Rydberg transitions induced by optical core dressing

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We study theoretically the behavior of two atomic Rydberg series, coupled by a light field that strongly drives the optical transition between the core states to which these series converge. It is shown that this strong-field version of isolated-core excitation leads to a strong modification of the photoionization spectrum and to transitions between adjacent Rydberg states when the Rabi frequency associated with the core transition becomes larger than the Rydberg spacing. The intensities and pulse durations (≈ 2 GW/cm^2 and 1 ps, respectively, for $n \approx 20$ Rydberg states) needed for the observation of these effects are within reach of present-day lasers.

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I. INTRODUCTION

When one of the electrons in an atom is promoted to a weakly bound Rydberg orbit, the interaction of the electron with the remaining ionic core is very weak due to the small spatial overlap between the extended Rydberg wave function (extending $2n^2$ Bohr radii, where n is the principal quantum number) and the core (confined to only a few Bohr radii). Hence the transitions of the ionic core are nearly undisturbed by the presence of the Rydberg electron. This forms the basis of the well-known method of isolated-core excitation (ICE) [1,2] (see also Fig. 1), which can be conveniently used to create and study doubly excited atomic states. In ICE an atomic electron is first promoted to a Rydberg level. Subsequently the core is excited by an optical transition. Since photoionization can only take place close to the core, the Rydberg electron is relatively insensitive to the light and essentially remains a spectator to the core transition. The properties (e.g., energies and autoionization rates) of the doubly excited states that are created in this way reveal information on the residual interaction of the Rydberg electron with the atomic core. ICE has proven to be a valuable means of determining the role of electron-electron interaction and correlation in atomic structure.

In this paper we study theoretically the case where the core transition is driven by an *intense* light source. The intense light field leads to a strong coupling between the two resonant core states. This coupling results in two "dressed" core states with an energy splitting proportional to the electric field amplitude of the light (an "Autler-Townes doublet"). One can also describe this as a rapid oscillation between the two original ("bare") core states, driven by the light field. The oscillation frequency, the so-called Rabi frequency, corresponds to the energy splitting between the dressed states.

In the absence of the intense light the atomic eigenstates form Rydberg series converging to the ionic core states. With the intense light on, these Rydberg series are strongly mixed and converge to the *dressed* ionic core states. This provides a means of externally controlling the atomic structure in a time-dependent manner by varying the intensity of the light. Of particular interest is the case where the Autler-Townes splitting of the core becomes comparable to the spacing of the Rydberg levels. Considered in the time domain instead of the frequency domain, this corresponds to a situation where the Rabi oscillation period of the core is similar to the Kepler orbit time ($2\pi n^3$ a.u.) of the Rydberg electron. We will show that this case leads to intriguing dynamics.

The system described above has been studied before by Robicheaux [3] for driving light of constant intensity. He calculated how the spectroscopy of the Rydberg series would change when these are coupled by a core-resonant intense light field. An experimental realization of such spectroscopy would be rather difficult (as pointed out in Ref. [3]) because the driving light needs to be both intense and of constant intensity. Otherwise the dressed Rydberg levels would shift appreciably during the presence of the probe laser.

We show in this paper that the atomic dynamics with photon-dressed core states can also be observed in a different way, using pulsed light fields. In particular we find that when the peak Rabi frequency in the light pulse becomes comparable to the Rydberg level spacing, this leads to strong modifications in the photoionization spectrum and to transitions between adjacent Rydberg levels. Since present-day laser systems can easily reach the intensities (≈ 2 GW/cm^2) and pulse durations (≈ 1 ps) used in the calculations, this approach makes an experimental realization feasible [4].

This paper focuses on the effects of a large core Rabi frequency in ICE, i.e., on the use of *intense* light pulses. We note here that the use of *short* light pulses in ICE has attracted considerable interest recently and the theoretical framework used in that case is very similar to the one adopted in this paper. Using light pulses much shorter than the Kepler orbit time of the Rydberg electron, one can excite a Rydberg radial wave packet [5–9] and study time-dependent autoionization [10–12] and time-dependent core excitation [13]. In such short pulses the frequency bandwidth of the light is much larger than the spacing between the Rydberg states, so that a coherent superposition of Rydberg states can be excited that con-

stitutes the wave packet. In contrast, in our case the necessary bandwidth is created by the large Rabi frequency of the core. Very recently, the use of short and intense pulses has been combined by Hanson and Lambropoulos [14] and by Zobay and Alber [15], in a study of the Rydberg wave-packet dynamics in the presence of core-resonant light, for the case that the Rabi frequency of the core becomes comparable to the Kepler orbit time.

In other related work it has been shown experimentally that for very high intensities ($\approx 10^{12}$ W/cm^2), when multiphoton ionization becomes important, one can observe (nonresonant) isolated-core *ionization* instead of (resonant) isolated-core excitation [16,17]. At such intensities the effects of the resonant core dressing on photoionization of the atomic ground state (instead of on the Rydberg series) also have been considered theoretically [18–22]. In this case the resonant core dressing has been predicted to yield a split peak in the photoelectron spectrum of multiphoton ionization of the atomic ground state. Experimental evidence for this effect was found recently [23,24].

In the following section of this paper we describe the theoretical model we have used and discuss the physical meaning of the parameters in this model. Next, in Sec. III the results of calculations using this model are described, illustrating the main features of this system and showing the effects that can be observed with a pulsed core-resonant light field. We also discuss how the effects studied here could induce "zero-photon ionization," i.e., ionization of Rydberg states by photon scattering instead of by photoabsorption.

II. THEORY

We study the dynamical behavior of the system depicted in Fig. 1 in a pulsed light field. In this field the

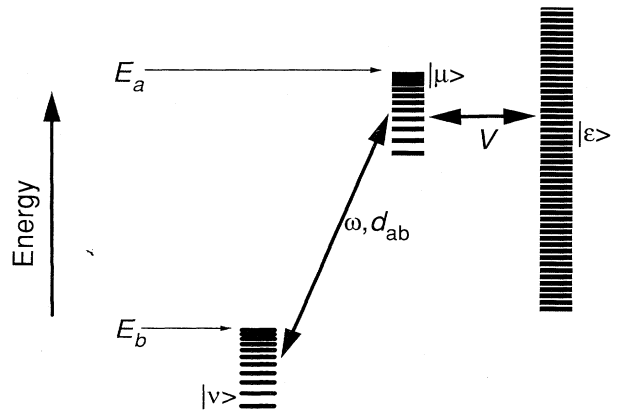

FIG. 1. Energy diagram schematically depicting the situation studied in this paper. Two atomic Rydberg series $|\mu\rangle$ and $|\nu\rangle$, converging to the ionic core states $|a\rangle$ and $|b\rangle$, respectively, are strongly coupled by light with frequency ω, resonant with the core transition $\omega \approx E_a - E_b$. The Rabi frequency of the core transition, induced by the light, is indicated by d_{ab}. The upper Rydberg series can autoionize by the coupling V to the continuum $|\epsilon\rangle$ above the ionic ground state.

various states involved shift in energy as a function of the field strength and population will be transferred from the initial state to other states at different energies. As a consequence, the traditional approach of quantum-defect theory [3,25], which calculates the atomic eigenstates for a given fixed energy, is not sufficient.

In order to calculate the evolution of the system in a time-dependent light field, it will be easiest to reduce the problem to a finite set of basis functions and then numerically integrate the time-dependent Schrödinger equation (TDSE) on that basis set. It turns out that a description in terms of such a finite basis set is possible, as we will show in this section. We start from the conventional description of ICE, within the framework of quantum-defect theory [1,25,26]. From this description we arrive at a TDSE for a finite basis set by eliminating the continuum, applying the rotating-wave approximation (RWA) (see, e.g., Ref. [27]), and selecting a range of relevant Rydberg states from each series. Atomic units (a.u.) [28] will be used throughout this paper, unless other units are explicitly given.

For simplicity we consider only two Rydberg series, each converging to one of the two core states. Since the Rydberg electron has a very small overlap (proportional to $n^{-3/2}$) with the core, it is usual in ICE to neglect the direct interaction between the two [1]. The atomic wave functions of the (bound) lower Rydberg series can thus be written as products of a core wave function $|b\rangle$ and Rydberg wave functions $|n_b\rangle$ [29]. The energies of the bound Rydberg states are given by $E_b - 1/2(n_b - \delta_b)^2$, where E_b is the energy of the ionic state $|b\rangle$. The quantum defect δ_b of the lower Rydberg series depends only on the electron configuration (i.e., the core state and the various angular momenta involved) and not on the principal quantum number n_b. If we disregard the autoionization of the upper Rydberg series for the moment, this upper series is also described by product wave functions $|a\rangle|n_a\rangle$ with energies $E_a - 1/2(n_a - \delta_a)^2$.

For convenience we now introduce effective quantum numbers μ and ν for the upper and lower Rydberg series, respectively,

$$\mu = n_a - \delta_a, \tag{1a}$$

$$\nu = n_b - \delta_b; \tag{1b}$$

a shorthand notation for the Rydberg states

$$|\mu\rangle = |a\rangle|n_a\rangle, \tag{2a}$$

$$|\nu\rangle = |b\rangle|n_b\rangle; \tag{2b}$$

and a shorthand for the Rydberg binding energy

$$W_\mu = \frac{-1}{2\mu^2}, \tag{3a}$$

$$W_\nu = \frac{-1}{2\nu^2}. \tag{3b}$$

For a driving light field that is near resonance with the core transition, the transition dipole moment between members of the two series is given by [1,26,30]

$$F\langle\nu|D|\mu\rangle = F\langle n_b|\langle b|D|a\rangle|n_a\rangle = d_{ab}\langle n_b|n_a\rangle \tag{4a}$$

$$= d_{ab}\frac{2\sin[\pi(\mu-\nu)]\sqrt{\mu\nu}}{\pi(\mu-\nu)(\mu+\nu)} = d_{ab}S_{\mu\nu}, \tag{4b}$$

where $d_{ab} = F\langle a|D|b\rangle$ is the Rabi frequency for the core states, determined by the matrix element of the dipole operator D and the the electric-field amplitude F of the light. The symbol $S_{\mu\nu}$ has been introduced to simplify the notation later. Note that Eq. (4b) contains the characteristic dependence of ICE on $\delta_a - \delta_b$ (mod 1) the *difference* in quantum defect between the two series. For $\delta_a = \delta_b$ (mod 1), $\mu - \nu$ is integer and $S_{\mu\nu}$ reduces to the Kronecker delta $\delta_{0,\mu-\nu}$. Physically, for $\delta_a = \delta_b$ (mod 1) the Rydberg wave functions of the upper and lower series are identical (except in the core region, which is negligibly small in the Rydberg overlap integral), so that the Rydberg overlap integral $\langle n_b|n_a\rangle$ is only nonzero for $n_a - \delta_a = n_b - \delta_b$.

For $\delta_a \neq \delta_b$ (mod 1), $S_{\mu\nu}$ is nonzero for all optical transitions between states $|\mu\rangle$ and $|\nu\rangle$ and has a maximum for $\mu \approx \nu$. This is well known in ICE spectroscopy and can be used to excite a range of autoionizing (upper series) Rydberg states from a single bound (lower series) Rydberg state with a (weak) probe laser, so-called shakeup.

Autoionization of the upper Rydberg series is introduced by a coupling $V_{\mu\epsilon}$ to a continuum, represented by the states $|\epsilon\rangle$ with energy ϵ. We have now introduced the states and couplings that are essential for the description of ICE and we neglect all further states and couplings that could be taken into account [31]. We can thus write the atomic state $|\psi\rangle$ as

$$|\psi\rangle = \sum_\mu a_\mu|\mu\rangle + \sum_\nu b_\nu|\nu\rangle + \int d\epsilon\, c_\epsilon|\epsilon\rangle \tag{5}$$

and the TDSE for the state $|\psi\rangle$ of Eq. (5) takes the form

$$i\frac{da_\mu}{dt} = (E_a + W_\mu)a_\mu + \sum_\nu S_{\mu\nu}b_\nu d_{ab}\cos\omega t + \int d\epsilon\, V_{\mu\epsilon}c_\epsilon, \tag{6a}$$

$$i\frac{db_\nu}{dt} = \sum_\mu S_{\mu\nu}a_\mu d_{ab}\cos\omega t + (E_b + W_\nu)b_\nu, \tag{6b}$$

$$i\frac{dc_\epsilon}{dt} = \sum_\mu V_{\mu\epsilon}a_\mu + \epsilon c_\epsilon, \tag{6c}$$

where ω is the angular frequency of the driving light.

The treatment so far is common in ICE calculations; now we continue by reducing the problem to a finite basis set. The continuum can be eliminated using very reasonable assumptions [32]: the continuum is taken to be flat in the (small) energy range of the autoionizing states we study, so that the autoionization matrix element does not depend on ϵ, $V_{\mu\epsilon} = V_\mu$. In addition, we assume the continuum to have infinite energy range (with respect the energy range of the upper Rydberg states we consider). Formally integrating Eq. (6c) and substituting the result in the integral over c_ϵ, which appears in Eq. (6a), results in

$$\int d\epsilon\, V_\mu c_\epsilon = \int d\epsilon\, V_\mu\left[-ie^{-i\epsilon t}\int_{-\infty}^{t} dt' \sum_{\mu'} V_{\mu'}a_{\mu'}(t')e^{i\epsilon t'}\right]$$

$$= -\pi i V_\mu \sum_{\mu'} V_{\mu'}a_{\mu'}(t), \tag{7}$$

which eliminates the continuum from Eq. (6a).

Since autoionization takes place close to the core, where the wave functions $|\mu\rangle$ are all similar, the autoionization coupling is given by

$$V_\mu = (-1)^{\mu+\delta_a}V\mu^{-3/2}, \tag{8}$$

where V is the coupling constant (taken to be real for convenience) and the dependence on μ is due to the scaling of the Rydberg wave function amplitude near the core. The alternating sign for consecutive Rydberg states is a consequence of our choice of the sign of the Rydberg states in Eq. (4a).

Finally, the optical frequency ω can be eliminated from Eq. (6) using the RWA [27]. This amounts to neglecting the nonresonant processes of photoemission from the lower series to the upper series and photoabsorption from the upper series to the lower series. The RWA is valid for a near-resonant light field as long as the Rabi frequency is much smaller than the optical frequency, i.e., under the conditions $d_{ab} \ll \omega$ and $\Delta \ll \omega$ for the detuning $\Delta = \omega - (E_a - E_b)$. Making the RWA [33] leads to the TDSE

$$i\frac{da_\mu}{dt} = (W_\mu - \Delta/2)a_\mu - \pi i V_\mu \sum_{\mu'} V_{\mu'}a_{\mu'} + \frac{1}{2}\sum_\nu d_{ab}S_{\mu\nu}b_\nu, \tag{9a}$$

$$i\frac{db_\nu}{dt} = \frac{1}{2}\sum_\mu d_{ab}S_{\mu\nu}a_\mu + (W_\nu + \Delta/2)b_\nu \tag{9b}$$

for the reduced wave function

$$|\phi\rangle = \sum_\mu a_\mu e^{-i(E_a+\Delta/2)t}|\mu\rangle + \sum_\nu b_\nu e^{-i(E_b-\Delta/2)t}|\nu\rangle. \tag{10}$$

Note that for this choice of phase factors in Eq. (10) the right-hand side of Eq. (9) does not explicitly depend on time anymore. Although Eq. (9) still contains an infinite number of Rydberg states, in most cases only a finite number of these is physically relevant. We can thus arrive at a TDSE for a finite basis set by selecting only the relevant states. Equations similar to our Eq. (9) were independently used by others for wave-packet calculations in the presence of an intense core-resonant light field [14,15].

The Hamiltonian matrix formed by the coefficients on the right-hand side of Eq. (9) is no longer Hermitian due to the autoionization of the upper Rydberg series to the continuum, hidden in the coupling terms V_μ. These terms not only cause decay, represented by the complex diagonal elements for the upper Rydberg series in Eq. (9a), but also induce an anti-Hermitian coupling between members

of the upper series. This type of coupling (also called "dissipative" or "resistive" coupling) is not nearly as well known as the usual Hermitian one, but has nevertheless been studied in quite different situations (see, e.g., Refs. [34,35]). As we will see in Sec. III A, this coupling has some interesting consequences.

As an additional aside we note here that direct photoionization of the bound Rydberg states $|\nu\rangle$ to the continuum $|\epsilon\rangle$ can be included in the finite-size Hamiltonian in a manner similar to the autoionization coupling of the upper Rydberg series [14]. For weak light fields the inclusion of this coupling leads to the well-known asymmetric Fano line shapes [1,36] due to the interference of this ionization path with the path via the autoionizing state. We have not studied the effects of the inclusion of the direct photoionization coupling in the present paper to keep the system as simple as possible and because in most ICE experiments such coupling turns out to be negligible [1].

III. CALCULATIONS

As indicated in the Introduction, we will be interested mainly in the regime where the Rabi frequency is of the order of the Rydberg spacing and in near-resonant excitation $\Delta \approx 0$. According to Eq. (4b), the light couples a state $|\nu\rangle$ mainly to states $|\mu\rangle$ with $\mu \approx \nu$. We thus expect that if the atom is initially prepared in a single Rydberg level, only a limited number of Rydberg states will be important for the atomic dynamics.

In our calculations we have therefore used a range of typically ten Rydberg states of each series, centered around $\mu \approx \nu \approx 20$. This center value was chosen since the relevant intensities and pulse durations are within reach experimentally in this case [4]. For $\nu \approx 20$ the Rydberg spacing is $W_{\nu+1} - W_\nu \approx \nu^{-3} \approx 1.25 \times 10^{-4}$ a.u., corresponding to a Kepler orbit time of $\tau_K = 2\pi\nu^3 \approx 5 \times 10^4$ a.u., i.e., 1.2 ps. A Rabi frequency that is equal

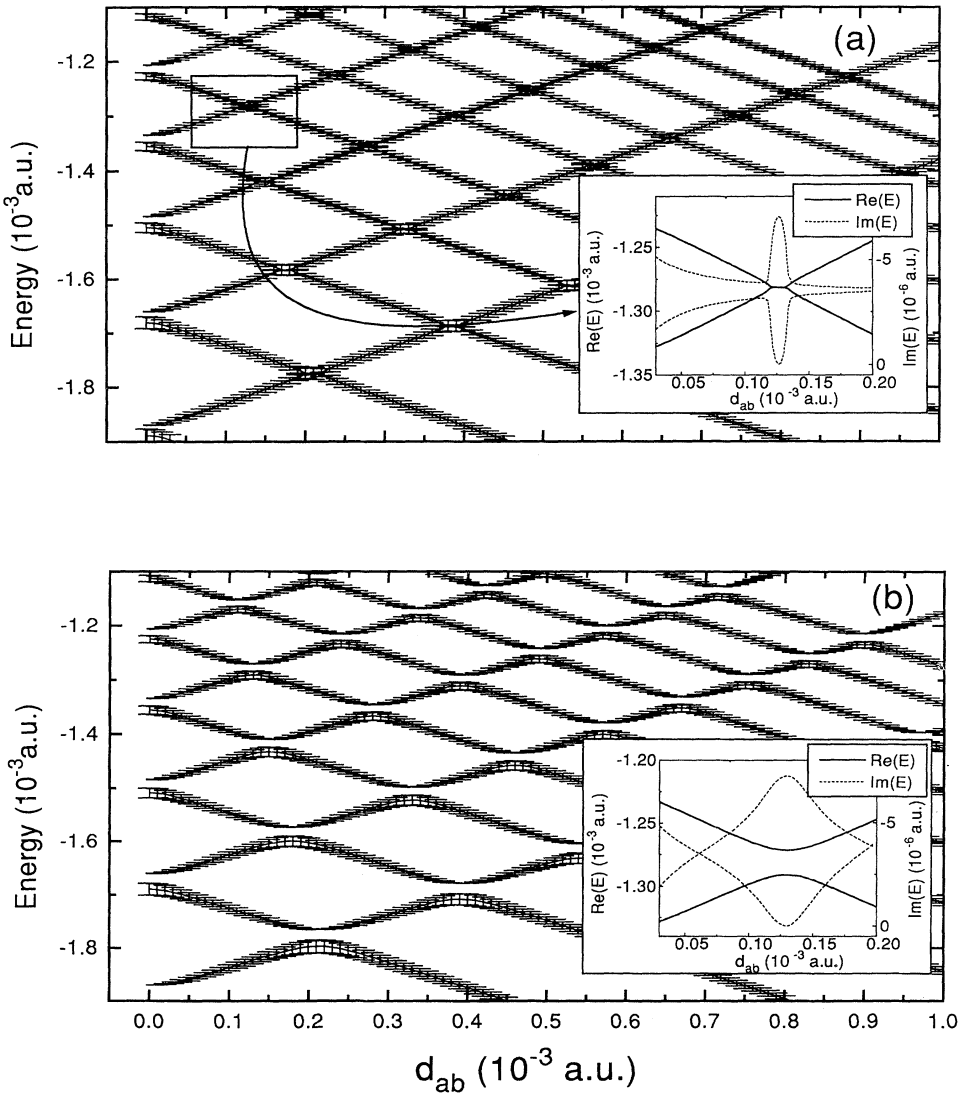

FIG. 2. Eigenenergies resulting from diagonalizing the Hamiltonian of Eq. (9) for ten Rydberg states of both the lower and upper series as a function of the Rabi frequency d_{ab}. The other parameters are listed in the text. The real part of the energies is plotted and the imaginary part (the decay rate) is drawn as an error bar. The insets show a particular (avoided) crossing on an expanded scale, with the real and the imaginary part of the eigenenergies plotted separately. (a) $\delta_a - \delta_b = 0$, $\Delta = 2 \times 10^{-5}$ a.u.; (b) $\delta_a - \delta_b = 0.15$, $\Delta = 0$; (c) $\delta_a - \delta_b = 0.5$, $\Delta = 0$; (d) $\delta_a - \delta_b = 0.15$, $\Delta = -3.3 \times 10^{-4}$ a.u.

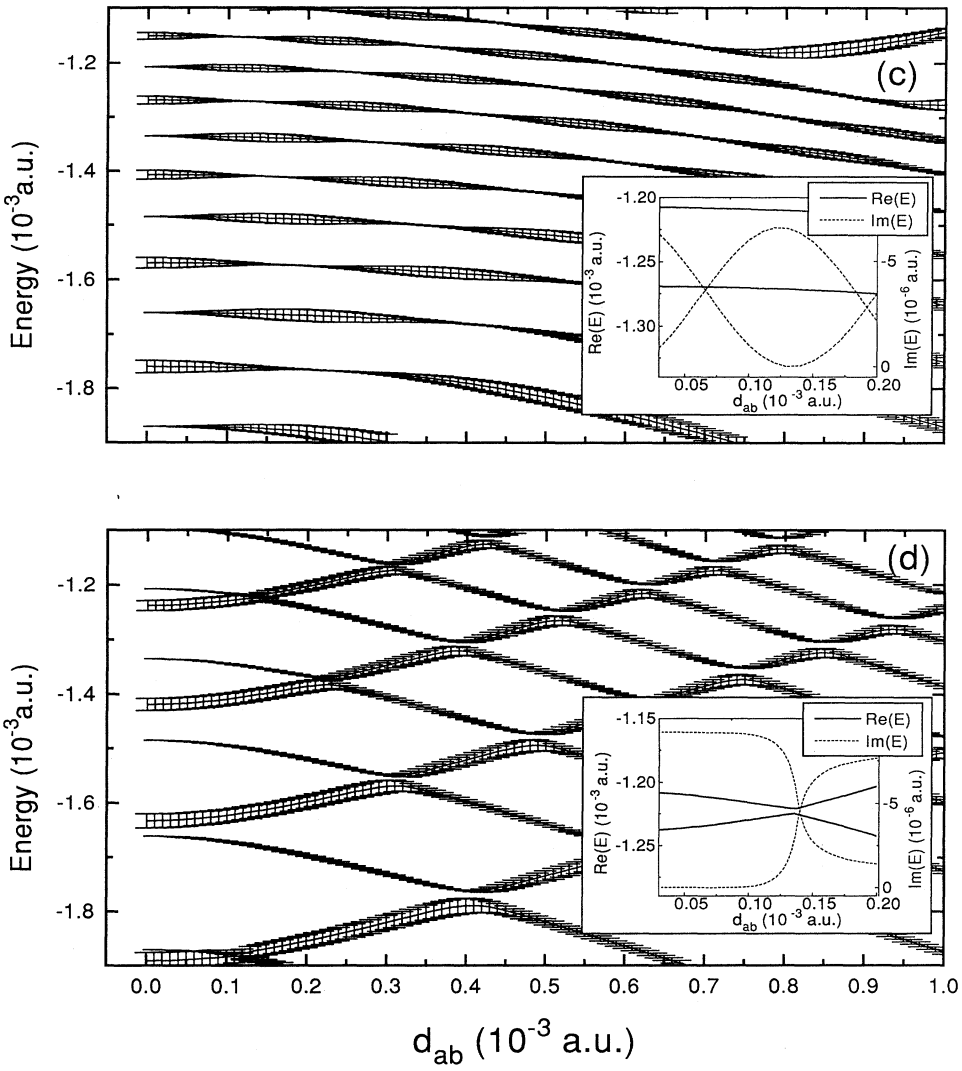

FIG. 2 *(Continued).*

to the Rydberg spacing implies $d_{ab} = 1.25 \times 10^{-4}$ a.u. $= 2\pi \times 0.8$ THz. Assuming a dipole matrix element $\langle a|D|b\rangle = 1$ a.u. (which is reasonable for a strong optical transition), the light intensity needed for this Rabi frequency is $I = F^2 = (d_{ab}/|\langle a|D|b\rangle|)^2 = 1.6 \times 10^{-8}$ a.u.= 0.5 GW/cm^2. The results are not expected to depend critically on the choice of ν once the light pulse duration and intensity are appropriately scaled (with ν^3 and ν^{-6}, respectively). The other parameters were chosen such that they reflect a typical Rydberg atom: $\delta_b = 0.65$, $\delta_a \approx \delta_b$, and $V = 0.132$ [corresponding to a population decay rate of $2\pi V_\mu^2 = 0.11\mu^{-3}$ and an autoionization probability of $1-\exp(-2\pi V_\mu^2 \tau_K) = 50\%$ per Kepler orbit time].

A. Eigenenergies

In order to gain insight into the behavior of the system we first calculate the eigenvalues of the (non-Hermitian) Hamiltonian defined by the coefficients on the right-hand side of Eq. (9), for fixed core Rabi frequency d_{ab}. The calculation was done numerically using standard methods of matrix diagonalization. The results are shown in Fig. 2 for different values of δ_a and ω as a function of d_{ab}. An instructive case is that of $\delta_a = \delta_b$, shown in Fig. 2(a). As noted in Sec. II, the transition dipole moment of Eq. (9) then reduces to $d_{ab}S_{\mu\nu}/2 = d_{ab}\delta_{0,\mu-\nu}/2$. If we ignore for the moment the (weak) autoionization coupling V, each member of the upper Rydberg series only interacts with the corresponding member of the lower Rydberg series and vice versa, and the Hamiltonian can be split into separate 2×2 matrices for each pair of states $|\nu\rangle$ and $|\mu\rangle$ with $\mu = \nu$. In other words, for equal quantum defects in the lower and upper Rydberg series the outer electron is not affected by the state of the core at all and stays in the same Rydberg state, while the interaction of the core with the light is described by the same simple two-level system for each Rydberg state. When the light is on resonance $\Delta = 0$, these levels are degenerate in dressed energy for zero light field strength. When the light is turned on, they are split in energy, with a separation of

d_{ab}, the Rabi frequency, and the dressed eigenstates are $(|\mu\rangle + |\nu\rangle)/\sqrt{2}$ and $(|\mu\rangle - |\nu\rangle)/\sqrt{2}$ (for each pair $\mu = \nu$). If we now allow for a small autoionization rate V, with $V_\mu^2 < d_{ab}$, the above dressed states will essentially remain eigenstates, while the autoionization rate, which only affects the states $|\mu\rangle$ for zero light field, will be distributed equally over the two new eigenstates. All these features are clearly illustrated in Fig. 2(a), where a small detuning Δ has been used to show the different decay rates for the two series at $d_{ab} = 0$.

As mentioned before, introducing the autoionization also introduces a coupling between the members of the upper Rydberg series. The influence of this is very small, except near the crossings of the dressed levels. The anti-Hermitian coupling between different $|\mu\rangle$ states causes a similar anti-Hermitian coupling between the two crossing dressed levels. The result is shown in the inset of Fig. 2(a), a blowup of one of the crossings. The anti-Hermitian coupling leads to a "contracted" level crossing for the (real part of the) eigenenergies, while the decay rates of the new eigenstates become zero and twice the original value, respectively. The interference of the two decay amplitudes of the crossing states is destructive and constructive, respectively. Thus there are states that are stable against autoionization at the level crossings, where the Rabi frequency is equal to the Rydberg spacing. Such states were also found by Robicheaux [3] in his theoretical analysis of the spectroscopy of this system.

Physically, at the level crossing the radial oscillation of the Rydberg electron and the Rabi oscillation of the core are "locked" [14,34,35] by the dissipative autoionization coupling. The new eigenstates at the crossing correspond to entangled states of the core and the Rydberg electron: in the long-lived state the Rydberg electron is far away from the core when the core is in the excited state $|a\rangle$ and it is near the core when the core is in the ground state $|b\rangle$, so that autoionization is inhibited. The short-lived state corresponds to a similar entanglement but with the role of the ground and excited core state reversed, i.e., now the Rydberg electron is close to the core when the core is in the excited state. The localized wave functions of the Rydberg electron in these entangled eigenstates are composed of superpositions of the (delocalized) ordinary Rydberg states that are present in the two crossing dressed levels.

It would be very interesting to observe the long-lived states and they might, for instance, reveal information about other decay mechanisms that are usually negligible compared to autoionization. Unfortunately, it is also quite difficult to observe these states experimentally since they require an intense light field with a constant amplitude for a time longer than the autoionization decay time.

As a final remark on the inset in Fig. 2(a) we note that the complex eigenenergies are nearly degenerate at the points where the real parts of the dressed eigenenergies meet. Such induced (near) degeneracies are typical for strongly driven systems involving decaying states [34,35]. For a recent theoretical example involving atoms driven by intense light, see Ref. [37].

In Fig. 2(b) the quantum defect of the upper series was changed so that $\delta_a - \delta_b = 0.15$. As noted in Sec. II, if $\delta_a \neq \delta_b$ (mod 1), a member of the lower Rydberg series $|\nu\rangle$ will not only be coupled by the light to the corresponding member of the upper series $|\mu\rangle$ with $\mu \approx \nu$ but also weakly to the neighboring states, i.e., shakeup is possible [cf. Eq. (4b)]. In our case, for a large core Rabi frequency, this leads to a (Hermitian) coupling between the dressed states and turns the contracted crossings of Fig. 2(a) into the usual avoided crossings, as is visible in Fig. 2(b). The size of the avoided crossings strongly depends on $\delta_a - \delta_b$ (mod 1), the difference in quantum defect between the two Rydberg series, through the overlap integral $S_{\mu\nu}$. The variation in decay rate of the eigenstates near the level crossing is still present, as is clearly visible in the inset of Fig. 2(b).

An interesting limiting case occurs if we choose $\delta_a - \delta_b = 0.5$ and tune the light exactly to the core resonance $\Delta = 0$. The result is shown in Fig. 2(c). The light now couples the Rydberg levels with comparable strength to the members of the other series just above and below and the net result is that the levels hardly shift their energy as a function of field strength. The levels are strongly mixed, as is evident from the oscillatory behavior of the decay rates as a function of field strength. Note that in experiments on ICE, where the principal quantum number of the inner electron is unchanged by the core transition, the quantum defects of two such Rydberg series usually do not differ by more than 0.2 [1], so that the case of Fig. 2(c) may be considered as rather unlikely in practice.

To conclude this section we show in Fig. 2(d) an example where the light is detuned significantly from the core resonance $\Delta = -3.3 \times 10^{-4}$ a.u. (i.e., approximately 2.6 Rydberg spacings). In this case the levels hardly shift until the Rabi frequency becomes comparable to the detuning. For lower Rabi frequencies, the decay rates only mix when members of the upper and lower Rydberg series are degenerate in (dressed) energy, so that the usual ICE spectrum of shakeup resonances is recovered for such large detunings.

B. Pulsed fields

As mentioned in the preceding subsection (and discussed by Robicheaux in Ref. [3]), measuring the energies and decay rates of the dressed eigenstates is difficult experimentally since it requires the use of an intense yet stable dressing light source. However, if one uses a pulsed light source, the effects of the dressing can also be observed, as we will show in this section. The basic idea is to start in a single bound Rydberg level and to use a light pulse that is sufficiently strong to pass one or more of the (avoided or contracted) level crossings of Fig. 2. Passing such a level crossing will in general cause a redistribution of the population over the dressed levels and (among other effects) will lead to population of *bound* Rydberg levels adjacent to the initial bound state once the light is off again. Such bound-state population transfer can subsequently be detected fairly easily [38].

As is well known [27], the actual amount of population

transferred when passing a level crossing will depend on the rate at which the crossing is passed (i.e., the rate of change of the Rabi frequency in our case) and on the shape of the crossing (determined in our case by the *difference* in quantum defect between the two series and the autoionization rate). We show below that considerable population transfer can be found for experimentally feasible light pulses, using the same atomic parameters as in Sec. III A.

Before we discuss the results of our calculations, we first emphasize that the population transfer is made possible by the large Rabi frequency of the core and that this effect can in principle also occur for very long light pulses [39]. It is thus quite different from the effect that population transfer can be observed when the pulse duration is much shorter than the Kepler orbit time (as discussed, e.g., in Ref. [8]).

The effects of a light pulse, nearly resonant with the core transition, were calculated by numerically integrating the time-dependent Schrödinger equation Eq. (9) using a fifth-order Runge-Kutta method with adjustable step size [40]. In order to verify the validity of our finite-basis-set Hamiltonian, we compared the results of the calculation for a narrow-bandwidth, low-intensity light pulse with those of perturbation theory [1]. As an example, Fig. 3(a) shows the total ionization yield, start-

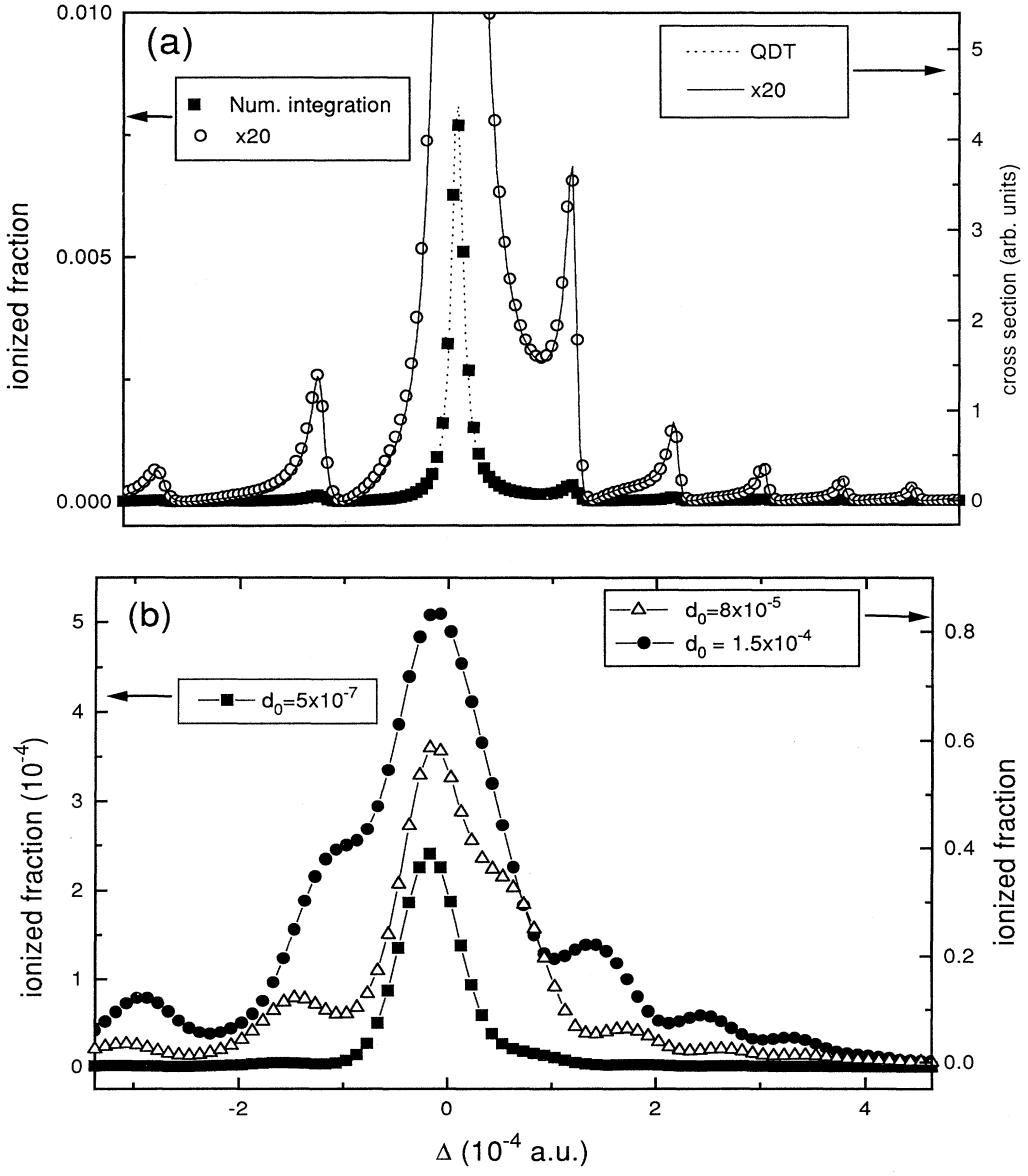

FIG. 3. Photoionization yield from initial state $\nu = 20.35$, as a function of light detuning Δ from the core resonance, for $\delta_a - \delta_b = 0.15$. (a) Perturbative regime, comparison of quantum-defect theory (QDT) to the numerical result, using Eq. (9) with a long ($\tau = 4 \times 10^5$ a.u.), weak ($d_0 = 5 \times 10^{-7}$ a.u.) pulse. (b) Nonperturbative regime, numerically calculated photoionization for three different pulse strengths, with a relatively short Gaussian pulse [Eq. (11), $\tau = 4 \times 10^4$ a.u. = 1 ps].

ing in the $\nu = 20.35$ bound state, for a weak, narrow-bandwidth Gaussian pulse. The same atomic parameters as in Fig. 2(b) were used and the light pulse was given by

$$d_{ab} = d_0 e^{-t^2/\tau^2}, \tag{11}$$

with $d_0 = 5 \times 10^{-7}$ a.u. and $1/\tau = 2.5 \times 10^{-6}$ a.u. (i.e., $\tau = 10$ ps). These numbers should be compared to the Rydberg spacing, which is $\mu^{-3} = 1.2 \times 10^{-4}$ a.u., and to the autoionization width $2\pi V_\mu^2 = 1.4 \times 10^{-5}$ for $\mu = 20$. Note that the ionization yield is given by the decrease in the norm of the wave function $|\phi\rangle$ (i.e., the population fraction transferred to the continuum during the light pulse) *plus* the population remaining in the upper Rydberg states at the end of the pulse. The latter states will decay by autoionization and will also be detected as photoionization.

The result is compared to the perturbative calculation of the photoionization cross section σ in standard quantum-defect theory [1]

$$\sigma \propto 2\pi^2 V^2 \frac{1 + \tan^2 \pi\mu}{(2\pi^2 V^2)^2 + \tan^2 \pi\mu} \mu^3 S_{\mu\nu}^2, \tag{12}$$

where μ is now taken as a continuous variable, defined by $-1/2\mu^2 = W_\mu = W_\nu + \Delta$. The agreement is excellent. Note that our numerical calculations reproduce quite well the shape of the ionization curve of Eq. (12) between the resonances, even though we have only used the quantum-defect result of the matrix elements for on-resonance excitation.

When the light pulse is shortened by a factor 10 ($\tau = 4 \times 10^4$), with d_0 fixed ($d_0 = 5 \times 10^{-7}$ a.u.), the ionization spectrum shown in Fig. 3(b) results. Clearly most of the details of the excitation structure are washed out due to the large bandwidth of the excitation pulse, but the general features remain unaltered. Increasing the peak Rabi frequency of the light significantly (to $d_0 = 8 \times 10^{-5}$ a.u.) changes the spectrum dramatically, as is also visible in Fig. 3(b). The main peak broadens and the shakeup and shakedown peaks, which were very weak initially, become clearly visible. Note that the broadening is due the large Rabi frequency and not only to ionization depletion of the bound states during the light pulse since the Rabi frequency is much higher than the autoionization decay rate. Thus this Rabi broadening is quite different from the depletion broadening observed in ICE using nanosecond lasers [1,41]. In the latter case the ground state is depleted during the laser pulse and the ionization rate is given by the perturbative rate. In our case, a major part of the initial population is transferred to other Rydberg states during the light pulse. Directly after the light pulse, there is still relatively little population transferred to the continuum and most of the population is still in the autoionizing (and in the bound) Rydberg series.

When the peak Rabi frequency is increased further ($d_0 = 1.5 \times 10^{-4}$ a.u.), the resonances broaden so much that the shakeup satellites are no longer recognizable. From the discussion of Fig. 2 we expect that for such high intensities transitions between the dressed levels occur. This is indeed the case and can be observed as population ending up in Rydberg states neighboring the initial Rydberg state. Examples of this for two different pulse strengths are shown in Fig. 4. Note that for $d_0 = 1.4 \times 10^{-4}$ a.u., more population is left in one of the neighboring bound Rydberg states than in the initial state, while for $d_0 = 1.8 \times 10^{-4}$ a.u. almost 40% of the population returns to the initial state. The final-state population distribution depends critically on the exact value of the parameters chosen. The size of the level crossing and the rate of change of the light amplitude determine the amount of population transferred. In addition, since during the light pulse each relevant dressed-level crossing will be passed on both the rising and the falling edge of the pulse, there will in general

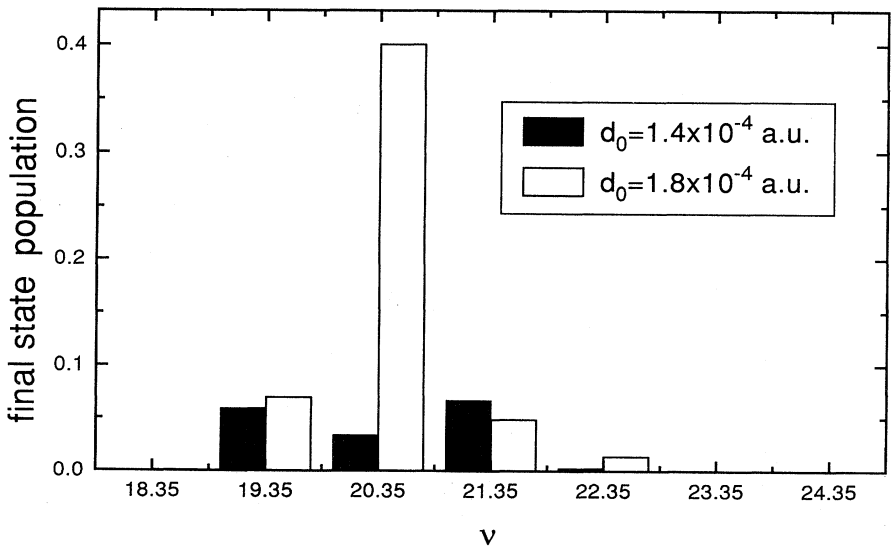

FIG. 4. Population distribution over the lower Rydberg series after a Gaussian light pulse [Eq. (11), $\tau = 4 \times 10^4$ a.u.] for two different peak intensities. The initial state is $\nu = 20.35$; the atomic parameters were chosen the same as in Fig. 2(b); the frequency of the light is on resonance ($\Delta = 0$).

be interference between these two crossings. We do not discuss these effects in detail here. The sensitivity to the parameters is illustrated in Fig. 4 by the large difference in final-state distribution in the two cases shown, where only the light pulse amplitude was changed by 30% (i.e., a change in peak intensity of 70%). These cases correspond to a peak light strength that reaches or just crosses the first level crossing. In an experiment using focused laser beams, the ionization yield and the final population distribution are averaged over the range of peak light strengths present in the laser focus and these interference effects are averaged out.

An interesting variation on the effects studied here would be reached when the peak Rabi frequency in the pulse would become larger than the binding energy of the initial state. For such Rabi frequencies, the coupled Rydberg series form two new Rydberg series, each converging to its own dressed core state [3]. The upper dressed series will be coupled to the continuum above the lower dressed series, giving rise to autoionization. This would result in electrons emitted at an energy corresponding to the Rabi frequency instead of to the photon frequency. Such low-energy electrons may be said to result from "zero-photon ionization" since photons are only scattered for this ionization process, not absorbed. Note that in this case our finite-basis approach would be inadequate since on the rising edge of the light pulse the atomic state should pass avoided crossings with dressed levels connected to the infinitely many Rydberg states with higher principal quantum number than the initial state.

IV. CONCLUSIONS

We have theoretically investigated the properties of two Rydberg series coupled by a strong light field resonant with the core transition. When the Rabi frequency equals the Rydberg level spacing, level crossings occur, which can be avoided or contracted. At these level crossings stable (dressed) states are found. The minimum separation in the avoided level crossings depends on the difference in quantum defect of the two bare series. For equal quantum defect contracted crossings show up due to the anti-Hermitian coupling associated with autoionization. Numerical integration of the time-dependent Schrödinger equation shows that a strong light pulse, shorter than the autoionization decay time, can lead to broadening of the photoionization spectrum and to transitions between the dressed levels, resulting in population of bound Rydberg states neighboring the initial state.

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