Quasibound States in a Chaotic Molecular System

Linda Reichl, Hoshik Lee, Alex Barr and Kyungsun Na
Center for Complex Quantum Systems
The University of Texas at Austin

Open quantum systems have a continuous spectrum, but underlying chaos can still profoundly affect the dynamics.

We consider two different types of open physical system in which chaos and resonances in the underlying classical dynamics largely determine the quantum dynamics:

1. Scattering states in ballistic electron waveguides.

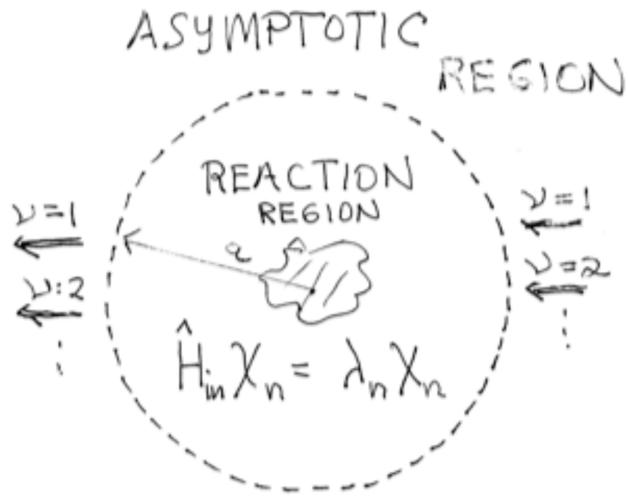
2. Scattering states of a molecule.

Wigner Eisenbud Scattering theory $\hat{H}\left|\Psi_E\right\rangle = E\left|\Psi_E\right\rangle$

Scattering channels $\nu = 1,\ldots,M$

Asymptotic incoming $\Phi_\nu^{in}(r)$ **and outgoing** $\Phi_\nu^{out}(r)$ **scattering state**

$$\Psi_E^{asymp}(\vec{r}) = \sum_\nu \left(A_\nu \Phi_\nu^{in}(\vec{r}) + B_\nu \Phi_\nu^{out}(\vec{r})\right)$$



Reaction region basis states $\chi_{n,\nu}(r), n = 1,\ldots,N$

Scattering state in the reaction region

$$\Psi_E^{reac}(\vec{r}) = \sum_{n,\nu} C_{n,\nu}\chi_{n,\nu}(\vec{r})$$

Reaction matrix $R_{\nu,\nu'}(E)$

$$\sum_{\nu'} R_{\nu,\nu'}(E)\left[A_{\nu'}\left(\frac{d\Phi_{\nu'}^{in}}{dr}\right)_{r=a} + B_{\nu'}\left(\frac{d\Phi_{\nu'}^{out}}{dr}\right)_{r=a}\right] = A_\nu \Phi_\nu^{in}(a) + B_\nu \Phi_\nu^{out}(a)$$

Wigner-Eisenbud reaction matrix

$$R_{\nu,\nu'}(E) = \frac{\hbar^2}{2ma}\sum_n \frac{\chi_{n,\nu}(a)\chi_{n,\nu'}^\dagger(a)}{\lambda_n - E} \quad \rightarrow \quad \bar{R}(E) = \bar{w}^\dagger \cdot \frac{1}{E\bar{1} - \bar{H}_{in}} \cdot \bar{w}, \quad where \quad w_{n,\nu} = \chi_{n,\nu}(a)$$

Scattering matrix S connects incoming to outgoing states $\bar{B} = \bar{S}\cdot\bar{A}$

$$\bar{S} = \frac{\bar{1} - \bar{R}}{\bar{1} + \bar{R}} = \bar{1} - i\bar{w}^\dagger \cdot \frac{1}{E\bar{1} - \bar{H}_{in} + i\bar{w}\cdot\bar{w}^\dagger}\cdot\bar{w}$$

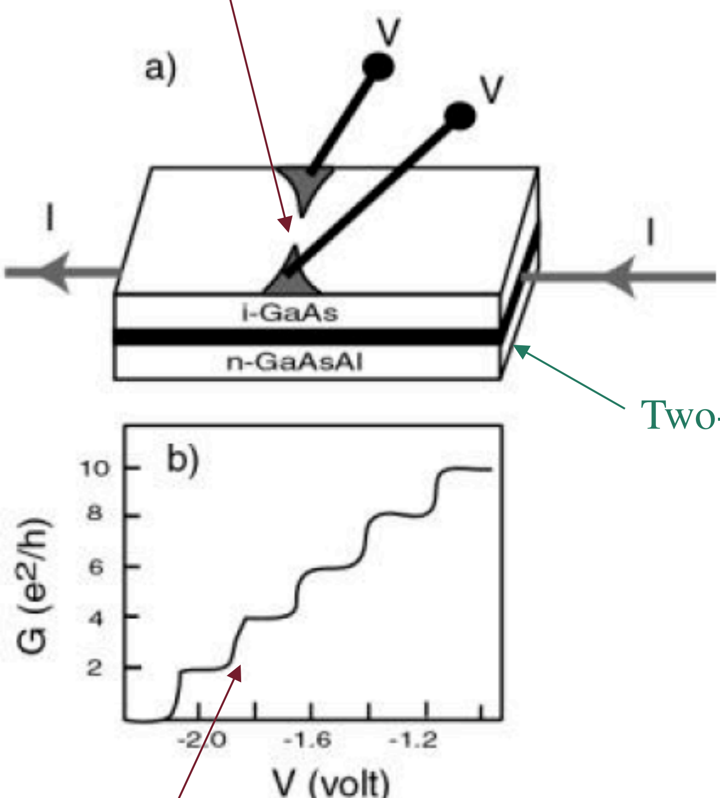
Effective Hamiltonian

$$\bar{H}_{eff} = \bar{H}_{in} + i\bar{w}\cdot\bar{w}^\dagger$$

Chaotic structures in ballistic electron waveguides.

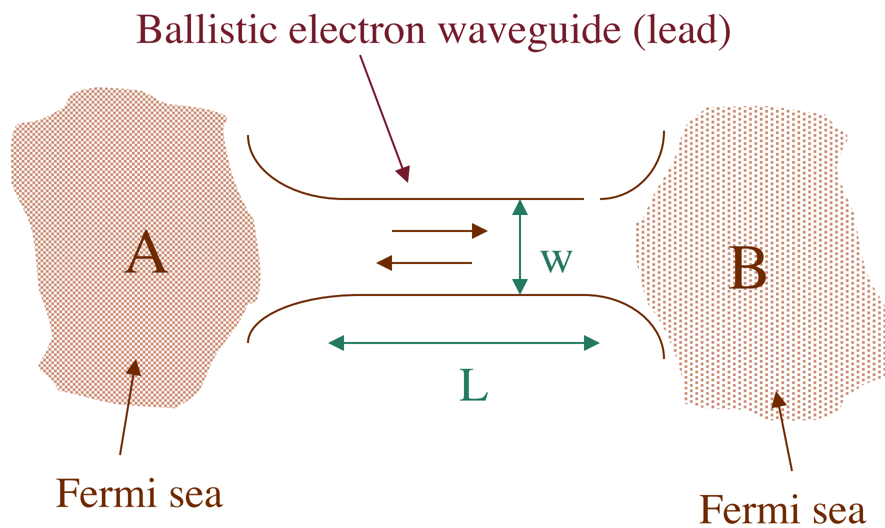
Ballistic electron waveguide

Quantum point contact



Two-dimensional electron gas

Conductance quantization



Energy of electron in *n*th longitudinal and νth transverse mode of the lead.

$$E_{n,\nu} = \frac{\hbar^2}{2m}\left[k_n^2 + \left(\frac{\pi\nu}{w}\right)^2\right]$$

Longitudinal wavevector

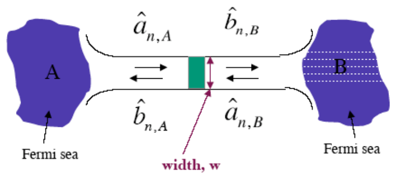
$$k_n = \frac{2\pi n}{L}$$

Quasibound states can live on dynamical structures in ballistic electron waveguides.

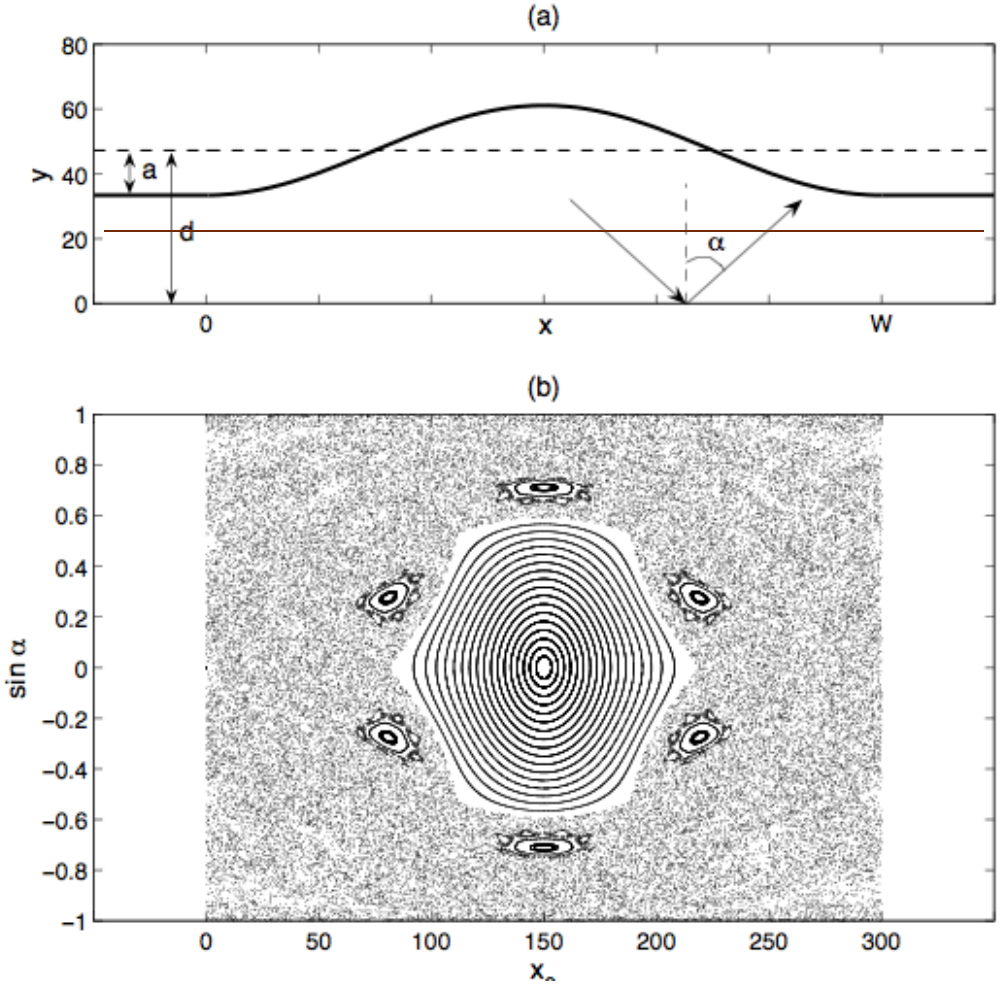
Quasibound states in ballistic electron waveguides

H. Lee, C. Jung, and L.E.Reichl, PRB 73 195315 (2006)

H. Lee and L.E.Reichl, PRB 77 205318 (2008

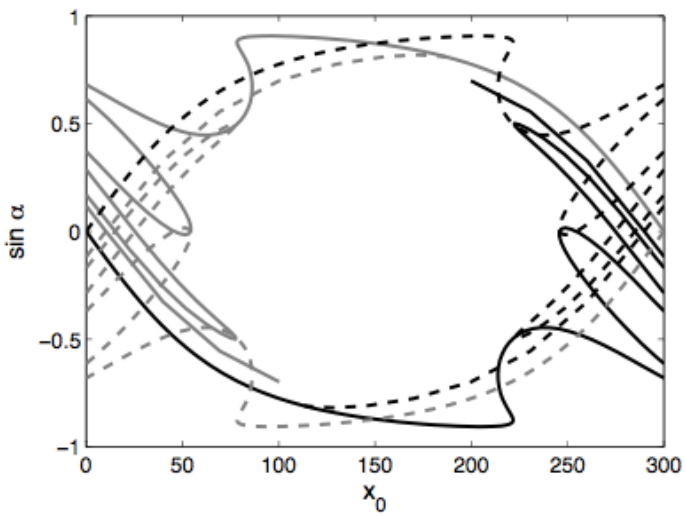


Waveguide channel

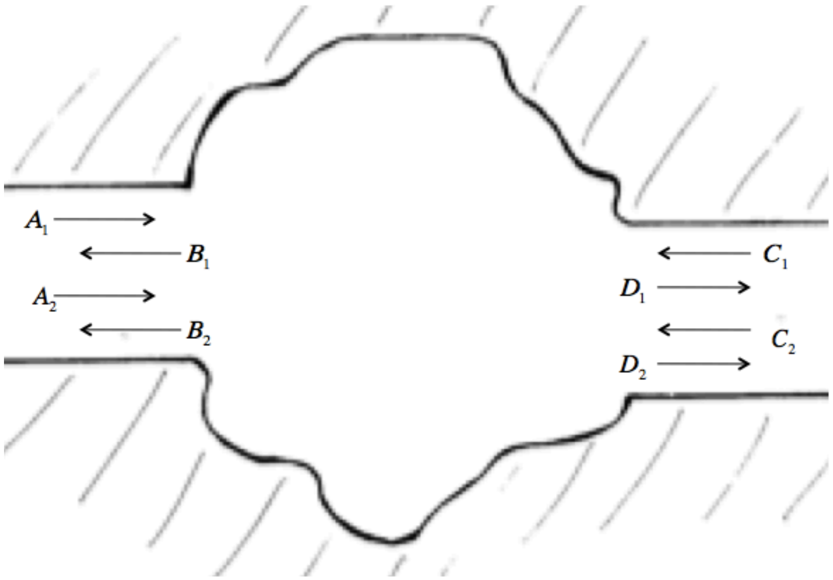


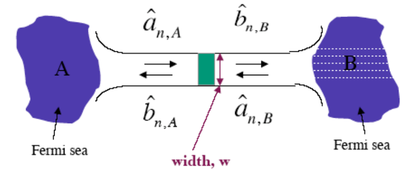
Birkhoff coordinates
off bottom of channel

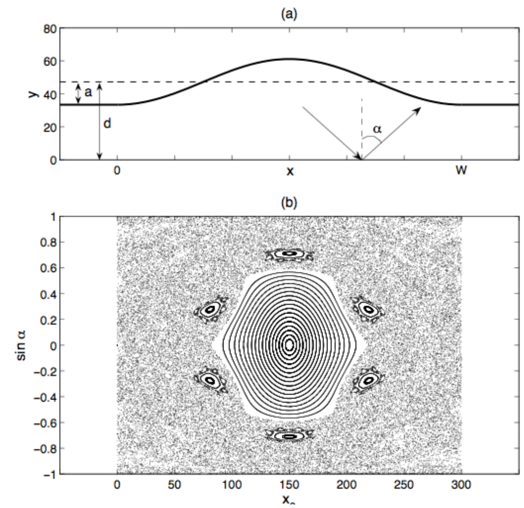
Hetero-clinic tangles



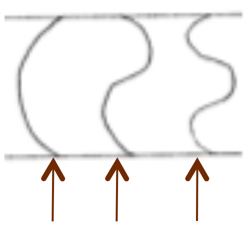
Quasibound states in ballistic electron waveguides







Channels



1st, 2nd, 3rd

Scattering matrix

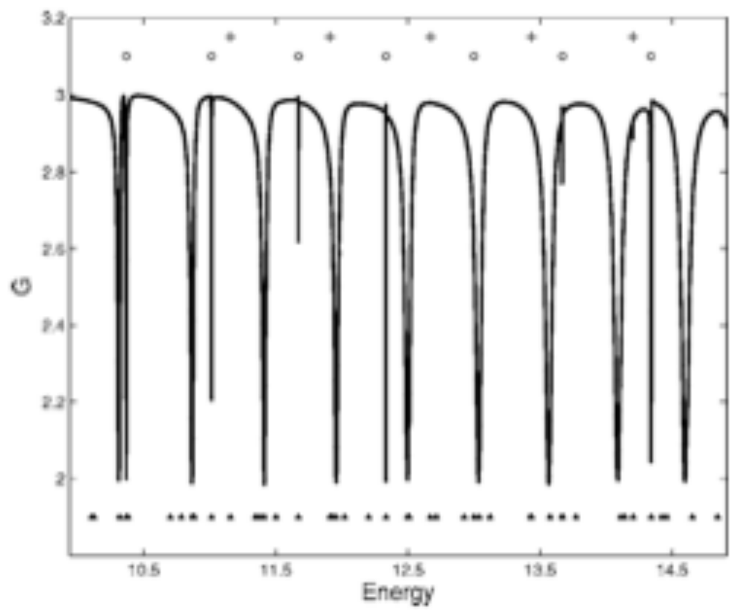
(*two propagating channels*)

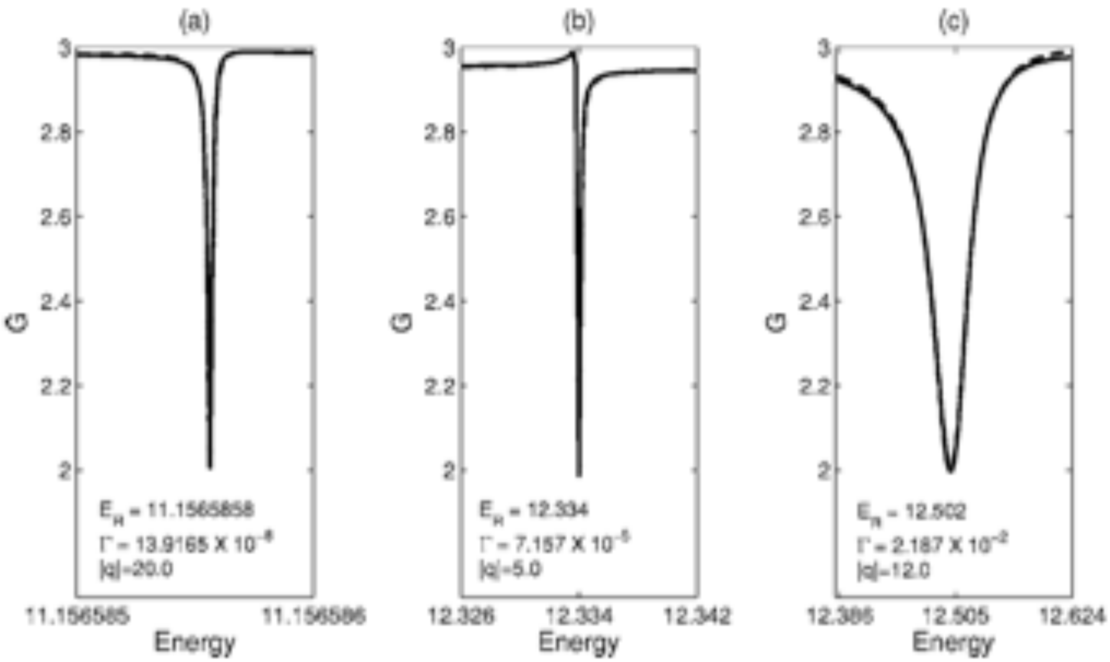
$$\begin{pmatrix} B_1 \\ B_2 \\ D_1 \\ D_2 \end{pmatrix} = \begin{pmatrix} r_{1,1} & r_{1,2} & t'_{1,1} & t'_{1,2} \\ r_{2,1} & r_{2,2} & t'_{2,1} & t'_{2,2} \\ t_{1,1} & t_{1,2} & r'_{1,1} & r'_{1,2} \\ t_{2,1} & t_{2,2} & r'_{2,1} & r'_{2,2} \end{pmatrix} \begin{pmatrix} A_1 \\ A_2 \\ C_1 \\ C_2 \end{pmatrix}$$

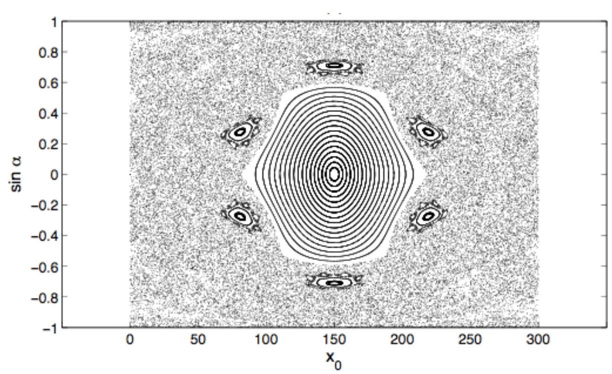
Landauer Conductance

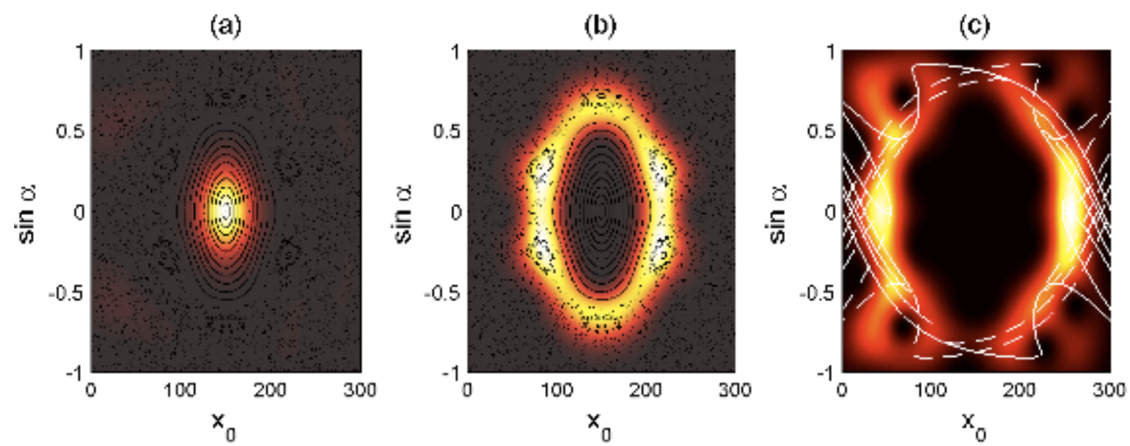
$$G = \frac{2e^2}{h} \sum_{i=1}^{n_p} \sum_{j=1}^{n_p} | t_{i,j} |^2$$

Quasibound states in ballistic electron waveguides









The Dicke effect - the coherent spontaneous radiation of a collection of noninteracting atoms which are coupled via a radiation field.

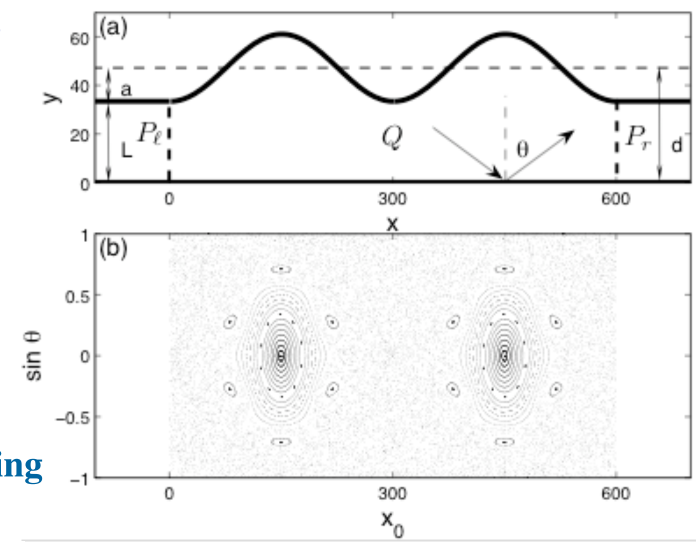
R.H. Dicke, Phys. Rev. **93** 99 (1954)

The coherent radiation of N atoms occurs N times faster than the radiation of a single atom and is called superradiance.

Similar effects have been observed in quantum dots. In two or three space dimensions, the superradiance intensity oscillates and decreases with increased spacing between dots.

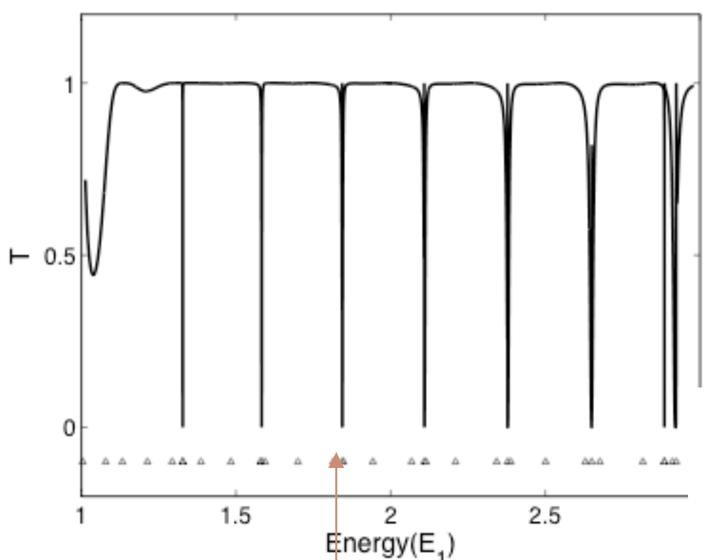
Bi-ripple GaAs waveguide (L=3.3nm, a=1.38nm, W=60nm)

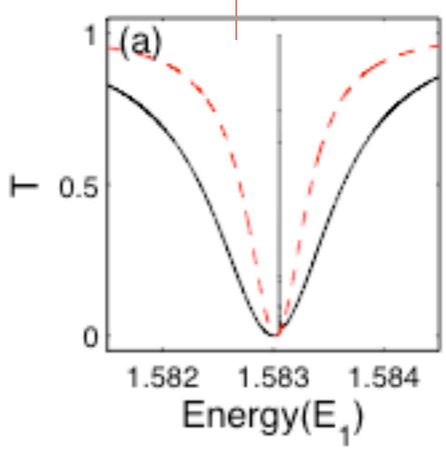
Superradiant and subradiant states



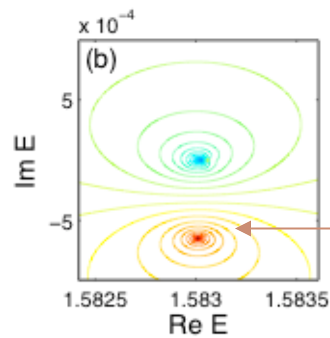
E_1=0.50eV

G=$2e^2T/h$

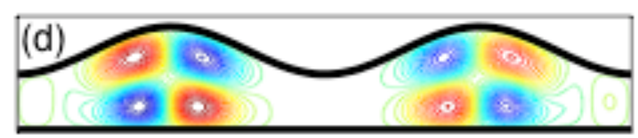


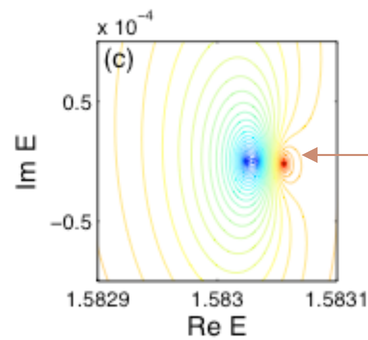


Poles of scattering matrix



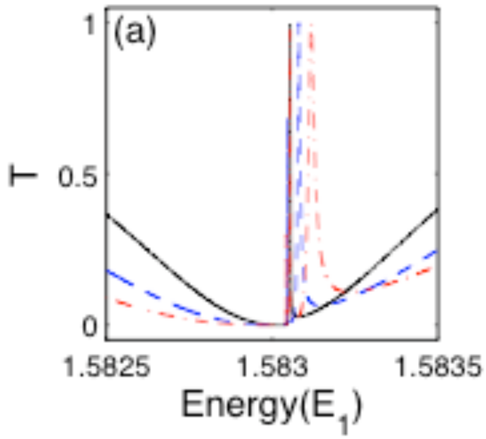
Symmetric quasibound scattering state

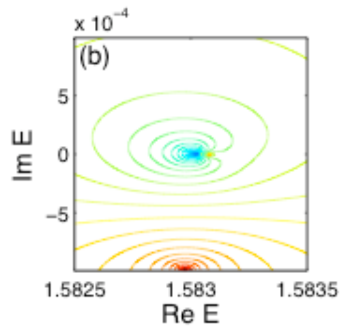




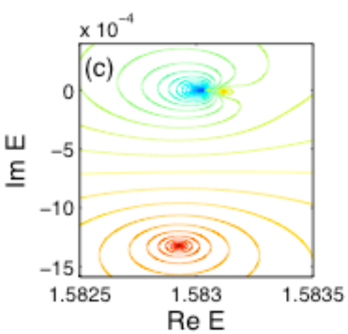
Anti-symmetric quasibound scattering state

The lifetime $\tau = 1/\Gamma$ of the superradiant state decreases as 1/N, where N is the number of quantum dots in the waveguide.

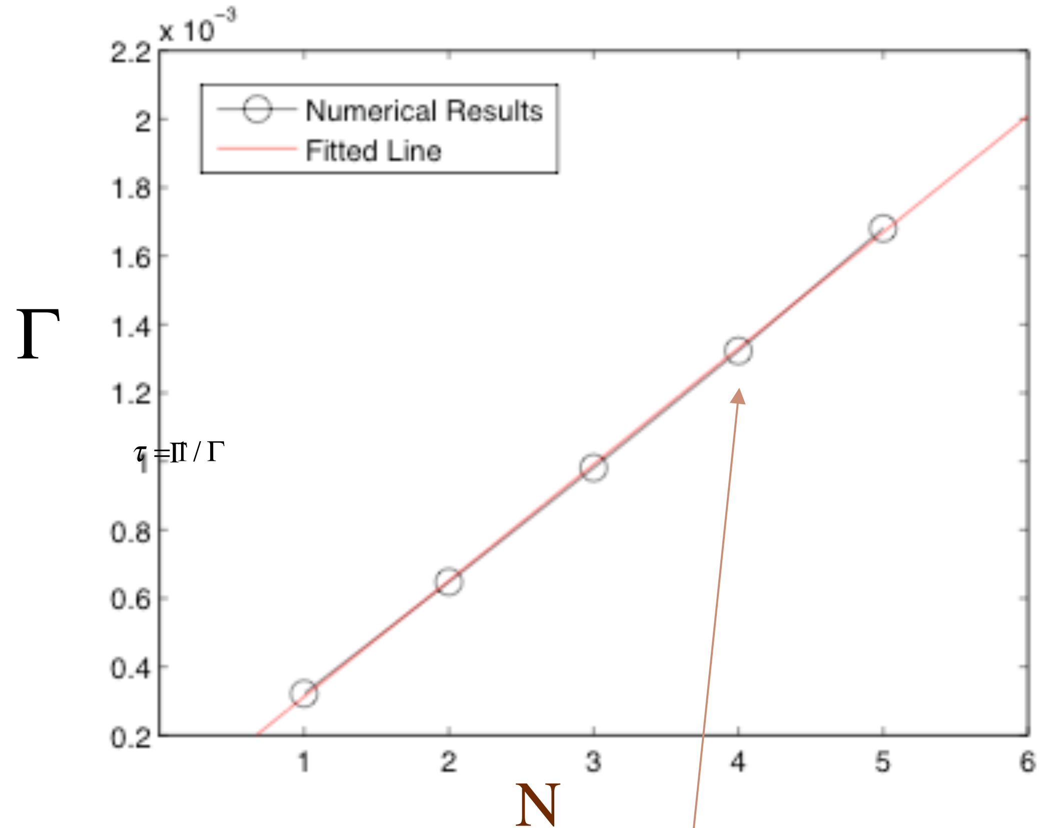


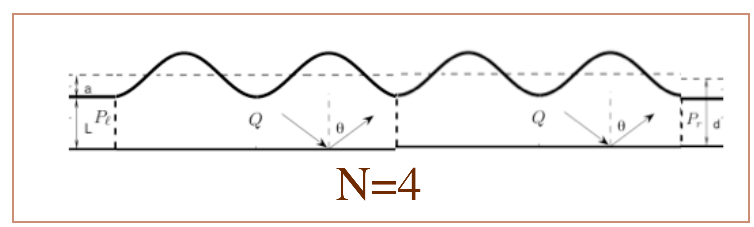


N=3



N=4



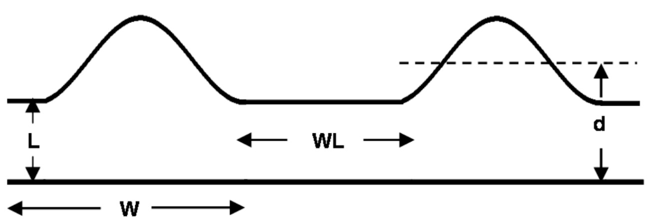


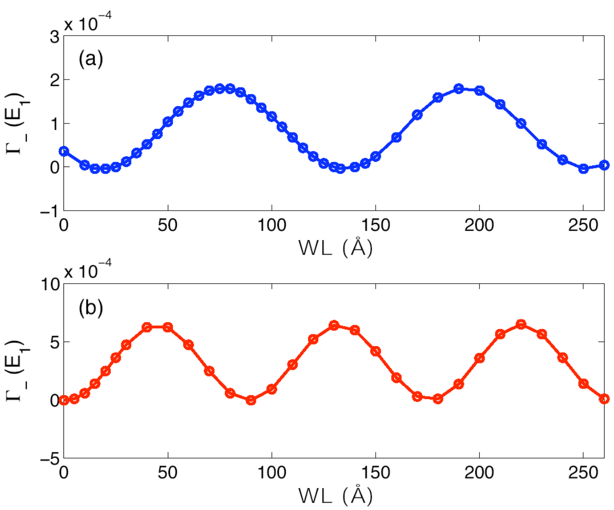
In the GaAs waveguide, the "superradiant" and "subradiant" poles oscillate in a circle (with π phase difference) in the complex energy plane (and change identities) as distance between quantum dots increases.

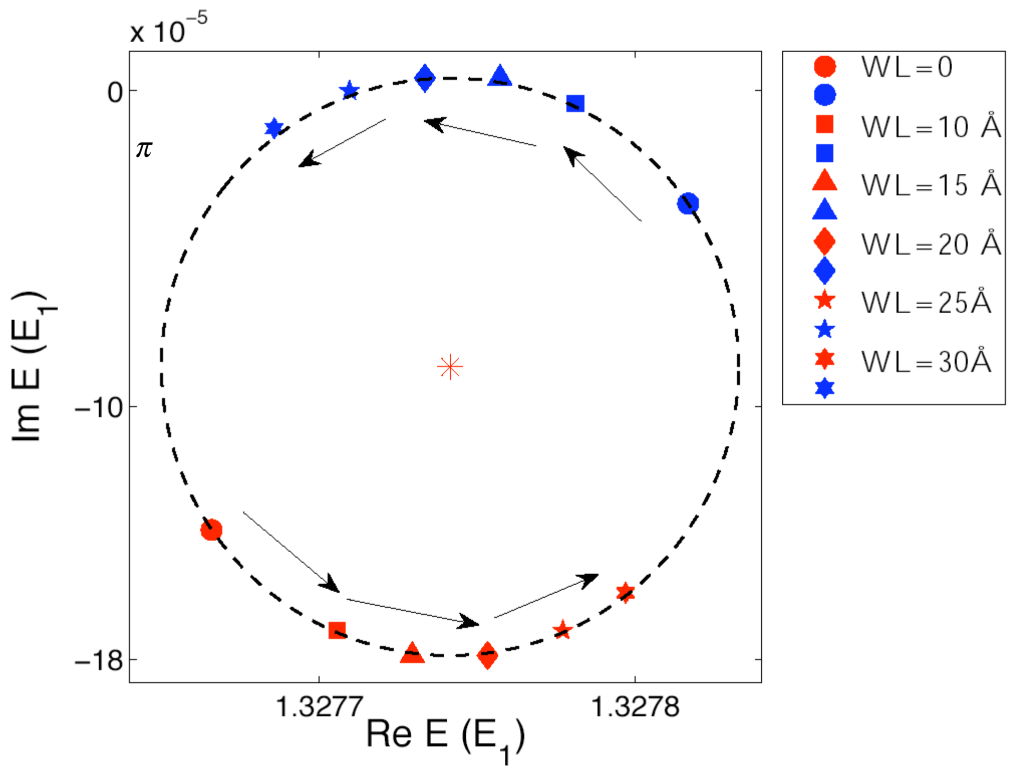
At some distances a "bound state in the continuum" can form.

In the GaAs waveguide, the Dicke effect does not decay with distance between the quantum dots.

H. Lee and L.E. Reichl PRB 79 193305 (2009)





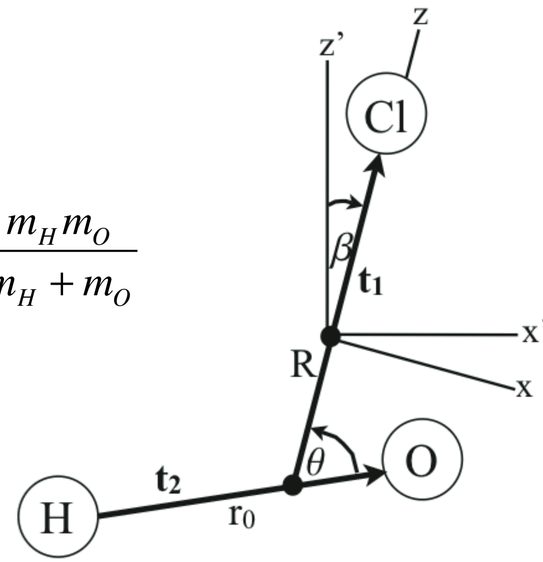


Quasibound states in the HOCl molecule

Dynamics of the HOCl molecule

Kinetic energy

$$T = \frac{1}{2}\mu_1 \dot{t}_1^2 + \frac{1}{2}\mu_2 \dot{t}_2^2 \quad \textbf{where} \quad \mu_1 = \frac{m_{Cl}\left(m_H + m_O\right)}{m_{Cl} + m_H + m_O} \qquad \mu_2 = \frac{m_H m_O}{m_H + m_O}$$



Restrict to total angular momentum L_{tot}=0

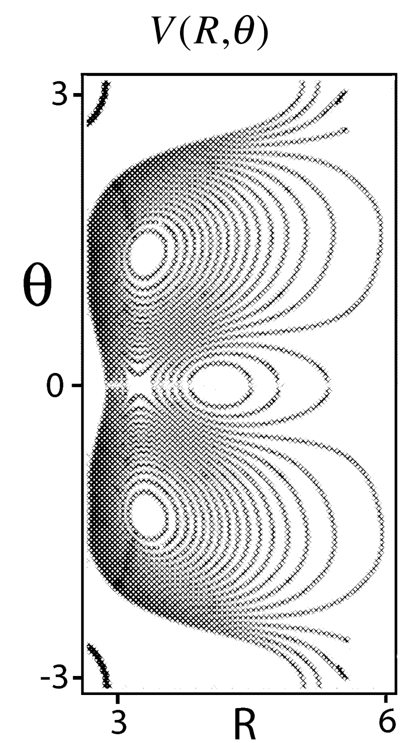
$$\vec{L}_{tot} = \mu_1 \vec{t}_1 \times \dot{\vec{t}}_1 + \mu_2 \vec{t}_2 \times \dot{\vec{t}}_2 = p_\beta \hat{y}$$

$$p_\theta = 0 \rightarrow \vec{L}_1 = \mu_1 \vec{t}_1 \times \dot{\vec{t}}_1 = -\vec{L}_2 = -\mu_2 \vec{t}_2 \times \dot{\vec{t}}_2$$

With L_{tot}=0, Cl and H-O can rotate relative to each other

Hamiltonian for HOCl

$$H = \frac{p_R^2}{2\mu_1} + \frac{p_\theta^2}{2\mu_2 r_0^2} + \frac{p_\theta^2}{2\mu_1 R^2} + V(R,\theta) = E$$

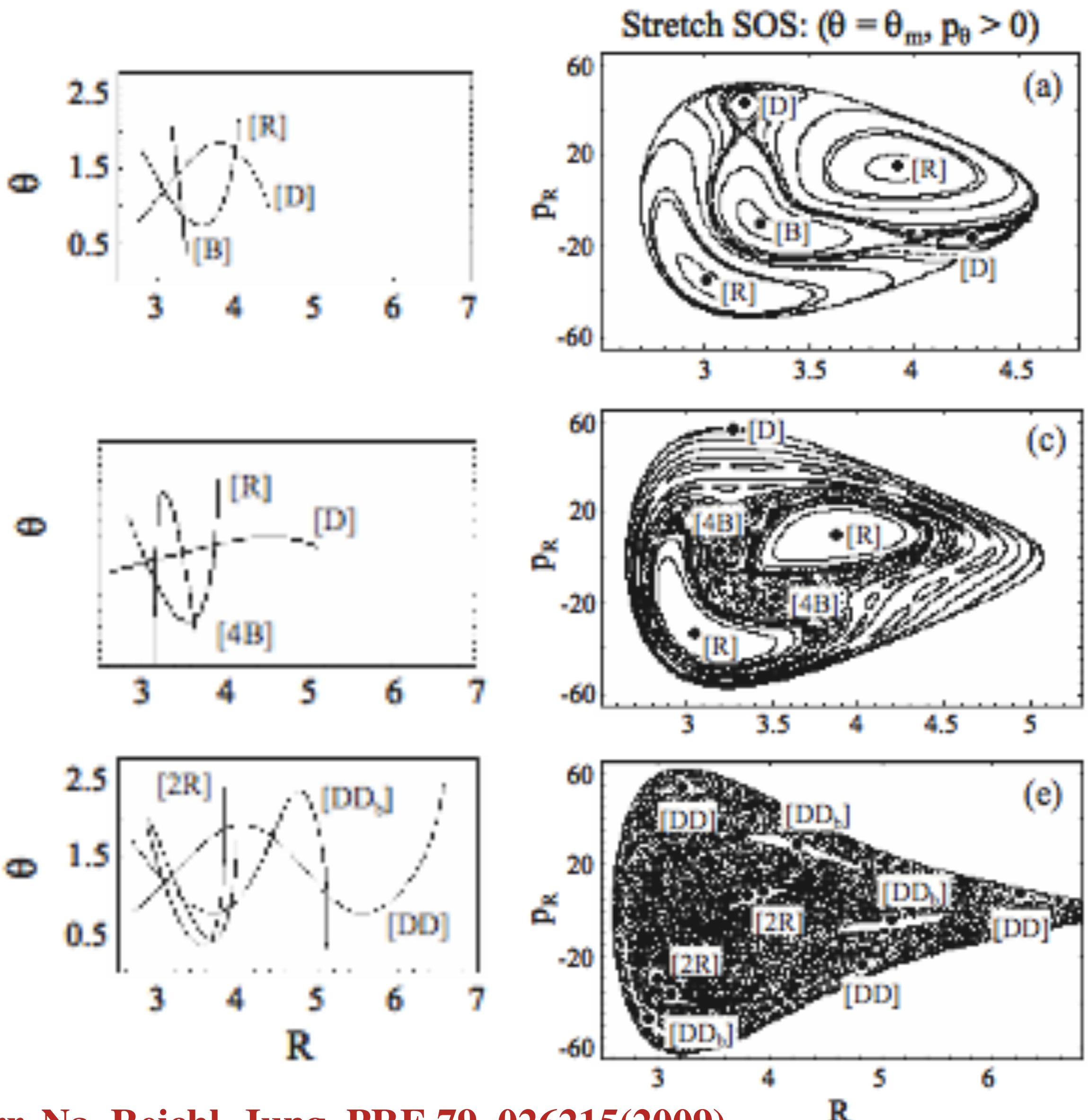


r_0=1.85 a, where a_0=0.529x10^{-10} m is Bohr radius.

Dissociation of Cl at energy E_d=20,312cm^{-1}=2.518eV

Classical bound motion of HOCl molecule

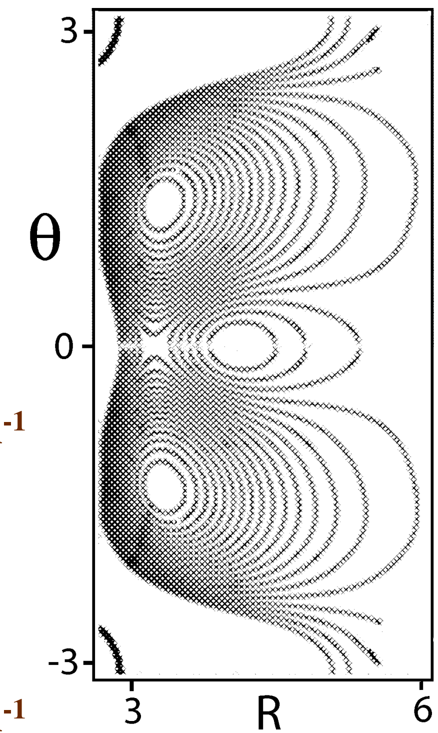
(below dissociation of Cl from H-O)

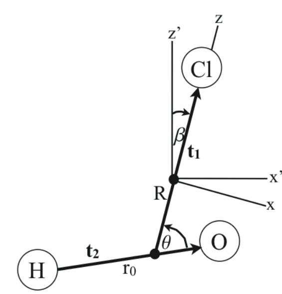


E=14,000cm^{-1}

E=17,020cm^{-1}

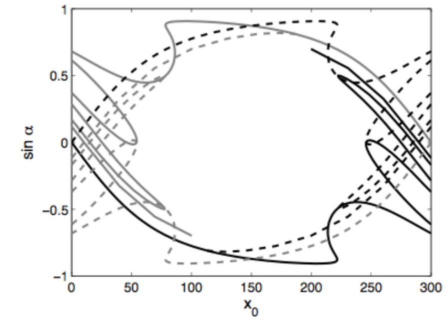
E=20,150cm^{-1}



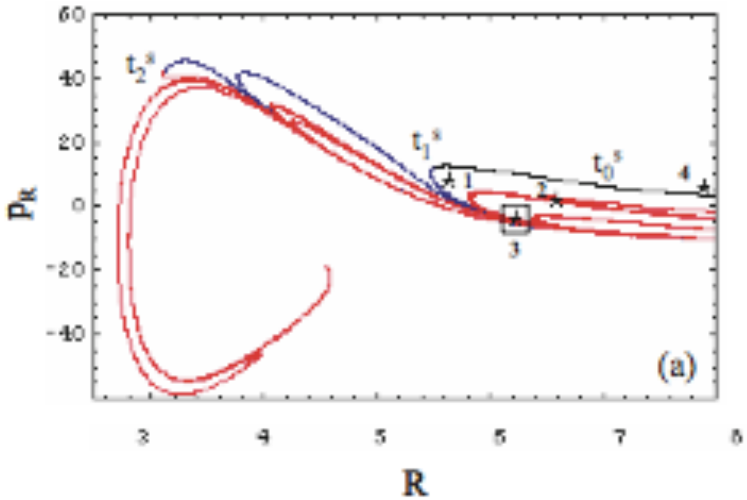


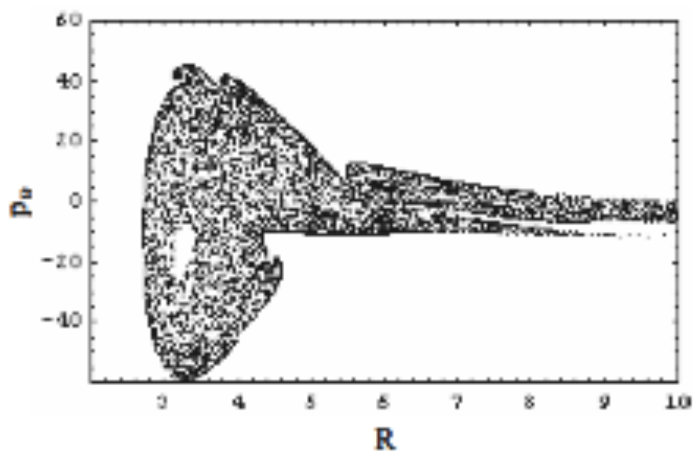
Barr, Na, Reichl, Jung, PRE 79, 026215(2009)

Classical Scattering of Cl from H-O
(above dissociation of Cl from H-O)

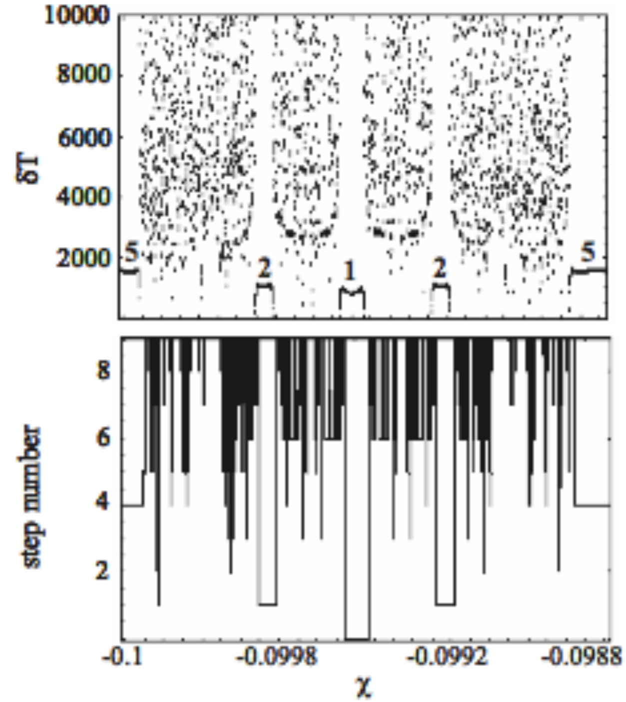


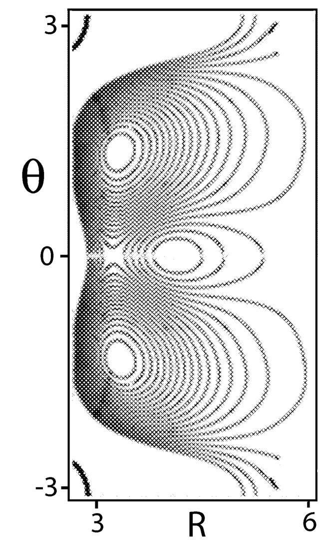
Two iterations of stable manifold

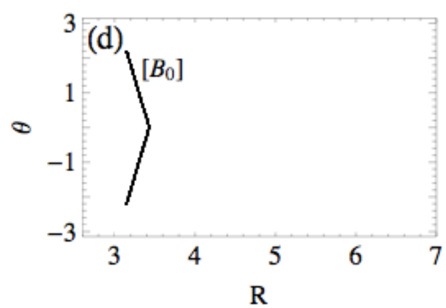


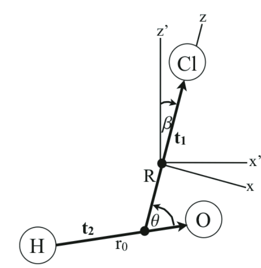


Scattering dynamics has fractal intervals









HOCl Molecule (Quantum)

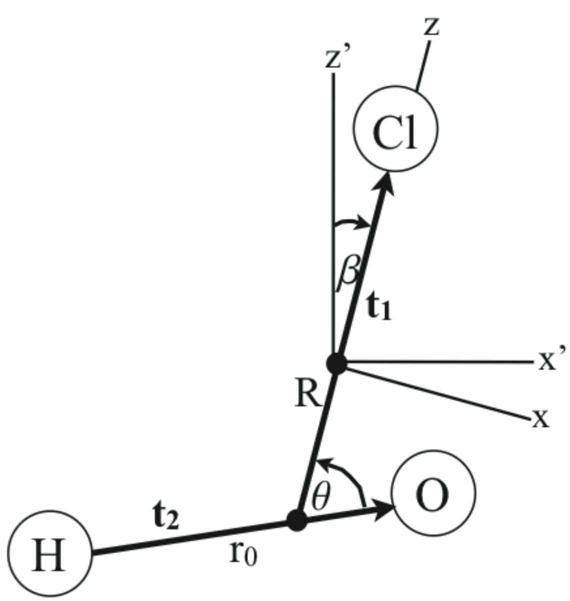
Barr, Na, Reichl, PRA 83, 062510 **(2011)**

Hamiltonian governing quantum dynamics of HOCl above and below dissociation assuming L_{tot}=0.

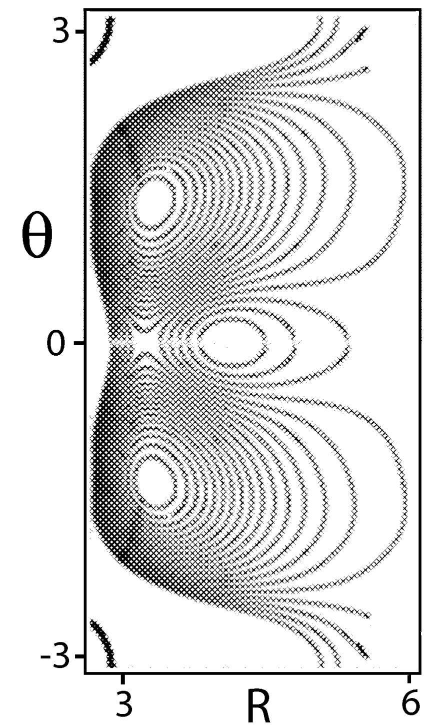
$$\hat{H} = -\frac{\hbar^2}{2}\Big[\frac{1}{\mu_1}\Big(\frac{\partial^2}{\partial R^2} + \frac{1}{R}\frac{\partial}{\partial R}\Big) + \Big(\frac{1}{\mu_1 R^2} + \frac{1}{\mu_2 r_0^2}\Big)\frac{\partial^2}{\partial \theta^2}\Big] + V(R,\theta)$$

r_0=1.85 a, where a_0=0.529x10^{-10} m is Bohr radius.

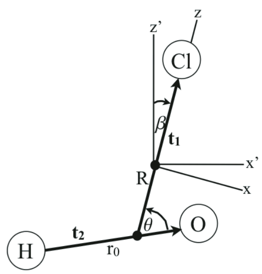
Dissociation of Cl at energy E_d=20,312cm^{-1}=2.518eV



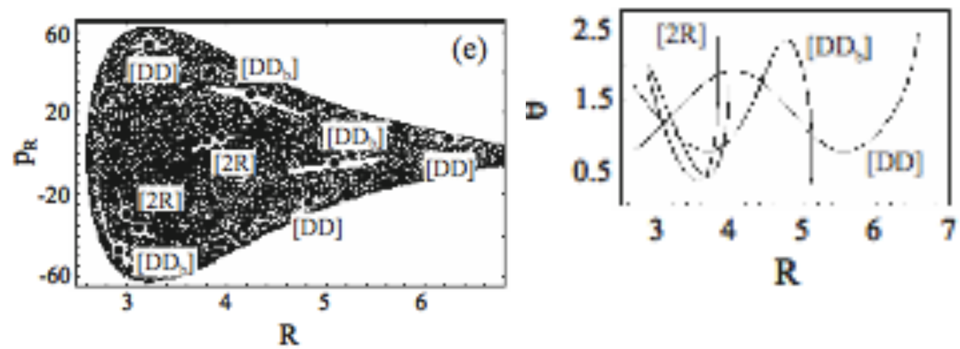
$V(R,\theta)$



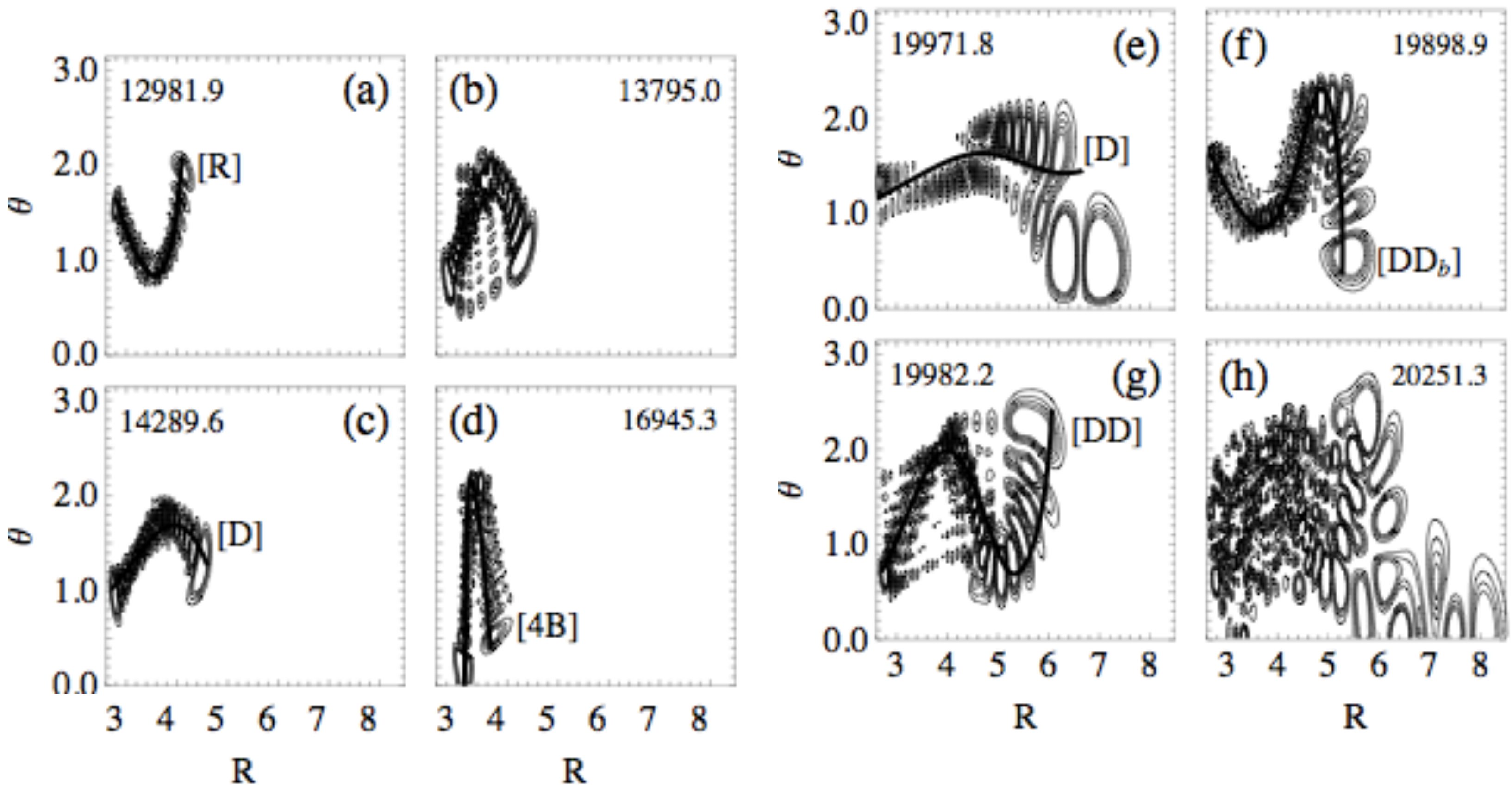
Bound states of HOCl Molecule



E=20,150cm^{-1}



Bound states tend to sit on periodic orbits



Wigner Eisenbud Scattering theory $\hat{H}|\Psi_E\rangle = E|\Psi_E\rangle$

Scattering channels m=1,…,M correspond to different incident angular momentum

Asymptotic incoming and outgoing scattering states are Hankel functions

$$\Psi_E^{asymp}(r) = \sum_m \left(A_m H_m^{(2)}(k_m r) + B_m H_m^{(1)}(k_m r)\right)\exp(im\theta)$$

Scattering state $\Psi_E^{reac}(r)$ in the reaction region is expanded in terms of a discrete set of basis states that are eigenstates of the full Hamiltonian in the reaction region and have zero slope boundary conditions at the interface of the reaction region and the asymptotic region. They are obtained numerically.

Scattering matrix

$$\mathbf{S} = -\mathbf{H}^{(2)} \cdot \left[\mathbf{1}_M + 2iR_{max}\mathbf{v}^\dagger \cdot (\mathbf{H}^{\text{eff}}(E) - E\mathbf{1}_N)^{-1} \cdot \mathbf{x}\right] \cdot \mathbf{H}^{(1)}$$

Here $\mathbf{H}^{(1,2)}$ are $M \times M$ diagonal matrices with elements $H_m^{(1,2)}(k_m R_{max})$, $\mathbf{1}_M$ is the $M \times M$ identity matrix, $\mathbf{v}^\dagger = (\mathbf{H}^{(1)}\mathbf{H}^{(2)})^{-1} \cdot \mathbf{w}^\dagger$ and $\mathbf{x} = \mathbf{w} \cdot \boldsymbol{\beta}$. The matrix $\mathbf{w}$ has elements $\mathbf{w}_{j,m} = \sqrt{\frac{\hbar^2}{2\mu_1}}\xi^*_{j,m}(R_{max})$ and $\boldsymbol{\beta}$ is the imaginary part of $\dot{\mathbf{H}}^{(1)}\mathbf{H}^{(2)}$. We have written the scattering matrix in terms of a non-hermitian effective Hamiltonian given by

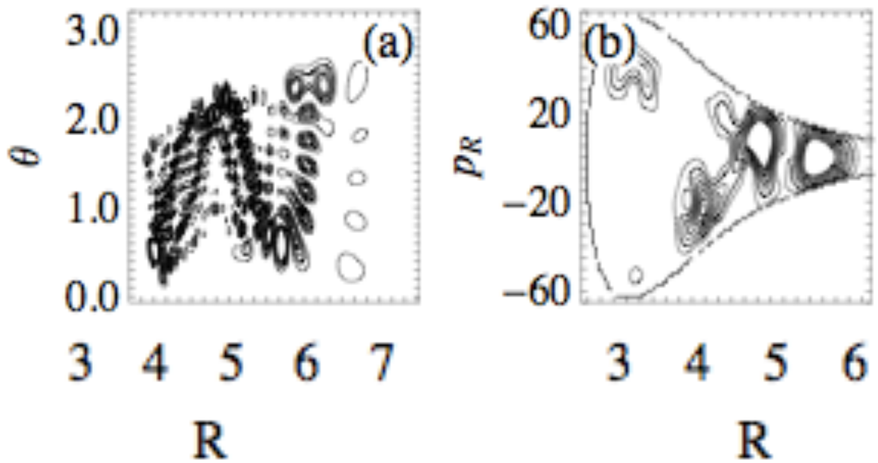
Effective Hamiltonian

$$\mathbf{H}^{\text{eff}}(E) = \mathbf{H}_{\text{in}} - R_{max}\mathbf{w} \cdot \dot{\mathbf{H}}^{(1)} \cdot (\mathbf{H}^{(2)})^{-1} \cdot \mathbf{w}^\dagger$$

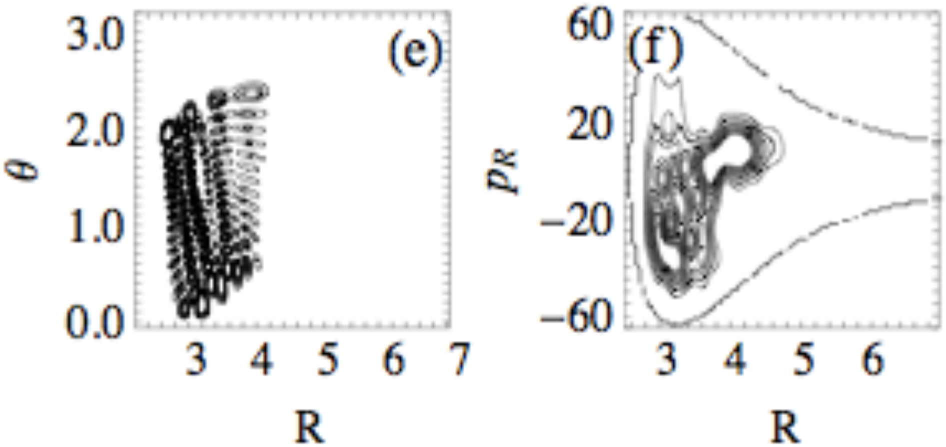
Barr and Reichl, PRA 81, 022707 (2010)

Some long lived quasibound states

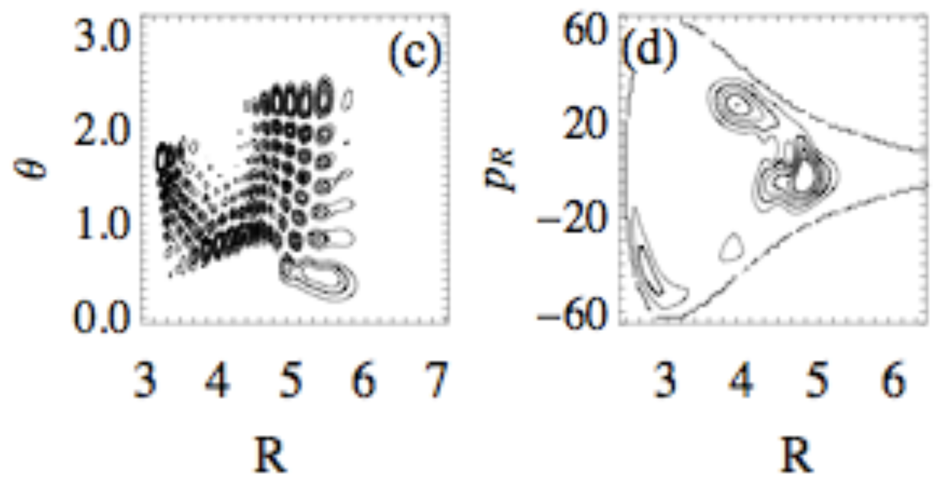
E=20,375 -i1.58x10^{-5}cm^{-1}

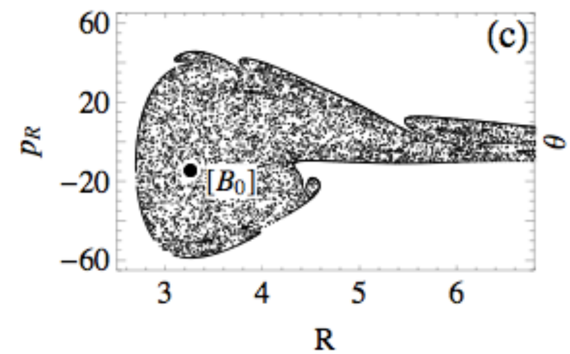


E=20,782-I3.36x10^{-4}cm^{-1}

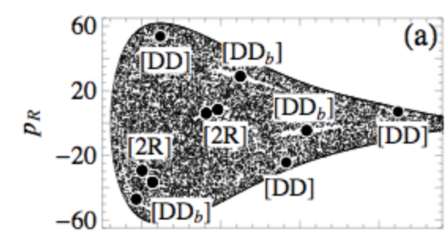


E=20,343 –i8.48x10^{-5}cm^{-1}





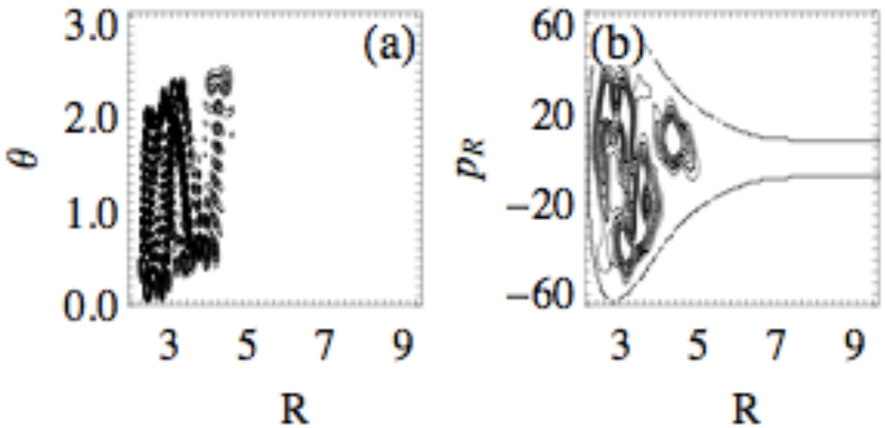
E=21,000



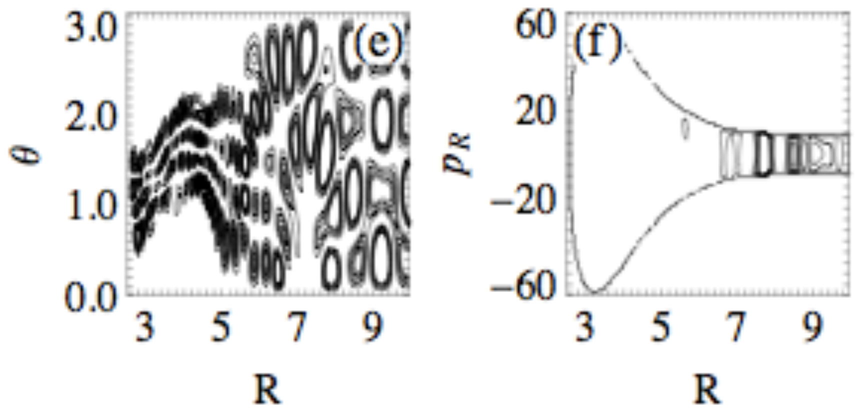
E=20,150

Some shorter lived quasibound states

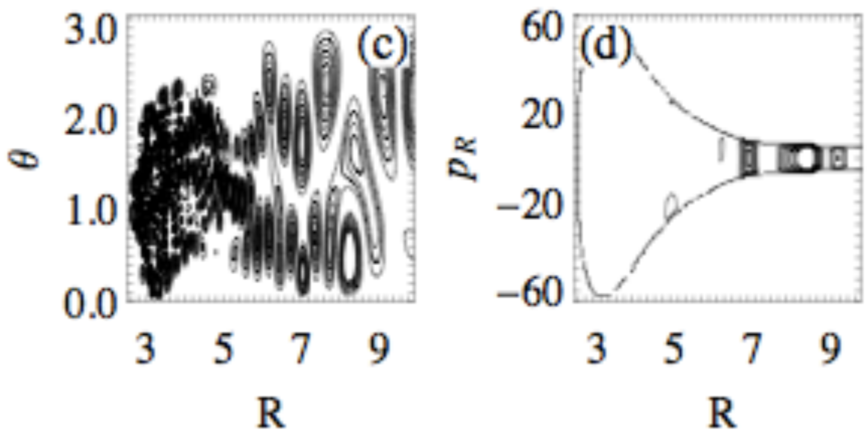
E=20,559 -i1.24x10^{-3}cm^{-1}

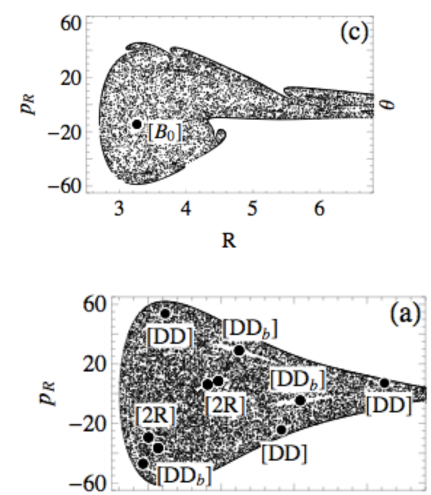


E=20,682-i2.0cm^{-1}

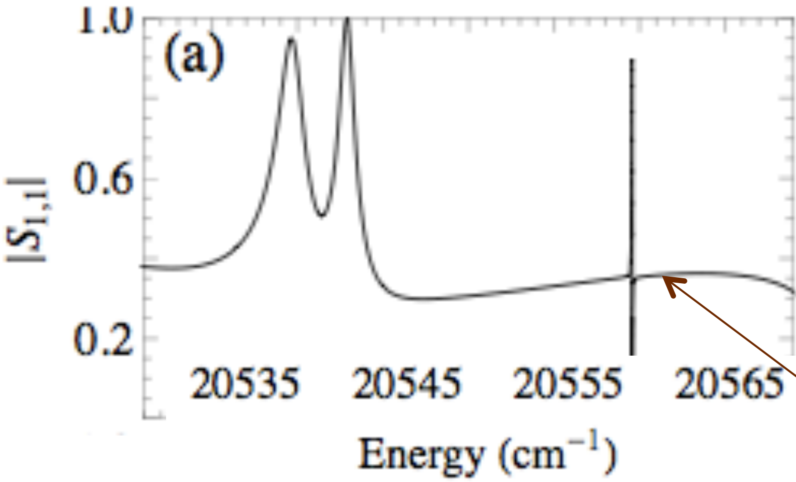


E=20,433 –i0.03cm^{-1}

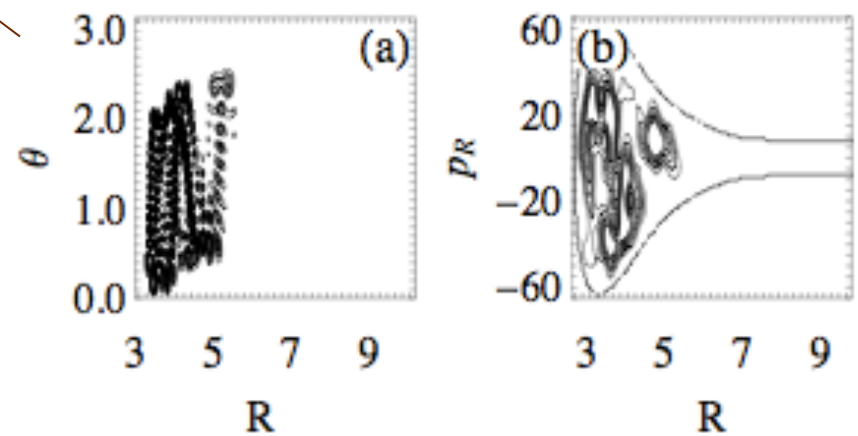




Scattering resonances



E=20,559 -i1.24x10^{-3}cm^{-1}



Conclusion

Chaotic structures can exist in open systems and provide an important platform to support quasibound state formation in the continuum.