Regular and chaotic multiphoton dissociation

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The rate of multiphoton dissociation of the BeH^{2+} molecule in its ground and first two excited vibrational levels has been computed via classical mechanics as a function of laser frequency. There is agreement with earlier quantum-mechanical results as regards the existence and magnitude of an optimal frequency, ω^*, for which the dissociation rate is maximized. This fact has been analyzed and understood via the application of the theory of chaotic scattering. Indeed, we find fractal singularities in the function $T_d(x)$ of the duration of photodissociation, and we compute their dimension to be equal to 1, in agreement with the conjecture of Lau, Finn, and Ott [Phys. Rev. Lett. **66**, 978 (1991)] that this must be a characteristic of systems exhibiting nonhyberbolic scattering. Turning to the problem of interpreting the appearance of an optimal ω^*, we propose the following two mechanisms for the reduction of the multiphoton dissociation rate. First is the increase of fractal singularities when the frequency ω attains values larger than ω^*. Second is the gradually increasing overlap of the classical initial state with the region of Kolmogorov-Arnold-Moser tori when $\omega<\omega^*$. Finally, as the intensity is increased there is a transition from chaotic to regular photodissociation, where the singularities in $T_d(x)$ are finite. It is conjectured that this reflects the emergence of the quantum-mechanical phenomenon of above-threshold dissociation. [S1050-2947(97)08302-9]

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I. INTRODUCTION

Given the rapid growth of the field of nonlinear classical dynamics (e.g., Ref. [1]), and the strong interest in making connections to results and features of quantum mechanics, it is important and challenging to adopt as objects of investigation real quantal systems and processes. One such case is the multiphoton dissociation (MPD) of diatomic molecules induced by strong infrared lasers, which has been studied by applying advanced quantal and classical methods and interpretations to the parametrized Morse oscillator [2–14]. These classical methods have their basis in the progress that has been made in the nonlinear dynamics of *bound* systems.

On the other hand, during the past decade, significant progress has been made in the understanding of the chaotic behavior of *unbound*, i.e., scattering, systems, where the interaction time is small [15–24]. Now, rather than emphasizing and analyzing phase-space characteristics, the basic theme is to achieve mappings of properties of final scattering states with details of initial conditions. One general result is that classical scattering is divided into regular and chaotic. The criteria for this division are the singularities that appear in the scattering functions. If their number is finite, then we have regular scattering, while chaotic scattering is associated with the appearance of fractal structure. By scattering function is meant ''*a plot of an output variable characterizing the trajectory after scattering versus an input variable characterizing the incident trajectory. In chaotic scattering the scattering function is singular on a Cantor set of values of the input variable*'' [23].

The theory of chaotic scattering has been developed using simple models as examples. As regards implementation to systems with quantal counterparts, noteworthy are recent results on one-dimensional atoms with model interactions, such as the work of Hillermeier, Blümel, and Smilansky [24] on ionization of H Rydberg atoms, of Gu and Yuan [25] on electron scattering from He^+, and of Handke [26] on autoionization of doubly excited He-like atoms.

Given the above, it appeared appropriate and timely to us to attempt an understanding of a possible connection of the current theory of chaotic scattering to the dynamics of MPD. Indeed, the object of this work was to study the MPD of BeH^{2+} into $Be^+ + H^+$ by the methods of chaotic scattering, having as a reference quantum-mechanical results that were obtained recently [27]. The BeH^{2+} potential supports only a few vibrational levels, and it was found to dissociate for intensities which are smaller by orders of magnitude than those needed for molecules such as HF (treated as a Morse oscillator).

In the following sections we will show how chaotic scattering applies to this problem, and we will present results demonstrating that MPD can be separated into regular and chaotic, and that the regular properties of the classical dissociation rate (CDR) emerge as a function of the laser frequency ω. (We note that in most publications on CDR, interest has focused on the dependence on the laser intensity.) For example, the quantum-mechanical result [27] that the rate of MPD is optimized for a value of ω which is about 0.9 times the frequency for the $v=0\rightarrow 1$ transition is verified here, and this agreement is used as input for a deeper analysis. (In Ref. [27] there were also interesting findings associated with tunneling. These do not concern us in the present work.)

In Sec. II we compute the CDR as a function of ω, where the molecule is in the ground vibrational level as well as in the first two excited ones. We conclude that the results offer

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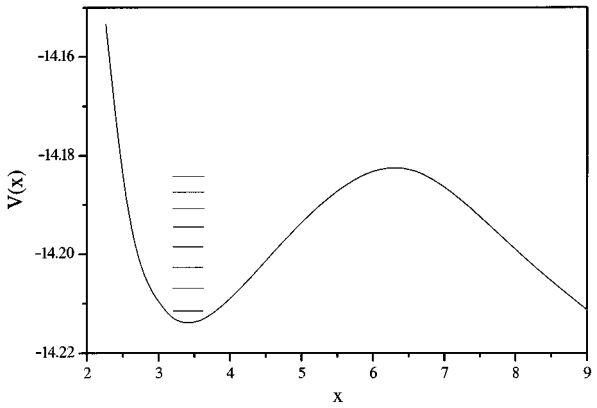

FIG. 1. The potential function $V(x)$ and its eight vibrational levels of the ground electronic state of BeH^{2+} (from Ref. [28]). For the needs of our calculations, we fit it to a seventh-degree polynomial.

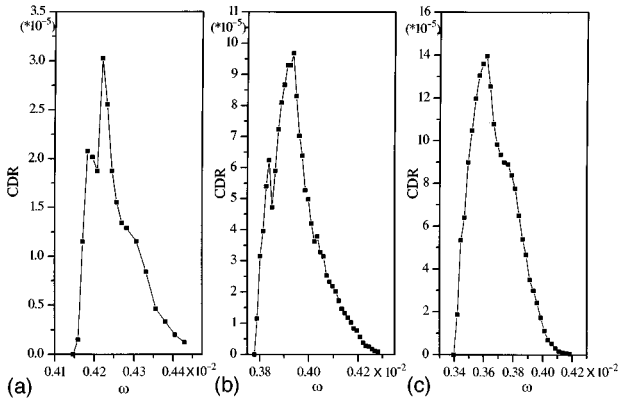

FIG. 2. The CDR as a function of the field frequency ω when the molecule is initially in the ground vibrational level (a), the first excited level (b), and the second excited level (c). In all cases $F=2\times10^{-3}$ a.u.

a sufficient basis for testing the relevance of the theory of chaotic scattering to MPD.

In Sec. III, we show the existence of fractal singularities in the function of the duration of MPD, and we compute their dimension. The results are in harmony with the results and conjectures of Lau, Finn, and Ott [23]. Furthermore, we find that the fractal structure is lost as we increase the field strength. In Sec. IV we utilize the finding of the existence of fractal singularities in order to propose two mechanisms for the reduction of the MPD rate as the frequency is increased or decreased from its optimal value. Finally, in Sec. V we summarize and discuss the perspectives of the present results.

II. CLASSICAL DISSOCIATION RATE VS LASER FREQUENCY

The calculation of the CDR for the purposes of the present study was carried out as follows: The classical Hamiltonian for BeH^{2+} plus the monochromatic laser field along the axis was taken to be (all quantities are in atomic units)

$$H(p,x)=\frac{p^2}{2\mu}+V(x)+Fx\,\cos(\omega t). \tag{1}$$

The reduced mass μ is 1651.8 a.u., and F is the field strength. Given the ionic character of the molecule, the dipole moment was assumed to be linear in the distance x. The potential function $V(x)$, which is presented in Fig. 1, was taken from the *ab initio* calculations of Ref. [28]. The figure also shows the eight vibrational levels supported by $V(x)$. The classical dynamics is obtained from the solution of Hamilton's equations

$$\frac{dp}{dt}=-\frac{\partial V(x)}{\partial x}-F\,\cos(\omega t), \tag{2a}$$

$$\frac{dx}{dt}=\frac{p}{m}. \tag{2b}$$

In previous calculations of CDR, e.g. [8], the approach is the following: One considers a set of trajectories with initial conditions of the coordinate x, where all points are equally distributed between the two turning points corresponding to the energy E of the level under consideration. Equations (2) are integrated for these trajectories and, assuming a criterion of dissociation, one obtains the number $N_b(t)$ of trajectories which at time t have not escaped from the potential. The CDR is then equated with the slope of the straight line expressing the short-time behavior of $N_b(t)$. The disadvantage of this method, when one is interested in obtaining CDR(ω), is that in many situations the linear fit of $N_b(t)$ is uncertain, regardless of whether the scales are linear [5,8] or semilogarithmic [14]. This fact has led us to the choice of a different method for the calculation of the CDR: Following Goggin and Milonni [8], we start with N equally spaced in coordinate x initial conditions, and we integrate Eqs. (2) while following the evolution of the trajectories for a fixed E. For a given trajectory starting from a point x at $t=0$, we define the duration of the photodissociation, $T_d(x)$, as the point in time where its compensated energy [5,8] E_c,

$$E_c=\frac{\left[p-\frac{F}{\omega}\sin(\omega t)\right]^2}{2\mu}+V(x), \tag{3}$$

becomes larger than the energy E_d of the saddle point of $V(x)$. By calculating T_d for the N initial conditions, we obtain the CDR from the relation

$$\mathcal{R}=\frac{1}{N}\sum_{i=1}^{N}(T_d^i)^{-1}. \tag{4}$$

The form of $\mathcal{R}(\omega)$ is almost independent of N, as long as its value is not very small. Our calculations were done for $N=500$, while few selected ones which were done for $N=1000$ produced the same results. The integration of Eqs. (2) was done by the fourth-order Runga-Kutta technique, and the accuracy was checked by reversing the velocity after sufficient time and verifying that the initial conditions are reproduced up to the fourth decimal digit.

The results are depicted in Fig. 2. When the molecule is in

the ground state, the value of ω for which CDR is largest is ${}^0\omega^*_{\rm cl}=4.22\times10^{-3}$ a.u. This value is very close to the one obtained from the quantal calculations [27], for which ${}^0\omega^*_{\rm qu}=4.1\times10^{-3}$ a.u. We note that the intensity was chosen to be larger than the one used before [27] by three orders of magnitude, in order to produce faster rates and thus make the calculations more economical. If we take the classical frequency which corresponds to the quantal frequency for the transition $v=0\rightarrow v=1$ to be the frequency $\omega_{\rm cl}$ of the trajectory with energy $E=(E_0+E_1)/2$ [29], then we find that ${}^0\omega^*_{\rm cl}\approx0.87\omega_{\rm cl}$, a relation which agrees with the quantal calculation [27] of 0.88, and in fact with earlier observations of frequency ''redshift'' for maximization of the dissociation rate [3,9]. The same conclusion can be drawn from the classical and quantal studies [13] of the MPD of the Morse oscillator representing the HF molecule: From Fig. 1(d) of Ref. [13], it follows that the redshift ratios are ≈0.89 and ≈0.90 for the quantal and the classical case, respectively.

For the first and second excited states, CDR is maximized when ${}^1\omega^*_{\rm cl}=3.94\times10^{-3}$ a.u. and ${}^2\omega^*_{\rm cl}=3.62\times10^{-3}$a.u.; in other words, there is a shift to smaller frequencies as we climb the energy spectrum. The same observation holds for the HF Morse oscillator [Fig. 3(a) of Ref. [13]].

We therefore conclude the following: First, that both quantum and classical mechanics predict a redshift of the single frequency for the maximization of the photodissociation rate of diatomic molecules, regardless of the details of the vibrational spectrum. Second, given the first conclusion, the presence of two factors, anharmonicity and the participation of the unbound scattering states, play a dominant role, and an additional understanding of the MPD process can be revealed by applying current methods of classical dynamics which account for these two factors. It is with this objective that we now turn to an application of the theory of chaotic scattering to molecular photodissociation.

III. FRACTAL SINGULARITIES IN PHOTODISSOCIATION DYNAMICS

One of the results of the theory of chaotic scattering is that the time delay function giving the sojourn time of the system in the scattering region shows fractal singularities as a function of an input variable [21]. When the singularities are finite, the scattering is called regular. By analogy, here we consider the photodissociation duration $T_d(x)$discussed in Sec. II, and ask whether there appear singularities, and whether we can determine the nature of these singularities.

To answer this question we calculated $T_d(x)$ for various values of the frequency ω and for the energies of the first three vibrational levels. A typical form of this function for the first excited level is shown in Fig. 3(a). A similar structure holds for $T_d(x)$ of the ground and second excited levels. We chose to analyze the first excited level instead of the ground level in order to reduce the computational time. Of course, the results and conclusions apply to the other two levels as well.

The existence of fractal singularities is obvious, while magnification of a region with width $\delta x\cong10^{-4}$ [Fig. 3(b)] shows that this situation persists in smaller scales. In fact, when magnification is performed, there is an increase in the number of singularities. As we discuss below, this is associated with the fact that the dynamics is nonhyperbolic.

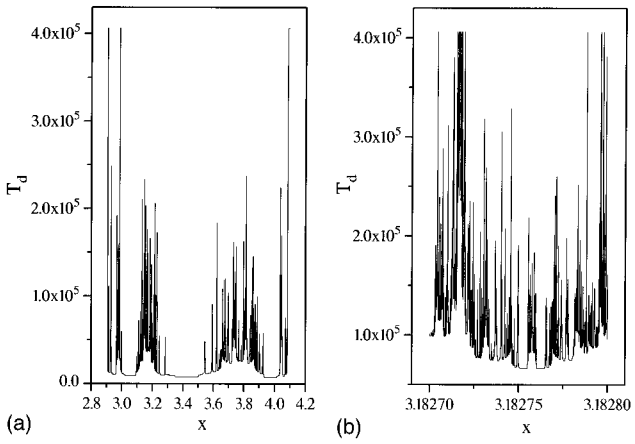

FIG. 3. (a) The duration of the photodissociation T_d as a function of the internuclear distance X when initial state is the first excited one $F=2\times10^{-3}$ a.u. and $\omega=3.9\times10^{-3}$ a.u. (b) Magnification of the interval (3.1827, 3.1828) of $T_d(x)$ of (a).

In the theory of chaotic scattering, a distinction is made between *hyperbolic* and *nonhyperbolic* chaotic scattering (Ref. [23] and references therein). The first appears when in the scattering region all the periodic trajectories are unstable and there are no Kolmogorov-Arnold-Moser (KAM) tori, while the second appears when stable periodic orbits do exist, so that the phase space of the system is simultaneously occupied by chaos and by islands of stability. In the case of hyperbolic chaotic scattering, the number of trajectories that remain in the scattering region is reduced exponentially with time and the fractal dimension d_f of the singularities in the delay function is $0<d_f<1$. Conversely, in the nonhyperbolic case, the number of bound orbits is reduced as a power law due to the sticking effect of the KAM tori. Furthermore, Lau, Finn, and Ott [23] proposed that the fractal dimension of the singularities is always 1, even though that the Lebesque measure remains zero.

In the problem of MPD which we study, the dynamics is nonhyperbolic, a conclusion which follows directly from the Poincaré surfaces of section for our system. Therefore, according to [23] the fractal dimension of the totality of the singularities of $T_d(x)$ should be equal to 1. It is this property which we decided to compute, and show whether the conjecture of Lau, Finn, and Ott [23] indeed holds for a physical process of a real system.

The dimension which should be evaluated is the ''uncertainty dimension'' d [23]. This quantity is given by $d=1-\beta$, where β appears as $f(\varepsilon)\approx\varepsilon^\beta$. $f(\varepsilon)$ is the percentage of uncertain points, for a given value of uncertainty, ε, and for randomly chosen points x_0. Each x_0 is considered to be uncertain if we find that the difference $|T_d(x_0)-T_d(x_0+\varepsilon)|$ is larger than a number of order 1. In our calculation we included as many random points as necessary to obtain 100 uncertain points per run. According to [23], the uncertainty dimension, whose computation is more economical than that of the ''*box-counting*'' dimension [23], will have the value of one only when it is computed for very small E, that is $d\rightarrow1$, as $\varepsilon\rightarrow0$. In practice, this implies that we must compute d for regions where ε becomes smaller and smaller, and check that d tends to 1 from below. Indeed, this is what is observed in our problem. In Fig. 4(a), the dependence of $f(\varepsilon)$

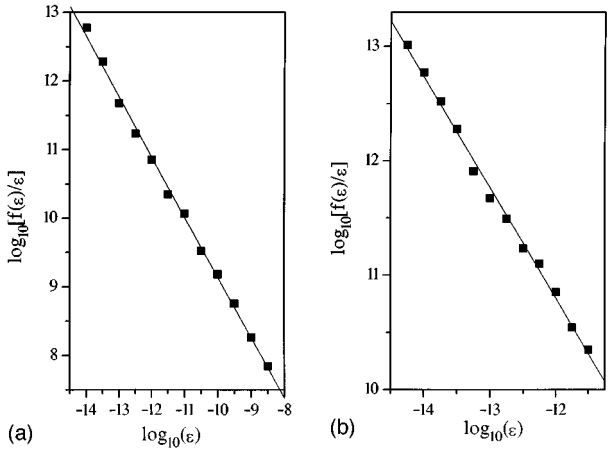

FIG. 4. (a) Evaluation of the uncertainty dimension of $T_d(x)$ of Fig. 3(b) for $10^{-8} \leqslant \varepsilon \leqslant 10^{-14}$. (b) The same as (a) but for $10^{-11.5} \leqslant \varepsilon \leqslant 10^{-14.25}$.

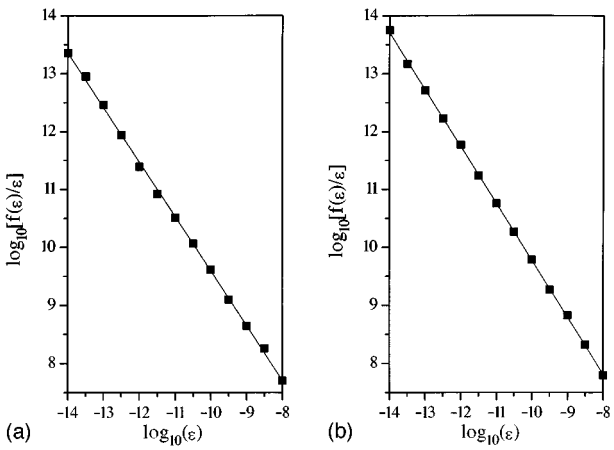

FIG. 5. (a) Evaluation of the uncertainty dimension of $T_d(x)$ when $3.1817 \leqslant x \leqslant 3.1828$ and $\omega = 4.04 \times 10^{-3}$ a.u. (b) The same as (a) but for $\omega = 4.23 \times 10^{-3}$ a.u.

on ε is plotted for $10^{-8} \leqslant \varepsilon \leqslant 10^{-14}$and $3.1827 \leqslant x \leqslant 3.1828$ a.u. From the slope we obtain $d \cong 0.87$. If, however, we restrict ourselves to the region $10^{11.5} \leqslant \varepsilon \leqslant 10^{14.25}$ for the same region of x, d becomes 0.97 [Fig. 4(b)]. Therefore, we can safely argue that the fractal dimension of the MPD process is consistent with its nonhyperbolic dynamics since, in the limit $\varepsilon \to 0$, d becomes equal to 1.

We now turn to another aspect of the results of Lau, Finn, and Ott [23], and see how they carry over to the problem of MPD. Lau, Finn, and Ott [23] gave a simple example of a Cantor set with zero Lebesque measure and fractal dimension $d=1$. In the general case, this set is constructed by successive subtractions from the interval [0,1] of parts which, at the nth stage of the process, have length

$$\eta_n = \frac{1}{n+c}, \tag{5}$$

where a and c are constants. The number of the remaining intervals N_r as a function of the length ε (of each one), is given by [23], Eq. (1), in the limits $\varepsilon \to 0$ (and $n \to \infty$),

$$N_r \approx \frac{1}{\varepsilon}\left[\ln\frac{1}{\varepsilon}\right]^{-a}. \tag{6}$$

Therefore, the dimension that is given by the slope of the curve $\ln N_r$ vs $\ln(1/\varepsilon)$,

$$\frac{d(\ln N_r)}{d\left(\ln\frac{1}{\varepsilon}\right)} = 1 - \frac{a}{\ln\frac{1}{\varepsilon}}, \tag{7}$$

is, for finite ε, always smaller than 1. The rate of approach of d to unity as $\varepsilon \to 0$ is proportional to the constant a. It is this dependence that we wish to verify for our problem.

Equation (5) shows that the constant α expresses the rate with which the interval [0,1] is emptied, and therefore, the corresponding quantity in the physics of dissociation is the CDR. Hence we expect that, as the CDR becomes smaller, the convergence of d toward 1 will appear faster. Indeed, this is verified if we compute the percentage of uncertain points, $f(\varepsilon)$, as a function of ε, for $\omega = 4.04 \times 10^{-3}$ and 4.23×10^{-3} a.u. [Figs. 5(a) and 5(b)], that is for frequencies larger than the optimal ${}^1\omega^*(=3.94 \times 10^{-3}$ a.u.). This calculation was done for the same interval of values of x and of ε as in Fig. 4(a). For $\omega = 4.04 \times 10^{-3}$ a.u., the resulting dimension is $d \cong 0.94$, while for $\omega = 4.23 \times 10^{-3}$ a.u., $d \cong 0.98$. In other words, as the CDR is reduced, the uncertainty dimension converges to 1 faster. Nevertheless, this occurs only for frequencies larger than ${}^1\omega^*$. For frequencies smaller than ${}^1\omega^*$, there appears to be some other mechanism of reduction of CDR which is not related to the structure of the fractal set. The explanation of this asymmetry in the mechanisms of reduction of CDR, as frequencies become smaller or larger than ω^*, is the object of Sec. IV.

Finally, we carried out calculations for various field strengths in the range 2×10^{-3}–4.5×10^{-3}a.u. As the highest value is approached, the fractal structure of the singularities is quickly replaced by only a small number of them (Fig. 6). Thus, by associating the appearance of fractal singularities in $T_d(x)$ with *chaotic photodissociation* and the singularities of finite number with *regular*, we observe that a change in the field strength causes a transition from regular to chaotic MPD. For smaller intensities, for which the decay rate is slower and the system can afford to spend more time in the inner part of the scattering region, chaotic behavior

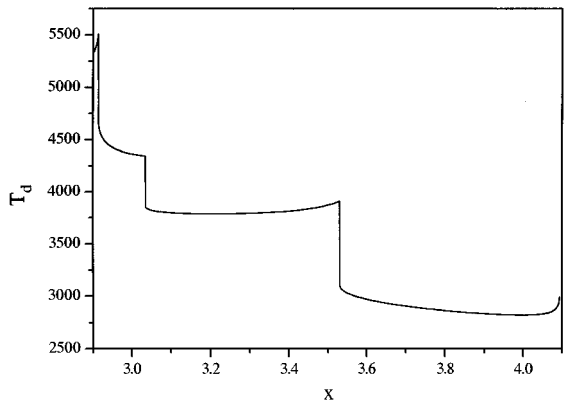

FIG. 6. The same as Fig. 3(a) but with larger field strength: $F = 4.5 \times 10^{-3}$ a.u.

appears. For larger intensities, the kinetic energy of the fragments is increased rapidly, while the $T_d(x)$ of the trajectories decrease analogously, thereby destroying the fractal structure. It is our conjecture that this transition to regular photodissociation reflects the emergence of above threshold dissociation (e.g., Ref. [27]), where, however, the increase in kinetic energy of the fragments is quantized.

IV. FRACTAL SINGULARITIES AND CDR

Among the results of the theory of chaotic scattering is that the fractal set of singularities that appear in the scattering and time-delay functions is caused by the intersection of the one-dimensional set of initial conditions with the stable manifolds of the uncountable unstable periodic orbits which are found in the scattering region [16]. The uncertain points which we counted in Sec. III for the determination of the fractal dimension are on this intersection, that is they are very near to the stable manifold of a periodic orbit. This is why they lead to orbits that are trapped for a long time in the scattering region. Therefore, in a scattering problem the trajectories can be divided into those that scatter without delay and into those that scatter with delay, because of the fact that they start from points very close to the stable manifold of a periodic orbit.

The photodissociation of a molecule, or the ionization, occurs from the chaotic regions of phase space, since a regular orbit always moves on the surface of a torus and cannot lead to fragmentation. It is therefore useful to look at the chaotic scattering characteristics of MPD in terms of the three categories of the N initial conditions that simulate classically an eigenstate. Category (i) contains the orbits that belong to KAM tori, and lead to quasiperiodic orbits that do not move away from the region of the potential (KAM orbits). Category (ii) contains the orbits that quickly move away through the chaotic region of phase space (short-lived orbits). Category (iii) contains the orbits that are long lived due to the reasons given in the first paragraph.

Given these three categories, we would like to see whether the decrease of the CDR away from ω^* is associated with the existence of orbits of categories (i) and (iii). Indeed, we will show that for values of ω smaller than ω^* the number of KAM orbits increases, while, for ω larger than ω^*, the number of long-lived orbits increases.

Let $N_{\mathrm{KAM}}(\omega)$ and $N_{\mathrm{LL}}(\omega)$ represent the number of KAM and long-lived orbits, respectively, for frequency ω. Figure 7 shows the case where $N=5000$ and $E=E_1$, where E_1 is the energy of the first excited level. We consider as long-lived orbits those corresponding to initial conditions for which $\varepsilon=10^{-10}$ makes them uncertain points. Calculations with different ε and N do not change the qualitative behavior of long lived (ω). Similar curves result for $E=E_0$ and $E=E_2$ as well. From Fig. 7 we conclude that the frequency ω^* which leads to maximum CDR, ($^1\omega^*_{\mathrm{cl}}=3.94\times10^{-3}$ a.u.), is at the edge of two different mechanisms of reduction of CDR. On the one hand, as the frequency is decreased to values smaller than ω^*, the CDR decreases due to the increase of KAM orbits (N_{LL} is negligible). This means that, as ω is reduced, an increasing part of the set of initial conditions simulating the quantum state enters the KAM region, where it is trapped. On the other hand, when $\omega>\omega^*$ the reduction of the

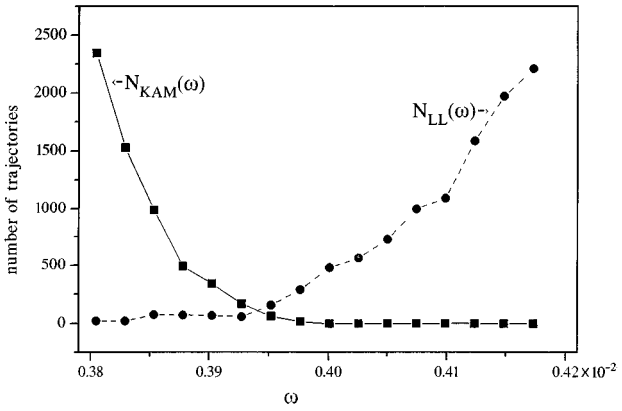

FIG. 7. Number of trajectories as a function of frequency ω, MKAM(ω) (solid line) and MLL(ω) (dash-dotted line), for the first excited state and field strength $F=2\times10^{-3}$ a.u. The optimal ω^* (3.94×10^{-3} a.u.) is a direct consequence of the behavior of the two curves (see text).

CDR is caused by the increase of N_{LL}. That is, in this case the region of phase space containing unstable periodic orbits is increased, so that the number of intersections of their stable manifolds with the initial classical state is also increased. In fact, this increase in the identity of intersections is accompanied by an increase of the duration for dissociation of the short-lived orbits, which also contributes to the decrease of CDR. The aforementioned two causes of reduction of the CDR influence the function $N_b(t)$ that gives the number of orbits that have not dissociated at time t. For $\omega<\omega^*$, $N_b(t)$ decreases rather rapidly until a certain value which subsequently remains constant. This value is the number of KAM orbits. For $\omega>\omega^*$, $N_b(t)$ decreases continuously at a slow rate.

The behavior of $N_{\mathrm{KAM}}(\omega)$ and $N_{\mathrm{LL}}(\omega)$ is similar for the ground and the second excited states. Thus, in all cases the value of ω^* is found when the number of KAM orbits becomes very small, and the number of LL orbits starts increasing.

V. SYNOPSIS

In this work we implemented techniques and results of classical nonlinear dynamics, and in particular of the theory of chaotic scattering, to the analysis of MPD, using as the testing ground the molecule BeH^{2+} for which quantum-mechanical calculations were recently published. The calculations and analysis were done not only for the ground vibrational level but also for the first two excited ones. Comparing the classical results with the quantal ones—which concerned the ground level—the findings were the same for two things: First, for infrared frequencies tuned around the resonance frequency of the $v=0\rightarrow v=1$ transition, there is an optimal frequency ω^* for which the dissociation rate is maximized. Second, the value of this ω^* is about 90% of the resonance frequency. An interesting question for future research is whether quantum-mechanical calculations confirm the classical predictions herein for the excited level as well and, in general, how the issue of optimizing the MPD rate with single-frequency infrared laser is revealed for each level and for different anharmonicities in the molecular potential. We

note that, given the results of Ref. [8] and [13] (Fig. 1), we adopted a uniform distribution as our initial conditions, a condition which is more suitable for the analysis of fractal structure. Since the basic conclusions are the same for the ground as well as the first and second excited states, it follows that the results in the present context are essentially independent of the initial energy and of the detailed structure of the initial-state distribution function.

Given that the frequency redshift phenomenon for maximizing the MPD rate has been found in the results of calculations on the ground vibrational level of other diatomic molecules as well, a fact implying some type of universality based on the existence of anharmonicity and on the continuous spectrum, we considered the possible connection of the CDR to the current theory of chaotic scattering: By analyzing the function of the duration of dissociation, $T_d(x)$, computed for a large collection of trajectories, we determined that as a function of field strength there is what we termed chaotic as well as regular MPD. In chaotic MPD there are singularities in $T_d(x)$ of fractal structure. This finding was made more basic and quantitative by showing that the fractal dimension for the MPD process is 1, thereby verifying the conjecture of Lau, Finn, and Ott [23] that this must be a characteristic of systems exhibiting nonhyperbolic scattering. In this context we pointed out that the CDR is related to the rate of convergence of the uncertainty dimension toward 1, as the uncertainty ε goes to zero.

Furthermore, the discovery of such fractal singularities was utilized for the implementation of the computationally observed optimal frequencies ω^*: When ω is increased, the collection of orbits simulating the quantum state leaves the region of KAM tori, and the CDR increases. However, as soon as the orbits exit completely, they start to intersect the stable manifolds of the unstable periodic orbits, and so the CDR stops increasing and starts decreasing. The frequency ω^* yielding the maximum CDR is the one for which the classical state has just excited the KAM region and has not managed yet to enter the region where the stable manifolds of the periodic orbits are sufficiently many.

Finally, there is the interesting finding of the transition as a function of the field intensity from conditions of chaotic MPD—for low intensities—to conditions of regular MPD, as revealed by the nature of the singularities in $T_d(x)$. This finding might be worth analyzing to determine whether it can be connected to an abrupt bifurcation [21] or to a saddle-center bifurcation [22]. In addition, further analysis might shed light on our conjecture that the appearance of regular MPD as the laser intensity is increased could be connected to the observed phenomenon of above threshold dissociation, which emerges naturally from quantum-mechanical calculations (e.g., [27]).

ACKNOWLEDGMENT

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