# Structure of autoionizing Rydberg series in strong laser fields: A multichannel-quantum-defect-theory approach

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We study one-photon resonant two-photon ionization into an autoionizing Rydberg series for the case of a weak excitation from the ground state to a resonant intermediate state, followed by a strong-field transition to a structured continuum. The dipole strength for the transition into the autoionizing series is thereby modeled by a multichannel-quantum-defect-theory expression taking into account the infinite number of autoionizing Rydberg states. Analytical expressions are derived for the total ionization rate in the limit of extremely overlapping resonances.

Recent multiphoton-ionization (MPhI) experiments in two-electron systems (alkaline earth)[1,2] have stimulated theoretical work on the dynamics of autoionizing transitions in strong laser fields. These autoionizing states appear as intermediate resonances and final states in these processes. The theoretical models studied so far are essentially all high-intensity generalizations of Fano's description of (weak-field) transitions of a bound state to an isolated autoionizing resonance, modeled by a bound (two-electron-excited) state coupled via configuration interaction to a structureless continuum (of single-excited electrons).[3-13] The characteristic feature of transitions to autoionizing states is the appearance of asymmetric line profiles due to interference between the direct photoionization to the continuum and the resonant excitation of the bound (two-electron excited) state followed by a decay to the continuum. A variety of novel effects in the total ionization probability, double resonance, and photoelectron spectrum, have been predicted for high laser intensities, when the autoionizing transition becomes saturated, which are all different manifestations of this basic interference pattern.

Present-day spectroscopic work parametrizes the configuration interaction in two-electron systems with the help of multichannel-quantum-defect-theory (MQDT),[14,15] allowing the interaction between different Rydberg *series* and dipole matrix elements to these states to be expressed in terms of a few physically significant parameters. The question therefore arises: To what extent can the high-intensity calculations based on the Fano model for a single (isolated) autoionizing resonance be extended to interacting Rydberg series and continua using the MQDT formalism? In principle, a complete dynamical calculation of MPhI for a Rydberg series involving an infinite number of states converging to a continuum threshold meets severe mathematical problems. There are, however, a variety of simple limiting cases in which the strong-field structure of autoionizing Rydberg series can be worked out even analytically.

In this Rapid Communication we study MPhI in a double optical resonance configuration (Fig. 1), where a weak laser of frequency $\omega_1$ excites atoms from the ground state $|g\rangle$ with energy $E_g$ to one of the (low-lying) excited states $|e\rangle$ (energy $E_e$), which is coupled by a strong second laser of frequency $\omega_2$ to an autoionizing Rydberg series. For a single isolated autoionizing state, this problem has been discussed in detail in the literature.[4,5,8,11,12] For weak excitation by the first laser the time-independent ionization rate in the long-time limit is given by[12]

$$R=-\tfrac{1}{2}|\Omega_{eg}|^2\,\mathrm{Im}\{[E_g+\omega_1-E_e-p|\epsilon_2|^2]^{-1}\} \tag{1}$$

and

$$p=\left\langle e\left|\left(\vec{\mu}\cdot\vec{e}_2^{\,*}\frac{1}{E_g+\omega_1+\omega_2-H_A+i\epsilon}\vec{\mu}\cdot\vec{e}_2+\vec{\mu}\cdot\vec{e}_2\frac{1}{E_e-\omega_2-H_A}\vec{\mu}\cdot\vec{e}_2^{\,*}\right)\right|e\right\rangle \tag{2}$$

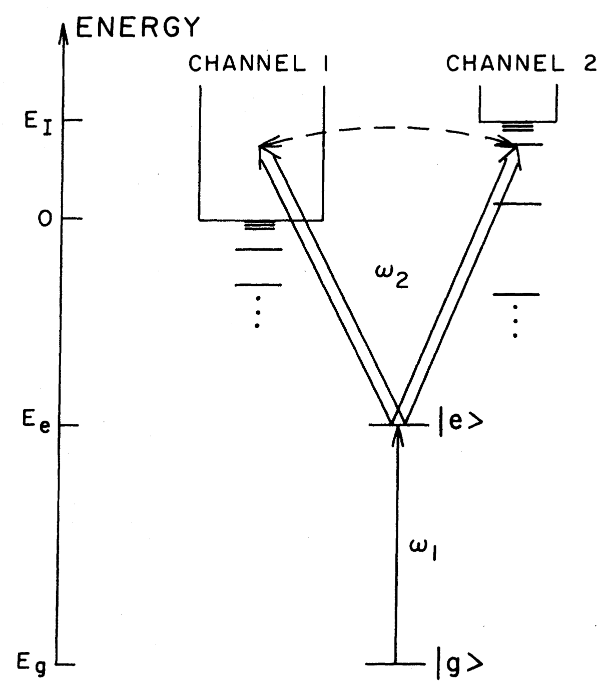


FIG. 1. Atomic configuration.

 

being a complex polarizability of level $|e\rangle$ due to the second (strong) laser with intensity $|\epsilon_2|^2$ and polarization $\vec{e}_2$. $H_A$ is the Hamiltonian of the free atom. $p$ describes the shift and width of $|e\rangle$ due to the second laser. We shall limit ourselves in the following to frequencies $\omega_2$ not too close to the threshold of the first channel. This allows us to split the polarizability (2) into a resonant (strongly energy-dependent) contribution

$$p_r = \int_0^{E_I} dE \frac{|\langle E|\vec{\mu}\cdot\vec{e}_2|e\rangle|^2}{E_g+\omega_1+\omega_2-E+i\epsilon} \tag{3}$$

coming from the autoionizing region extending from $0 \leq E \leq E_I$, with $E_I$ the second ionization threshold (Fig. 1), and a slowly varying contribution. The $|E\rangle$ are the exact (diagonalized) continuum states. Our model ignores any incoherent decay mechanisms and laser-induced ionization from the autoionizing states to higher channels.

In a two-channel MQDT approximation the dipole density of the transition from the excited state $|e\rangle$ to the structured continuum $|E\rangle$ has the form

$$|\langle E|\vec{\mu}\cdot\vec{e}_2|e\rangle|^2 = \frac{|P(E)|^2}{\mathcal{R}} \frac{[q(E)+x(E)]^2}{1+x(E)^2} , \tag{4}$$

as discussed in detail by Seaton.[15] The function $x(E) = \tan\pi(\nu+\alpha)/\tanh\pi\beta$, with $\nu$ an effective quantum number in the second (bound) channel, defined by

$$E = E_I - \frac{\mathcal{R}}{\nu^2} ,$$

leads to a repeated resonance structure in Eq. (4) reflecting the autoionizing Rydberg series. The parameter $\alpha$ may be identified with the quantum defect of the unperturbed Rydberg series in channel two.

The dipole strength $|P(E)|^2$ is related to the transition from $|e\rangle$ to the structureless continuum of channel one; $q(E)$ is a generalized Fano parameter, while $\beta$ describes the configuration interaction between the Rydberg series and the continuum. For $\beta$ small it is twice the ratio between the autoionization rate and the level spacing in the series.

In the limit $\beta \to 0$, Eq. (4) reduces to a Rydberg series of isolated autoionizing resonances, each of which is described by the standard Fano formula.[16] The opposite limit $\beta \to \infty$ corresponds to a Rydberg series of extremely overlapping resonances. According to MQDT, $\alpha$, $\beta$, $P$ and $q$ are slowly varying functions of the energy. Substituting the MQDT formula (4) into Eqs. (3) and (1) gives an expression for the MPhI probability into a continuum with an (infinite) series of autoionizing Rydberg states. Because the double-resonance ionization profiles have been discussed in the literature for well-separated autoionizing states (i.e., $\beta \to 0$), we concentrate here on the opposite limit of strongly overlapping resonance ($\beta \to \infty$). If we ignore the energy dependence of the MQDT parameters, Eq. (1) can, after straightforward albeit tedious manipulations, be written in the form

$$R = -\frac{|\Omega_{eg}|^2}{\Gamma}\mathrm{Im}\{[\Delta+\sin\phi(\bar{\nu})-A(\bar{\nu}) + i(1+\cos\phi(\bar{\nu}))]^{-1}\} , \tag{5}$$

with $\Delta$ a dynamical detuning of the first laser from the first transition frequency, including slowly energy-dependent contributions to the Stark shift, measured in units of the laser-induced ionization rate

$$\tfrac{1}{2}\Gamma = \pi \frac{|P|^2|\epsilon_2|^2}{\mathcal{R}} \frac{q^2+1}{2} .$$

The effective quantum number $\bar{\nu}$ contains the dependence on the laser frequencies,

$$E = E_g + \omega + \omega_2 = E_I - \frac{\mathcal{R}}{\bar{\nu}^2} .$$

The phase $\phi(\nu)$ is given by

$$\phi(\nu) = -2\pi(\nu+\alpha) + \arcsin 2q/(q^2+1)$$

with $\nu_0 = \sqrt{\mathcal{R}/E_I}$. Furthermore, we have defined a function

$$A(\bar{\nu}) = \frac{1}{\pi}[f(2\pi(\bar{\nu}-\nu_0)) - f(2\pi(\bar{\nu}+\nu_0))]\sin\phi(\nu_0) - \frac{1}{\pi}[g(2\pi(\bar{\nu}-\nu_0)) + g(2\pi(\bar{\nu}+\nu_0))]\cos\phi(\nu_0) , \tag{6}$$

where $f$ and $g$ are combinations of integral-sine and -cosine

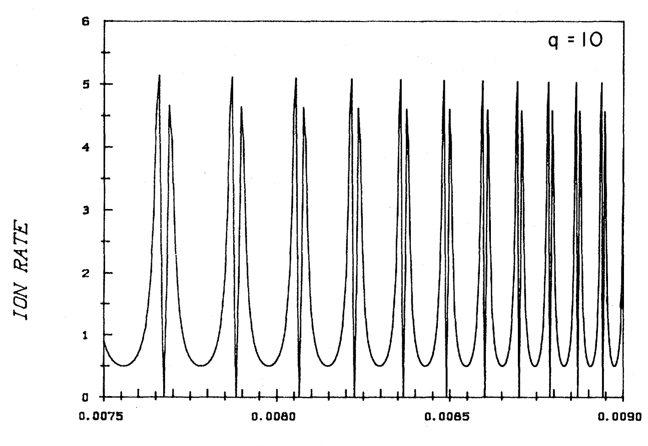


FIG. 2. Total ionization rate $R$ in units of $|\Omega_{eg}|^2/\Gamma$ as a function of the energy $E = E_g + \omega_1 + \omega_2$ (in Ry) for $\Delta = 0$ and $q = 10$.

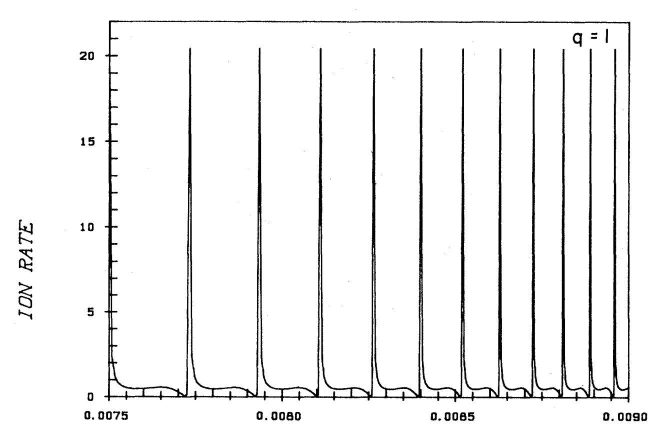


FIG. 3. Total ionization rate $R$ in units of $|\Omega_{eg}|^2/\Gamma$ as a function of the energy $E = E_g + \omega_1 + \omega_2$ (in Ry) for $\Delta = 0$ and $q = 1$.

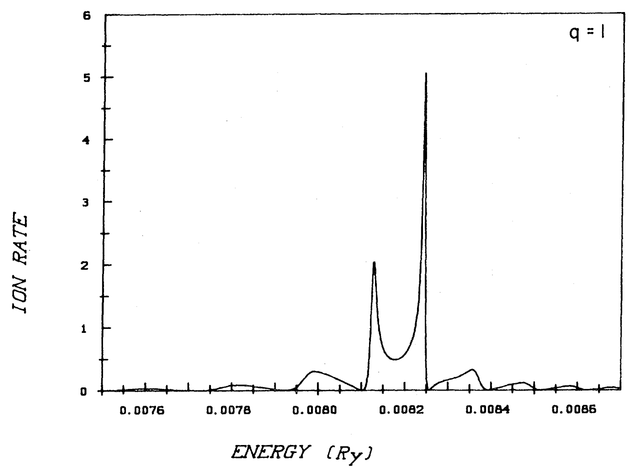


FIG. 4. Total ionization rate $R$ in units of $|\Omega_{eg}|^2/\Gamma$ as a function of the energy $E=E_g+\omega_1+\omega_2$ (in Ry) for $\omega_2=$const and $\frac{1}{2}\Gamma=\Delta E\times 0.5$.

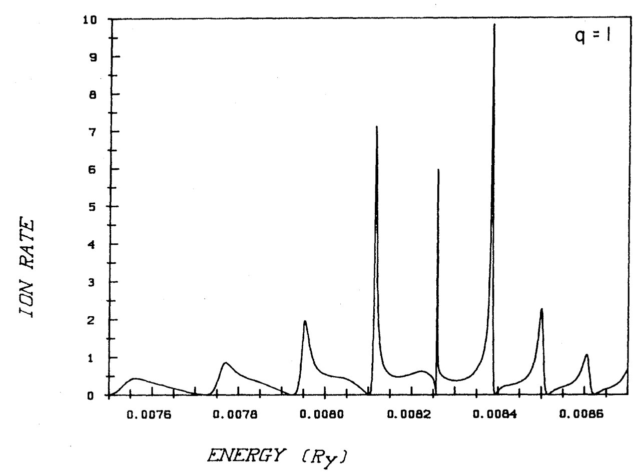


FIG. 5. Total ionization rate $R$ in units of $|\Omega_{eg}|^2/\Gamma$ as a function of the energy $E=E_g+\omega_1+\omega_2$ (in Ry) for $\omega_2=$const and $\frac{1}{2}\Gamma=\Delta E\times 2$.

functions as defined by Abramowitz and Stegun.[17]

In order to exhibit the strong-field structure of the extremely overlapping autoionizing Rydberg series inherent in formula (5) we consider a model atom with $E_I=0.01$ Ry and $\alpha=2.8$. In Figs. 2 and 3 we have plotted the total ionization rate $R$ in units of $|\Omega_{eg}|^2/\Gamma$ as a function of $E=E_g+\omega_1+\omega_2$ (in rydbergs) for fixed laser frequency $\omega_1$ and different $q$ values. The frequency of the first laser is chosen so that the dynamical detuning from resonance $\Delta$ is zero. The considered energy range extends from $20\leq\bar{\nu}\leq 31$. Contrary to the off-resonance case corresponding to large detunings $\Delta$, where the ionization rate is proportional to

$$R\sim[q\cos\pi(\bar{\nu}+\alpha)+\sin\pi(\bar{\nu}+\alpha)]^2/\Delta^2$$

with zeros at energies corresponding to $\tan\pi(\bar{\nu}+\alpha)=-q$ and minima at $\tan\pi(\bar{\nu}+\alpha)=1/q$, the character of the two-step-excitation profiles is quite different. Whereas the zeros remain at the same positions, local minima with a value of $R\cong\frac{1}{2}(|\Omega_{eg}|^2/\Gamma)$ appear approximately at the positions of the maxima of the corresponding off-resonance profiles. Each of these minima is surrounded by two maxima whose relative height is strongly dependent on the value of the function $A(\bar{\nu})$ and, in particular, on the phase $\phi(\nu_0)$ as is apparent from Figs. 2 and 3. The small but nonzero value of the slowly energy-dependent function $A(\bar{\nu})$ gives rise to these effects. Note that $R=\frac{1}{2}(|\Omega_{eg}|^2/\Gamma)$ independent of the laser frequency $\omega_2$ if $\Delta-A(\bar{\nu})=0$.

In Figs. 4 and 5, we study the dependence of the ionization rate $R$ on the laser frequency $\omega_1$ for fixed frequency $\omega_2$ and $q=1$. In particular, we are interested in the intensity dependence of these resonance profiles. The frequency of the second laser is chosen so that $\tilde{E}_e+\omega_2$ is equal to the energy at which we would observe a maximum in the off-resonance profile corresponding to $\bar{\nu}=26.25-\alpha$. Thereby the slowly energy-dependent contribution to the quadratic Stark shift has been absorbed in the modified energy of the excited state $\tilde{E}_e$. At intensities of the second laser at which the field-induced ionization rate $\frac{1}{2}\Gamma$ is much less than the energy separation between the adjacent resonances $\Delta E$ we would observe a single peak in these resonance profiles at an energy $E$ which corresponds to the resonant one-photon ionization from $|g\rangle$ to $|e\rangle$, i.e., $E=\tilde{E}_e+\omega_2$. Increasing the intensity of the second laser we observe ac-Stark splitting of the autoionizing resonance located at $\tilde{E}_e+\omega_2$ as soon as $\frac{1}{2}\Gamma$ becomes comparable with the energy separation $\Delta E$. Due to the interference effects involved in the excitation of the autoionizing Rydberg series both ac-Stark-split states have different widths, as is evident from Fig. 4. Contrary to the case of isolated resonances the adjacent autoionizing states give rise to a sideband structure. In Fig. 5 the intensity of the second laser is so high that two resonances are involved in the ac-Stark-splitting process. Now each of these resonances is split and all four ac-Stark-split states have different widths.

In this Rapid Communication we have investigated resonance profiles of two-step ionization processes via a series of strongly overlapping autoionizing resonances. It should be noted that the form of the resonance profiles obtained in this paper strongly depends on the appearance of zeros in the total ionization rate at certain energies because we assumed that the autoionizing Rydberg series may decay into only *one* continuum. The consideration of more than one continuum in the excitation process causes an imperfect correlation[16] between the laser-induced ionization from the excited state $|e\rangle$ and the autoionization process and will tend to wash out this structure.

## ACKNOWLEDGMENTS

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