances in the Rydberg-to-Rydberg Raman migration

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It is shown that in the $|\Delta l|=2$, resonant Raman migration of the population between Rydberg states of comparable principal quantum numbers $n \gg 1$, the coupling of the resonant intermediate state to a lower-lying one acts as a suppressor of the migration due to the shift splitting of the resonant state.

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I. INTRODUCTION

When an atom in a high-lying Rydberg state with definite angular momentum interacts with intense enough laser field, Raman migrations of the population can occur to other high-lying states, but of different angular momenta [1]. However, those Raman migrations will be shown in this paper to be suppressed in some cases due to the intermediate-state Stark shift from resonance. To be more specific, we consider a hydrogen atom prepared in the 40*d* state (Fig. 1). We subject the atom to the interaction with the laser field of linear polarization and frequency equal to the atomic transition frequency between the states of the principal quantum numbers 40 and 7. What we could expect in such a case is an effective Raman transition through the intermediate state $7f$ to the high-lying Rydberg states of the angular-momentum quantum number equal to 4. As will be shown, however, such a Raman migration does not occur that is related to the presence of the low-lying states of principal quantum numbers $n=5$ of large dipole matrix elements with the state of $n=7$. The coupling between these states results in their Stark shift in the presence of the laser field [2]. So the $7f$ state becomes pushed from the resonance with the 40*d* state causing the suppression of the expected $d \rightarrow g$ Raman migration. However, when we choose the laser frequency in a way compensating the Stark shift of the $7f$ state, the $40d$-$7f$ resonance will be supported giving rise to the $d \rightarrow g$ Raman migration in question.

II. THE MODEL AND ITS SOLUTION

Let the hydrogen atom be initially in the high $|n,l,m\rangle = |40,2,2\rangle$ Rydberg state (Fig. 1). We allow this atom to interact with the laser field of linear polarization along the z axis and frequency comparable to that of the $n=40 \rightarrow 7$ transition. However, we do not assume perfect resonance for purposes that will become clear later on. Under these assumptions, a transition is possible to the $|n,l,m\rangle = |7,3,2\rangle$ state and then to high-Rydberg states around $n=40$ of the angular-momentum quantum number equal to 4. From the latter states, the population can be moved to the $|n,l,m\rangle = |7,5,2\rangle$ state and then again to the high-lying states, but of the angular-momentum quantum number higher by one. However, the above coupling path is perturbed by the presence of the $|n,l,m\rangle = |5,2,2\rangle$ and $|n,l,m\rangle = |5,4,2\rangle$ states, because the laser frequency that ensures the transition $n=40 \rightarrow 7$ is accidentally in near-resonance with the $n=7$ to $n=5$ transition. To describe the generalized coupling scheme of Fig. 1, we make standard approximations, namely the rotating-wave and rectangular pulse approximations that allow us to transform the Schrödinger equation to the Laplace domain $(t \rightarrow s)$.

We denote the high-Rydberg states by $|n,j\rangle$, where $n \cong 40$ and the angular quantum number is equal to $2j$. Let $|1\rangle$ and $|2\rangle$ stand for the states $|n,l,m\rangle = |7,3,2\rangle$ and $|n,l,m\rangle = |7,5,2\rangle$, respectively, while the lower-lying states $|n,l,m\rangle = |5,2,2\rangle$ and $|n,l,m\rangle = |5,4,2\rangle$ are denoted by $|3\rangle$ and $|4\rangle$, respectively. By the selection rules, all states of interest have the same magnetic quantum number equal to 2. Let $\Omega_{j,j'}$ stand for the matrix element of the interaction Hamiltonian (we use the units in which $\hbar=1$) between states $|j\rangle$ and $|j'\rangle$. The corresponding matrix element between states $|j'\rangle$ ($j'=1,2$) and $|n,j\rangle$ is denoted by $V_{j',jn}$. The last one scales as $n^{-3/2}$ [3], so for n of an order of 40, we can safely replace this matrix element by some representative one, namely that for $n=40$ ($V_{j',jn} \cong V_{j',j40} e^{-i\Delta_n t} \equiv V_{j',j} e^{-i\Delta_n t}$). Finally, $D_{jn,jn'}$ stands for the Raman coupling through the continuum between two high-Rydberg states of the same angular-momentum quantum number. We do not include Raman couplings between two high-lying states of different angular momenta because of their negligible role. We take

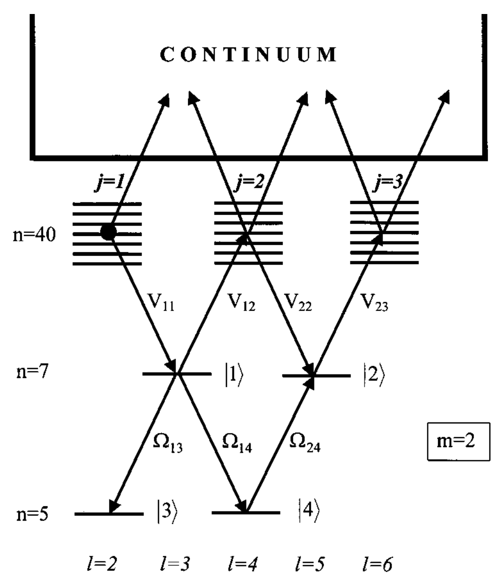

FIG. 1. Rydberg-atom photoionization from the 40d state with the possible transitions to states of principal quantum numbers 7 and 5.

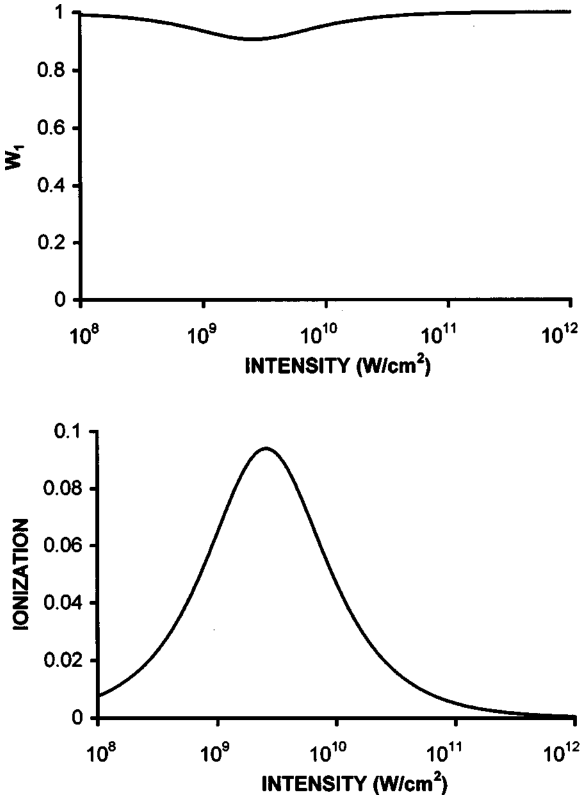

FIG. 2. Population vs laser intensity in the first quasicontinuum (W_1), and atomic continuum for the laser frequency equal to the $n=40\rightarrow 7$ transition frequency. Practically no population migrates to the second quasicontinuum.

advantage of the fact that $D_{jn,jn'}$ scales as $(nn')^{-3/2}$ [4], so for a high n, we also replace this coupling by that for $n=n'=40$ $(D_{jn',jn}\cong D_{j40,j40}e^{-i(\Delta_n-\Delta_{n'})t}\equiv D_j e^{-i(\Delta_n-\Delta_{n'})t})$. We also assume the equidistant Bixon-Jortner structure for these states [5], so the energy difference between the initial state and that of the principal quantum number equal to n simply reads $\Delta_n=(n-40)\Delta$, where Δ is the frequency difference between the states of $n=40$ and $n=41$. The time-dependent Schrödinger amplitude for the state $|j\rangle$ is denoted by b_j and that for the state $|n,j\rangle$ by b_{jn}. With this approximation, the Schrödinger equation for these amplitudes takes the form

$$\dot{b}_3=-i\Omega_{13}^{*}b_1, \tag{1}$$

$$\dot{b}_4=-i\Omega_{14}^{*}b_1-i\Omega_{24}^{*}b_2, \tag{2}$$

$$\dot{b}_1=-i\Omega_{13}b_3-i\Omega_{14}b_4-iV_{11}\sum_n b_{1n}e^{-i\Delta_n t} -iV_{12}\sum_n b_{2n}e^{-i\Delta_n t}, \tag{3}$$

$$\dot{b}_2=-i\Omega_{24}b_4-iV_{22}\sum_n b_{2n}e^{-i\Delta_n t}-iV_{23}\sum_n b_{3n}e^{-i\Delta_n t}, \tag{4}$$

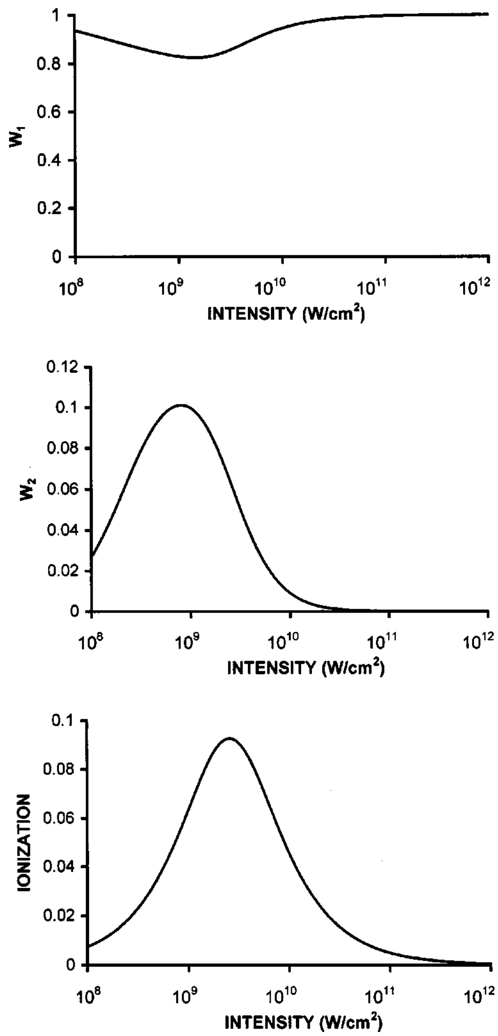

FIG. 3. Same as in Fig. 2, ignoring the $n=7\rightarrow 5$ couplings $(\Omega_{13}=\Omega_{14}=\Omega_{24}=0)$.

$$\dot{b}_{1n}=-iV_{11}^{*}b_1e^{i\Delta_n t}-D_1\sum_{n'} b_{1n'}e^{i(\Delta_n-\Delta_{n'})t}, \tag{5}$$

$$\dot{b}_{2n}=-iV_{12}b_1e^{i\Delta_n t}-iV_{22}^{*}b_2e^{i\Delta_n t}-D_2\sum_{n'} b_{2n'}e^{i(\Delta_n-\Delta_{n'}}t, \tag{6}$$

$$\dot{b}_{3n}=-iV_{23}^{*}b_2e^{i\Delta_n t}-D_3\sum_{n'} b_{3n'}e^{i(\Delta_n-\Delta_{n'})t}, \tag{7}$$

with the boundary conditions $b_1(0)=1$ and zero for all other amplitudes. We solved the above set in time scale shorter than the classical Kepler period of the initial state, $T=2\pi/\Delta$, following the line of our previous paper [6].

III. RESULTS

To obtain representative numerical results, we use the expression $D_{jn,jn'}=(1+iq_{jn,jn'})\sqrt{\gamma_{jn}\gamma_{jn'}}/2e^{i(\Delta_n-\Delta_{n'})t}$ [7] for Raman coupling between two high-Rydberg states, where $\gamma_{jn}(\gamma_{jn'})$ is Fermi's golden rule ionization rate for the state $|j,n\rangle(|j,n'\rangle)$, and $q_{jn,jn'}$ stands for the Fano parameter. We

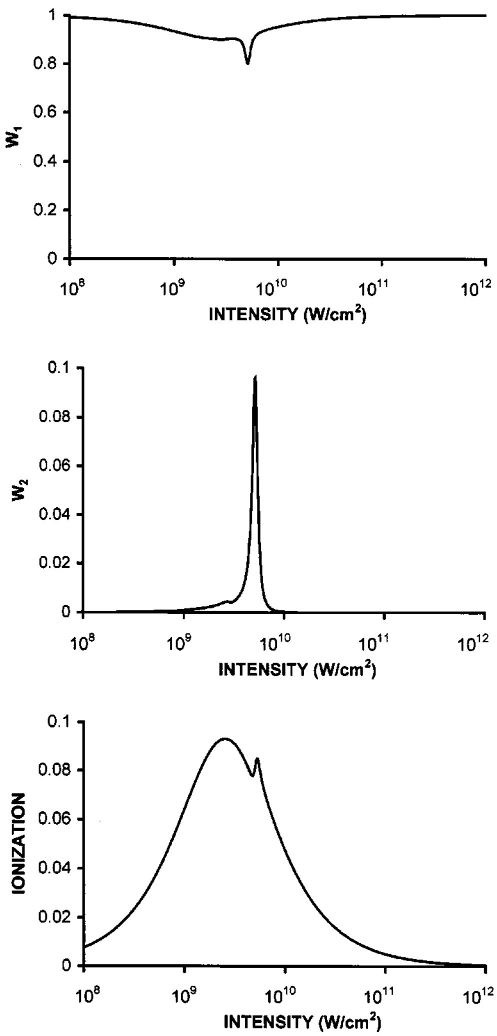

FIG. 4. Same as in Fig. 2, but for the laser frequency detuned from the $n=40\rightarrow7$ transition frequency by an amount $\varepsilon=1.5\times10^{13}\,\mathrm{s}^{-1}$.

choose this parameter equal to 20, in analogy with [1]. In order to calculate the bound-bound and bound-free matrix elements, we use the prescription based on the Laplace transform [8]. The matrix element for the coupling of a high-lying state ($n\gg1$) with any other state was checked to be in agreement with the quasiclassical approximation [3] and exhibited weak n dependence, which justifies our approximation. The pulse duration time was chosen equal to 10 ps corresponding to the Kepler period of the $n=40$ state.

We aimed at studying the dependence of the population of bound states and ionization on laser intensity under different frequency conditions. Figure 2 presents the results for the case of the laser frequency coinciding with the free-of-field $n=40\rightarrow7$ transition frequency. We see that nearly the whole population remains in the first quasicontinuum of the angular-momentum quantum number equal to 2, i.e., in that including the initial state, and a little of the population is released to the continuum. We do not observe any significant Raman migration of the population to the second quasicontinuum. The latter result differs from the strongly effective migration observed in [1] when considering a simpler model without the extra near resonances of the type $n=7\rightarrow5$. We also see that for high enough intensities the ionization decreases with intensity, which testifies to the atomic stabilization [9].

To see the importance of the extra $n=7\rightarrow5$ near resonance, we show in Fig. 3 the analogous results, but there obtained arbitrarily ignoring the $n=7\rightarrow5$ couplings ($\Omega_{13}=\Omega_{14}=\Omega_{24}=0$). The striking difference between Fig. 3 and Fig. 2 is the effective Raman migration of the population to high-Rydberg states of the angular-momentum quantum number equal to 4, shown in Fig. 3. The following interpretation of this behavior is proposed. The actual near-resonant coupling between the states of $n=7$ and $n=5$ results in the Stark shift splitting of these states. For intense enough laser field, this shift splitting cancels, in fact, the $n=40\rightarrow7$ resonance, making the Raman transfer of the population from the first to the second quasicontinua impossible. The estimated Stark shift of the state $|n,l,m\rangle=|7,3,2\rangle$ for the intensity of $I=2.6\times10^{9}\,\mathrm{W/cm}^2$ i.e., the one for which there is the minimum population of the first quasicontinuum in Fig. 2, amounts to $\varepsilon_{\mathrm{Stark}}\approx1.5\times10^{13}\,\mathrm{s}^{-1}$.

Figure 4 is made for the general model shown in Fig. 1, but now for the frequency compensating the above estimated shift, i.e., the frequency ensuring the dynamical $n=40\rightarrow7$ resonance. As distinct from Fig. 2, we observe in Fig. 4 significant Raman migration of the population to the second quasicontinuum ($l=4$) at the intensity of about $I=5.2\times10^{9}\,\mathrm{W/cm}^2$, accompanied by the dip in the populations of both the first quasicontinuum ($l=2$) and the continuum. This supports our prediction that the absence of the Raman migration in Fig. 2 is caused by the Stark shift of the state $|n,l,m\rangle=|7,3,2\rangle$ due to its coupling to the $|n,l,m\rangle=|5,2,2\rangle$ state.

Such an interpretation is also supported by Fig. 5, which shows the population of the second quasicontinuum versus

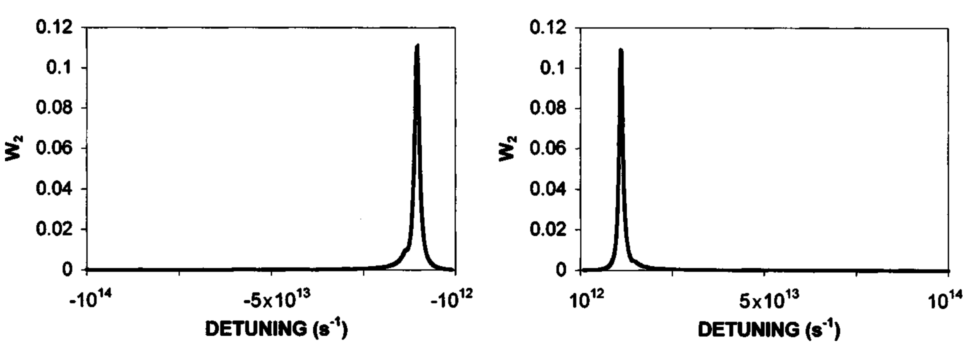

FIG. 5. Population in the second quasicontinuum vs detuning of the laser frequency from the $n=40\rightarrow7$ transition frequency.

the laser frequency at the intensity of $I=2.6\times10^9$ W/cm^2. The two peaks observed are the counterpart of the Autler-Townes splitting of the $|n,l,m\rangle=|7,3,2\rangle$ state as the result of its near-resonant coupling to the lower-lying states, predominantly to the $|n,l,m\rangle=|5,2,2\rangle$ state.

IV. SUMMARY

We studied nominal one-photon ionization of the $40d$ Rydberg state accompanied by higher-order processes such as the resonant Raman migration via the $7f$ state towards higher-angular-momentum Rydberg states of $n\cong40$ and the double optical resonance from the initial state down to the $5d$ state via the $7f$ state. It was shown that the strong $7f$-$5d$ coupling is able to suppress completely the Raman migration to the ng states of $n\cong40$. The reason for this suppression was found to be the dynamical shift splitting of the $7f$ state pushing out this state from the resonance with the initial $40d$ state. This finding extends the results of a recent paper [1].

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[1] R. Parzyński and S. Wieczorek, Phys. Rev. A **58**, 3051 (1998).
[2] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, 1997)
[3] N. B. Delone, S. P. Goreslavsky, and V. P. Krainov, J. Phys. B **27**, 4403 (1994).
[4] M. S. Adams, M. V. Fedorov, V. P. Krainov, and D. D. Meyerhofer, Phys. Rev. A **52**, 125 (1994).
[5] H. G. Muller and L. D. Noordam, Phys. Rev. Lett. **82**, 5024 (1999).
[6] A. Wójcik, R. Parzyński, and A. Grudka, Phys. Rev. A **55**, 2144 (1997).
[7] R. Grobe, G. Leuchs, and K. Rzażewski, Phys. Rev. A **34**, 1188 (1986).
[8] G. Feldman, T. Fulton, and B. R. Judd, Phys. Rev. A **51**, 2762 (1995).
[9] *Super-Intense Laser-Atom Physics IV*, Vol. 13 of *NATO ASI Series 3 on High Technology*, edited by H. G. Muller and M. V. Fedorov (Kluwer Academic, Dordrecht, 1996).