Fano interference and cross-section fluctuations in molecular photodissociation

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We derive an expression for the total photodissociation cross section of a molecule incorporating both direct and indirect processes that proceed through excited resonances, and show that it exhibits generalized Beutler-Fano line shapes. Assuming that the closed system can be modeled by random-matrix theory, we derive the statistical properties of the photodissociation cross section and find that they are significantly affected by the direct processes. In the limit of isolated resonances, we find that direct processes suppress the correlation hole of the cross-section autocorrelation function and lead to a maximum in the cross-section distribution.

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Spectral correlations of closed quantum systems, whose associated classical dynamics are chaotic, are known to be nearly universal and can be modeled by the Gaussian invariant ensembles of random-matrix theory [1–3]. When such systems become open through their coupling to continuum channels, their bound states acquire decay widths and become resonances, but they are still expected to exhibit universal statistics [4]. Examples are the conductance fluctuations in quantum dots [5] and the statistics of the indirect molecular photodissociation cross section [6,7]. If the coupling is weak, the corresponding resonances are isolated and are often characterized by a Lorentzian line shape. For a classically chaotic system, the statistics of these resonances can be derived from random-matrix theory. In recent years, such a random-matrix approach has been successfully used to model the statistics of resonances [4] and cross-section fluctuations in the photodissociation of classically chaotic molecules [8]. A semiclassical treatment was discussed in Refs. [9,10].

However, as first observed by Beutler [11] and later interpreted by Fano [12], the line shape of an individual resonance may differ substantially from a Lorentzian: interference between the indirect decay via the quasibound state and direct (fast) decay to the continuum gives rise to a so-called Beutler-Fano line shape. For real bound-state wave functions, the line shape (versus energy E) is proportional to $(q_n+\varepsilon_n)^2/(1+\varepsilon_n^2)$, where $\varepsilon_n=2(E-E_n)/\Gamma_n$. Here, E_n and Γ_n are, respectively, the energy and width of the nth resonance, while q_n is the Fano parameter characterizing the line shape. Beutler-Fano profiles have been observed in molecular photodissociation [13], autoionization [14], conductance through quantum wells [15] and quantum dots [16], scanning tunneling microscope spectroscopy of surface states [17], semiconductor superlattices [18], Aharonov-Bohm rings [19], and neutron scattering from nuclei (see, for example, Ref. [20]).

In systems that are classically chaotic, the Fano parameter q_n is expected to fluctuate from one resonance to another. Distributions of q_n have been calculated for transmission through a quantum dot [21] and for the photodissociation of molecules [22] in the one-channel case.

Here we derive an expression (2) for the total photodissociation cross section for any number of open channels in the presence of direct decay processes (see Figs. 1 and 2). In the limit of isolated resonances, we show that their line shapes have the form of generalized Beutler-Fano profiles $|q_n+\varepsilon_n|^2/(1+\varepsilon_n^2)$ with a *complex* Fano parameter [21] q_n.

Given that direct photodissociation processes affect the line shapes so dramatically, it is interesting to find out how they affect the statistics of the photodissociation cross section when the closed system is classically chaotic. We derive a closed expression for the cross-section autocorrelation function (in energy) and find that it is universal provided that the excitation process and the continuum coupling are spa-

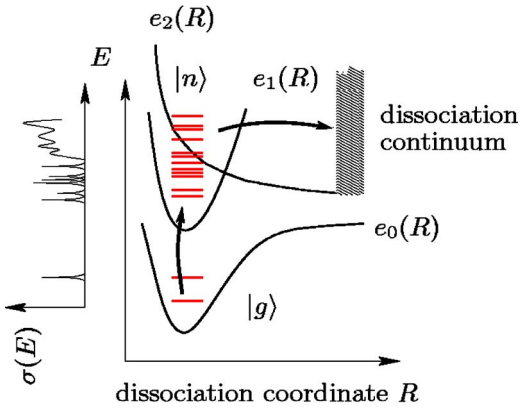

FIG. 1. (Color online) Indirect molecular photodissociation. Shown are three electronic surfaces: the ground-state surface e_0 (in which the ground state $|g\rangle$ resides), a binding surface e_1, and a repulsive surface e_2. The surfaces e_1 and e_2 give rise to an effective electronic surface with a barrier near their crossing (not shown), and the bound vibrational states $|n\rangle$ in e_1 become resonances. Indirect photodissociation proceeds through these resonances with a total cross section $\sigma(E)$ (shown on the left) that is a sum of Lorentzian line shapes.

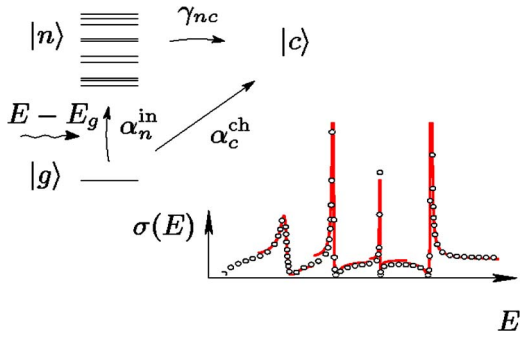

FIG. 2. (Color online) Top: states and transition matrix elements in the random-matrix model discussed in the text. Bottom: the photodissociation cross section $\sigma(E)$ (arb. units) for one time-reversal invariant realization of H_0 (circles), with $N=128$ and $\Lambda=1$. The solid lines describe fitted Beutler-Fano line shapes.

tially well-separated. System-specific information enters only in the values of the direct and indirect coupling constants and the transmission coefficients describing the coupling of the resonances to the continuum. We find that the direct processes suppress the correlation hole (see Fig. 3), which is a characteristic feature of the spectral autocorrelation function for a closed chaotic system [23]. We also calculate the cross-section distribution, and find that the direct decay gives rise to a characteristic maximum in the distribution (see Fig. 4) in the regime of isolated resonances, in contrast to the monotonically decreasing behavior of the distribution in the absence of direct coupling. Our results also apply to atomic autoionization [14,22].

A molecule can dissociate into several channels c by absorbing a photon. In the dipole approximation, the total photodissociation cross section at energy E is given by

$$\sigma(E) = \sigma_0(E) \sum_{c=1}^{\Lambda} |\langle \Phi_c^{(-)}(E) | \hat{\mu} | g \rangle|^2, \tag{1}$$

where $\hat{\mu} = \hat{\boldsymbol{\mu}} \cdot \mathbf{e}$ is the component of the dipole moment $\hat{\boldsymbol{\mu}}$ of the molecule along the polarization $\mathbf{e}$ of the absorbed light and $\sigma_0(E) \propto (E - E_g)$. Here $|g\rangle$ is the ground state with energy E_g, and $|\Phi_c^{(-)}(E)\rangle$ $(c = 1, \ldots, \Lambda)$ is a dissociation solution at

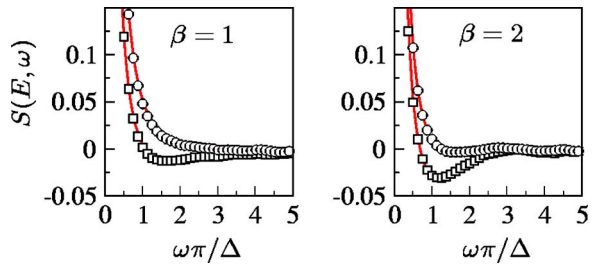

FIG. 3. (Color online) Cross-section autocorrelation functions for GOE (left panel) and GUE (right panel). The symbols are from simulations of random matrices ($N=128$) at the center of the band ($E=0$) and for $\Lambda=10$ and $\lambda_c=10^{-2}$ for all channels c. The autocorrelation functions without direct coupling ($\theta=0$, open squares) and with direct coupling ($\theta=1.26$, open circles) are compared to Eq. (12) (solid lines).

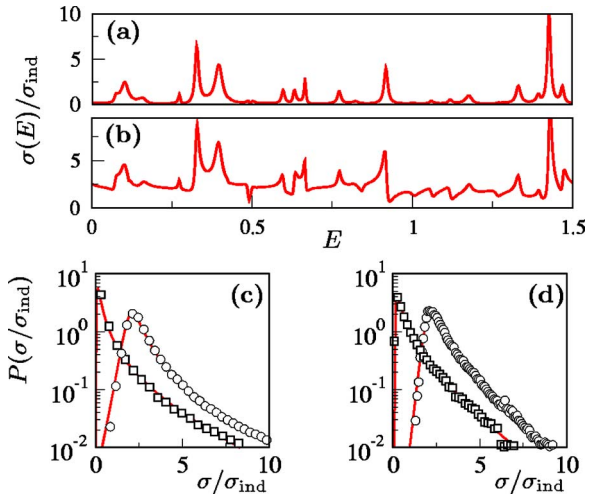

FIG. 4. (Color online) Cross-section distributions. (a) Cross section vs energy E in the absence of direct coupling ($\theta=0$) and for one random-matrix realization with H_0 from the GOE. Here $N=64$, $\Lambda=10$, and $\lambda_c=5\times10^{-2}$ for all c. (b) Same as in (a) but in the presence of direct processes ($\theta=0.125$). (c) Cross-section distributions at $E=0$ for $\theta=0$ (squares) and $\theta=0.125$ (circles). (d) as in (c) but for GUE. The solid lines in (c) and (d) are inverse Fourier transforms of $F_\beta(s)$, obtained by computing the average in Eq. (13) numerically.

energy E defined by an outgoing wave in channel c and incoming waves in all other channels. We consider a model [4,6,7] in which the Hilbert space is divided into two parts: an internal "interacting" region, and an external "channel" region (cf. Fig. 2). The internal region is described by the Hamiltonian $\hat{H}_0$ represented by an $N\times N$ matrix $\boldsymbol{H}_0$ with eigenstates $|n\rangle$ $(n=1,\ldots,N)$. The external region is spanned by the Λ open dissociation channels $|c\rangle$. The two regions are coupled by an operator $\hat{W}$ that can be represented by an $N\times\Lambda$ matrix $\boldsymbol{W}$ with matrix elements $\langle c|\hat{W}|n\rangle = \gamma_{nc}$. In general, the dipole operator $\hat{\mu}$ can couple the ground state to both the internal states $|n\rangle$ and the external channels $|c\rangle$. We define $|\alpha\rangle = \hat{\mu}|g\rangle$, and introduce two vectors $\boldsymbol{\alpha}^{\rm in}$ and $\boldsymbol{\alpha}^{\rm ch}$. The first has N components $\alpha_n^{\rm in} \equiv \langle n|\alpha\rangle$, describing the dipole coupling to the internal states, and the second has Λ components $\alpha_c^{\rm ch} \equiv \langle c|\alpha\rangle$, describing the dipole coupling to the continuum channels.

An explicit expression for the photodissociation cross section can be derived from Eq. (1) by separating the channel and internal components of the Green function [24]. Ignoring the energy dependence of $\alpha_c^{\rm ch}$ and γ_{nc}, we obtain

$$\sigma(E)/\sigma_0(E) = \|\boldsymbol{\alpha}^{\rm ch}\|^2 - \frac{1}{\pi}\,{\rm Im}\left[(\boldsymbol{\alpha}^{\rm in} + i\pi \boldsymbol{W}\boldsymbol{\alpha}^{\rm ch})^\dagger \frac{1}{E - \boldsymbol{H}_{\rm eff}} (\boldsymbol{\alpha}^{\rm in} - i\pi \boldsymbol{W}\boldsymbol{\alpha}^{\rm ch})\right]. \tag{2}$$

Here $\boldsymbol{H}_{\rm eff} = \boldsymbol{H}_0 - i\pi \boldsymbol{W}\boldsymbol{W}^\dagger$ is an effective (non-Hermitian) $N\times N$ Hamiltonian in the internal space. In the absence of direct photodissociation, $\boldsymbol{\alpha}^{\rm ch}=0$, and Eq. (2) reduces to the result of Refs. [6,7].

Fano resonances with complex q. In general, the operator $\boldsymbol{H}_{\rm eff}$ is characterized by bi-orthonormal right and left eigenvectors $|R_n\rangle$ and $|L_n\rangle$ with complex eigenvalues $\mathcal{E}_n$. However, in the regime of isolated resonances, $|R_n\rangle \approx |L_n\rangle \approx |n\rangle$ and $\mathrm{Im}\,\mathcal{E}_n = -\Gamma_n/2 \approx -\pi\Sigma_c|\gamma_{nc}|^2$. The cross section (2) can then be written as a sum over resonances. In the presence of direct photodissociation, the contribution from each of these resonances has a generalized Fano line shape $|q_n+\varepsilon_n|^2/(1+\varepsilon_n^2)$ with a *complex* parameter q_n whose real part and modulus are given by

$$\mathrm{Re}\; q_n = \frac{\mathrm{Re}[\alpha_n^{\mathrm{in}^*}\sum_{c=1}^{\Lambda}\alpha_c^{\mathrm{ch}}\gamma_{nc}]}{\pi\sum_{c=1}^{\Lambda}|\alpha_c^{\mathrm{ch}}|^2\sum_{c=1}^{\Lambda}|\gamma_{nc}|^2},$$

$$|q_n|^2 = 1 + \frac{|\alpha_n^{\mathrm{in}}|^2 - \pi^2\left|\sum_{c=1}^{\Lambda}\alpha_c^{\mathrm{ch}}\gamma_{nc}\right|^2}{\pi^2\sum_{c=1}^{\Lambda}|\alpha_c^{\mathrm{ch}}|^2\sum_{c=1}^{\Lambda}|\gamma_{nc}|^2}. \tag{3}$$

In general, $\mathrm{Im}\,q_n \neq 0$, and there is no energy for which the cross section vanishes. In the special case of time-reversal symmetric $\boldsymbol{H}_0$ and $\Lambda=1$, Eq. (3) simplifies to Fano's expression $\mathrm{Re}\,q_n = \alpha_n^{\mathrm{in}}/(\pi\alpha_c^{\mathrm{ch}}\gamma_{nc})$ and $\mathrm{Im}\,q_n = 0$ (assuming that all matrix elements are real).

Statistical model. Assuming the dynamics in the closed interaction region are fully chaotic, we model the matrix $\boldsymbol{H}_0$ by an $N\times N$ random matrix [1], which belongs to the Gaussian orthogonal ensemble (GOE) or to the Gaussian unitary ensemble (GUE). To fix the energy scale, we require the mean level density to be equal to $1/\pi$ in the center of the spectrum in the limit of large N. In the same limit, the quantities α_n^{in} and γ_{nc} $(c=1,\ldots,\Lambda)$ are uncorrelated Gaussian random variables for different values of n, and, for each n [5],

$$P(\alpha_n^{\mathrm{in}},\gamma_n) \propto \exp\left[-\frac{\beta}{2}(\alpha_n^{\mathrm{in}^*},\gamma_n^{\dagger})\boldsymbol{M}^{-1}\begin{pmatrix}\alpha_n^{\mathrm{in}}\\ \gamma_n\end{pmatrix}\right], \tag{4}$$

where $\beta=1,2$ for the GOE and the GUE, respectively. Here $\gamma_n^{\dagger}=(\gamma_{n1}^*,\ldots,\gamma_{n\Lambda}^*)$, and

$$\boldsymbol{M} = N^{-1}\boldsymbol{V}^{\dagger}\boldsymbol{V} \quad \text{with } \boldsymbol{V}=(\alpha^{\mathrm{in}},\boldsymbol{W}) \tag{5}$$

is an $(\Lambda+1)\times(\Lambda+1)$ matrix. In the following, we assume the channel vectors (columns of $\boldsymbol{W}$) to be mutually orthogonal. This can always be achieved by a suitable orthogonal (unitary) transformation in channel space.

Average cross section. In the center of the band $(E=0)$, we find for the average cross section

$$\langle\sigma\rangle = \sigma_{\mathrm{ind}} + \sum_{c=1}^{\Lambda}\sigma_c^{\mathrm{dir}} - \sum_{c=1}^{\Lambda}\sigma_c^{\mathrm{dir}}\lambda_c/(1+\lambda_c), \tag{6}$$

where $\sigma_{\mathrm{ind}} = \sigma_0\|\boldsymbol{\alpha}^{\mathrm{in}}\|^2/\pi$ and $\sigma_c^{\mathrm{dir}} = \sigma_0|\alpha_c^{\mathrm{ch}}|^2$ are the average cross sections in the limiting cases of purely indirect and purely direct dissociation, respectively, and $\lambda_1,\ldots,\lambda_\Lambda$ are the eigenvalues of the matrix $\pi\boldsymbol{W}^{\dagger}\boldsymbol{W}$. The coupling to the continuum is often parametrized by transmission coefficients $T_c = 4\lambda_c/(1+\lambda_c)^2$. The last term on the right-hand side of Eq. (6) describes the effect of "backscattering" from the continuum states into the quasibound states, and it exists even when the photoexcited state has no overlap with the quasibound states.

Cross-section autocorrelation function. We define a dimensionless cross-section autocorrelation function [6,7]

$$S(E,\omega) = \frac{1}{\sigma_{\mathrm{ind}}^2}[\langle\sigma(E-\omega/2)\sigma(E+\omega/2)\rangle - \langle\sigma\rangle^2]. \tag{7}$$

In the Breit-Wigner approximation, this correlation is most conveniently calculated in the time domain [25]. Defining $C(E,t)=\int_{-\infty}^{\infty}d\omega e^{i\omega t}S(E,\omega)$, and using Eq. (4), we find

$$C(E,t) = \frac{1}{4\pi^2}[A_\beta(t) - B_\beta^2(t)b_{2,\beta}(t)], \tag{8}$$

where $b_{2,\beta}(t)$ is the two-level form factor [1], and $A_\beta(t), B_\beta(t)$ are functions that depend in general on the matrix $\boldsymbol{M}$ in Eq. (5) and the dipole coupling coefficients to the continuum α_c^{ch}.

The expressions for $A_\beta(t)$ and $B_\beta(t)$ simplify when the dipole "channel" is orthogonal to all channel vectors, i.e., $\boldsymbol{W}^{\dagger}\boldsymbol{\alpha}^{\mathrm{in}}=0$. This is the case when the excitation process and the continuum coupling are spatially well separated. We introduce the parameters $\tau_c = T_c\sigma_c^{\mathrm{dir}}/\sigma_{\mathrm{ind}}$, which characterize the relative strength of the direct dissociation channels. Using the so-called rescaled Breit-Wigner approximation [7,26], we find

$$A_1(t) = \prod_{c=1}^{\Lambda}(1+2T_ct)^{-1/2}\left[3 + \frac{1}{2}\sum_c \tau_c(1+2T_ct)^{-1} + \frac{3}{16}\left(\sum_c \tau_c(1+2T_ct)^{-1}\right)^2\right],$$

$$B_1(t) = \prod_{c=1}^{\Lambda}(1+T_ct)^{-1/2}\left[1 - \frac{1}{4}\sum_c \tau_c(1+T_ct)^{-1}\right] \tag{9}$$

for $\beta=1$. A similar result is obtained for $\beta=2$,

$$A_2(t) = \prod_{c=1}^{\Lambda}(1+T_ct)^{-1}\left[2 + \frac{1}{8}\left(\sum_c \tau_c(1+T_ct)^{-1}\right)^2\right],$$

$$B_2(t) = \prod_{c=1}^{\Lambda}(1+T_ct/2)^{-1}\left[1 - \frac{1}{4}\sum_c \tau_c(1+T_ct/2)^{-1}\right]. \tag{10}$$

The results (9) and (10) describe universal correlations; they depend on the coefficients T_c and on the parameters τ_c, but they are independent of the microscopic details of the system such as the ground state or the nature of the excitation mechanism. Two special cases are of interest. First, in the limit of $\tau_c=0$ (no direct coupling to the continuum), the results of Ref. [7] are recovered. Second, consider the case of Λ equivalent open channels $T_c=T$ and $\tau_c=\tau$ (i.e., $\sigma_c^{\mathrm{dir}}=\sigma^{\mathrm{dir}}$). In the limit $\Lambda\to\infty, T\to 0$ with $\Lambda T\equiv\kappa$ const, the expressions (9) and (10) simplify to

$$A_\beta(t) = A_\beta e^{-\kappa|t|}, \quad B_\beta(t) = Be^{-\kappa|t|/2} \tag{11}$$

with $A_1 = 3 + \theta/2 + 3\theta^2$, $A_2 = 2(1 + \theta/16)$, $B = (1 - \theta/4)$, and θ is defined by $\theta = \Lambda\tau = \kappa\sigma^{\rm dir}/\sigma_{\rm ind}$. We obtain

$$S(E,\omega) = \frac{1}{4\pi^2}\left[A_\beta f(\omega) - B^2 \int \frac{d\omega'}{\pi} f(\omega - \omega') Y_{2,\beta}(\omega')\right], \tag{12}$$

where $Y_{2,\beta}(\omega)$ is the two-level cluster function [1], and $f(\omega) = (\kappa/2)/(\omega^2 + \kappa^2/4)$. Figure 3 shows the autocorrelation function (12) (solid lines) together with results from random-matrix simulations (symbols). In the absence of direct decay ($\theta = 0$), we observe a "correlation hole" (i.e., a minimum in the autocorrelation), which originates in level repulsion and is described by the term containing $Y_{2,\beta}$ in Eq. (12). In the presence of direct decay ($\theta > 0$), the coefficient B decreases while A_β increases, and the correlation hole is suppressed. This suppression is stronger for $\beta = 1$ ($A_1 > A_2$), and in the case shown in Fig. 3, the correlation hole has completely disappeared for $\beta = 1$.

Cross-section distribution. The distribution $P(\sigma/\sigma_{\rm ind})$ is calculated from its Fourier transform $F_\beta(s) = \langle e^{-is\sigma/\sigma_{\rm ind}}\rangle$ within the Breit-Wigner approximation. We have determined $F_\beta(s)$ for Λ equivalent open channels in the limit of $\Lambda \to \infty$ with $\Lambda T \equiv \kappa$ kept constant. In this case, $\Gamma_n \simeq \Gamma = 2\Lambda\lambda/N$, and using Eq. (4) we obtain

$$F_\beta(s) = e^{is\theta/(2\Gamma N)}\left\langle \left(\frac{\det[(E - \boldsymbol{H}_0)^2 + \Gamma^2/4]}{\det[(E - \boldsymbol{H}_0)^2 + \tilde{\Gamma}_\beta^2/4]}\right)^{\beta/2}\right\rangle \tag{13}$$

with $\tilde{\Gamma}_\beta^2 = [\Gamma + 4\pi i s/(N\beta)][\Gamma - 2\pi i s\theta/(N\beta)]$. Equation (13) can be evaluated further using the results of Refs. [27,28]. For $\beta = 1$, the result is given in [27]. Figures 4(c) and 4(d) show inverse Fourier transforms of $F_\beta(s)$, which were obtained by numerically averaging the ratio of determinants in Eq. (13) (solid lines). Also shown are histograms of the total cross section obtained from random-matrix simulations (symbols). In the presence of direct coupling, the cross-section distribution exhibits a maximum. In the limit of isolated resonances, this maximum is a clear signature of the direct processes.

In summary, we have derived an expression for the total molecular photodissociation cross section in the presence of direct processes interfering with indirect dissociation. In the case of isolated resonances, this interference gives rise to generalized Fano line shapes. Within a random-matrix model, we have studied the fluctuations of the photodissociation cross section in cases where the associated classical molecular dynamics is fully chaotic. Our results are derived within the rescaled Breit-Wigner approximation. In particular, we have studied the case of many weakly coupled channels (allowing for overlapping resonances).

We have identified statistical signatures of the interference between direct and indirect processes: the suppression of the correlation hole in the cross-section autocorrelation function, and—for isolated resonances—the existence of a maximum in the cross-section distribution. We have also found that the contribution of the direct processes to the average cross section is reduced by a backscattering effect.

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