

Dissociation quenching using exceptional points

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Abstract We examine a short way to reach an exceptional point that corresponds to a coalescence of two resonance energies. The application concerns the photodissociation of the Na_2 molecule exposed to a laser field. In this case, the resonances can be correlated with the field-free vibrational states of the diatomic species. The resonances are due to the field-induced coupling with the continuum of a repulsive potential. We also draw attention to a new kind of exceptional point involving a resonance originating from a vibrational state coalescing with a shape-type resonance of the repulsive potential. A laser control scenario, aiming at the adiabatic transport from this field-free decaying state to a stable field-free vibrational state, is discussed in terms of field-induced dissociation quenching.

Keywords Photodissociation · Resonances · Exceptional points · Population transfer · Laser control · Dissociation quenching · Floquet formalism

Introduction

The description of quantum decaying processes generally involves the determination of resonance states with complex eigenenergies. These energies are in fact the poles of the scattering matrix [1]. The rate of decay of an initially prepared state

is derived from the imaginary part of such energies. One recent development in this context is the possibility to produce the degeneracy of two of these energies. This means that both real and imaginary parts of the resonance energies can merge. This is achieved by an appropriate choice of the parameters governing the dynamics of the system. The corresponding point in the parameter plane is called an exceptional point (EP) [2, 3]. As a matter of fact, at such a point, there exists a unique resonance wavefunction with the property of self-orthogonality [4]. Among the consequences of the existence of such EPs, there is the possibility to produce adiabatically a transfer from one resonance to another, provided the parameters are varied along a loop encircling the corresponding EP [5, 6]. Many situations have been created where EPs have been produced, with all the consequences expected from the formalism. Among the experimental confirmations, there is the study of the field distribution in a microwave cavity [7]. EPs have been detected for a hydrogen atom subjected to electric and magnetic fields [8]. The collision of an electron with an hydrogen molecule is affected by EPs under some conditions [9].

Here, we consider laser-induced photodissociation of a diatomic molecular species treated within Floquet formalism [10]. The resonances are due to the radiative coupling between the vibrational states of an attractive potential with the continuum of a repulsive one. We have already proved the existence of EPs in the photodissociation of the H_2^+ species and showed that they could form the basis of a transfer from one of the field-free vibrational states to another [11]. In the case of Na_2 , we have proposed a cooling scenario to bring the molecules to their lowest vibrational state [12]. We pursue this problem with two aims: (1) to describe a novel way to reach, in a very simple manner, the coordinates of an EP in the laser wavelength-intensity parameter plane; (2) to examine the possibility of a dissociation quenching mechanism by the laser-controlled transfer of a field-free shape-type resonance of the repulsive electronic state onto a field-free vibrational level of the binding electronic state.

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The paper is organized as follows: in the following Section, we recall the formalism developed by Hernández et al. [6, 13] to discuss the behavior of resonance energies in the vicinity of an EP. This leads to the formulation of a simple way to reach EPs. The Floquet theory needed to describe molecular photo-dissociation is summarized in “Floquet formalism and its application to Na₂”. A section on “EP localization in the laser parameter plane” proceeds to the determination of an EP of Na₂. We then describe a new type of resonance found for the special case of a repulsive potential of Na₂ and which leads to EPs of a new type. We end with a laser control scenario based on such EPs, aiming at the adiabatic transfer of population temporarily prepared on these resonances towards field-free vibrational bound states (Laser controlled dissociation quenching). More precisely, this is a dissociation quenching mechanism. The otherwise naturally decaying population prepared by laser excitation from a lower third electronic state to the resonance state is trapped momentarily into a metastable vibrational state.

Resonance behavior close to an exceptional point

Hereafter, we follow the analysis given by Hernández et al. [13] for the resonance behavior close to an EP. We take as the starting point the identity of the complex energies E_{\pm} of the pair of resonances close to an EP:

$$E_{\pm} \equiv \frac{1}{2}[E_+ + E_-] \pm \left[\frac{1}{4}(E_+ - E_-)^2 \right]^{\frac{1}{2}} \quad (1)$$

The energies are assumed to depend on two control parameters, x_1 and x_2 , present in the Hamiltonian. In the following, these parameters are the wavelength and the intensity of an external laser field. The particular values leading to the coalescence of two complex energies are denoted x_1^{EP} and x_2^{EP} . We are looking for the conditions under which there is either equality of the real parts of the energies, or of their imaginary parts. We assume that we are close enough to the EP to justify a Taylor expansion. The only quantity to be considered in such an expansion is

$$\epsilon_{\pm} = \pm \left[\frac{1}{4}(E_+ - E_-)^2 \right]^{\frac{1}{2}} \quad (2)$$

since only this quantity can be close to zero near an EP and therefore be at the origin of a discrepancy between the two energies. Following [13] we write an approximation to ϵ_{\pm} as

$$\epsilon_{\pm} = \pm \sqrt{\frac{1}{4}[C_1(x_1 - x_1^{EP}) + C_2(x_2 - x_2^{EP})]} \quad (3)$$

Hernández et al. [6, 13] introduce three column vectors

$$\vec{\xi} = \begin{pmatrix} \xi_1 \\ \xi_2 \end{pmatrix} = \begin{pmatrix} x_1 - x_1^{EP} \\ x_2 - x_2^{EP} \end{pmatrix} \quad \vec{R} = \begin{pmatrix} ReC_1 \\ ReC_2 \end{pmatrix} \quad \vec{I} = \begin{pmatrix} ImC_1 \\ ImC_2 \end{pmatrix} \quad (4)$$

An analysis of the energy splitting shows that the real and imaginary parts of the resonance energies close to the EP differ respectively by

$$Re(\epsilon_{\pm}) = \pm \frac{1}{2\sqrt{2}} \left[\sqrt{(\vec{R} \cdot \vec{\xi})^2 + (\vec{I} \cdot \vec{\xi})^2} + \vec{R} \cdot \vec{\xi} \right]^{\frac{1}{2}} \quad (5)$$

and

$$Im(\epsilon_{\pm}) = \pm \frac{1}{2\sqrt{2}} \left[\sqrt{(\vec{R} \cdot \vec{\xi})^2 + (\vec{I} \cdot \vec{\xi})^2} - \vec{R} \cdot \vec{\xi} \right]^{\frac{1}{2}} \quad (6)$$

The cancellation of the energy difference ϵ_{\pm} (i.e., the fulfillment of the resonance coalescence condition) is obtained by a specific choice of two unit vectors $\vec{\xi}_R$ and $\vec{\xi}_I$ in the parameter plane. More precisely, the real parts (Eq. 5) are canceled with the choice:

$$\vec{I} \cdot \vec{\xi}_R = 0 \quad \vec{R} \cdot \vec{\xi}_R = -|\vec{R} \cdot \vec{\xi}_R| \quad (7)$$

while the imaginary parts (Eq. 6) are canceled with the choice:

$$\vec{I} \cdot \vec{\xi}_I = 0 \quad \vec{R} \cdot \vec{\xi}_I = |\vec{R} \cdot \vec{\xi}_I| \quad (8)$$

Equations 7 and 8 lead to the following important consequence:

$$\vec{\xi}_R = -\vec{\xi}_I \quad (9)$$

the interpretation being that the half-axis starting from the EP defined by $\vec{\xi}_R$ is a branch cut for the real parts of the resonance energies, while the half-axis supported by $\vec{\xi}_I$ and lying in the opposite direction, is a branch cut for their imaginary parts [6, 13]. Finally, the common point of these two half-axes is precisely the EP, where both real and imaginary parts of the resonance energies are equal.

Floquet formalism and its application to Na₂

We consider a 1D molecular system interacting with an external electromagnetic field. This is justified when the pulse duration is less than the rotational period, such that

the rotational motion is approximately frozen. With a time-periodic Hamiltonian $\mathcal{H}(R, t)$ the Floquet ansatz consists in writing the wave function in the form

$$\Phi(R, t) = e^{-iE_F t/\hbar} \Phi_F(R, t) \tag{10}$$

where $\Phi_F(R, t)$ is a periodic function of time fulfilling the eigenvalue equation

$$\mathcal{H}_F(R, t)\Phi_F(R, t) = E_F\Phi_F(R, t) \tag{11}$$

the Floquet Hamiltonian being

$$\mathcal{H}_F(R, t) = \mathcal{H}(R, t) - i\hbar \frac{\partial}{\partial t} \tag{12}$$

E_F is called a quasi-energy. For time-periodic Hamiltonians, such energies play the same role as the eigenenergies of time-independent Hamiltonians. Our model implies two electronic states. Na_2 is an illustrative example of this situation with electronic states (a) $^3\Sigma_u^+$ and (1) $^3\Pi_g$, hereafter abbreviated as $|u\rangle$ and $|g\rangle$ respectively, and depicted in Fig. 1 after the field-dressing operation described below. The lowest electronic state accommodates 14 bound vibrational levels, while the other potential is repulsive.

The wave function is written

$$|\Phi_F(R, t)\rangle = \chi_u(R, t)|u\rangle + \chi_g(R, t)|g\rangle \tag{13}$$

R is the nuclear coordinate. With the length-gauge and the long-wavelength approximation, the nuclear wave functions are solutions of the time-dependent Schrödinger equation

$$[\mathbf{H} - i\hbar \frac{\partial}{\partial t} \mathbf{1}] \Xi(R, t) = 0 \tag{14}$$

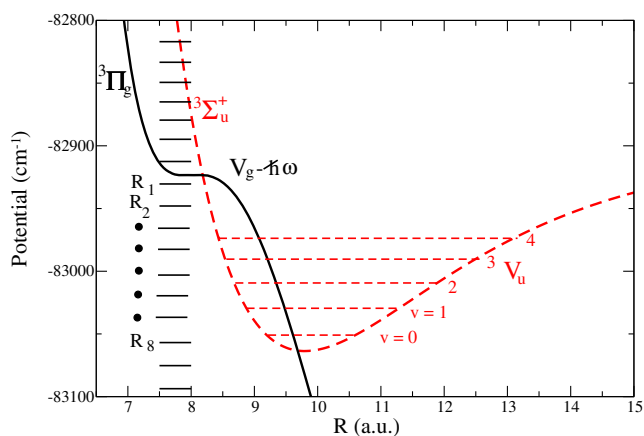


Fig. 1 The field-dressed potentials of the Na_2 molecule for a wavelength $\lambda=552$ nm. The horizontal segments (solid black lines) indicate the positions of the resonances (R_1, R_2, \dots) associated with the repulsive potential $V_g - \hbar\omega$. The dotted red lines indicate the positions of the bound levels of the attractive potential V_u . Figure in color online

Ξ is the column vector made of χ_g and χ_u . \mathbf{H} is the matrix operator

$$H = 1T_R + \begin{bmatrix} V_u(R) & \mu(R)\mathcal{E}(t) \\ \mu(R)\mathcal{E}(t) & V_g(R) \end{bmatrix} \tag{15}$$

$T_R = -\hbar^2/2\mathcal{M}(d^2/dR^2)$ is the vibrational kinetic energy operator, with \mathcal{M} the reduced mass. $V_g(R)$ and $V_u(R)$ are the two Born-Oppenheimer potentials. $\mu(R)$ is the electronic transition dipole moment between states $|u\rangle$ and $|g\rangle$. $\mathcal{E}(t)$ is the linearly polarized laser electric field amplitude of the form $\mathcal{E}_0 \cos(\omega t)$, with a frequency ω , wavelength $\lambda = 2\pi c/\omega$ and intensity $I \propto \mathcal{E}_0^2$. The Floquet ansatz applied to this case consists in writing the vector wave function as

$$\begin{bmatrix} \chi_u(R, t) \\ \chi_g(R, t) \end{bmatrix} = e^{-iE_F t/\hbar} \begin{bmatrix} \phi_u(R, t) \\ \phi_g(R, t) \end{bmatrix} \tag{16}$$

$\phi_k(R, t)$ ($k=u, g$), being time periodic, can be Fourier expanded as

$$\phi_k(R, t) = \sum_{n=-\infty}^{+\infty} e^{in\omega t} \varphi_k^n(R) \tag{17}$$

Two channel wavefunctions are enough for convergence when the laser intensity is such that only single photon processes are present. The two channel wave functions needed in this situation can be taken as $\varphi_u^n(R)$ and $\varphi_g^{n-1}(R)$ or in short $\varphi_u(R)$ and $\varphi_g(R)$. They are solutions of the coupled equations

$$[T_R + V_u(R) - E_F]\varphi_u(R) - 1/2 \mathcal{E}_0 \mu(R)\varphi_g(R) = 0 \tag{18}$$

$$[T_R + V_g(R) - \hbar\omega - E_F]\varphi_g(R) - 1/2 \mathcal{E}_0 \mu(R)\varphi_u(R) = 0 \tag{19}$$

The complex resonance energies result from an iterative matching procedure of outward and inward propagations using the Fox-Goodwin algorithm [14]. Complex rotation of the spatial coordinate R in its exterior version is used. This procedure has the advantage of introducing automatic L^2 boundary conditions for a resonance wave function [15, 16]. Finally, it should be noted that this formalism, although strictly valid for a continuous wave laser, is well known to apply successfully in cases of pulsed fields accommodating even only a few optical cycles. Figure 1 gives the potentials $V_u(R)$ and $V_g(R) - \hbar\omega$ corresponding to a wavelength $\lambda=552$ nm. The Born-Oppenheimer potentials $V_u(R)$ and $V_g(R)$ are taken from [17] and the electronic transition moment $\mu(R)$ from [18].

EP localization in the laser parameter plane

Our example is the EP resulting from the merging of the resonances issued from the levels $\nu=3$ and $\nu=4$ of the Na_2 molecule in its $(a)^3\Sigma_u^+$ electronic state. A photodissociation laser couples these channels to the continuum of the $(1)^3\Pi_g$ electronic state. We first determine the privileged axis in the $I-\lambda$ parameter plane along the direction of the unit vectors $\vec{\xi}_R$ and $\vec{\xi}_I$ defined in the section [Resonance behavior close to an exceptional point](#). Two points are enough for this purpose. We calculate the resonance energies for two wavelengths, namely $\lambda=562.4$ nm and $\lambda=562.6$ nm. The real and imaginary parts of the resonance energies are shown in Fig. 2a,b for the smallest of the two wavelengths, and Fig. 2c,d for the highest. The intensity goes from $I=0.3$ GW/cm^2 to $I=0.4$ GW/cm^2 . On the left there is a crossing of the real parts and an avoided crossing of the imaginary parts. On the right, it is just the reverse. This is a strong indication that there is an EP for an intermediate wavelength. From these graphs, it is possible to determine the intensities at which there is a crossing. This intensity is $I=0.369330$ GW/cm^2 in Fig. 2a and $I=0.313314$ GW/cm^2 in Fig. 2d. The equation of the axis we are looking for can be given the form:

$$I = a\lambda + b \quad (20)$$

Solving the two equations:

$$0.36933 = a \cdot 562.4 + b \quad 0.313314 = a \cdot 562.6 + b \quad (21)$$

provides the values

$$a = -0.28008, b = 157.886322 \quad (22)$$

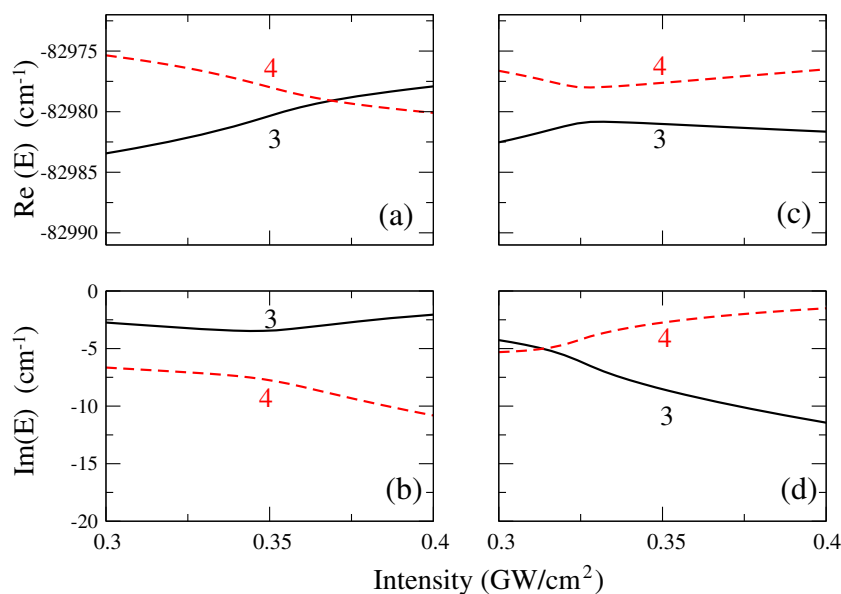
The next operation is to follow this axis and to determine for each point along the axis the real and imaginary parts of

the resonances originating from $\nu=3$ and $\nu=4$. The result is shown in Fig. 3. Increasing the wavelength means going from right to left in the graph, since this amounts to a decrease in intensity. In Fig. 3a, we first see an equality of the real parts, followed suddenly by a split. The reverse situation is seen in Fig. 3b, where the imaginary parts are different and merge suddenly. The critical intensities at which these events occur can be estimated as $I=0.3328$ GW/cm^2 in Fig. 3a and $I=0.3326$ GW/cm^2 in Fig. 3b. These values, when used as inputs in the relation between intensity and wavelength, produce the two wavelengths $\lambda=562.530$ nm and $\lambda=562.531$ nm. The coordinates of the EP in the parameter plane derived from this analysis can be taken as $\lambda_{\text{EP}}=562.530$ nm and $I_{\text{EP}}=0.3327$ GW/cm^2 .

Resonances associated with the $(1)^3\Pi_g$ state

Rather surprisingly, we found that the potential of the $(1)^3\Pi_g$ state used in this study can accommodate resonances. Some examples are known in the literature of resonances associated with potentials without barrier. Such is the case for an exponential potential of the form Ae^{-aR} [19], with resonances below the asymptotic limit. Another example is the step potential where resonances are above or below the threshold energy [4]. The positions of some of the shape-type resonances under consideration are shown in Fig. 1. Some of the widths are given in Fig. 4. There are two noticeable characteristics. One is that the real parts of the resonance energies are spaced very regularly (differences of ca. 20 cm^{-1}). The other is that the imaginary parts vary slowly (around 25 cm^{-1}). Figure 4 illustrates these two properties. In order to examine these resonances further, we refer

Fig. 2 Crossings between real or imaginary parts of the resonance energies are used to determine the axis passing through the exceptional point (EP). **a, b** Wavelength, $\lambda=562.4$ nm; **c, d** $\lambda=562.6$ nm. Figure in color online



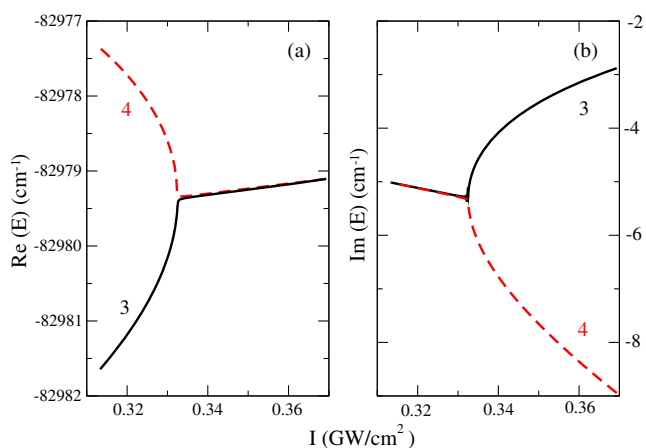


Fig. 3 Going along the axis passing through the EP leads to the localization of the EP, either as the point at which there is divergence of the real parts (a) or as the point where there is merging of the imaginary parts (b) as the intensity is increased (as a consequence of decreasing the wavelength in Eq. 20). Figure in color online

to a method originating from the work of Humblet and Rosenfeld [20], devoted to a definition of partial widths when there is more than one open channel, but which can also be applied to the present single channel problem. Consider the wave equation and assume that it has been solved with the Siegert outgoing wave boundary conditions appropriate for a resonance [21]:

$$-\frac{\hbar^2}{2\mathcal{M}}\Psi''(R) + V(R)\Psi(R) = E\Psi(R) \quad (23)$$

We use the notations $\Psi'(R)$ and $\Psi''(R)$ for the first and second derivatives of $\Psi(R)$ with respect to R . Assuming that $V(R)$ is real, multiplying this equation by $\Psi(R)^*$ (the star * is

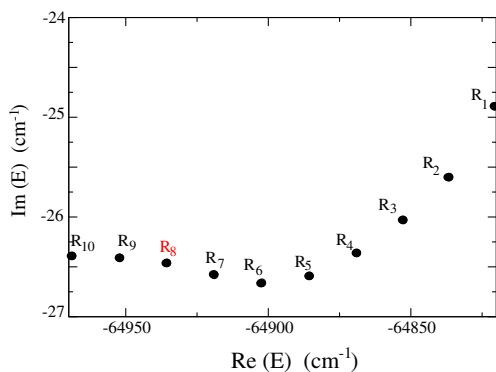


Fig. 4 Complex energies of the resonances of the $(1)^3\Pi_g$ potential. The resonances are spaced regularly, with widths all of the same order of magnitude. R8 is the resonance involved in the transfer studied in Laser controlled dissociation quenching. Figure in color online

for the complex conjugate) and subtracting from this expression the product of $\Psi(R)$ by the complex conjugate of the wave equation (Eq. 23) yields

$$-\frac{\hbar^2}{2\mathcal{M}}[\Psi^*(R)\Psi''(R) - \Psi(R)\Psi''^*(R)] = (E - E^*) |\Psi(R)|^2 \quad (24)$$

Writing the energy in the form $E = E_R - i\Gamma/2$, we have $(E - E^*) = -i\Gamma$. The bracket on the left is

$$\begin{aligned} &[\Psi^*(R)\Psi''(R) - \Psi(R)\Psi''^*(R)] \\ &= d[\Psi^*(R)\Psi'(R) - \Psi(R)\Psi'^*(R)] \end{aligned} \quad (25)$$

Integrating both sides from 0 to R , and taking into account that at $R=0$ the wave function is vanishing for a molecular problem, we obtain

$$\frac{\hbar^2}{2\mathcal{M}} [\Psi'^*(R)\Psi'(R) - \Psi(R)\Psi'^*(R)] = i \Gamma \int_0^R |\Psi(R)|^2 dR \quad (26)$$

From this relation, the width is obtained as

$$\Gamma = \frac{\hbar^2}{\mathcal{M}} \frac{Im(\Psi^*(R)\Psi'(R))}{\int_0^R |\Psi(R)|^2 dR} \quad (27)$$

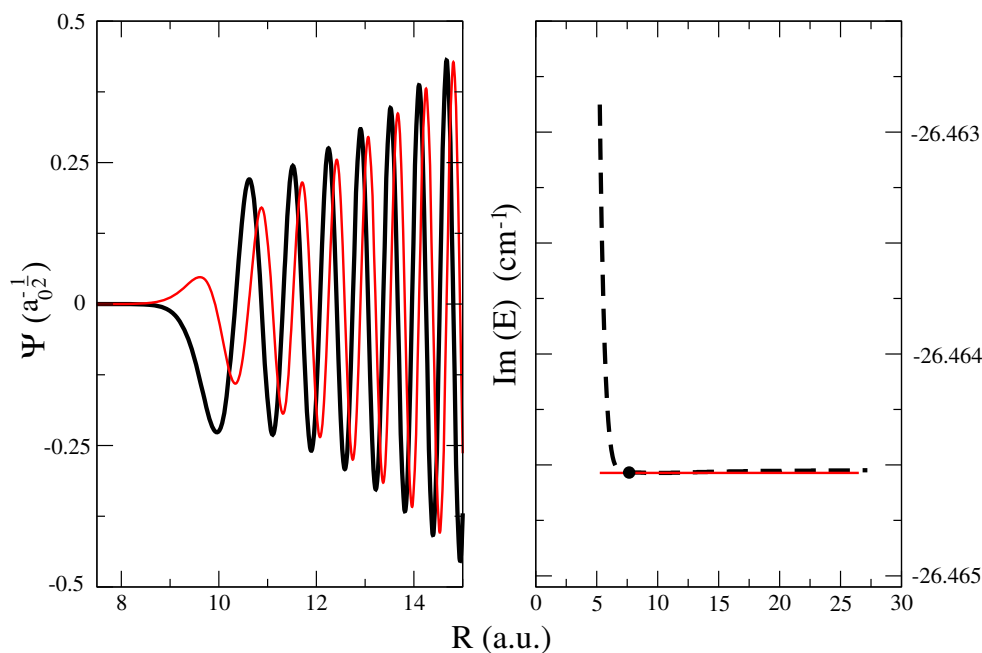
Figure 5 compares the result of this analysis, giving the width from the wave function to the value obtained from the quantization condition. This is done for the resonance R_8 of Fig. 1. The agreement is almost perfect, even in the non-classical region, which is on the left of the turning point. Far inside the non-classical region, the error on the width is surprisingly small (about 0.001 cm^{-1}). This shows that the Humblet-Rosenfeld formula goes far beyond the result of a flux analysis in the asymptotic region.

Laser-controlled dissociation quenching

The last application in our search for EPs in this model for Na_2 is the observation that the occurrence of a degeneracy of complex energies can involve a pair of levels mixing the two kinds of resonances described in this work: the resonances originating from the field-free vibrational states of the $(a^3\Sigma_u)$ state, with the resonances of the $(1)^3\Pi_g$ state as modified by the application of a laser field. To give an example, we found that with a chirped laser pulse of the form

$$\lambda = \lambda_0 + \delta\lambda \sin(\phi); I = I_{max}\sin(\phi/2) \quad (28)$$

Fig. 5 **a** Resonance wave function (in atomic units) used in this analysis. The *black thick* and the *red thin* curves represent the real and imaginary parts, respectively. **b** A comparison of the imaginary part (*solid red line*) of the R8 resonance energy of the $(1)^3\Pi_g$ electronic state (see Fig. 1) as obtained from energy quantization using Floquet formalism with the result of Humblet-Rosenfeld formula Eq. (27) (*black dashed line*). The *black bullet* shows the position of the turning point on the potential. Figure in color online

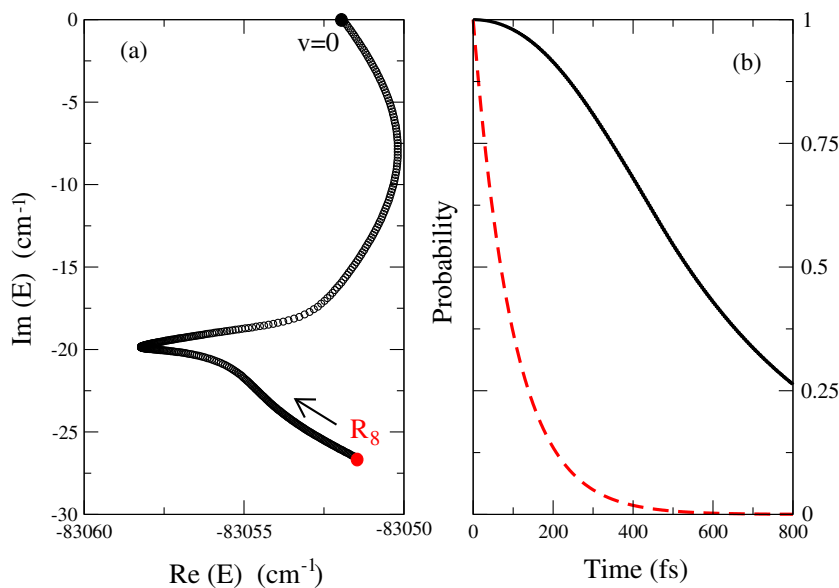


it is possible to produce an adiabatic transfer from a field-free vibrational state of the $(a)^3\Sigma_u$ state to a field-free resonance state of the $(1)^3\Pi_g$ state. ϕ varies from 0 to 2π . The pulse parameters are $\lambda_0=552$ nm, $\delta\lambda=0.5$ nm and $I_{\max}=0.3$ GW/cm². It is worthwhile noting that, in the vicinity of an EP, the adiabaticity conditions validating the Floquet approach, which is referred to have to be checked very carefully through the choice of laser parameters (in particular pulse duration). The transfer is from $v=0$ to the resonance R_8 of Fig. 4. This is a proof that there is an EP involving the two kinds of resonances inside this loop. This suggests a different experiment. Such transfers are reversible. It is sufficient to change $\delta\lambda$ into $-\delta\lambda$.

Figure 6 presents the transfer from R_8 to $v=0$. It must be emphasized that, even in the absence of a field, a complex eigenenergy corresponds initially to the former state. To the latter state there corresponds a real eigenenergy at the end of the pulse. It is possible to calculate the survival probability P_s of the initial state from an expression based on an adiabatic formulation of the Floquet theory [22, 23]:

$$P_s = \exp \left[-\hbar^{-1} \int_0^{t_f} \Gamma(t') dt' \right] \quad (29)$$

Fig. 6 Transfer from resonance R_8 of the $(1)^3\Pi_g$ electronic state to the bound vibrational level $v=0$ of the $(a)^3\Sigma_u^+$ state (see Fig. 1). **a** Trajectory in the complex energy plane leading from R_8 (*red bullet*) to $v=0$ (*black bullet*). **b** Survival probabilities of the state originating from R_8 calculated in two ways. The *red dashed curve* is the decay of R_8 in the absence of radiative coupling. The *black solid curve* corresponds to an adiabatic transfer along the loop as discussed in the text. Figure in color online



t_f is the pulse duration. $\Gamma(t')$ is the width calculated at time t' with the field parameters at that time. t' is related to the pulse phase ϕ of Eq. (28) by

$$t' = \frac{\phi}{2\pi} t_f \quad (30)$$

The survival probability is shown by the continuous black curve in Fig. 6b, calculated with $t_f=800$ fs. This choice ensures an adiabatic transfer, since, from the energy interval between the two states $v=3,4$ (17 cm^{-1}), it is estimated that the pulse duration has to exceed 400 fs, such that its energy broadening be less than this interval. Comparison is made with the decay of R_8 in the absence of a field (dashed red curve). We conclude that the application of the laser pulse has, to a large extent, quenched the decay of R_8 , with 25 % of the molecules being led to the bound state $v=0$ at time t_f . This state belongs to an excited electronic state and has a lifetime of the order of a few ns, while we are concerned with processes lasting less than a picosecond.

Conclusion

In this work we have proven that the purely repulsive potential of the $(1)^3\Pi_g$ electronic state of the Na_2 molecule unexpectedly accommodates a full series of shape-type resonances with regular spacings and almost constant widths. This is presumably in relation to the specific form of this potential curve, which shows, in particular, a plateau behavior extending up to about 2 \AA in terms of internuclear distances. The laser parameters (wavelength and intensity) inducing the coalescence of two resonances originating from this shape-type resonance and a nearby field-dressed bound vibrational state define a new type of EP. Moreover, it is shown that, following the well known properties of EPs, a laser loop encircling such an EP offers the possibility of adiabatic transport of a population from the field-free shape-type resonance to the field-free vibrational state. Our claim is that such a control scheme is a dissociation quenching strategy. Actually, the shape-type resonance, prepared initially by photoexcitation, is trapped into a metastable vibrational state, the mechanism involving three electronic states.

We have shown that, with a convenient choice of laser parameters, up to one-fourth of the population prepared on the shape-type resonance can be preserved from dissociation.

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