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Multiple occurrence of zero-width resonances in photodissociation: Effect of laser field intensity and frequency

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Critical intensities at which the strong field photodissociation rates of a molecule are vanishing could play an important role in control scenarios to filter a given vibrational state. We have previously shown [O. Atabek, R. Lefebvre, and F. Gadéa, Phys. Rev. A **74**, 063412 (2006)] that there is a multiplicity of such intensities when the diabatic curve crossing point is on the left of the minimum of the attractive potential in a photon dressed picture (c^- case). A semiclassical explanation was given. We complete this study of the multiple occurrence of zero-width resonances (ZWRs) by addressing, both numerically and semiclassically, the more common c^+ case of a crossing point on the right of the potential minimum. Coincidences between modified diabatic and adiabatic levels are the fingerprints of such events. They occur several times because the adiabatic energies are moving upward more rapidly than the diabatic ones. Advantage can be taken of the frequency dependence of the critical intensities to produce the phenomenon even for weak fields, this circumstance leading to more favorable control issues. This is illustrated on the example of H_2^+. The wave functions associated with the ZWRs reflect the fact that the adiabatic picture is the best zeroth order description in the limit of high fields.

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I. INTRODUCTION

The existence of zero-width resonances (ZWRs) has been demonstrated in both predissociation [1,2] and photodissociation [3–5]. The two problems have much in common because in a dressed picture photodissociation leads also to a pattern with a potential supporting bound states crossed by a repulsive potential. The purpose of this paper is to investigate further the properties of the ZWR's in the context of molecular photodissociation in a wide range of laser intensities. The model is one dimensional and involves two Born-Oppenheimer electronic states, one attractive, the other repulsive, an assumption which is commented below. This is a rather general frame, our example being H_2^+ for which there is considerable information for its behavior in an intense laser field [6]. Two different languages are appropriate: In the case of a small interchannel coupling, the states associated with the original potentials make up a good zeroth order basis. This is the diabatic picture. But if the interchannel coupling is very strong, it could be more significant to start the study with the potentials which diagonalize the potential matrix. This is the adiabatic limit. If the wave function is accurately calculated with coupled channel equations, both sets of potentials can be used and should lead to identical results. This was shown explicitly in the case of predissociation by van Dishoek and Dalgarno [7]. In our study, although the wave equation is formulated in the diabatic picture, we are able to easily transform it to get the wave functions which could be obtained in the adiabatic approach. This is very useful for interpretative purposes because in the photodissociation problem we are led to calculate the wave functions over a large range of interchannel couplings.

We have already pointed out [4] that it is possible for more than one intensity to lead to a ZWR for a given initial field-free vibrational state. This was done in the context of a c^- case (curve crossing on the left of the potential minimum). The diabatic crossing point is between the two left turning points of the adiabatic potentials. It is possible to explain the occurrence of ZWR's through an interference mechanism for the vanishing of the outgoing amplitude in the lower adiabatic channel. An integer is involved in this condition so that the intensities at which the photodissociation rate is zero can be labeled with this integer. We prove here that there is also a sequence of critical intensities leading to zero width in the c^+ case, that is to say with a crossing point to the right of the minimum of the attractive potential. For H_2^+ this occurs for a wavelength beyond ~105 nm. This situation is similar to that analyzed in the predissociation problem [1,2]. A different interference process is now at work. In Sec. II we recall the basic formulas for the semiclassical treatment of the ZWR's in the c^+ case. Section III gives the basic formulas for a Floquet approach to the calculation of the photodissociation rate for a molecule exposed to a continuous wave (cw) field. The semiclassical explanation for the multiplicity of critical intensities is given in Sec. IV. The problem is so far treated with a two-channel formalism which assumes that the process is limited to only one net photon absorption. We devote Sec. V to the following question arising from the rather high intensities which are present: Do the ZWR's survive when introducing additional channels amounting to two- and three-photon absorptions? Some words of conclusion on control scenarios involving ZWRs follow in Sec. VI.

II. THE SEMICLASSICAL FORMALISM

Let us now comment the interference mechanism illustrated in Fig. 1 and give its semiclassical formulation. The upper panel shows both the diabatic and the adiabatic poten-

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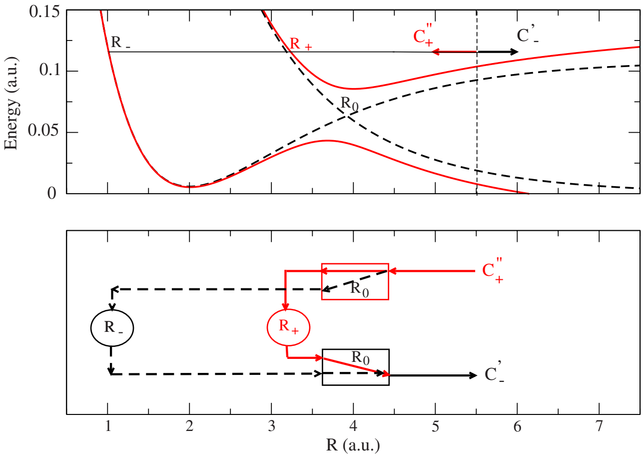

FIG. 1. (Color online) Interfering paths leading to a ZWR in a semiclassical description. The upper panel shows the diabatic (dashed black curves) and adiabatic potentials (solid red curves). A wavelet of amplitude C''_+ travels on the upper adiabatic potential toward R_0, the position of the avoided crossing between adiabatic potentials. The two resultant wavelets are reflected at the positions of the turning points and come back to be splitted again. The outgoing amplitude in the open channel C'_- is made of two interfering contributions symbolized by the dashed black and solid red paths.

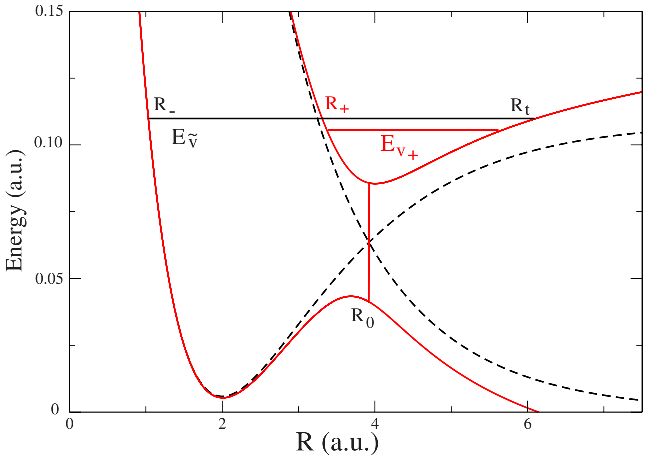

FIG. 2. (Color online) The two energies involved in the coincidence criterion for producing a ZWR. The energy E_{v_+} belongs to the upper adiabatic potential (upper solid red curve), but with a phase modification in the quantization condition [see Eq. (5)]. The energy $E_{\tilde{v}}$ belongs to the modified diabatic potential made of two branches: The lower adiabatic potential (lower solid red curve) for $R < R_0$ and the upper adiabatic potential (upper solid red curve) for $R > R_0$.

tials. The latter come from a diagonalization of the potential matrix. For predissociation the coupling is of internal nature (electronic, spin-orbit,…). For photodissociation the coupling is the matter-field interaction. The diabatic crossing point R_0 is on the right of the minimum of the attractive potential. In a semiclassical approach all relevant quantities such as potentials and wave numbers are to be evaluated in the adiabatic approach [8]. We imagine a wavelet of amplitude C''_+ traveling inward on the upper (closed) potential [9,10]. It meets the upper box symbolizing the region of avoided crossing which acts as a beam splitter. The two waves produced by the box meet the left turning points, as shown in the lower panel of Fig. 1, and come back toward the lower box which again splits each of the two waves. We are interested by the amplitude of the wave following the lower (open) adiabatic channel. This wave is made up from two contributions, one coming from the path in dashed black, the other from the path in solid red. At some characteristic energies which will now be determined, this amplitude can be zero. The system is trapped, and the resonance width is zero.

The outgoing scattering amplitude C'_- in the lower (open) adiabatic channel at position R can be written as [4,11]

$$C'_- = (\mathcal{P}_1 e^{i\Phi_1} + \mathcal{P}_2 e^{i\Phi_2})C''_+ \tag{1}$$

with

$$\Phi_1 = -\int_R^{R_0} dR' k_+(R') - 2\int_{R_0}^{R_-} dR' k_-(R') - \pi/2 - \chi + \int_{R_0}^{R} dR' k_-(R'), \tag{2}$$

$$\Phi_2 = -\int_R^{R_0} dR' k_+(R') - 2\int_{R_0}^{R_+} dR' k_+(R') - \pi/2 + \chi + \int_{R_0}^{R} dR' k_-(R'). \tag{3}$$

Φ_1 and Φ_2 represent the phases accumulated along the two pathways. The relevant turning points R_- and R_+ are given on Fig. 2 as well as R_0, the crossing point of the diabatic curves. The quantities $\mathcal{P}_1$ and $\mathcal{P}_2$ are transition amplitudes for a change of adiabatic channel through a passage by the complex intersection points of the two adiabatic potentials. They are such that $\mathcal{P}_1 = -\mathcal{P}_2$ [8]. χ is an additional phase occurring when a wave passes through R_0 while staying on the same channel [8]. In Φ_1 the first term is the phase accumulated until the wave reaches the avoided crossing. There is then a change from the upper to the lower channel. The excursion to R_- and back is given by the second term, with a phase correction $-\pi/2$ due to the turning point. There is then (third term) progression outward on the lower channel. A different story is told by Φ_2. The progression inward is followed by the excursion to R_+ and back. The channel change occurs when reaching R_0 in this outward motion. This is followed by free motion on the lower channel. After multiplication by $\exp[2i\int_{R_0}^{R_t} dR' k_+(R')]$ a modified outgoing amplitude is obtained, which accounts for the reflections taking place on the right turning point R_t of the upper adiabatic potential (see Fig. 2). The new amplitude is

$$\tilde{C}'_- = C'_- \exp\left[2i\int_{R_0}^{R_t} dR' k_+(R')\right]. \tag{4}$$

The cancellation of $\tilde{C}'_-$ is a sufficient condition for that of C'_-. A way to achieve this is to satisfy simultaneously the two relations [8]

$$\int_{R_+}^{R_0} dR' k_+(R') + \int_{R_0}^{R_t} dR' k_+(R') + \chi = \left(\tilde{v}_+ + \frac{1}{2}\right)\pi \tag{5}$$

and

$$\int_{R_-}^{R_0} dR' k_-(R') + \int_{R_0}^{R_t} dR' k_+(R') = \left(\tilde{v} + \frac{1}{2}\right)\pi. \tag{6}$$

The predissociation is quenched. Since these conditions are nothing but Bohr-Sommerfeld quantization conditions, this means that there is coincidence of two energies, one, say $E_{\tilde{v}^+}$, which is associated with the upper adiabatic potential, but with a phase correction χ, which in weak coupling is $-\pi/4$ [8]. This amounts to a shift which is approximately one quarter of the difference between successive adiabatic levels. The second condition means that the potential is made by joining two branches (see Fig. 2). On the left of R_0, the potential is the lower adiabatic potential, while on the right of this crossing point it is the upper adiabatic potential. For a weak coupling, this is practically the diabatic attractive potential. Let $E_{\tilde{v}}$ be an energy of this potential. The coincidence condition is well displayed by the expression for the resonance width Γ_R which emphasizes the role of the proximity of the two kinds of energies in the form $\Gamma_R \propto (E_{\tilde{v}} - E_{\tilde{v}_+})^2$ [1,2]

In the predissociation problem the coincidence can only be accidental since there is no easy and continuous way to modify the potentials and the coupling. In the case of IBr studied by Child [2], it is by looking at a series of rotational lines that some near degeneracy could be predicted and confirmed by the survival of these lines despite the strong interchannel coupling. For a diatomic molecule submitted to an electromagnetic field the wavelength and the intensity are two parameters which allow such coincidences and therefore produce at will the ZWRs.

III. THE FLOQUET FORMALISM

The rate calculation for a molecule submitted to a cw field is made with the Floquet formalism [6] which gives the solution of wave equations with periodic Hamiltonians. With two electronic states being involved, the wave function is written

$$|\Psi(R,t)\rangle = \chi_g(R,t)|g\rangle + \chi_u(R,t)|u\rangle \tag{7}$$

with $|g\rangle$ and $|u\rangle$ representing the ground and excited electronic states, respectively. The nuclear wave functions are solutions of the time-dependent Schrödinger equation, written in the length gauge as

$$i\hbar\frac{\partial}{\partial t}\begin{bmatrix}\chi_g(R,t)\\ \chi_u(R,t)\end{bmatrix} = \left(T_N + \begin{bmatrix}V_g(R) & 0\\ 0 & V_u(R)\end{bmatrix} - \mu(R)\mathcal{E}(t) \times \begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix}\right)\begin{bmatrix}\chi_g(R,t)\\ \chi_u(R,t)\end{bmatrix}, \tag{8}$$

where T_N is the nuclear kinetic energy operator. $V_g(R)$ and $V_u(R)$ are the Born-Oppenheimer potentials, and $\mu(R)$ is the electronic transition moment between states $|g\rangle$ and $|u\rangle$. $\mathcal{E}(t)$ is the laser electric field amplitude of the form $\mathcal{E}_0 \cos(\omega t)$ with a wavelength $\lambda = 2\pi c/\omega$. The Floquet ansatz consists in writing the wave function as

$$\begin{bmatrix}\chi_g(R,t)\\ \chi_u(R,t)\end{bmatrix} = e^{-iEt/\hbar}\begin{bmatrix}\varphi_g(R,t)\\ \varphi_u(R,t)\end{bmatrix}. \tag{9}$$

E is called the quasienergy. $\varphi_k(R,t), (k=g,u)$, being time periodic, can be Fourier expanded as

$$\varphi_k(R,t) = \sum_{n=-M}^{+M} e^{in\omega t}\Psi_{k,n}(R). \tag{10}$$

M is the maximum number of exchanged photons (either absorbed, for M positive or emitted, for M negative) chosen to lead to converged results. The time-dependent exponentials represent basis functions in an extended space. Identification of the coefficients of these basis functions leads to the coupled equations

$$[T_N + V_g + n\hbar\omega - E]\Psi_{g,n}(R) - 1/2\mathcal{E}_0\mu(R)[\Psi_{u,n-1}(R) + \Psi_{u,n+1}] = 0, \tag{11}$$

$$[T_N + V_u + n\hbar\omega - E]\Psi_{u,n}(R) - 1/2\mathcal{E}_0\mu(R)[\Psi_{g,n-1}(R) + \Psi_{g,n+1}] = 0. \tag{12}$$

The solution with Siegert outgoing wave boundary conditions in the open channels produces a complex quasienergy $E_R - i\Gamma_R/2$. Γ_R is the decay rate called below the Floquet rate. If the intensity of the field is such as to allow only for a one-photon net absorption, the coupled equations reduce to

$$[T_N + V_g(R) - E]\Psi_{g,0}(R) - 1/2\mathcal{E}_0\mu(R)\Psi_{u,-1}(R) = 0, \tag{13}$$

$$[T_N + V_u(R) - \hbar\omega - E]\Psi_{u,-1}(R) - 1/2\mathcal{E}_0\mu(R)\Psi_{g,0}(R) = 0. \tag{14}$$

This situation is illustrated in Fig. 2 which shows the two potentials now identified with $V_g(R)$ and $V_u(R) - \hbar\omega$. Dressing one of the potentials changes a situation with asymptotically degenerate potentials into one with a crossing of the two potential curves. The equations are solved with the Fox-Goodwin propagator, with exterior complex scaling [12,13] and the potentials of H_2^+ given by Bunkin and Tugov [14].

IV. MULTIPLICITY OF CRITICAL INTENSITIES

Figure 3 is showing for two different wavelengths ($\lambda = 400$ and 420 nm) that the Floquet rate calculated as a function of the field intensity passes twice by the value zero. We can make a number of comments. (i) the critical intensities are very dependent on wavelength. We have managed to have with $\lambda = 420$ nm a rather small critical intensity $I_c = 0.13 \times 10^{13}$ W/cm^2. This could be exploited to follow adiabatically a ZWR by modifying progressively the frequency (chirped pulse), as suggested before in Ref. [5]. In this way high intensities could be achieved, with the molecule still in a stable state. (ii) In order to show that beyond the second critical intensity the rate is increasing, we have to amplify it.

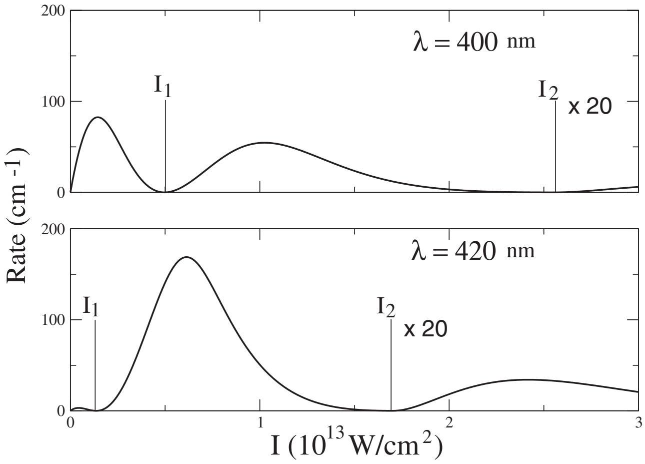

FIG. 3. As the field intensity grows the rate passes twice through zero. Two ZWR's are born from the initial field-free vibrational state $v=8$ of H_2^+. Note that the critical intensities are very different for the two wavelengths. For readability, the rates beyond the second critical intensities are magnified by a factor 20.

This is clearly related to the fact that, at such intensities, the resonance is more and more dominated by the upper adiabatic channel, with attenuation of the interchannel coupling which is of kinetic type. (iii) It is possible to repeat the calculations of the ZWR's by setting θ, the rotation angle used in the exterior complex scaling procedure, equal to zero. This is rather unexpected at such intensities. Figure 4 illustrates this observation. It represents in polar coordinates the wave functions obtained for two intensities in the case $\lambda=400$ nm. The first intensity ($I=0.140\times10^{13}$ W/cm^2) corresponds to the maximum rate reached before the first critical intensity ($I_c=0.500\times10^{13}$ W/cm^2). We recognize that in the lower channel there is first a number of oscillations followed by a behavior typical of a Siegert wave. The wave functions at the critical intensity in both channels are aligned with the real axis because they are those of a bound state. Table I is summarizing our findings concerning the first two critical intensities at the two wavelengths $\lambda=400$ and 420 nm.

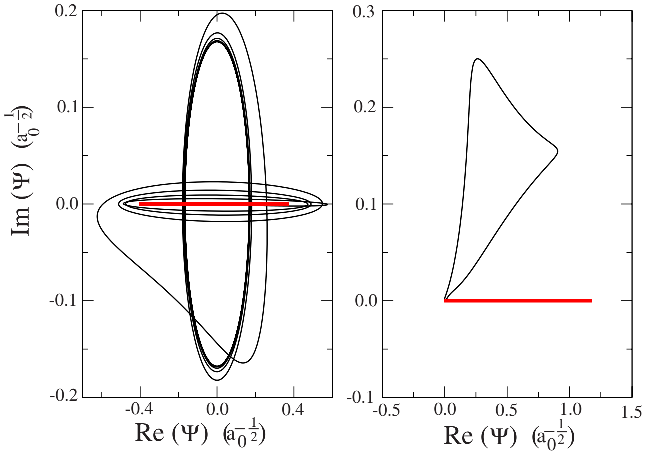

FIG. 4. (Color online) The polar representation of the two adiabatic channel functions for two intensities, with the left panel for the lower channel and the right panel for the higher one. In solid thin black with $I=0.140\times10^{13}$ W/cm^2, giving the maximum rate before the first critical intensity. In solid thick red, at the first critical intensity $I_c=0.500\times10^{13}$ W/cm^2. At such an intensity the two component functions are real and therefore aligned with the real axis. a_0 is the Bohr radius.

TABLE I. Critical intensities I_c and corresponding energies E_{ZWR} for the two wavelengths $\lambda=400$ and 420 nm. The first two lines correspond to the first critical intensity calculated with a number of channels N ($N=2$ or 8). The last two lines give the same information for the second critical intensity.

	$\lambda=400$ nm		$\lambda=420$ nm	
N	I_c (10^{13} W/cm^2)	E_{ZWR} (cm^{-1})	I_c (10^{13} W/cm^2)	E_{ZWR} (cm^{-1})
2	0.500	−6534.71	0.136	−6815.79
8	0.480	−6629.63	0.135	−6840.93
2	2.556	−3971.28	1.703	−4454.29
8	2.286	−4507.43	1.545	−4841.70

We would like to draw attention to the fact that in a two-channel Floquet calculation the critical intensity can be tuned to obtain a rate which is zero within the numerical accuracy of the algorithm. This means a rate Γ of the order of 10^{-10} cm^{-1}, that is, 10 orders of magnitude less than a typical Γ. This observation goes beyond the semiclassical explanation and leads us to think that the ZWR is a strict property of the wave equation.

We test now the adequacy of the semiclassical approach to explain the multiplicity of critical intensities. Figures 5 and 6 show in the lower panel how the modified diabatic energies $E_{\tilde{v}}$ for $\tilde{v}=8$ and 9 are affected by a change of the intensity. Also shown is the trajectory of the modified adiabatic energy $E_{\tilde{0}_+}$. Because the modified diabatic energies are moving upward slower than the modified adiabatic energy two crossings are occurring in Fig. 5 which determine the critical intensities I^{sc}. They correlate rather well with the critical intensities calculated from coupled equations for $\lambda=400$ nm. The semiclassical interpretation is less accurate for $\lambda=420$ nm as illustrated in Fig. 6. The expected first intersection to explain the lowest (and weak) critical intensity I_1^{sc} is missing, although the two curves are close to each other at zero intensity. Also displayed in the upper panels of Figs. 5 and 6 are the wave functions at the first and second critical intensities. It is interesting to note that the amplitude in the upper adiabatic channel is enhanced relative to that of the lower channel when going from the first to the second critical intensity. This reflects again the increasing validity of the states of the upper potential as a zeroth order approximation. The kinetic coupling to the lower channel is decreasing. The number of nodes is precisely that of the states $v_+=0$ in the upper channel and $v=8$ in the lower one. We can note that in the predissociation problem [15] it is also possible to get several crossings between modified diabatic and adiabatic energies, the parameter being $J(J+1)$. The centrifugal correction to both potentials also produces an upward motion of the modified adiabatic energies which is faster than that of the modified diabatic energies.

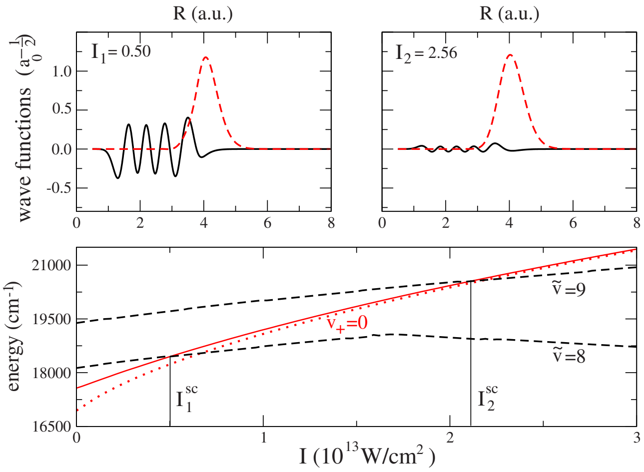

FIG. 5. (Color online) Lower panel: An explanation for the successive occurrence of two critical intensities for the Floquet state derived from the field-free state $v=8$ at the wavelength $\lambda=400$ nm. The modified adiabatic energy for $\tilde{v}_+=0$ [solid red curve of the lower panel, the dotted red curve being for the adiabatic energy, without the phase correction χ in Eq. (5)] is moving upward faster than the modified diabatic energies for $\tilde{v}=8$ and 9 (dashed black curves of the lower panel). Two coincidences occur, which can be related to the critical intensities of Fig. 3. The upper panels show the component wave functions for the two critical intensities. For the largest critical intensity, the lower channel wave function in solid black is considerably reduced with respect to the higher channel wave function in dashed red. This reflects the fact that the adiabatic upper channel becomes a better zeroth order approximation.

V. THE MULTICHANNEL CASE

It is useful to check whether at the second critical intensity we are still allowed to maintain the conclusions of the study limited to two channels (with its semiclassical explanation for the occurrence of critical intensities). Two points need to be investigated. (i) Is the rate still going to zero at some intensity and how close is this intensity to that of the two-channel picture? The converged calculations are made with eight channels: two Floquet blocks above the reference one (that is that of the two-channel study) and one Floquet block below. There are three open channels, corresponding to, respectively, one, two, and three photon absorptions. Table I is giving a comparison for the two wavelengths of the results for either two or eight channels for the first two critical intensities. It was found that indeed the rates are still passing through zero even with eight channels. (ii) Since now, with the advent of additional open channels, there are partial rates, what happens to these rates? A vanishing of the total rate implies the simultaneous vanishing of all partial rates. This would be very surprising. Figure 7 gives the rates on a logarithmic scale and provides the answer. On the left panel we see that the one-photon absorption process is still the dominant one in the case treated here. It is again possible to refer to a critical intensity since at $I_c=2.286\times10^{13}$ W/cm^2 (instead of $I_c=2.556\times10^{13}$ W/cm^2 in the two-channel calculation) there is a collapse by five orders of magnitude of the one-photon rate, while the two other rates are down by one order of magnitude at a slightly different intensity. We conclude that the interference phenomenon is still at work for the one-photon process, while probably another interference process is occurring for the two other rates, but starting from values considerably smaller in this intensity range.

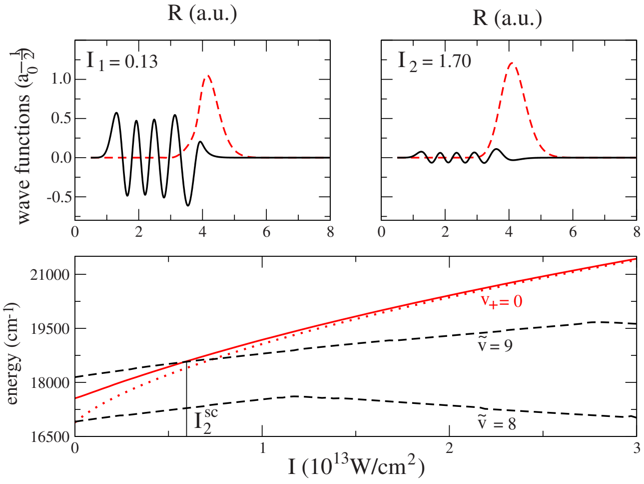

FIG. 6. (Color online) The same situation as in Fig. 3, now for $\lambda=420$ nm. The semiclassical approach is not as successful as in the case $\lambda=400$ nm to predict the critical intensities.

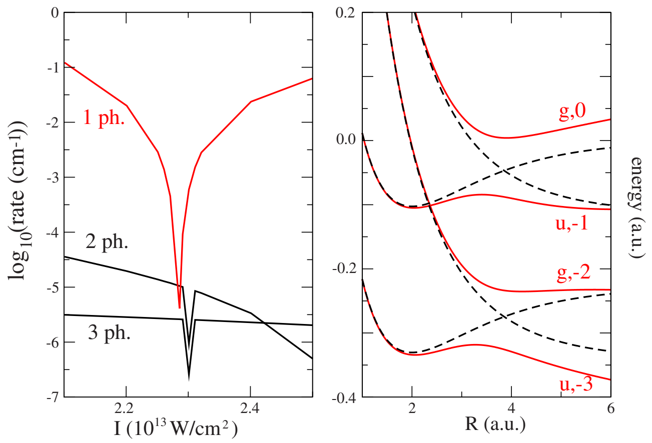

FIG. 7. (Color online) Left panel: The partial rates on a logarithmic scale close to the second critical intensity for $\lambda=400$ nm in a eight-channel calculation. The one-photon dominant partial rate collapses by five orders of magnitudes in the vicinity of the critical intensity. Right panel: The diabatic (dashed black) and adiabatic (solid red) potentials of the two lowest Floquet blocks to show the channels involved in the one-, two- and three-photon absorption processes.

An additional concern is the transposability to molecular systems involving more dimensionality (rotational or electronic). We have shown before [5] that the introduction of the rotational degree of freedom is still leading to critical intensities, with a scaling effect due to the modification of the interchannel couplings. Because the curve crossing is monitored by the laser frequency one could expect favorable situations for the advent of ZWRs even in systems involving several excited electronic states.

VI. CONCLUSIONS

An extension of the study made before [3–5] of the phenomenon of zero-width resonances in the context of a molecule interacting with a laser field has revealed several interesting aspects. There exists a sequence of intensities at which the rate calculated with the Floquet formalism passes through zero. Although a semiclassical treatment gives an explanation for the sequence, the existence of ZWR's appears to be a fundamental property of the two-channel wave equation. These critical intensities are very sensitive to the choice of laser frequency. It is possible to obtain, with an appropriate choice of the frequency, a ZWR even at low intensity. This could be the basis for a scenario where, with a conveniently chirped pulse, the molecule initially in a given field-free state, would be maintained adiabatically, as time is evolving, in a ZWR which has all the properties of a bound state. The dissociation would be completely quenched, while a laser field is present. This could be done in conditions which validate fully the two-channel approximation where arbitrarily small values of the rate can be attained. From a more technical point of view, this means that the molecular wave function strictly obeys bound state boundary conditions, without having to resort to complex scaling of the dissociative coordinate, although the energy is well above the dissociation limit. Work is in progress on the adiabatic driving of a ZWR.

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