

Zero-width resonances in intense-field molecular photodissociation

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We provide additional evidence for the existence of zero-width resonances in the intense-field photodissociation of H_2^+ . In a previous investigation [Atabek, Chrysos, and Lefebvre, Phys. Rev. A **49**, R8 (1994)] the situation, in a two-channel dressed picture, corresponded to a diabatic crossing point classically reachable in both adiabatic potentials. A semiclassical explanation could be modeled after that developed for predissociation in the intermediate-coupling regime. At higher frequencies the crossing point lies between the two turning points. A numerical study shows that zero-width resonances exist also in such a case. An extension of the semiclassical approach provides an explanation for the occurrence of these resonances. It is shown that they survive even when going to a multichannel description. The associated wave functions and probability densities are studied: they are very similar to those of the upper adiabatic potential, with a minor component in the lower adiabatic channel. Some conditions for the production of such long-lived dressed molecular species are stated.

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I. INTRODUCTION

When a molecular species is subjected to a high-intensity laser field, some spectacular effects can arise: they are called bond softening [1,2], vibrational trapping [3,4] or dynamical dissociation quenching [5]. Most of these studies, both theoretical and experimental, have been performed on the H_2^+ species and the theory was initiated by one-dimensional models. It has also been shown [6] that it is possible to choose the laser intensity and frequency in order to reduce the photodissociation resonance widths, of this ion or of its isotopic parent D_2^+ , to zero. This is similar to an effect observed in the predissociation of the molecule IBr and given a semiclassical explanation by Bandrauk and Child [7,8]. In the latter case one may say, as a first rough statement, that this arises when there is a near coincidence of a diabatic level with an adiabatic level. One may follow the change in these energies as one varies the rotational quantum number in order to provoke such a near coincidence [8,9]. Resonances of zero width (sometimes called bound states in the continuum) have also been discussed when there is an interference between two resonances belonging to two different channels [10]. In this work only one resonance is present in the energy range under study. The description of strong-field photodissociation is based on dressed potential energy curves with positions (and therefore crossing points) depending on the frequency, while the coupling depends on the intensity. It is therefore very easy to reach the conditions for a vanishing

width. In the previous study [6] the semiclassical approach could be immediately adapted to this situation because in all cases examined in [6] the crossing points, as in the formalism developed by Child [8], could be reached classically in the different channels. Another way to state this condition is that, with the usual conventions to depict the potential energy curves of a dressed diatomic species, the turning points are on the left of the crossing point. The study concluded by pointing out that, for a given frequency, the conditions for zero width for D_2^+ and H_2^+ correspond to significantly different intensities so that one species would dissociate while the other could survive. We have here in view the case of higher laser frequencies. As shown below, the crossing point of the diabatic potential curves, in a two-channel description, is now *between* the adiabatic turning points. We first produce convincing evidence that, for a given frequency and in a two-channel description, it is still possible to produce resonances of practically zero widths by properly choosing the intensity (Sec. II). In Sec. III we first give a simplified semiclassical explanation of the zero width for the case where the crossing point can be reached classically in both channels. This short-range picture is then applied to the present case. An extension of the semiclassical formalism gives an explanation based on a destructive interference between two paths. Section IV shows that these resonances can be obtained even when going to a multichannel Floquet picture. The semiclassical argument provides an explanation. In Sec. V we examine a few wave functions and probability densities of these particular states. The conditions for production and detection of such dressed species are discussed in Sec. VI.

II. NUMERICAL RESULTS

As in the previous study [6] the potential energy curves of the electronic ground state $^1\Sigma_g^+$ and of the first electronic

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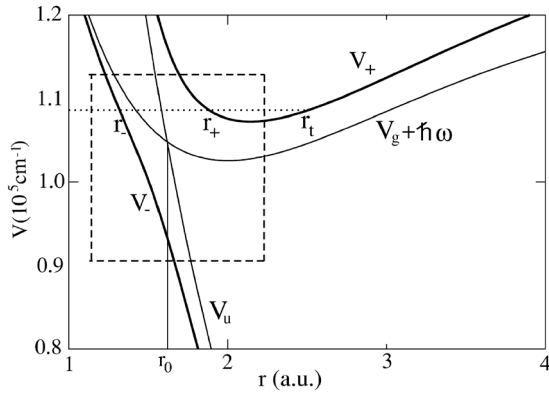


FIG. 1. The two adiabatic potentials V_+ and V_- (in units of 10^5 cm^{-1}) resulting from the interaction of the dressed ground-state potential $V_g + \hbar\omega$ with the excited state potential V_u of H_2^+ . The wavelength is $\lambda=80 \text{ nm}$ and the intensity $I=11.19 \times 10^{13} \text{ W/cm}^2$. The dotted horizontal line shows at which energy there is occurrence of a zero-width resonance. The thin vertical line shows the position of the diabatic crossing point r_0 . This point is between the left turning points of both adiabatic potentials r_- and r_+ . Short-range scattering theory examines the events occurring in the dashed rectangle. r_t is the right turning point in the upper adiabatic potential. The origin of the energies is at the asymptotic value of the repulsive potential V_u .

excited state $^1\Sigma_u^+$ of H_2^+ , as well as the transition moment between the two states, are taken from the work of Bunkin and Tugov [11]. The wavelength of the laser is taken equal to 80 nm , giving $125\,000 \text{ cm}^{-1}$ for the energy of a light quantum. A preliminary study is based on a two-channel representation of the Floquet problem [12,13], with the ground-state potential dressed by the photon energy. The methodology to get resonance energies includes exterior complex scaling [14] of the nuclear coordinate and asymptotic analysis of the open-channel amplitudes, after a diabatic-to-adiabatic transformation to eliminate the persistent interchannel coupling due to the form chosen for the transition moment [15].

Figure 1 gives the two dressed diabatic potentials and the two adiabatic potentials obtained at the intensity producing a zero-width resonance originating from the vibrational ground state of the free molecule. This shows clearly that the crossing point of the diabatic potentials is between the two classical turning points in the two adiabatic channels. The calculations are performed by progressively increasing the laser intensity with, at low intensity, a trial energy which is a field-free energy with vibrational quantum number ν_{diab} . We have followed in this way the five lowest diabatic energies. For $\nu_{diab}=0, 1, 3$, and 4 the width starts, as expected, as a linear function of the intensity, then passes through a maximum. This is followed by a decrease to zero for an intensity which will be hereafter called a critical intensity. There are then oscillations leading to further critical intensities. This is illustrated in Fig. 2 which gives the variation of the quantity $-2 \text{Im}(E_R)$ (the rate in cm^{-1}) as a function of the intensity for $\nu_{diab}=0$. E_R is the complex resonance energy; “Im” stands for the imaginary part. It is useful to check how sensitive is the rate in the vicinity of a critical intensity. From the data

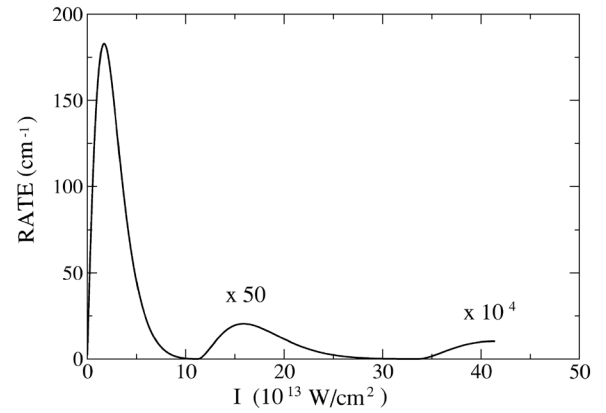


FIG. 2. The variation with intensity of the rate (in cm^{-1}) for the resonance originating from the ground vibrational state of the free molecule. This is a two-channel calculation. Near intensities $I \sim 11.19 \times 10^{13} \text{ W/cm}^2$ and $I \sim 33.65 \times 10^{13} \text{ W/cm}^2$ the width can be made arbitrarily small. Some amplification factors (50 times or 10^4 times) are needed to see the oscillations of the rate.

represented in Fig. 2 the rate remains less than 0.2 cm^{-1} for a range of the field amplitude extending from 0.0539 to 0.0609 a.u. , that is, a 13% change. This shows that there is robustness around the critical intensity. For $\nu_{diab}=2$ we were unable to get plausible results: the calculation soon went to unphysical numbers with extremely high width. In a parallel study of D_2^+ we observed exactly the same circumstance. We will come back to this point when discussing the probability densities of the zero-width resonances (Sec. V).

Table I gives the initial dressed field-free energies, the energies of the first zero-width resonance, the corresponding intensities, and also, for future discussion, the nearest energy belonging to the upper adiabatic potential calculated for these intensities. The table gives also a quantity to be derived

TABLE I. Numerical study of zero-width resonances issuing from the dressed zero-field ground vibrational states of H_2^+ , with vibrational quantum numbers and energies given in columns 1 and 2. The resonance energies and the corresponding intensities are in columns 3 and 4. Columns 5 and 6 give the vibrational quantum number and the energy closest to the resonance energy of the upper adiabatic potential for these intensities. No width is given for the resonance energies because they can be made arbitrarily small. For the intensities given in the table they are generally of the order of 10^{-6} cm^{-1} . No physically acceptable result could be obtained when starting from the field-free state with $\nu_{diab}=2$. Column 7 gives a quantity occurring in the semiclassical treatment of Sec. III. According to this analysis Q should be close to $3\pi/4 \sim 2.356$. All energies are in cm^{-1} with the origin at the asymptotic value of the repulsive potential. Intensities I are in units of 10^{13} W/cm^2 .

| ν_{diab} | E_{diab} | E_R | I | ν_{adiab} | E_{adiab} | Q |
|--------------|------------|-----------|-------|---------------|-------------|-------|
| 0 | 103701.26 | 108561.48 | 11.19 | 0 | 108537.80 | 2.220 |
| 1 | 105937.19 | 110296.17 | 9.05 | 1 | 110242.23 | 2.341 |
| 2 | 108049.17 | | | | | |
| 3 | 110037.21 | 111878.17 | 6.97 | 2 | 111779.73 | 2.621 |
| 4 | 111901.29 | 113304.35 | 4.97 | 3 | 113148.57 | 2.803 |

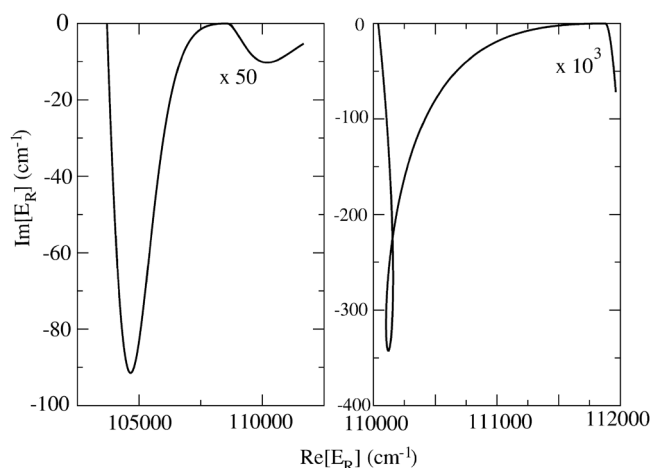


FIG. 3. The trajectories in the complex plane of the resonance energies issued from the field-free vibrational states $v_{diab}=0$ (left panel) and 3 (right panel). The starting point of each curve corresponds to a diabatic energy with no width. After reaching the critical intensities an amplification factor (50 times or 10^3 times) helps in revealing that this is followed by a range of rather small widths. For $v_{diab}=3$ the crossing corresponds to two resonance energies equal to $E_R=110\,154.20-i225.03\text{ cm}^{-1}$, but for two different intensities (0.738×10^{13} and $2.153 \times 10^{13}\text{ W/cm}^2$) so that no phenomenon can be associated with this “degeneracy.”

in the semiclassical treatment of Sec. III. The change undergone by the resonance energies as the field is switched on may also be followed by looking at the trajectories of the energy in the complex plane. This is shown in Fig. 3 for two initial states with $v_{diab}=0$ and 3.

III. SEMICLASSICAL APPROACH

We will first of all give a simplified version of the semiclassical method when applied to the previously treated case of two turning points on the left of the diabatic crossing point [8]. The situation is depicted in Fig. 4. We consider the two adiabatic potentials in the neighborhood of the diabatic

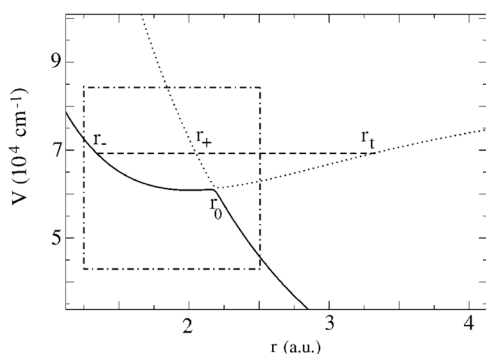


FIG. 4. Two adiabatic potentials with the left turning points both on the left of the diabatic crossing point. Short-range scattering theory examines the events occurring in the dash-dotted rectangle, ignoring that at the energy symbolized by a horizontal line there is also a right turning point for a state supported by the upper adiabatic potential.

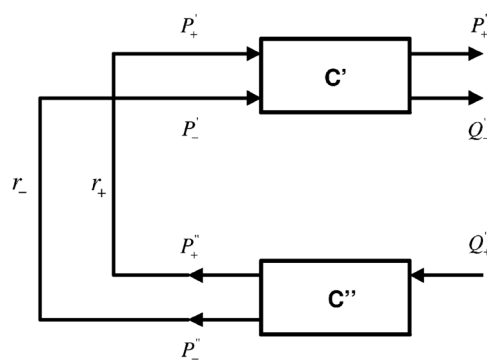


FIG. 5. Diagrammatic representation of the short-range inelastic transition from the upper to the lower adiabatic potential. An incoming wave of amplitude Q'_+ in the upper adiabatic potential reaches the crossing region symbolized by the box C'' , which produces two wavelets reflected by the turning points r_- and r_+ . On the way back each wavelet is again split into two by the box C' . The final amplitude of interest is Q'_- .

crossing point (represented by the dash-dotted rectangle), without paying attention, for the moment, to the fact that the upper adiabatic potential can accommodate bound states. Imagine the system to be originally in the upper adiabatic potential and described by a wave traveling toward the critical region where a transition to the lower adiabatic potential can take place. We make use of the diagrammatic representation developed by Child (see, for example, [16]). Figure 5 shows how the crossing region symbolized by the box C'' (in fact as a result of complex crossing points in the adiabatic picture) splits the original wave. After reflection on turning points r_- and r_+ , there is further splitting by the box C' so that the amplitude in the lower adiabatic channel has two contributions: it is possible to either stay in the upper potential at the first crossing encounter and then go to the lower potential at the back encounter, or vice versa. The consequence is that there is an interference between the two routes. Following the expressions given by Child [16], we find for the inelastic scattering amplitude Q'_- :

$$Q'_- = -i\lambda\sqrt{(1-\lambda^2)} \left[\exp\left(-2i \int_{r_0}^{r_+} dr k_+(r)\right) e^{i\chi} - \exp\left(-2i \int_{r_0}^{r_-} dr k_-(r)\right) e^{-i\chi} \right]. \quad (1)$$

λ is $\exp(-\pi\nu)$ where, for linear potentials, ν is the familiar Landau-Zener parameter $V_{12}^2/v|\Delta F|$, with V_{12} for the constant diabatic coupling, v for the classical velocity at the crossing point r_0 , and ΔF for the difference in slopes at the crossing point. χ is a phase depending on the strength of the coupling. The adiabatic wave numbers $k_{\pm}(r)$ are $\{2m[E - V_{\pm}(r)]\}^{1/2}$. In fact the upper adiabatic potential supports bound states which are, in a semiclassical picture, trapped between left and right turning points. Let us call such a turning point r_r . We multiply the amplitude Q'_- by $\exp[2i \int_{r_0}^{r_r} dr k_+(r)]$ and obtain a modified amplitude \tilde{Q}'_- which can be written

$$\begin{aligned} \tilde{Q}'_- = & -i\lambda\sqrt{(1-\lambda^2)}e^{-i\chi} \left\{ \exp \left[2i \left(\int_{r_+}^{r_0} dr k_+(r) \right. \right. \right. \\ & \left. \left. \left. + \int_{r_0}^{r_t} dr k_+(r) + \chi \right) \right] - \exp \left[2i \left(\int_{r_-}^{r_0} dr k_-(r) \right. \right. \right. \\ & \left. \left. \left. + \int_{r_0}^{r_t} dr k_+(r) \right) \right] \right\}. \end{aligned} \quad (2)$$

Let us assume now that the energy E is such that the two following conditions can be simultaneously satisfied:

$$\int_{r_+}^{r_0} dr k_+(r) + \int_{r_0}^{r_t} dr k_+(r) + \chi = \left(v_+ + \frac{1}{2} \right) \pi \quad (3)$$

and

$$\int_{r_-}^{r_0} dr k_-(r) + \int_{r_0}^{r_t} dr k_+(r) = \left(v_d + \frac{1}{2} \right) \pi, \quad (4)$$

where v_+ and v_d are two integers. v_d is distinct from v_{diab} since the levels defined by Eq. (4) are supported by a piecewise adiabatic potential with a discontinuity at r_0 . In the limit of a very weak coupling v_d becomes v_{diab} . v_+ is different from the vibrational quantum v_{adiab} of the upper adiabatic potential because of the phase factor χ . In the limit of a strong coupling v_+ becomes v_{adiab} . We get now for the scattering amplitude

$$\begin{aligned} \tilde{Q}'_- = & -i\lambda\sqrt{(1-\lambda^2)}e^{-i\chi} \left\{ \exp \left[2i \left(v_+ + \frac{1}{2} \right) \pi \right] \right. \\ & \left. - \exp \left[2i \left(v_d + \frac{1}{2} \right) \pi \right] \right\} \\ = & -i\lambda\sqrt{(1-\lambda^2)}e^{-i\chi}(e^{i\pi} - e^{i\pi}) = 0. \end{aligned} \quad (5)$$

The particle is then trapped on the upper adiabatic potential. Dissociation is completely quenched. This quenching is due to a destructive interference which in a scattering situation gives rise to the so-called Stückelberg oscillations [17]. A coupled-channel calculation has confirmed this semiclassical analysis [9].

It is interesting to give another view of this phenomenon based on the identification of the semiclassical paths that allow the transition from one potential to the other to take place. This somewhat alternative treatment will be useful to treat the situation where the crossing point is not classically reachable in both channels. The formalism has been developed to describe low-energy atomic and molecular collisions [18–20]. We will refer more specifically to the work of Laing *et al.* [20] since they give simple descriptions for the three possible configurations of the turning points: (a) both left-turning points to the left of the diabatic crossing point; (b) one to the left, one to the right; (c) both turning points to the right. We recall that according to Fig. 1 we are dealing with case (b). Let us recall first of all how this formalism works for case (a). This is again, in the present context, a short-range scattering view of the process which has to be later completed by a boundary condition about the upper adiabatic potential. It is important to stress (cf. [18]) that the semiclass-

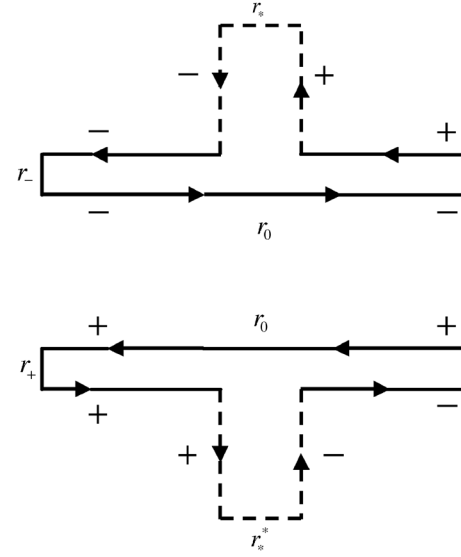


FIG. 6. The two semiclassical paths contributing to the short-range scattering amplitude for a transition from the upper to the lower adiabatic potential. r_0 is the real part of the complex crossing point r_* and its complex conjugate r_*^* . Damping takes place along the dashed paths toward and away from the complex crossing points. Along the real parts of the path different phases are accumulated, which result in an interference that may produce a vanishing of the amplitude at some particular energies. The upper path can be translated as \mathcal{S}_1 of Eq. (7), while the lower path produces \mathcal{S}_2 of Eq. (8).

sical theory is entirely formulated with the adiabatic potentials and their continuations in the complex plane, taking no account of the residual nonadiabatic couplings. We look for situations with a strong radiative interaction, meaning a strong diabatic coupling and therefore a small nonadiabatic transition probability. This implies that the prefactor $[p(1-p)]^{1/2}$, present in the transition amplitude [20], where p is the transition probability in the crossing region, can be approximated as $p^{1/2}$. The transition amplitude takes the simple form

$$S_{-,+} = \mathcal{S}_1 e^{-i\Delta} + \mathcal{S}_2 e^{i\Delta}. \quad (6)$$

The phase Δ is to be determined later. \mathcal{S}_1 and \mathcal{S}_2 correspond to the two paths of Fig. 6 and are given by

$$\begin{aligned} \mathcal{S}_1 = & \exp \left(-i \int_r^{r_0} dr' k_+(r') \right) \mathcal{P}_1 \exp \left[-2i \int_{r_0}^{r_-} dr k_-(r) \right] \\ & \times e^{-i\pi/2} \exp \left(i \int_{r_0}^r dr' k_-(r') \right), \end{aligned} \quad (7)$$

$$\begin{aligned} \mathcal{S}_2 = & \exp \left(-i \int_r^{r_0} dr' k_+(r') \right) \exp \left(-2i \int_{r_0}^{r_+} dr k_+(r) \right) \\ & \times e^{-i\pi/2} \mathcal{P}_2 \exp \left(i \int_{r_0}^r dr' k_-(r') \right). \end{aligned} \quad (8)$$

\mathcal{P}_1 and \mathcal{P}_2 represent the contributions along complex segments shown in Fig. 6, with the crossing point chosen in

order to produce a damping of the waves [20]. They are given by

$$\mathcal{P}_1 = \exp\left(-i \int_{r_0}^{r_*} dr [k_+(r) - k_-(r)]\right) \quad (9)$$

and

$$\mathcal{P}_2 = \exp\left(i \int_{r_0}^{r_*} dr [k_+(r) - k_-(r)]\right). \quad (10)$$

We have introduced in each amplitude a factor $e^{-i\pi/2}$ not present in the expression given in [20] to account for the reflection on each turning point [16]. Transition occurs either at r_* for \mathcal{S}_1 or at r_*^* for \mathcal{S}_2 . It can be proven [16,19] that, if a linear model is adopted for the region close to the crossing,

$$\mathcal{P}_1 = \mathcal{P}_2 = \mathcal{P}. \quad (11)$$

There is therefore a common factor in the two terms of the transition amplitude which expresses the fact that the damping of the wave is the same along the two complex paths. Ignoring common factors of modulus unity, the effective transition amplitude is

$$S_{-,+} \sim -i\mathcal{P} \left[\exp\left(-2i \int_{r_0}^{r_-} dr k_-(r)\right) e^{-i\Delta} + \exp\left(-2i \int_{r_0}^{r_+} dr k_+(r)\right) e^{i\Delta} \right]. \quad (12)$$

We multiply this amplitude by $\exp[2i \int_{r_0}^{r_+} dr k_+(r)]$ and obtain

$$\begin{aligned} \tilde{S}_{-,+} = & -i\mathcal{P} \left\{ \exp\left(2i \int_{r_-}^{r_0} dr k_-(r)\right) \exp\left(2i \int_{r_0}^{r_+} dr k_+(r)\right) e^{-i\Delta} \right. \\ & \left. + \exp\left(2i \int_{r_+}^{r_0} dr k_+(r)\right) \exp\left(2i \int_{r_0}^{r_+} dr k_+(r)\right) e^{i\Delta} \right\}. \end{aligned} \quad (13)$$

Comparing $\tilde{S}_{-,+}$ of Eq. (13) with \tilde{Q}'_- of Eq. (2) when $\lambda \ll 1$, we see that they are identical (except for a trivial factor) if we make the choice $\Delta = \chi + \pi/2$ [which leads to a difference of two terms rather than a sum as in Eq. (13)] and if \mathcal{P} is identified with λ . According to Child χ is $-\pi/4$ in the weak-diabatic-coupling limit and goes to zero as the coupling is increased. The discussion about the occurrence of zero-width resonances follows that given previously.

We now turn to the case (b), with a diabatic crossing point between the two left-turning points. The short-range disposition of the two adiabatic potentials and all the relevant points along the real axis are shown on Fig. 1. We indicate on Fig. 7 the semiclassical paths to be followed in order to describe a transition from the upper to the lower adiabatic potential. The dashed parts of the paths indicate where damping takes place, instead of just phase accumulation. The damping takes place now not only along the excursions toward or away from the complex crossing point, but also along a real segment (from r_+ to r_0), but with an imaginary wave number. There are again two contributions

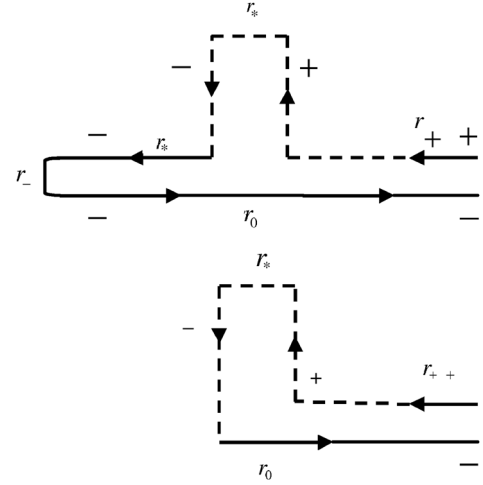


FIG. 7. The two interfering paths of the semiclassical treatment to be followed when the diabatic crossing point is between the two left turning points of the adiabatic potentials. The upper path corresponds to \mathcal{S}_1 of Eq. (14), while the lower path produces \mathcal{S}_2 of Eq. (15).

$$\begin{aligned} S_1 = & \exp\left(-i \int_r^{r_+} dr' k_+(r')\right) \exp\left(-i \int_{r_+}^{r_0} dr k_+(r)\right) \mathcal{P}_1 \\ & \times \exp\left(-i \int_{r_0}^{r_-} dr k_-(r)\right) \times e^{-i\pi/2} \exp\left(i \int_{r_-}^{r_0} dr k_-(r)\right) \\ & \times \exp\left(i \int_{r_0}^r dr' k_-(r')\right) e^{-i\Delta} \end{aligned} \quad (14)$$

and

$$\begin{aligned} S_2 = & \exp\left(-i \int_r^{r_+} dr' k_+(r')\right) \exp\left(-i \int_{r_+}^{r_0} dr k_+(r)\right) \mathcal{P}_1 \\ & \times \exp\left(i \int_{r_0}^r dr' k_-(r')\right) e^{i\Delta}. \end{aligned} \quad (15)$$

\mathcal{P}_1 is given by Eq. (9). We note that the same excursion toward a complex intersection point is implied in the two paths. The damping along the two paths is the same and we can write for the effective transition amplitude, after ignoring identical contributions of modulus unity,

$$\begin{aligned} \tilde{S}_{-,+} = & \exp\left(-i \int_{r_+}^{r_0} dr k_+(r)\right) \mathcal{P}_1 \\ & \times \left[e^{-i\pi/2} \exp\left(2i \int_{r_-}^{r_0} dr k_-(r)\right) e^{-i\Delta} + e^{i\Delta} \right]. \end{aligned} \quad (16)$$

The condition for a vanishing of this amplitude can be written as

$$\left[1 + \exp\left(2i \int_{r_-}^{r_0} dr k_-(r)\right) e^{-2i\Delta} e^{-i\pi/2} \right] = 0 \quad (17)$$

or

$$2 \int_{r_-}^{r_0} dr k_-(r) - 2\Delta - \frac{\pi}{2} = (2n+1)\pi. \quad (18)$$

For $n=0$ this is

$$\int_{r_-}^{r_0} dr k_-(r) = \Delta + \frac{3\pi}{4}. \quad (19)$$

In Table I we give for each zero-width resonance the quantity denoted Q , which is

$$Q = \int_{r_-}^{r_0} dr k_-(r). \quad (20)$$

All values of Q are close to $3\pi/4=2.356$. This can be obtained if one takes $\Delta \sim 0$. A necessary condition for a zero width is therefore, again, that a destructive interference is at work to cancel the transition amplitude. However, trapping can occur only if at this energy there is effectively a bound state in the upper adiabatic potential. Table I shows that each time we observe a zero-width resonance the energy of that resonance is indeed close to that of a bound state of the upper adiabatic potential. This is confirmed in Sec. V by a comparison between probability densities of the Floquet approach and those supported by the upper adiabatic potential. The semiclassical treatment also provides an explanation for the additional values for the critical intensities, as displayed in Fig. 2. For $n=1$, we should have, to produce a destructive interference between the two paths,

$$Q = \int_{r_-}^{r_0} dr k_-(r) = \Delta + \frac{7\pi}{4}. \quad (21)$$

Accepting again $\Delta \sim 0$, this gives Q close to 5.50. Numerical integration provides $Q=5.23$.

Finally, it is useful to stress that, if a situation with the two turning points to the right of the crossing point could be produced, no interference could operate since in that case there is only one path to go from the upper to the lower adiabatic potential [19,20].

IV. THE MULTICHANNEL CASE

It is well known that in the presence of a high-intensity laser it is often necessary to go beyond a two-channel approach in the Floquet picture. The two-channel formulation describes the absorption of a single photon. Multiphoton processes may also occur, which require additional channels to be included [21], according to the following scheme:

$$|g, n\rangle \leftrightarrow |u, n-1\rangle \leftrightarrow |g, n-2\rangle \leftrightarrow |u, n-3\rangle \cdots \quad (22)$$

Figure 8 shows the rate versus intensity for the two- and four-channel approaches applied to the resonance issued from the fourth field-free vibrational state of the molecule ($v_{diab}=3$). The two additional channels $|g, n-2\rangle$ and $|u, n-3\rangle$ correspond to the absorption of two and three photons. The rates are shown in the neighborhood of the critical intensities. Beyond four channels there is no significant change of the results. The four-channel description can therefore be

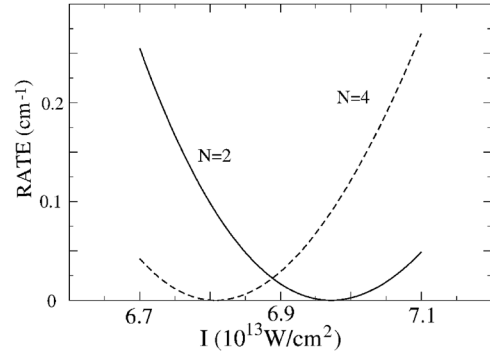


FIG. 8. Rate versus intensity for a two-channel approach ($N=2$) or a four-channel approach ($N=4$). The resonance under investigation is issued from the field-free $v_{diab}=3$ vibrational state. The critical intensities for a zero-width resonance are, respectively, $I=6.972 \times 10^{13}$ W/cm² for $N=2$ and $I=6.808 \times 10^{13}$ W/cm² for $N=4$.

considered as the converged limit as far as the number of channels is concerned. The critical intensity yielding a zero-width resonance is somewhat smaller when going to the converged study, but the width can again be made very small by “zooming” near this intensity. We conclude that such resonances are present, irrespective of the number of channels. A detailed semiclassical study based on four or more channels appears to be rather complicated, due to the number of paths that can lead to dissociation. Each open channel adds its contribution to the total width. Our argument will be based on a circumstance which has been stressed in [21]: each path leading to dissociation has to go through the crossing present in the two-channel description. If for some reason the amplitude for a transition through this first crossing vanishes, the entire process leading to dissociation is quenched. The first crossing plays the part of a doorway crossing. Therefore the analysis showing that the short-range transition amplitude can be made to vanish as a result of an interference of two terms is still valid. It remains to explain why there is a change in the critical intensity. We attribute this to the distortion suffered by the two primitive potentials (those of the two-channel Floquet picture) as a result of the interaction with the additional channels. The argument about the fact that in a multichannel approach the transition at the first crossing met by the system is essential to eventually quench the dissociation can also be applied to an extension including rotational effects. This was explained in [6] and is valid for the present case. This is due to the chainlike structure of the coupling with the channels with higher rotational quantum numbers,

$$|g, n, J=0\rangle \leftrightarrow |u, n-1, J=1\rangle \leftrightarrow |g, n-2, J=2\rangle \leftrightarrow \cdots \quad (23)$$

The two-channel Floquet picture is making use of the first two elements of this chain which are sufficient, to a good approximation, to describe the trapping.

V. WAVE FUNCTIONS AND PROBABILITY DENSITIES

The study of the wave functions shows that at each critical intensity the component wave function in the upper chan-

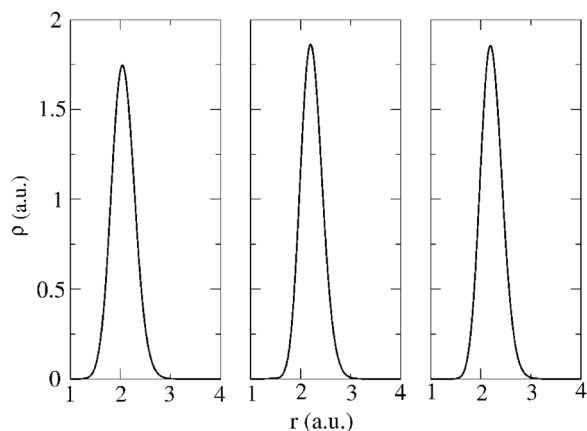


FIG. 9. The probability densities $\rho(r)$ obtained when using the field-free ground vibrational state as a starting point. Left panel: the field-free state. Middle panel: the density of the two-channel Floquet function at the first critical intensity producing a vanishing of the width. Right panel: the density calculated for the lowest state of the upper adiabatic potential at the critical intensity.

nel of the Floquet treatment is extremely close to the wave function of the upper adiabatic channel at this same intensity. This is in agreement with the analysis showing that for the width to vanish we must satisfy two conditions: (a) we must be at an energy where there is vanishing of the short-range transition amplitude, but also (b) at this energy there must be a level of the upper adiabatic potential. This is illustrated by the probability densities $\rho(r)$ given in Figs. 9 and 10 for an initial choice $v_{diab}=0$ and 3. These densities are defined as the sum over the channels of the square moduli of the component wave functions. This is justified by the fact that a zero-width resonance wave function is square integrable. One observes a remarkable agreement between the densities of the two approaches, except in Fig. 10 where there is on the left a small hump in the Floquet density. This hump is associated with the lowest adiabatic channel, with a turning point to the left of the equilibrium distance of the field-free molecule. The study of the densities in Fig. 10 reveals a curious circumstance which may be related to the failure to follow the fate of the diabatic state with $v_{diab}=2$. While for $v_{diab}=0$ and 1 there is no change in nodal structure when going from a null intensity to the first critical intensity, for $v_{diab}=3$ and 4, there is a decrease by 1 of the number of nodes. This is also observed in D_2^+ . We note that for $v_{diab}=0$ the dressed state corresponds to a slight lengthening of the bond. This is due to the fact that the equilibrium distance of the upper adiabatic potential is to the right of that of the field-free molecule. There is in this case only a very weak component of the wave function in the lower channel.

VI. PRODUCTION AND DETECTION

We first of all examine the possibility of an adiabatic transfer from a field-free vibrational state to a dressed zero-width resonant state. Our example will be again the resonance state issued from the $v_{diab}=0$ field-free state. Figure 2 shows that, before reaching the zero-width resonant state, the

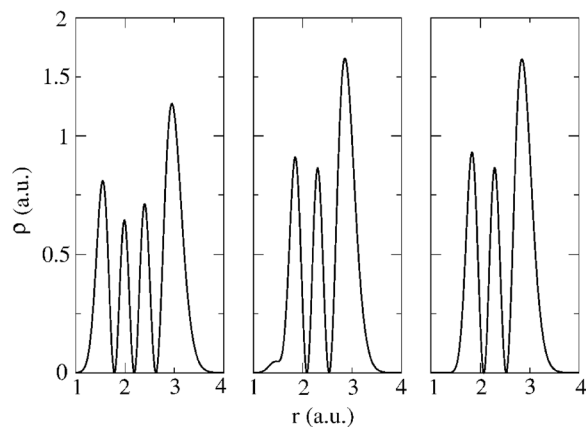


FIG. 10. The probability densities $\rho(r)$ obtained when using the field-free vibrational state with $v_{diab}=3$ as a starting point. The three panels have the same meaning as in Fig. 9. We note the change in the number of nodes when going from the field-free treatment to the Floquet and adiabatic approaches at the critical intensity. The change is progressive along the trajectory. The difference between the Floquet wave function and the adiabatic wave function resides in the hump around $r=1.4$ a.u., which is associated with the lowest channel.

rate passes by a maximum of the order of 200 cm^{-1} . The lifetime is at this intensity $\sim 2.65 \times 10^{-14} \text{ s}$. It is therefore required that the excitation process be significantly shorter than this time in order for some molecules to survive along the way. Let us say ten times shorter. The period of the field, with $\lambda=80 \text{ nm}$, is $\sim 2.67 \times 10^{-16} \text{ s}$. This corresponds to about ten oscillations during that time. Thus we are within the limits for the applicability of Floquet theory [22]. A criterion for adiabatic transport is that the ratio of the Rabi frequency to the field frequency is at the intensity of the maximum rate much lower than 1. For the present example, near the crossing region, we have $\mu\sqrt{I}/\omega \approx 0.04$.

We can also imagine a sudden pulse such that the molecule is exposed in a very short time to the critical intensity leading to the zero-width resonant state. Let us describe the excitation process in the simplest possible way: the various dressed adiabatic states are populated with probabilities

TABLE II. Franck-Condon amplitudes and factors between the field-free ground vibrational state of H_2^+ and the states of the upper dressed adiabatic potential at the intensity $I=11.19 \times 10^{13} \text{ W/cm}^2$ producing a zero-width resonance.

| v_{diab} | $\langle 0_{diab} v_{diab} \rangle$ | $ \langle 0_{diab} v_{diab} \rangle ^2$ |
|------------|---------------------------------------|---|
| 0 | 0.9320 | 0.8686 |
| 1 | -0.2834 | 0.0803 |
| 2 | 0.1586 | 0.02516 |
| 3 | 0.1027 | 0.01054 |
| 4 | -0.07304 | 0.005335 |
| 5 | 0.05518 | 0.003045 |

proportional to the Franck-Condon factors between the original diabatic vibrational state and the final dressed adiabatic states.

We give in Table II these Franck-Condon factors. It is clear that the lowest adiabatic state is favored. Since at this intensity only one of the adiabatic states has a small width, all other states should disappear quickly. This means that in the detection of the dissociation products there should be, as a signature of the existence of molecules raised to a zero-width resonant state, a signal with a significant delay with respect to the signal following immediately the excitation. The fact that after the critical intensity there is generally a range of intensities where the width remains rather small (cf. Fig. 2) means that there is a certain flexibility in the choice of the laser intensity.

VII. CONCLUSIONS

The present work has some points in common with the study of so-called vibrational trapping (cf. for instance, [4]). In such studies one examines the fate of the molecule when a pulse is applied. One or several resonances may be excited, depending on whether the pulse is smooth or sudden. The main difference is that the choice of the intensity is not tailored to match the potential structure of the dressed molecule. We have shown that there exist in the case studied here, as well as in the previous study [6], very special intensities that may facilitate the trapping of the molecule in a dressed adiabatic state. A test is under way to examine how the molecule responds when the peak intensity is chosen to be precisely a critical intensity.

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