Laser-Induced Degeneracies Involving Autoionizing States in Complex Atoms

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An initial bound state and an autoionizing state of a complex atom (or ion) which are resonantly coupled by a radiation field can both be characterized in a Floquet approach by complex quasienergies. For certain values of the field's intensity and frequency, the two quasienergies become equal, giving rise to laser-induced degenerate states (LIDS). This phenomenon is demonstrated by applying the *ab initio,* fully nonperturbative R-matrix Floquet theory to Ar, H^-, and He. The general case is considered by constructing models containing discrete states and continua. Results for one- and for two-photon resonances are presented and suggestions for experimental observation are given.

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In this paper we demonstrate that laser-induced degeneracies exist between the ground state and an autoionizing state of an atomic system in a strong laser field. This constitutes an experimentally accessible application of the pioneering work of Berry [1], where the adiabatic passage around degeneracies in a parameter space was described and which sparked intensive study, particularly with respect to the associated geometric phase [2,3]. In the simplest case, going around a circuit about a degeneracy leads back to the original state apart from a dynamical and a geometrical phase factor. Atoms in laser fields offer the additional feature that, since they are decaying in the field, the dynamical phase has an imaginary part, which complicates the topology [4]. The appearance of laser-induced degenerate states (LIDS) in atomic hydrogen in a two-color laser field has been reported in [4,5]. In our case, a single laser is needed to produce a degeneracy, and thus the parameter space is two-dimensional and characterized by the laser frequency and intensity.

Let us first recall that autoionizing or doubly excited states [6] of atoms or molecules will produce characteristic resonance structures in the yield not only in photoionization [7], but also in multiphoton ionization (e.g., [8,9]). Atomic and molecular states may also be embedded in the continuum by dressing an atom or molecule by an intense laser field. This process, reviewed by, e.g., Knight *et al.* [10], is referred to as laser-induced continuum structure or LICS. Unlike the autoionizing states mentioned above, the decay of these LICS states is caused by the laser field coupling a bound state with the continuum so that a second laser is required for the observation of LIDS.

In our calculations we use the recently developed R-matrix Floquet theory [9,11] which is both nonperturbative and also includes electron-electron correlation effects for complex atomic systems. In this way the mechanisms that produce the autoionizing states and the laser dressing can both be accurately included. We study in detail the situation where, by tuning the frequency of the laser and varying its intensity, the dressed autoionizing state and the dressed ground state can be made degenerate, i.e., have *the same position and width,* giving a pair of LIDS. These are the first *ab initio* calculations of LIDS involving a bound and an autoionizing state for a real atomic system. In our examples we study both a one- and a two-photon coupling. Atomic units (a.u.) will be used unless otherwise stated.

The R-matrix Floquet theory is discussed in detail in [11]. Here we just remark that the Floquet ansatz yields a complex quasienergy for an atomic system in a monochromatic laser field of frequency ω and intensity I, where the imaginary part of the energy, $-i\Gamma/2$, is proportional to the width, or decay rate, Γ. This represents a quasistationary picture of a system that decays in time with the usual exponential decay law [12]. It is implied here that the preparation of the atomic system can be achieved by an adiabatic turn-on of the laser (however, on a time scale short enough compared to the lifetime of the atomic system). For the diabatic case, in which there can be significant transfer of population into other levels, a full time-dependent analysis must be performed (see, e.g., [10]).

As a first example, we have applied the R-matrix Floquet theory to Ar. Using the inner region configuration-interaction basis adopted by Burke and Taylor [13], we have obtained very accurate results for the $2p^6 3s 3p^6 np\,^1P^\circ$ resonance series in Ar for weak fields in agreement with [13], and we choose its lowest ($n = 4$) member as the autoionizing state, tuning the frequency around 0.99 a.u., corresponding to a one-photon resonance.

In Fig. 1 we show the resulting complex energies, for a few fixed values of the frequency, for an intensity up to 5×10^3 W/cm^2. The zero-field position of the ground state on the real energy axis is $E_g = -0.57816$,

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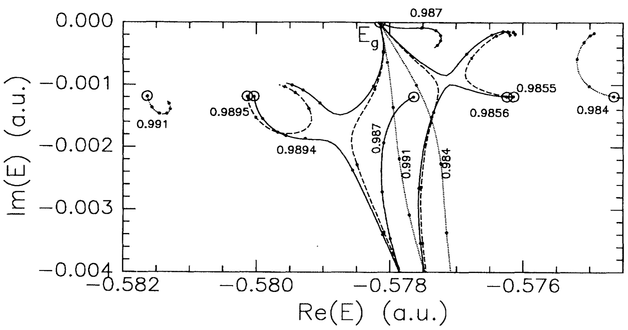

FIG. 1. Trajectories of the complex Floquet quasienergies for the ground state and the $2p^6 3s 3p^6 4p\ ^1P^\circ$ autoionizing state of Ar, connected by a one-photon transition, for an intensity varying from 0 to 5×10^{13} W/cm^2. The detunings are indicated (in a.u.).

while the energy of the autoionizing state is shifted by $-\omega$. Thus the zero field position of the autoionizing state (denoted by the big circles) changes with ω, and is at the complex energy $-\omega + 0.40936 - i0.00119 = -\omega + E_a - i\Gamma_a/2$, where Γ_a is the field-free width of the autoionizing state. For each frequency there are two curves, one connected adiabatically with the ground state, the other with the autoionizing state; corresponding curves have the same linetype (solid, dashed, or dotted) and some are labeled by the frequency. The small dots give the intensity in steps of 9×10^{12} W/cm^2. The corresponding detuning is defined as $d = E_a - E_g - \omega$. At large (positive or negative) detunings (e.g., $\omega = 0.991$) the autoionizing state does not move much from its position while the width of the ground state increases with intensity. At very small detunings ($\omega = 0.987$), just the opposite happens: the curve connected to the autoionizing state increases in width with intensity, while the ground state is "trapped" close to the real axis. For intermediate detunings, both on the positive and on the negative side, two structures are visible, about which the curves of the ground state and of the autoionizing state exchange their roles. There is a "critical point" at the center of each of these two structures, at which the two complex energies are exactly degenerate such that LIDS occur.

In order to understand the general features of these LIDS structures, we have used a model that retains the essential ingredients of the full R-matrix Floquet calculation, namely, a bound state coupled nonperturbatively by the field to an autoionizing state and to the continuum. Models of this type have been widely used to study various aspects of LICS and of autoionization in strong fields [14,15]. The complex energies of the ground and the autoionizing states as a function of field intensity I and frequency ω are given by [10] (ignoring the small field-induced shifts)

$$2E_{1,2} = E_a + E_g - \omega - i(\Gamma_a + I\gamma)/2 \pm \Omega\,, \qquad (1)$$
$$\Omega = \{[d - i(\Gamma_a - I\gamma)/2]^2 + \Gamma_a I\gamma[q - i]^2\}^{1/2}.$$

Here γ is the coupling parameter of the ground state to the unstructured continuum (so that the off-resonance width at low intensity I is equal to γI—neglecting any variation of γ with ω) and q is the Fano line profile index for the resonance transition. For the results shown in Fig. 2, we have chosen $q = -0.3$ (the correct value for the transition to the autoionizing state considered in Fig. 1), and we have taken units in which $\Gamma_a = 2$ and $\gamma = 2$.

The good qualitative agreement of Figs. 1 and 2 indicates that the LIDS and thus the form of the trajectories are a general phenomenon for such an arrangement. In particular, LIDS can also be inferred from the model of Agarwal *et al.* [15]. The full R-matrix Floquet calculation does reveal a few important corrections to the model, however: (i) In Fig. 1 the curves exhibit no "perfect trapping" zeros at $I > 0$, since there are more continua into which the system can decay by multiphoton ionization (in particular, for the degeneracy point on the left, where the intensity is higher). (ii) The picture in Fig. 1 is distorted due to the laser-induced shifts in the real part of the energies of the two states, which can be considerable but have been neglected in the model of Fig. 2. This is also the reason why we have refrained from scaling the parameters γ and Γ_a in the model to the values of the calculation of Fig. 1.

The model can give an indication for finding systems in which LIDS are experimentally accessible. At the degeneracy we must have $\Omega = 0$, which gives a fourth-order equation in I (or in d). This is readily solved to yield two unphysical solutions ($I_{3,4} = -\Gamma_a/\gamma$), while the two physical solutions are

$$d_{1,2} = \Gamma_a(q \pm \sqrt{q^2 + 1})\,, \qquad (2)$$
$$I_{1,2} = \frac{1}{\gamma\Gamma_a} d_{1,2}^2\,.$$

Therefore, in order to obtain a degeneracy at low intensity, it is desirable to take a system for which Γ_a/γ is small. This could be achieved by using (i) a higher-n member of the autoionizing Rydberg series, since Γ_a scales with

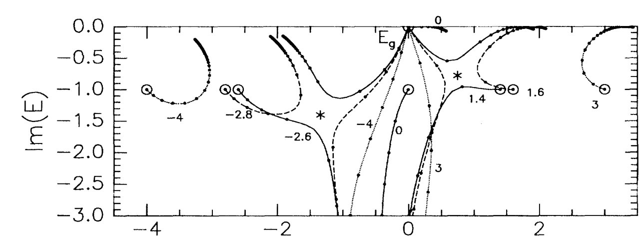

FIG. 2. Trajectories of the complex energies of Eq. 1, for an intensity varying from 0 to 20 (the dots indicate intensity steps of 0.5), for the detunings indicated next to the curves. The units are arbitrary (see text), and the energy scale is chosen such that $E_g = 0$. The stars denote the points where the two complex energies are exactly degenerate.

n as $(n-\mu)^{-3}$ (where μ is the quantum defect) while γ varies much more slowly, or (ii) an autoionizing state with large q. For $q = 0$ the two degeneracy points are at the same intensity, but for large q one of the intensities $I_{1,2}$ becomes small (the other becomes large and their product is constant). In the usual LICS case, another bound state is coupled to the continuum by a second laser so that Γ_a can be varied by changing the intensity of this laser.

It would be even more interesting if the LIDS were to appear also in a multiphoton context, which is the usual one in practical laser-atom interaction experiments. As a second example, we present in Fig. 3 the results for a two-photon resonant case, obtained from an R-matrix Floquet calculation for the H^- ion (using the same $1s, 2s, 2p$ basis as in [9]), where the ground state is coupled by two photons with the lowest-lying ${}^1S^e$ autoionizing state. In this case, one photon is sufficient to detach the electron, and therefore at weak fields the detachment rate, $\Gamma = -2\,\mathrm{Im}(E)$, is proportional to the field intensity and structureless over the resonance region (see also [9] for an example in He). The trajectories of the complex energies of the ground state and of the autoionizing state (the latter shifted by -2ω) are shown, for an intensity up to 1.6×10^{13} W/cm^2 along the curves. The dashed curves correspond to $\omega = 0.187\,50$ and the solid curves to $\omega = 0.187\,52$ (the detuning is now $d = E_a - E_g - 2\omega$). A degeneracy point can be seen at $I = 7 \times 10^{12}$ W/cm^2, which in its close surroundings is very similar to the left one observed for the one-photon resonance case. However, its upper left (high-intensity) "arm" is bent down, toward larger widths due to the importance of the extra continuum channels.

We have found a very similar degeneracy point for the He atom in a field coupling the ground state by two photons with the lowest ${}^1S^e$ resonance.

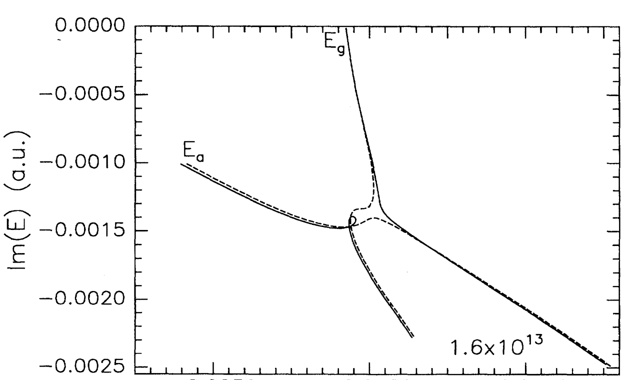

FIG. 3. Trajectories of the complex Floquet quasienergies for the ground state and the $2s^2\,{}^1S^e$ autoionizing state of H^-, connected by a two-photon transition, for an intensity varying from 0 to 1.5×10^{13} W/cm^2. The detunings are indicated (in a.u.).

We have constructed a new two-photon model, which demonstrates the generality of the LIDS, just like in the one-photon case. This model involves the ground state, a first continuum (single photon ionization), and a second continuum, in which the autoionizing state is embedded. Both discrete states are thus coupled by a two-photon transition via the first continuum. The solution of this model is not as simple as for the one-photon case, and the resulting expressions for the quasienergies are too cumbersome to offer much analytical insight. In Fig. 4 we show the resulting trajectories for the quasienergies of the ground state and the autoionizing state for intensities up to 7×10^{14} W/cm^2, for four different detunings, indicated next to the zero-field positions $E_a - 2\omega - i\Gamma_a/2$. The qualitative agreement with the R-matrix Floquet results of Fig. 3 for the curves around the upper left degeneracy point is again very good. One must bear in mind that the model does not include any of the several other continua which are present for the full R-matrix Floquet H^- calculation. The "lower right" degeneracy point is at a much higher intensity and would therefore require a much larger Floquet calculation.

One of the most striking consequences of the one-photon LIDS is that "trapping" of population can occur at some nonzero field intensity [10,14]. From Fig. 1 one can see that for a fixed frequency, which must lie within the two detunings given by Eq. (2), the rate of ionization of the ground state will first increase with intensity and then exhibit a typical "stabilization," namely, a decrease of the

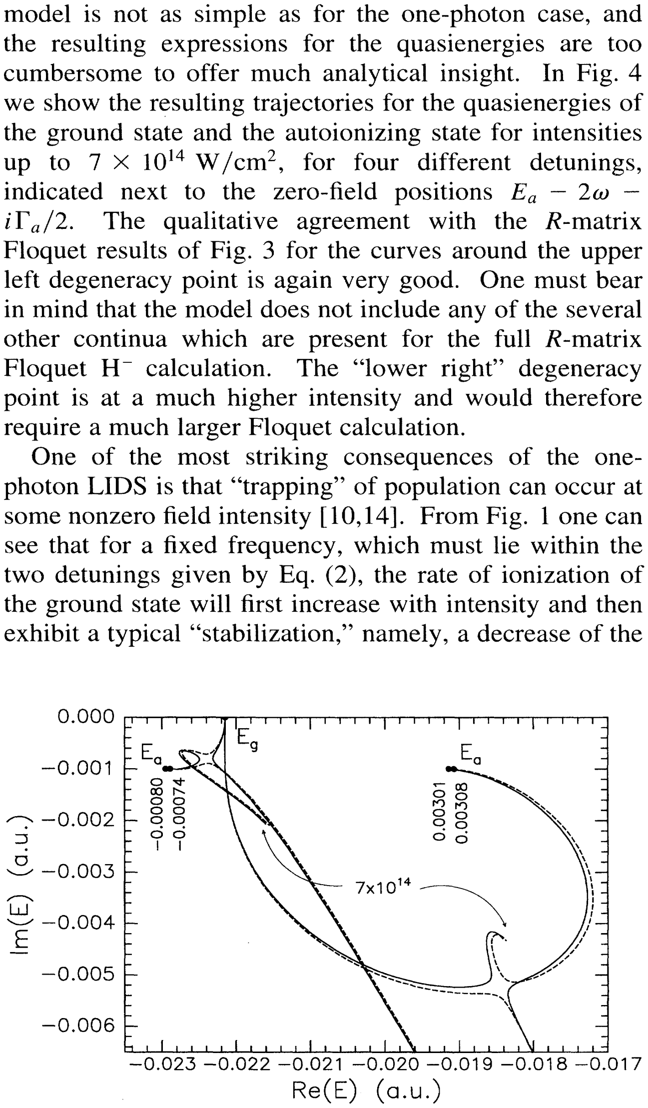

FIG. 4. Trajectories of the complex Floquet quasienergies for the ground state and the autoionizing state of the two-photon model discussed in the text. The parameters used in this two-photon model are the line profile index of the resonance ($q = 6$) in the two-photon channel, the zero-field width ($\Gamma_a = 2 \times 10^{-3}$) of the autoionizing state, the one-photon ionization coupling parameter of the ground state ($\gamma_1 = 3 \times 10^{-16}$), the free-free dipole coupling ($\gamma_2 = 5 \times 10^{-17}$), the field-induced shift of the autoionizing level (2.9×10^{-18}), and the real part of the dipole coupling between the two discrete levels (7.3×10^{-19}). The units of the last four quantities are such that multiplication by the intensity in W/cm^2 gives a value in a.u.

ionization rate with increasing intensity. This behavior should be experimentally verifiable.

A direct experimental probe of LIDS would be to first produce a degeneracy by coupling a bound (possibly excited) state with a strong laser to an autoionizing state. This distorted atom could then be probed by scanning with a weak laser or with synchrotron radiation that connects another bound state resonantly with the position of the LIDS.

A possible application would be population transfer from the ground state to an autoionizing state. In the case of Ar (Fig. 1) this could be achieved by (i) choosing $\omega = 0.9856$ and increasing I from 0 to 2×10^{13} W/cm^2, (ii) changing ω slightly, from 0.9856 to 0.9855, and (iii) decreasing I back to 0. The trajectories must be timed precisely due on one side to the constraint of adiabaticity (the changes in frequency and intensity must not be too fast) and on the other side by the fact that the atomic system is decaying in the field. Instead of step (iii) above, further adjustments in frequency and intensity allow a circuit to be completed around the degeneracy. These trajectories in the parameter space are quite natural in laser physics since intensity variations are the rule for strong (pulsed) lasers, while frequency variations (chirps) are routinely controlled experimentally. Our two-photon example in H^- involves a frequency close to existing lasers, while the example in Ar may be accessible with new high-frequency synchrotron light sources.

In conclusion, we have investigated degeneracies induced by a single laser field in a real atomic system around a resonance between the ground state and an autoionizing state. Their generality is shown by a simple model, while their appearance in real physical systems is shown by full R-matrix Floquet calculations. These degeneracies appear for one-photon and for two-photon resonances. It is probable that such degeneracies are also present for higher multiphoton resonances involving frequencies corresponding to existing strong lasers, and work is under way to explore these, considering atoms with lower lying and narrower autoionizing states. Suggestions for experimental observation are given, which might lead to novel ways of studying autoionizing states.

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