Scaling of the average survival probability for low dimensional systems

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Received 15 October 1998

Abstract

Recent conjectures put forward by Schofield and Wolynes regarding the scaling behaviour of averaged survival probabilities are tested in an extreme limit. The system of choice is the quasi 3-dimensional Baggot’s spectroscopic Hamiltonian for H$_2$O. Despite the low density of states for the system, the survival probability when averaged over the states of a given polyad shows the expected scaling with the resonance coupling. Power law behaviour, for intermediate times, is observed for both classically integrable and non-integrable systems. The power law behaviour persists even when the accompanying fluctuations, possibly due to quantum twinkling in the system, are very large. However, among the integrable subsystems of the full Hamiltonian the 1:1 resonance case exhibits an anomalously large exponent. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

Intramolecular vibrational energy redistribution (IVR) in polyatomic molecules has been a focal point for experimentalists and theorists alike in order to understand the complexities of chemical reaction dynamics [1–3]. This fundamental problem impacts the very fabric of reaction dynamics since IVR is inevitably linked to the rates of chemical reactions. In this sense then, the aim of understanding (and perhaps even controlling) chemical reactions will be partially achieved if one can understand the important features of energy flow in molecules [3,4].

The non-trivial nature of the problem, theoretically, is clear from the kind of deep questions that emerge in the course of attempting to address the central problem in a direct fashion. For example, the various classical structures (transport in phase space due to resonances in particular) and their quantum incarnations play a significant role in the phenomenon of IVR. The fact that our understanding of classical–quantum correspondence in systems with three or more degrees of freedom (most of the molecules of interest!) is incomplete makes this a hard problem to tackle. Nevertheless, quite a few approximate theories have been proposed in the literature which directly or indirectly take on the issue of IVR [2,3,5]. A crucial lesson learnt from a number of studies on various molecules and model Hamiltonians is that the specific nature of mode couplings is very important in understanding the phenomenon of IVR [6–10].

It is apparent that one can adopt two different strategies to study the flow of energy in molecules.

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One is to study specific molecules in detail and the other is much less detailed but aims to uncover the generic features in a large class of molecules. The former approach is best suited for small molecules since the detailed dynamical and spectroscopic signatures of IVR require solutions to Hamilton and Schrödinger equations for relatively longer times. In the latter category, to which the present work is a contribution, the size of the molecules is not a factor since the objective is to construct models which capture enough of the underlying physics in order to be able to detect possible universal features. Thus, the two approaches usefully complement each other with the exciting possibility of an appropriately sized molecule providing deep insights from this dual perspective.

In a large number of experimental [1] and theoretical studies [2] of IVR, the survival probability $P(t)$ of a zeroth-order state is the central object of interest. One of the reasons for this has to do with the fact that $P(t)$ is directly related to the fourier transform of the spectrum $\mathcal{F}(\omega)$. On the theoretical side, considerable efforts have been devoted towards studying the behaviour of suitably averaged survival probabilities with time and uncovering universal features. Pechukas [11] and others [12,13] showed that $P(t)$ averaged over both Hamiltonian ensembles and initial wavefunction conditions exhibits a difference for all times between systems with underlying regular and irregular classical dynamics. Pavlov–Verevkin and Lorquet [14] have used ideas from an earlier work of Heller [15] to obtain, in the limit of long times, an expression for the number of phase space cells sampled as a function of time. They observed different asymptotics depending on the nature of the classical dynamics, i.e. regular or chaotic. More recently, Lakshminarayanan [16] has studied the quasiperiodic fluctuations associated with the correlation between two density operators in the long time limit. It was shown that the variance associated with the fluctuations show universal scaling with $\hbar$ when the underlying classical dynamics is completely chaotic.

In this Letter, we are interested in the theory of dynamical scalings for averaged $P(t)$ suggested recently by Schofield and Wolynes [17]. The scalings proposed are based on similarities between the problem of energy flow in molecules and the phenomenon of Anderson localization [18]. Earlier studies on the mechanism of energy flow via Fermi resonances [19] pointed to the importance of the local couplings between zeroth-order quantum states. From a classical standpoint, it is the resonances between the various zeroth-order modes which lead to energy flow through the entire molecule. The parallels between models with resonant interactions and the Anderson model point towards possible eigenstate localization or delocalization as some parameters in the Hamiltonian are varied. Exploiting the analogy, Schofield and Wolynes [17] showed that on the delocalized side (diffusional scaling)

$$\langle P(t) \rangle \sim (Dt)^{-\gamma},$$  \hspace{1cm} (1.1)

where $D$ is a diffusion constant, $\gamma \equiv d_s/\alpha$ with $d_s$ being some effective dimensionality ($= s - 1$, with $s$ being the number of oscillators) and $\alpha$ is the degree of the random walk in the quantum number space. The averaging is performed over a group of states sharing the same number of total quanta among the various modes (polyad). On the other hand, near the localization transition (critical scaling)

$$\langle P(t) \rangle \sim (\omega t)^{-\frac{1}{2}},$$  \hspace{1cm} (1.2)

where $\omega$ is a microscopic frequency depending on the local density of states. The results are appropriate only for times less than the Poincaré recurrence time for the system which arises due to the finiteness of the quantum state space. One of the interesting observations, apart from the universality, is the prediction of a linear, as opposed to the quadratic golden rule, dependence of the decay rate on the couplings. These results have been seen in different systems before [3,20] and were confirmed for a highly degenerate 6-dimensional resonantly coupled morse oscillator system [21]. Bigwood and Gruebele [20] have systematically studied large systems and observed a wide range of behaviours for the survival probability with deviations from exponential decay and/or golden rule predictions. The role of higher-order anharmonic couplings, in order to understand IVR for long times and large density of states, has also been emphasized recently by Pearman and Gruebele [10].

In this work, we test the scaling ideas of Schofield and Wolynes [17] in an extreme limit. As originally proposed, the scaling results are to be expected for
systems whose underlying classical dynamics is completely chaotic and have high quantum density of states. However, the arguments used seem mostly "geometric" in nature and it is natural to ask if the scalings would manifest themselves even in systems where the density of states is small with the corresponding classical dynamics being sufficiently chaotic. In order to explore the limits, a quasi 3-dimensional spectroscopic Hamiltonian for H₂O is chosen [22] for which very detailed classical phase space structure and classical–quantum correspondence are known [23]. This provides us with the opportunity to correlate various classical phase space structures with the behaviour of \( \langle P(t) \rangle \). Moreover, the Hamiltonian used in the current studies is a fairly realistic one for water, although it will be extended beyond the limit of strict applicability as far as this work is concerned. This is done in order to gain insight about the scaling of \( \langle P(t) \rangle \) in various limits which also might be indicative of a class of molecules. It is shown that even for this small system the \( \langle P(t) \rangle \) exhibit a universal power law behaviour for intermediate times. Various integrable subsystems are also studied which highlights a particular integrable limit demonstrating anomalous behaviour.

In Section 2, the Baggot spectroscopic Hamiltonian for H₂O is described. The computed average survival probabilities are presented and discussed in Section 3. The Section 4 concludes with a summary and future directions.

2. Model Hamiltonian

The quantum spectroscopic Hamiltonian due to Baggot [22] is a three degree of freedom local-mode/bend Hamiltonian

\[
H = H_0 + H^{1:1} + H^{2:2} + \sum_{\sigma=1,2} H^{2:1}_\sigma ,
\]

where \( H_0 \) is the zeroth-order Hamiltonian

\[
H_0 = \sum_{\sigma=1,2} \left[ \Omega_\sigma n_\sigma + \alpha_\sigma n_\sigma (n_\sigma + 1) + \varepsilon_{\sigma b} \left( n_\sigma \left( n_\sigma + \frac{1}{2} \right) + \frac{1}{2} n_\sigma \right) + \Omega_b n_b + \alpha_b n_b (n_b + 1) + \varepsilon_{\sigma a} (n_\sigma n_\sigma + \frac{1}{2} (n_\sigma + n_\sigma)) \right],
\]

and the resonant interaction terms are

\[
H^{1:1} = \beta_{12} \left( a^*_1 a_2 + a^*_2 a_1 \right), \\
H^{2:2} = \beta_{22} \left( a^*_1 a_1 a_2 a_2 + a^*_2 a_2 a_1 a_1 \right), \\
H^{2:1}_r = \beta_{rb} \left( a^*_b a_0 a_b + a_0 a^*_b a_b \right).
\]

The operators \( n_1, a^*_1 \) and \( a_1 \) represent the number, creation and destruction operators for the two local mode stretches (\( \sigma = 1,2 \)) and the bend (\( b \)), respectively. In the actual fit [22], the 1:1 resonance coefficient included a vibrational-level-dependent interbond coupling which we chose to ignore in this work. The results obtained are very similar if the extra term is included. In addition, the 2:2 resonance term will not be considered as far as this Letter is concerned. Despite the presence of the resonant couplings, there is a linear combination of the number operators known as the superpolyad number \( P = n_1 + n_2 + n_b/2 \) which commutes with the Hamiltonian.

Thus, \( P \) is a good quantum number and the system has effectively two degrees of freedom. As a result, the quantum Hamiltonian is block diagonal in \( P \) and for a given \( P \) there are \( N_P \equiv (P+1)(P+2)/2 \) states. The classical–quantum correspondence for this system has been studied in detail in an earlier work [23].

The survival probability of a zeroth-order state \( |n_1,n_2,n_b \rangle \equiv |n \rangle \) is defined in the usual fashion as

\[
P(n,t) = |\langle n | \exp(-iHt/\hbar) | n \rangle|^2 = \sum_{\alpha,\beta} I_{\alpha} I_{\beta} \cos(\omega_{\alpha,\beta} t),
\]

where \( \alpha, \beta \) refer to the eigenstates of the Hamiltonian, \( I_{\alpha} \equiv |\langle n | \alpha \rangle|^2 \) are the spectral intensities and \( \omega_{\alpha,\beta} \equiv (E_{\alpha} - E_{\beta})/\hbar \). The average is computed using a simple ‘microcanonical’ weighting

\[
\langle P(t) \rangle = \frac{1}{N_P} \sum_{n \in P} P(n,t).
\]

In Section 3, we will compute \( \langle P(t) \rangle \) for various values of the parameters. One class of parameters corresponds to the usual scenario wherein the Chirikov analysis [24] of the classical resonances holds. In this case, it is already known [23] that the quantum eigenstates organize themselves around the classical resonance zones. For the other class of
Table 1
The two classes of parameters used in this Letter

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Class A (cm⁻¹)</th>
<th>Class B (cm⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Omega_x )</td>
<td>3885.57</td>
<td>3885.57</td>
</tr>
<tr>
<td>( \Omega_y )</td>
<td>1651.72</td>
<td>1951.72</td>
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<tr>
<td>( \alpha_x )</td>
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<td>-1.91</td>
</tr>
<tr>
<td>( \alpha_y )</td>
<td>-1.91</td>
<td>-1.17</td>
</tr>
<tr>
<td>( \varepsilon_{xy} )</td>
<td>-12.17</td>
<td>-1.17</td>
</tr>
<tr>
<td>( \varepsilon_{yb} )</td>
<td>-19.12</td>
<td>-1.12</td>
</tr>
</tbody>
</table>

parameters, the Chirikov approximation fails and the quantum states bear no resemblance to the states in the former class. Although only these two-parameter cases are dealt with in this work, similar results were seen for almost any choice of parameters and the superpolyad number \( P \) for the three-mode system. A particular integrable subsystem is obtained by turning off the rest of the resonance strengths. In Table 1, the two classes of parameters are indicated with class B being a system having a weakly anharmonic \( HH \). The reduced anharmonicity of the bend mode invalidates a Chirikov-type analysis in this case. Table 2 provides information about the mean level spacings \( \langle \Delta E \rangle \) and time \( \tau_0 = \pi \hbar \langle \Delta E \rangle^{-1} \) in units of the average harmonic time period at which the quantum dynamics become sensitive to individual pairs of energy levels as a function of the various resonance parameters.

3. Results and discussion

In the results presented in this section, the value of the superpolyad number is fixed at \( P = 16 \) which corresponds to a total of \( N_p = 153 \) states. As shown in Table 2, the density of states can vary widely depending on the choice of \( \mathcal{H}_0 \) and the various resonance parameters. The survival probabilities of each of the individual states are calculated to a time less then \( \tau_0 \) and averaged over the entire polyad to obtain \( \langle P(t) \rangle \). All of the figures concerning \( \langle P(t) \rangle \) are shown on a \( \log \) scale. In addition, results for both case A and case B are shown together in the same figure.

3.1. Non-integrable case

In Fig. 1, the \( \langle P(t) \rangle \) is shown for the classically non-integrable case of two 2:1 resonances for increasing values of \( \beta_{ib} \). It is immediately evident from Fig. 1 that after a critical value for the coupling constant \( \beta_{ib} \), the slope of the linear portion in the figure settles down to a constant value. The value of this slope is close to \( \gamma = -0.95 \) which is in accordance with the scaling picture as there are three oscillators and a constant polyad leading to an effective dimensionality of two (assuming a random walk which is uncorrelated). A power law fit with the

\[ \ln(P(t)) = \ln(\beta_{ib}/\hbar) \]

Fig. 1. Variation of \( \langle P(t) \rangle \) with \( \beta_{ib}/\hbar \) for increasing values of \( \beta_{ib} \) in the non-integrable case. Results for case A and case B (26.57 cm⁻¹ (solid), 226.57 cm⁻¹ (long dashed), 326.57 cm⁻¹ (short dashed)) are superimposed. The symbols correspond to case A with \( \beta_{ib} = 26.57 \) cm⁻¹. The straight line fit indicated with * has a slope of \(-0.95\).
value of $\eta$ close to $-1$ provides agreement for longer times than an exponential fit, as has been noted before [21,29]. Moreover, the decay rate extracted from an exponential fit, i.e. obtained as the inverse of the time required for $\langle P(t) \rangle$ to decay to $1/e$, agrees well with the expression $D = 14\beta_{\Delta b}/\hbar$ as shown in Fig. 2. The universality of the power law behaviour is clear from the fact that all the curves in Fig. 1, irrespective of the nature of $\mathcal{H}_0$ for $\beta_{\Delta b} > \beta_{\Delta b}^c$, can be fit with the same straight line. Although not shown here, it was seen that the variance of the averaged survival probability tracks $P(t)$ itself and attains a fairly constant value, scaling as $N_0^{1/2}$, for times $\tau$ such that $\beta_{\Delta b} \tau \sim 1$. Notice that the linear scaling of $P(t)$ in the figure approximately holds until $t \sim \tau$. However, the root mean square relative fluctuations for the system under study exceed unity and becomes roughly constant for long times. This is in contrast to the observations in an earlier work [21]. Interestingly, the higher moments of the average survival probability, for larger coupling strengths, defined by

$$M^{(k)} = \frac{\langle P^{(k)}(t) \rangle}{\langle P(t) \rangle},$$

(3.1)

become very large with $k$ at $t = \tau$ despite a steady decrease in $P^{(k)} \equiv \langle P^{(k)}(t) - \langle P(t) \rangle^2$. In Fig. 3, the moments for $k = 2,3,4$ are shown for case A as a function of the resonance coupling strength for $t = \tau$. Two important observations are the initial steep increase in the moments and the levelling off for $\beta > \beta_{\Delta b}^c$. The variance of $\langle P(t) \rangle$ can be written as

$$P^{(2)} = \sum_{\alpha,\beta,\mu,\nu} \mathcal{C}_{\alpha\beta,\mu,\nu} \cos(\omega_{\alpha}\mu t) \cos(\omega_{\mu,\nu} t),$$

(3.2)

where $\mathcal{C}_{\alpha\beta,\mu,\nu} \equiv \langle I_{n\alpha} I_{n\beta} I_{n\mu} I_{n\nu} \rangle - \langle I_{n\alpha} I_{n\beta} \rangle \langle I_{n\mu} I_{n\nu} \rangle$ can be identified as a correlation function involving the spectral intensities $I_{n\alpha}$ in a pairwise fashion. The element $\mathcal{C}_{\alpha\beta,\mu,\nu}$ is clearly related to the correlation between the inverse participation ratios (IPR) [25] of the two eigenstates labelled by $\alpha$ and $\mu$. Preliminary results clearly indicate that even though $\mathcal{C}_{\alpha\beta,\mu,\nu}$ itself is very small (of the order of $10^{-5}$), the quantity $\langle I_{n\alpha}^2 I_{n\beta}^2 \rangle / \langle I_{n\alpha}^2 \rangle \langle I_{n\beta}^2 \rangle$ is very large (of the order of 10). This behaviour is reminis-
cent of a similar phenomenon, involving very strong intensity fluctuations of light, in the short wavelength limit, passing through a random medium, which is called as twinkling [26]. In an earlier work, Berry et al. [27] studied the variations in probability density $|\psi(q)|^2$ across configuration space of a given quantum state, in the semiclassical limit, as embodied in the higher moments of $|\psi(q)|^2$. In the semiclassical limit, it was shown that for $\psi$ corresponding to regular classical dynamics, the higher moments diverge whereas for states corresponding to chaotic classical dynamics the moments converge to some constant value. Note that the spectral intensities in our case can be interpreted as the variation of a given eigenstate over the zeroth-order basis in order to make contact with the earlier [27] analysis. Thus, the large moments being observed here could be the manifestation of quantum twinkling [28]. For the non-integrable system the moments do saturate to some large value with increasing strength of the resonant interactions as evident from Fig. 3.

It was also observed that even for very high resonance coupling values, the asymptotic value, i.e. for $t < t_r$, of $\langle P(t) \rangle$ is quite high compared to what had been predicted earlier [11]. Computation of the inverse participation ratios for increasing $\beta_{s\theta}$ indicate the delocalized nature of almost all of the eigenstates for high values of the coupling. The infinite time average of $\langle P(t) \rangle$ is the IPR averaged over the eigenstates and one cannot expect it to distinguish between regular and irregular regimes [11]. However, the average IPR is an indication of the number of states, on the average, over which the eigenstates are delocalized. For the Baggot Hamiltonian, the average IPR in the large coupling limit saturates to a value of around 0.034 irrespective of the nature of $\mathcal{H}_0$. This is larger than one would expect if the eigenstates were delocalized over the entire basis of 153 states. The $\langle P(t) \rangle$ itself oscillates around the average IPR value.

In the analysis by Schofield, Wyatt and Wolynes [21], the states over which the survival probability was averaged were roughly of the same energy. Thus, the diffusional scaling observed corresponded well with the picture of action diffusion constrained to constant total energy. In our case, especially for the class A parameters, the states in the polyad are of varying energies. However, the exact constancy of

The superpolyad number ensures that one is looking at action diffusion constrained to constant $P$. In this sense, the random walk in action space, beyond $\beta_{s\theta}$, again has an effective dimensionality of $d_s = s - 1$ with $s$ being the number of oscillators being coupled. It is precisely for this reason that the density of states being low for the Baggot Hamiltonian is not a factor in the observed scaling of $\langle P(t) \rangle$. A possible generalization of the arguments above can now be stated. If one computes $\langle P(t) \rangle$ for a group of zeroth-order states corresponding to a system consisting of $s$ oscillators then the expected maximum value of the exponent is $\gamma = (s - l)/\alpha$. Here, $l$ is the total number of constraints on the group of states. The constraints themselves could be due to constant energy, polyad or other constants of the motion which survive at those specific values of the coupling strengths. Note that the ‘geometry’ is in the factor $(s - l)$ whereas signatures of ‘dynamics’ could manifest in the factor $\alpha$.

3.2. Integrable cases

In Fig. 4, the results for the classically integrable single 2:1 resonance are shown. The important observation is that a universal power law behaviour is again evident (until $t \sim \tau$) after a critical coupling constant. However, the accompanying fluctuations

![Fig. 4. Variation of $\langle P(t) \rangle$ with $\beta_{s\theta}t/\hbar$ for the integrable 2:1 case. The symbols correspond to case A with $\beta_{s\theta} = 26.57 \text{ cm}^{-1}$. The straight line fit indicated with $\ast$ has a slope of $-0.65$.](image-url)
are rather large compared to the non-integrable case discussed before. The value of the slope is now close to $\gamma = -0.65$. This value corresponds to an effective dimensionality of larger than one but certainly much less than two. A simple dimensional counting would lead to $\gamma \sim -0.5$ which arises from the fact that there are two oscillators ($s = 2$) and one constraint of constant $P$. The IPR value, averaged over the eigenstates, saturates around 0.2 indicating the localized character of the states.

Based on the above observation it is fair to expect any integrable subsystem of the Baggot Hamiltonian to show similar behaviour. In Fig. 5, the $\langle P(t) \rangle$ integrable 1:1 resonance is shown. The qualitative features are the same, however with one important exception – the constant slope has the value of about $\gamma = -0.95$. This anomalous result is seen only for the 1:1 resonance situation whereas the integrable 2:2 subsystem again shows a slope closer to the integrable 2:1 system. Thus, the 1:1 integrable system has an exponent $\gamma$ which is almost two times larger than one would expect for a classically integrable system. The average IPR value, perhaps not surprisingly, saturates to a value close to 0.17 which is comparable to the integrable 2:1 case. It is of interest to note that anomalously low exponents have been seen by Gruebele [29] which, perhaps, could be due to localization effects. For the present 1:1 integrable situation an anomalously high exponent is observed despite the average IPR being reasonably high. One possibility is that the nature of the random walk, as characterized by $\alpha$ with the exponent $\gamma = d_1/\alpha$, could be different. The relevant things to explore in this context are the classical correlation functions $\Phi(t) = \langle \xi(0) \xi(t) \rangle / \langle \xi \rangle^2$ for dynamical variables $\xi$. A Lévy flight behaviour for $\Phi(t)$, as has been observed in classical systems before (see articles in Ref. [30]), could explain the anomalous exponent for the 1:1 system, provided the effect survives quantization.

For both the integrable cases, the higher moments $M^{(k)}$ increase with increasing coupling strengths. This is in contrast to the non-integrable case discussed in the previous section where the moments of $\langle P(t) \rangle$ become constant beyond some coupling strength. This observation is in agreement with the work of Berry et al. [27].

4. Conclusions

The results of this Letter clearly show that power law behaviour in the averaged survival probabilities can be observed for low dimensional, sparse density of states resonant Hamiltonians. The power law behaviour extends for times larger than the $1/e$ fall time for $\langle P(t) \rangle$ and up to the time scale of the inverse of the coupling constant. Thus, the universality of power law behaviours at intermediate times does not seem to be restricted to large dimensional systems. The accompanying large fluctuations in $\langle P(t) \rangle$, perhaps due to quantum twinkling, do not seem to alter the power law behaviour. One of the limitations of the present study lies in the fact that the largest exponent that could be expected is $\gamma = -1$. This implies that, from a scaling perspective, the separation of diffusive and critical regimes is not very sharp. This limitation can be removed by breaking the constancy of the superpolyad number. This and a detailed study of the classical–quantum correspondence is in progress. Interestingly, the Poincaré surface of sections even for very high resonance

Fig. 5. Variation of $\langle P(t) \rangle$ with $\beta_{21} t/\hbar$ for the integrable 1:1 case. Results for both case A and B (156.48 cm$^{-1}$ (solid), 256.48 cm$^{-1}$ (long dashed), 356.48 cm$^{-1}$ (short dashed)) are superimposed. The symbols correspond to case A with $\beta_{12} = 56.48$ cm$^{-1}$. The straight line fit indicated with * has a slope of $-0.95$. There is evidence that the effect survives quantization at least in the kicked rotor case. See Ref. [31]
couplings do show some amount of regularity for lower energy values. However, this is not surprising since power law tails have been observed classically, in mixed phase space, for time–time autocorrelation functions of dynamical variables [32–34]. More relevant, perhaps, to the subject of the present investigation is the universal power law dependence of the classical probability $P(t)$ in mixed phase space. $P(t)$ is the probability that an initial point in phase space close to an island remains in the neighbourhood at time $t$. It definitely seems worthwhile to investigate such classical objects for the Baggot Hamiltonian. The anomalous behaviour shown by the integrable 1:1 resonant system is puzzling and the fact that it shows a slope close to the critical scaling limit is disconcerting. Even the integrable 2:1 case does show an effective dimensionality greater than one, although the deviations are not as severe as in the 1:1 subsystem. Thus, as emphasized before [29], it is important to understand the connection between the exponent $\gamma$ and the various molecular properties. Work is in progress to understand these features and the importance of quantum interference, in general, to the scaling picture from semiclassical viewpoints.

Acknowledgements

I am grateful to Greg Ezra for useful discussions regarding this topic and Peter Wolynes for pointing out the possible connections to quantum twinkling.

References