Quantum Control in the Classical Limit: Can the Interferences Survive?

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TALK OUTLINE

General Structure and Issues (GENERALLY CONCEPTUAL):

Theme A: Control in closed systems

1. Coherent Control provides a powerful method for controlling molecular processes --- highly successful for isolated molecular processes.

2. Coherent Control is based on the interference between pathways to the same final state. Such control is often manifest via a dependence on eatures such as relative phases of incident laser fields.

3. But there are classical analogs (limits?) that show similar dependences.

a. Are they the same phenomena?

b. If they are the same phenomena then how are we to understand the survival of the quantum interferences in the classical limit?

c. When is the language of classical or of quantum mechanics appropriate? [Note qualitative but not necessarily quantitative.]

4. Can we propose an experiment to examine this quantum to classical transition of the control?

General Structure and Issues:

Theme B: Control in Open systems

1. Coherent Control provides a powerful method for controlling molecular processes --- highly successful for isolated molecular processes.

2. Coherent Control is based on the interference between pathways to the same final state. Such control is generally manifest via a dependence on features such as relative phases of incident laser fields.

3. But in open systems decoherence is expected to cause quantum → classical, and (quantum) control is lost.

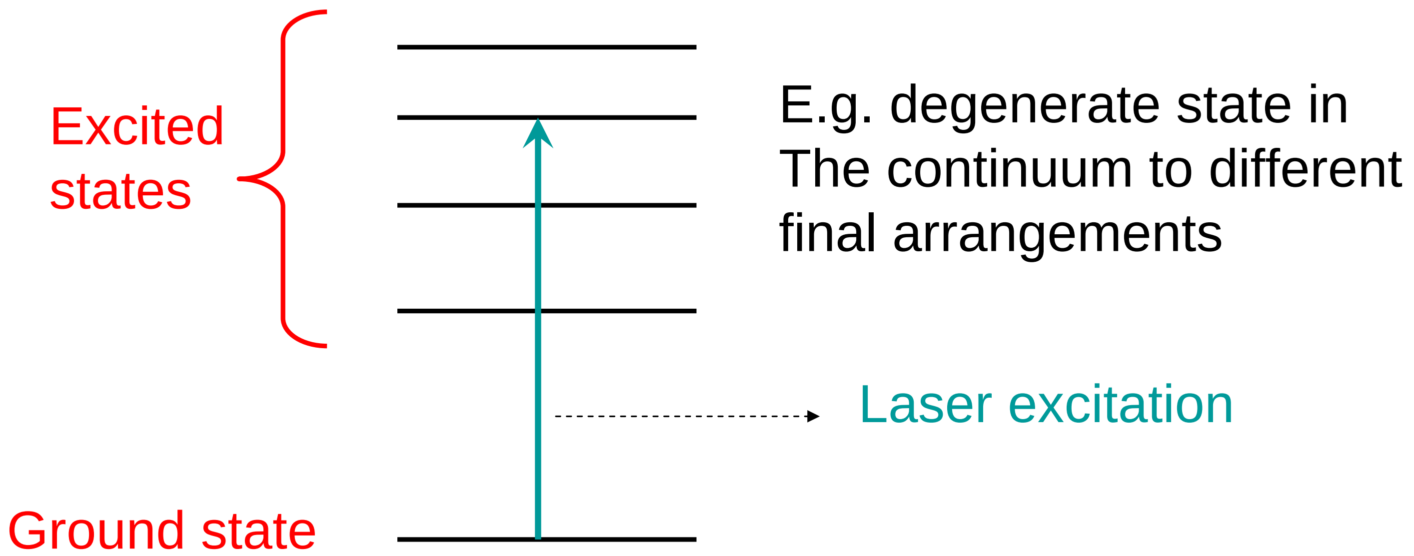
But if classical control still exists then maybe control, as a tool, will not disappear due to decoherence?

4. Can we propose an experiment to examine the outcome of these open system considerations?

EXAMINE BOTH OF THESE THEMES USING ONE EXAMPLE: 1vs2 absorption

Only Theme A today.

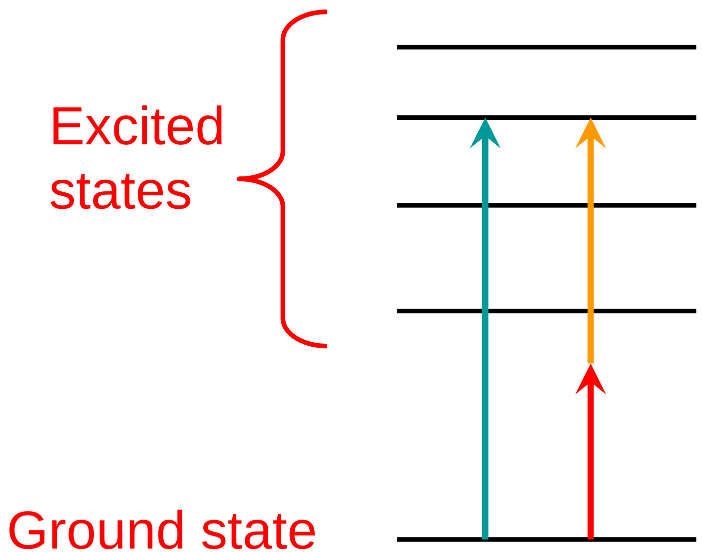
Traditional Photoexcitation in Photochemistry/Photophysics



That is, one route to the final state of interest

Associated Coherent Control Scenario

Coherent Control and "Double Slits" in Photochemistry/Photophysics

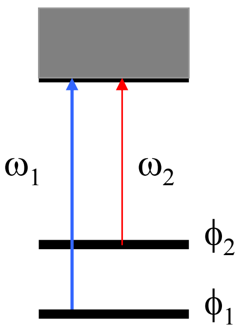


Two (or more) indistinguishable interfering routes to the desired products.

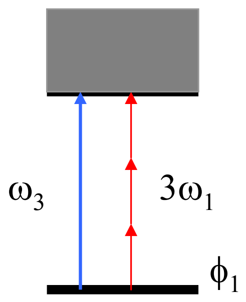
Control laser characteristics ➔

Control Interferences →

Control relative cross sections



Bichromatic Control



1 vs 3

Hence typical successful coherent control scenarios rely upon multiple pathway interferences such as those below. This is the essence of quantum control, and (hopefully) of many of the as-yet-to-be-characterized optimal control schemes.

Common to rely upon analogy of double slit experiment.

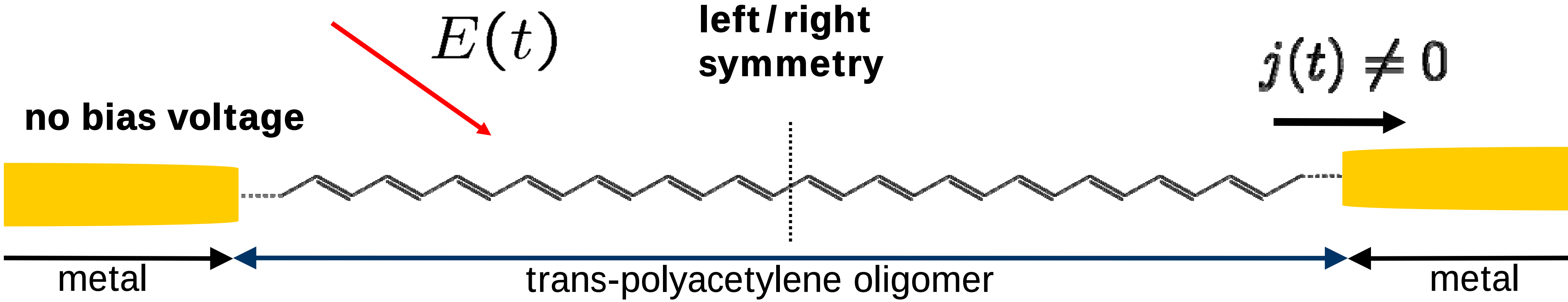
Obvious reminder – double slit interference pattern disappears as hbar → 0.

Consider now as an example of interest: symmetry breaking in driven currents.

That is, omega + 2 omega excitation --- which, e.g.

$$E(t) = \epsilon_\omega \cos(\omega t + \phi_\omega) + \epsilon_{2\omega} \cos(2\omega t + \phi_{2\omega})$$

Laser-induced symmetry breaking



This is a type of rectification:

AC source → **DC response!**

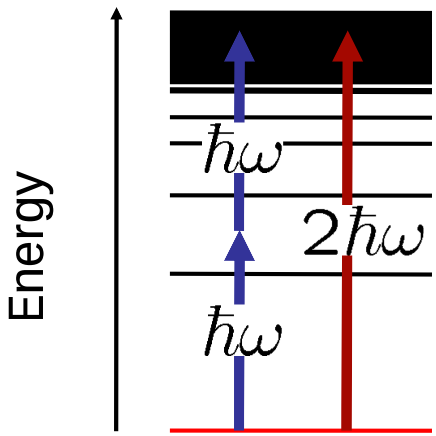
$$\langle j \rangle \sim \overline{E(t)^3} \sim \epsilon_\omega^2 \epsilon_{2\omega} \cos(2\phi_\omega - \phi_{2\omega} + \alpha)$$

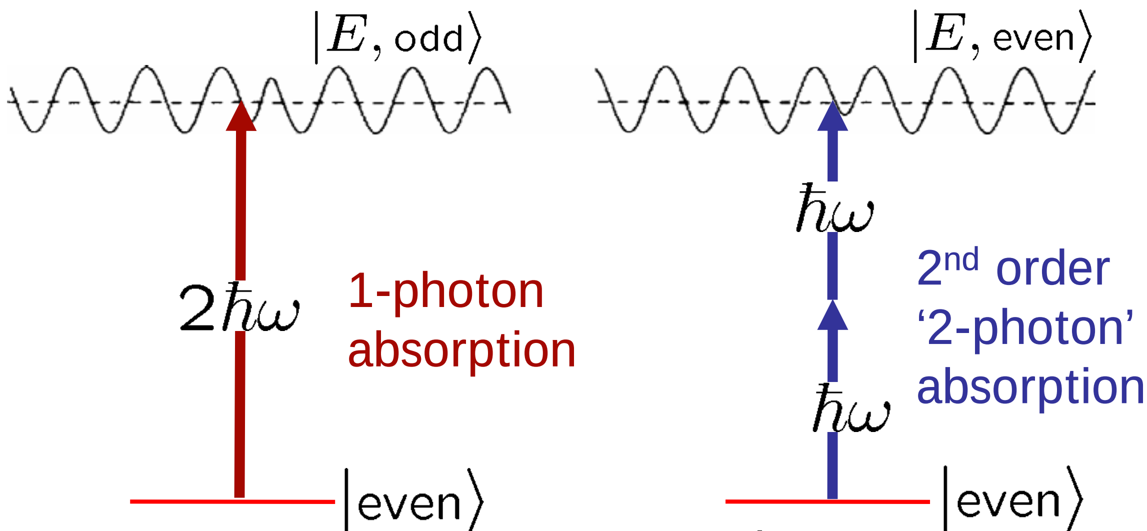
Control current direction by varying relative laser phase.

The 1 vs. 2 scenario and symmetry breaking

$$E(t) = \epsilon_\omega \cos(\omega t + \phi_\omega) + \epsilon_{2\omega} \cos(2\omega t + \phi_{2\omega})$$

$$\langle j \rangle \sim \overline{E(t)^3} \sim \epsilon_\omega^2 \epsilon_{2\omega} \cos(2\phi_\omega - \phi_{2\omega} + \alpha)$$

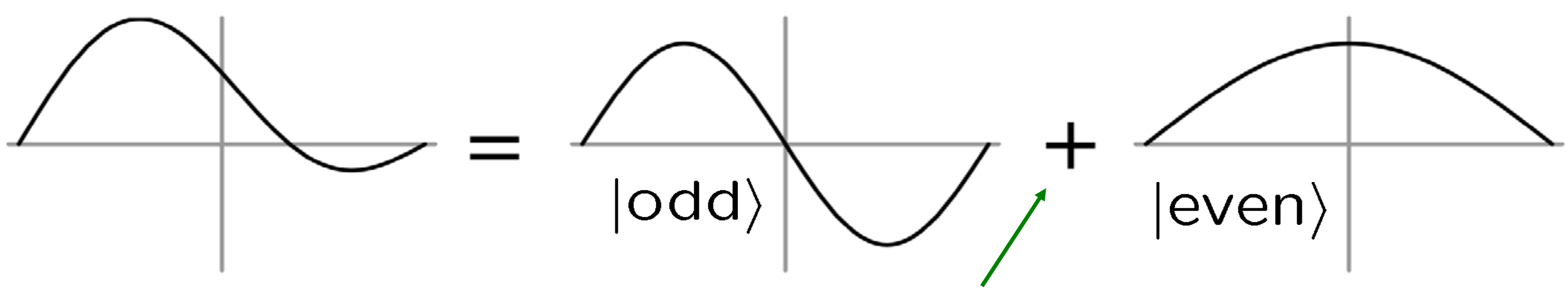




couples states with **opposite** parity

couples states with the **same** parity

Final State:



Not a parity eigenstate:
Broken symmetry

Laser controllable

The 1 vs. 2 scenario: role of interference

I.e, :

After the ω + 2ω field, the excitation left on the system:

$$|\Psi(t)\rangle = \underbrace{c_1}_{\text{from the 1-photon absorption}} \mathrm{e}^{-iEt/\hbar}|E,\mathrm{odd}\rangle + \underbrace{c_2}_{\text{from the 2-photon absorption}} \mathrm{e}^{-iEt/\hbar}|E,\mathrm{even}\rangle$$

Net photoinduced momentum:

$$\langle p\rangle = |c_1|^2\langle E,\mathrm{odd}|p|E,\mathrm{odd}\rangle + |c_2|^2\langle E,\mathrm{even}|p|E,\mathrm{even}\rangle + \underbrace{2\mathrm{Re}\{c_1c_2^*\langle E,\mathrm{even}|p|E,\mathrm{odd}\rangle\}}_{\text{Interference contribution}}$$

Direct terms

Only the interference contribution survives:

$$\langle p\rangle = 2\mathrm{Re}\{c_1c_2^*\langle E,\mathrm{even}|p|E,\mathrm{odd}\rangle\} \sim A\cos(\phi_{2\omega} - 2\phi_\omega + \alpha)$$

Laser control: **Changing the relative phase of the lasers changes the magnitude and sign of the current.**

E.g. done exptly in quantum wells by Corkum's group, PRL 74, 3596 (1995)

The classical correspondence issue--

Quantum interpretation of laser-induced symmetry breaking

- Quantum interference
- Parity

These concepts do not have a classical analogue and the effect seems completely quantum mechanical.

However

An $\omega + 2\omega$ field generates phase-controllable symmetry breaking in completely classical systems as well!

See, for example,
S. Flach, O. Yevtushenko, and Y. Zolotaryuk, Phys. Rev. Lett. 84, 2358 (2000)

Or papers on classical ratchet transport, e.g. Gong and Brumer

How are the classical and quantum versions of symmetry breaking related, if at all?

The classical correspondence issue--

Quantum interpretation of laser-induced symmetry breaking

Quantum interference

Parity

These concepts do not have a classical analogue and the effect seems completely quantum mechanical.

How are the classical and quantum versions of symmetry breaking related, if at all?

What happened to the double slit analog where, no doubt, the interference terms vanish in the classical limit?

The Earlier Strategy (Franco and Brumer, PRL 97,040402, 2006)

Quantum Control $\xrightarrow{\text{Classical Limit}}$?

Analytically consider the quantum-to-classical transition of the net dipole induced by an ω + 2ω field in a quartic oscillator

$$H = \frac{p^2}{2m} + \frac{1}{2} m\omega_0^2 x^2 + \lambda m \epsilon x^4 - qxE(t)$$

Simplest model with well-defined classical analog wherein induced symmetry breaking is manifest

(a) time-dependent perturbation theory in the Heisenberg picture that admits an analytic classical ($\hbar \to 0$) limit in the response of the oscillator to the field.

(b) Anharmonicities included to minimal order in a multiple-scale approximation; interaction with the radiation field is taken to third order.

Advantages

1. The result of the perturbation is independent of the initial state

$$\hat{O}_{\mathsf{H}}(\hat{x}_{\mathsf{H}}(t), \hat{p}_{\mathsf{H}}(t)) = U^{\dagger}(t)\hat{O}(\hat{x}, \hat{p})U(t)$$

2. The classical limit of the solution coincides with true classical result.

Osborn and Molzahn, Ann. Phys. 241, 79-127 (1995)

$$\hat{O}_{\mathsf{H}}(\hat{x}_{\mathsf{H}}(t), \hat{p}_{\mathsf{H}}(t)) \xrightarrow[\hbar \to 0]{\hat{x}\to x, \hat{p}\to p} O(x(t), p(t))$$

Main drawbacks

1. Operators and their algebraic manipulation (not always easy)

2. One needs to begin with a system for which an exact solution in Heisenberg picture exists (e.g. harmonic oscillator)

Calculation

Symmetry breaking is characterized through the long-time average of the position operator in Heisenberg representation

$$\overline{\widehat{x}_{\mathrm{H}}(t)} = \overline{\widehat{U}(t)^{\dagger}\widehat{x}\widehat{U}(t)}$$

We employ the Interaction picture where

$$\widehat{U}(t) = \widehat{U}_0(t)\widehat{U}_{\mathrm{I}}(t)$$

Evolution operator in the absence of the field

Captures the effects induced by the field

This splits the problem into two steps

$$\widehat{x}_{\mathrm{I}}(t) = \widehat{U}_0^{\dagger}(t)\widehat{x}\widehat{U}_0(t)$$

Perturbative analysis to include the oscillator anharmonicities; C. M. Bender and L. M. A Bettencourt, Phys. Rev. Lett. 77, 4114 (1996)

$$\widehat{x}_{\mathrm{H}}(t) = \widehat{U}_{\mathrm{I}}^{\dagger}(t)\widehat{x}_{\mathrm{I}}(t)\widehat{U}_{\mathrm{I}}(t)$$

Subsequent perturbation to incorporate the effect of the field (to third order in the field)

Calculation-II

The perturbative expansion for $\hat{U}_{\mathrm{I}}(t)$ is given by

$$\hat{U}_{\mathrm{I}}(t) = \hat{U}_{\mathrm{I}}^{(0)} + \hat{U}_{\mathrm{I}}^{(1)}(t) + \hat{U}_{\mathrm{I}}^{(2)}(t) + \hat{U}_{\mathrm{I}}^{(3)}(t) + \cdots$$

$$\hat{U}_{\mathrm{I}}^{(0)}(t) = \hat{1}$$

zeroth order term

$$\hat{U}_{\mathrm{I}}^{(n)}(t) = -\frac{i}{\hbar}\int_0^t \mathrm{d}t' \, \hat{V}_{\mathrm{I}}(t')\hat{U}_{\mathrm{I}}^{(n-1)}(t') \ (n \geq 1)$$

***n*th order correction**

$$\hat{V}_{\mathrm{I}}(t) = -q\hat{x}_{\mathrm{I}}(t)E(t)$$

The result up to third order in the field (34370 Oscillatory operator terms)

$$\hat{x}_{\mathrm{H}}(t) = \hat{x}_{\mathrm{I}} + \hat{U}_{\mathrm{I}}^{(1)\dagger}\hat{x}_{\mathrm{I}}\hat{U}_{\mathrm{I}}^{(1)} + \left[\hat{U}_{\mathrm{I}}^{(0)\dagger}\hat{x}_{\mathrm{I}}\left(\hat{U}_{\mathrm{I}}^{(1)\dagger} + \hat{U}_{\mathrm{I}}^{(2)} + \hat{U}_{\mathrm{I}}^{(3)}\right) + \hat{U}_{\mathrm{I}}^{(1)\dagger}\hat{x}_{\mathrm{I}}\hat{U}_{\mathrm{I}}^{(2)} + \mathrm{H.c.}\right]$$

Note that the terms : $\hat{U}_{\mathrm{I}}^{(i)\dagger}(t)\hat{x}_{\mathrm{I}}(t)\hat{U}_{\mathrm{I}}^{(j)}(t) + \mathrm{H.c}$
describe the contribution to the dipole coming from the interference between an i-th order and a j-th order optical route

Calculation-III

Which terms contribute to symmetry breaking?

1. Only those terms allowed by the symmetry of the initial state

$\langle \hat{x}_{\mathrm{H}}(t) \rangle = \mathrm{Tr}(\hat{x}_{\mathrm{H}}(t)\hat{\rho}_0)$ $\quad$ $\langle x(t) \rangle_{\mathrm{C}} = \mathrm{Tr}(x(t)\rho_0(x,p))$

Parity $\quad \xleftrightarrow{?} \quad$ **Reflection symmetry**

Using reflection symmetry and not parity:

$\hat{U}_{\mathrm{I}}^{(i)\dagger}(t)\hat{x}_{\mathrm{I}}(t)\hat{U}_{\mathrm{I}}^{(j)}(t)$ $\quad$ $i+j$ odd $\quad$ **have a non-zero contribution to the trace**

Symmetry breaking comes from the interference between an even-order and an odd-order response to the field

2. Only those terms that have a zero-frequency (DC) component

The remaining terms, with a residual frequency dependence, average out to zero in time

Final Result

Operator expression for the net dipole:

$$\overline{\hat{x}_{\mathrm{H}}(t)} = \overline{\hat{x}_{\mathrm{I}}(t)\hat{U}_{\mathrm{I}}^{(3)}(t)} + \overline{\hat{U}_{\mathrm{I}}^{(1)\dagger}(t)\hat{x}_{\mathrm{I}}(t)\hat{U}_{\mathrm{I}}^{(2)}(t)} + \text{ H.c.},$$
$$= \frac{q^3\epsilon_\omega^2\epsilon_{2\omega}}{16m^2\omega_0}\hat{\Gamma}(\hat{\mathcal{H}},\omega,\lambda,\hbar)\cos(2\phi_\omega - \phi_{2\omega}),$$

where $\hat{\mathcal{H}} = \frac{\hat{p}^2}{2m} + \frac{1}{2}m\omega_0^2\hat{x}^2$

Some properties:

1. **The sign and magnitude of the dipole can be manipulated by varying the relative phase between the frequency components of the laser -- irrespective of the initial state.**

2. **In the zero-anharmonicity limit all symmetry breaking effects are lost**

$$\lim_{\lambda\to 0}\hat{\Gamma}(\hat{\mathcal{H}},\omega,\lambda,\hbar) = 0$$

It is precisely because of the anharmonicities that the system can exhibit a nonlinear response to the laser, mix the frequencies of the field and generate a zero harmonic component in the response.

The Classical Limit

The $\hbar \to 0$ limit is analytic and nonzero, despite the fact that individual perturbative terms can exhibit singular behavior as $\hbar \to 0$

$$\overline{\hat{x}_{\mathrm{H}}}(\hat{\mathcal{H}},\omega,\lambda,\hbar) \xrightarrow[\hbar\to 0]{\hat{x}\to x,\hat{p}\to p} \overline{x}(\mathcal{H},\omega,\lambda) = \frac{q^3\epsilon_\omega^2\epsilon_{2\omega}}{16m^2\omega_0}\Gamma_{\mathrm{C}}(\mathcal{H},\omega,\lambda)\cos(2\phi_\omega-\phi_{2\omega})$$

The field induced interferences responsible for symmetry breaking survive in the classical limit and are the source of classical control.

Quantum Corrections

In the quantum case, the net dipole can be written as

$$\overline{\hat{x}_{\mathrm{H}}(t)} = \overline{\hat{x}_{\mathrm{C}}(t)} + \overline{\hat{x}_{\mathrm{q}}(t)}$$

$$\overline{\hat{x}_{\mathrm{C}}(t)} = \lim_{\hbar \to 0} \overline{\hat{x}_{\mathrm{H}}(t)}$$

$\hbar$-independent classical-like contribution

Quantum Corrections

The nature of the quantum corrections can be associated with the $\hbar$ dependence of the resonance structure of the oscillator

Resonances sampled by the ω +2ω field

Quantum Case

$$\frac{1}{2}(\omega_0 + \xi(\hat{\mathcal{H}} \pm \frac{\hbar\omega_0}{2}))$$
$$\omega_0 + \xi(\hat{\mathcal{H}} \pm \frac{\hbar\omega_0}{2})$$
$$\omega_0 + \xi(\hat{\mathcal{H}} \pm \hbar\omega_0)$$
$$2(\omega_0 + \xi(\hat{\mathcal{H}} \pm \hbar\omega_0))$$

$$\hbar \longrightarrow 0$$

Classical Case

$$\frac{1}{2}(\omega_0 + \xi\hat{\mathcal{H}})$$
$$\omega_0 + \xi\hat{\mathcal{H}}$$
$$2(\omega_0 + \xi\hat{\mathcal{H}})$$

$$\xi = \frac{3\epsilon\lambda}{m\omega_0^3}$$

The fine $\hbar$-dependent structure can change the magnitude and sign of the effect

Quantum Corrections-II

Different initial states emphasize the classical part of the solution or the quantum corrections depending on the nature of the state

$$\langle \hat{x}_{\mathrm{H}}(t)\rangle = \mathrm{Tr}([\hat{x}_{\mathrm{C}}(t) + \hat{x}_{\mathrm{q}}(t)]\hat{\rho}_0)$$

Classical **Quantum solution with increasing energy**

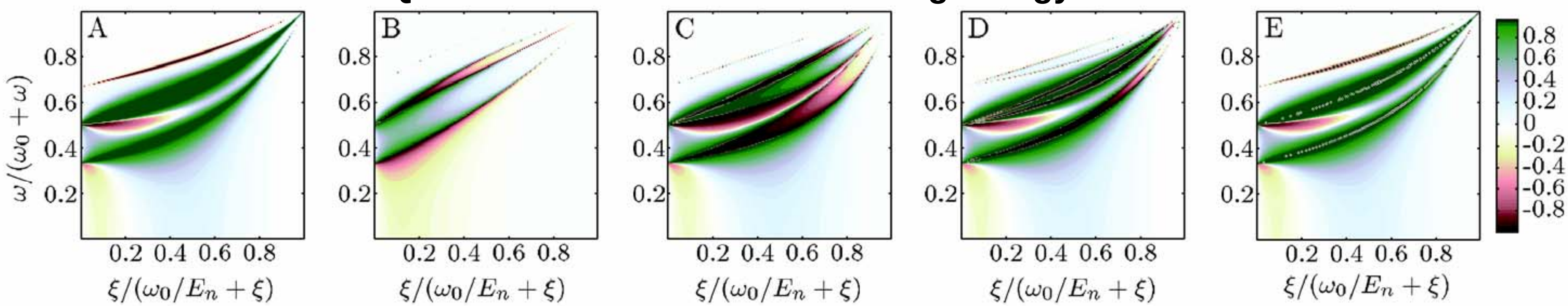


FIG. 2 (color). Net dipole induced by an $\omega + 2\omega$ field in a quantum and classical anharmonic oscillator. The contour plots show the dependence of $\frac{2}{\pi}\arctan[(16m^2\omega_0^4E_n/[q^3\epsilon_\omega^2\epsilon_{2\omega}\cos(2\phi_\omega - \phi_{2\omega})])\overline{\langle\hat{x}_{\mathrm{H}}(t)\rangle}]$ on the anharmonicities of the potential (x axis) and the frequency of the field (y axis). The system is initially prepared in the nth eigenstate of $\hat{\mathcal{H}}$ with energy $E_n = \hbar\omega_0(n+\frac{1}{2})$. Panel (a) shows the classical part of the solution which in this parameter space is the same for all E_n. The remaining panels show the full quantum-mechanical solution for: (b) $n = 0$; (c) $n = 1$; (d) $n = 3$; and (e) $n = 40$. The color code is given at the far right.

Classical limit reached as quantum state level increased.

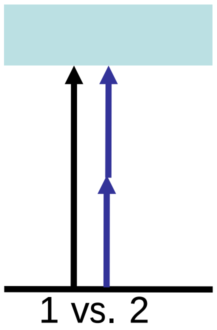
Note then --- the 1 vs 2 scenario can persist classically ---

Spatial symmetry breaking using $\omega + 2\omega$ fields

Related experimental and theoretical references.

Theoretically:

Note the general phenomenon can be accounted for from:



1) The coherent control perspective of interfering optical pathways

G. Kurizki, M. Shapiro and P. Brumer Phys. Rev. B **39**, 3435 (1989);
M. Shapiro and P. Brumer, Principles of the Quantum Control of Molecular Processes (Wiley, 2003)

2) Nonlinear response theory arguments

I. Franco and P. Brumer Phys. Rev. Lett. **97**, 040402 (2006);
Goychuk and P. Hänggi, Europhys. Lett. **43**, 503 (1998)

$$\langle \mu(t) \rangle = \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \cdots$$

3) Space-time symmetry analyses of the equations of motion

I. Franco and P. Brumer J. Phys. B 41, 074003 (2008)
S. Flach, O. Yevtushenko, and Y. Zolotaryuk, Phys. Rev. Lett. **84**, 2358 (2000)

$$\rho_{\mathsf{C/W}}(\mathbf{x}, \mathbf{p}, t_0) \rightsquigarrow \rho_{\mathsf{C/W}}(\mathbf{x}, \mathbf{p}, t)$$

$$\mathcal{T}_\alpha \rho_{\mathsf{C/W}}(\mathbf{x}, \mathbf{p}, t_0) \rightsquigarrow \rho^{(\alpha)}_{\mathsf{C/W}}(\mathbf{x}, \mathbf{p}, t) = \mathcal{T}_\alpha \rho_{\mathsf{C/W}}(\mathbf{x}, \mathbf{p}, t)$$

Asides:

1. hence if you see (experimentally) dependence of features -on relative laser phase, this does not necessarily imply that it is quantum effect.

2. It connects to classical language “here and there”. E.g. early work and language of Bucksbaum/Corkum

3. On (often major) quantitative difference in classical vs. quantum response functions --- see several papers by Roger Loring.

Key conceptual issue:

So the question becomes --- is the quantum interference, and if it is, how/why does it survive in the classical limit? Is the standard analogy with the double slit in need of supplementing?

Then: can we do an experiment that shows these features clearly?

Return to origin of the symmetry breaking

I.e, :

After the ω + 2ω field, the excitation left on the system:

$$|\Psi(t)\rangle = c_1 e^{-iEt/\hbar}|E,\mathrm{odd}\rangle + c_2 e^{-iEt/\hbar}|E,\mathrm{even}\rangle$$

from the 1-photon absorption   from the 2-photon absorption

c_1 proportional to $\epsilon_\omega^{\ 2}$

c_2 proportional to $\epsilon_{2\omega}$

Crucial difference from the double slit analog is that the interference term is driven by external fields.

Significantly --- driven interference terms need not vanish in the classical limit

Analysis substantiated by recent Heisenberg representation analysis of interference processes (I. Franco, Ph.D. Dissertation, U of Toronto, Franco and Brumer, in prep) – not a competition between terms

Proposed experimental examination of the quantum – classical transition

(M. Spanner and P. Brumer, in prep)

Consider an atom interacting with a longitudinally shaken 1D optical lattice. Hamiltonian is:

$$H = \frac{P^2}{2m} + U f_1(t)\cos(2kx - \beta f_2(t)),$$

$$f_2(t) = \cos(\omega t + \phi_{\mathrm{rel}}) + s\cos(2\omega t),$$

by rescaling the coordinates as

$$\begin{aligned} \theta &= 2kx, \\ P_\theta &= P(2k/\omega m), \\ \tau &= \omega t, \end{aligned}$$

and defining

$$\mathcal{U} = (2k/\omega)^2 (U/m),$$

the Hamiltonian becomes

$$\mathcal{H} = \frac{{P_\theta}^2}{2} + \mathcal{U} f_1(\tau)\cos(\theta - \beta f_2(\tau)),$$

Gives Schrodinger equation with effective, controllable, "hbar"

$$i\hbar_e \frac{\partial \Psi(\theta,\tau)}{\partial \tau} = \left[-\frac{\hbar_e^2}{2}\frac{\partial^2}{\partial\theta^2} + \mathcal{U} f_1(\tau)\cos(\theta - \beta f_2(\tau))\right]\Psi(\theta,\tau),$$

$$\hbar_e = \hbar(2k)^2/(\omega m)$$

Related to standard dipole driven form by defining:

$$z = \theta - \beta f_2(\tau) \tag{10}$$

and employ the gauge transformation

$$\Psi'(z, \tau) = e^{izA(\tau)}\Psi(z, \tau), \tag{11}$$

where

$$A(\tau) = -\beta \dot{f}_2(\tau). \tag{12}$$

The Schrödinger equation then takes the form $i\hbar_e \partial_\tau \Psi'(z, \tau) = \mathcal{H}'(\tau)\Psi'(z, \tau)$, where

$$\mathcal{H}'(\tau) = \frac{P_z^2}{2} + \mathcal{U} f_1(\tau)\cos(z) + zE(\tau), \tag{13}$$

P_z is the momentum conjugate to z, and

$$E(\tau) = \beta \ddot{f}_2(\tau). \tag{14}$$

$$f_2(t) = \cos(\omega t + \phi_{\rm rel}) + s\cos(2\omega t),$$

Sample numerical results --- (Spanner and Brumer, in prep)

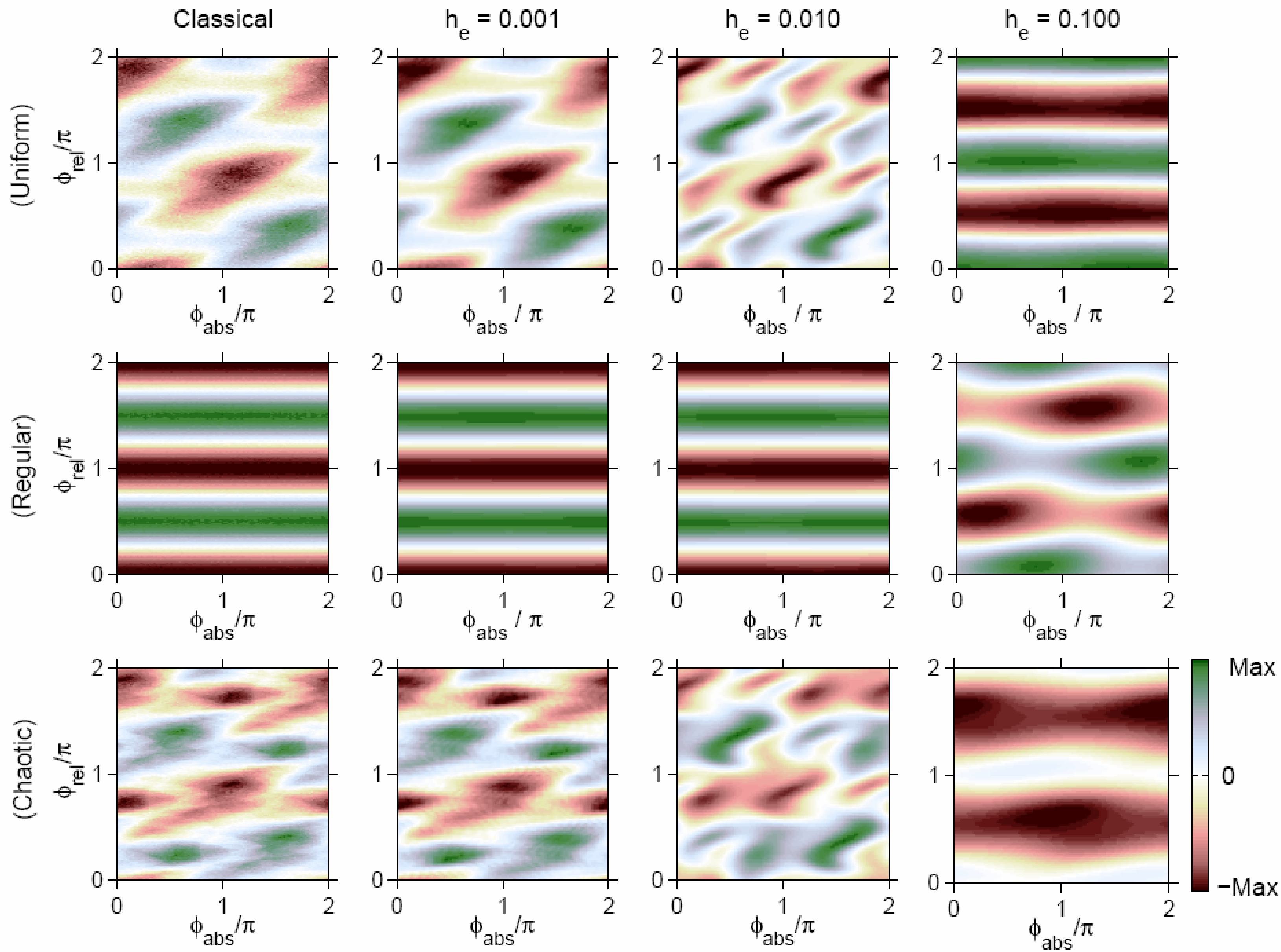
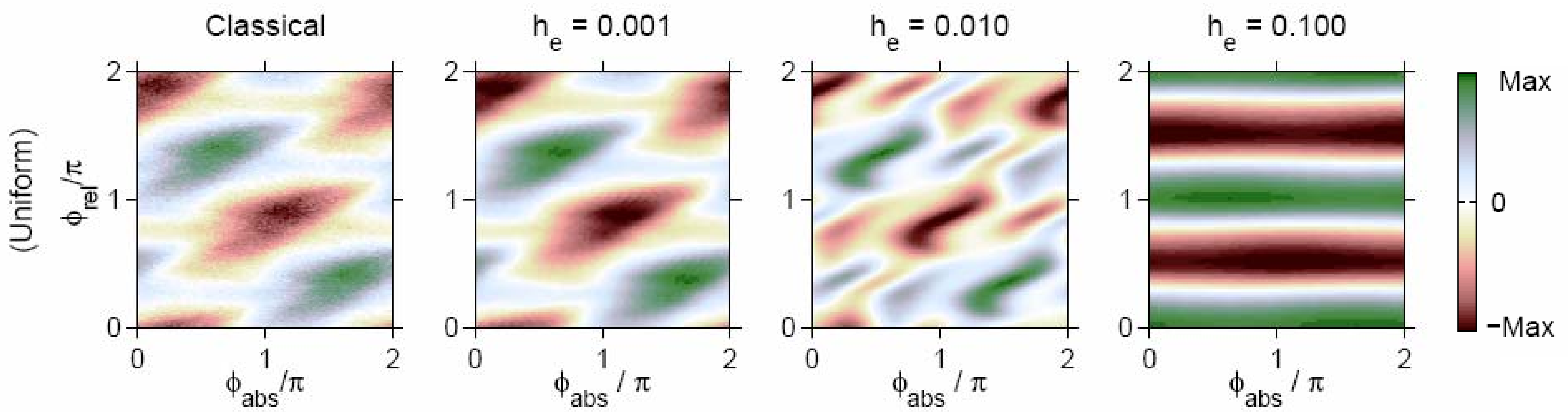


FIG. 1: Control dynamics. These plots show the final average momentum for the classical and quantum systems. The top row is for the uniform initial state, while the middle and bottom row correspond to the regular and chaotic initial state. The first column plot the classical results, while the remaining columns plot the quantum results for three values of $\hbar_e = 0.001$, 0.01, and 0.1.

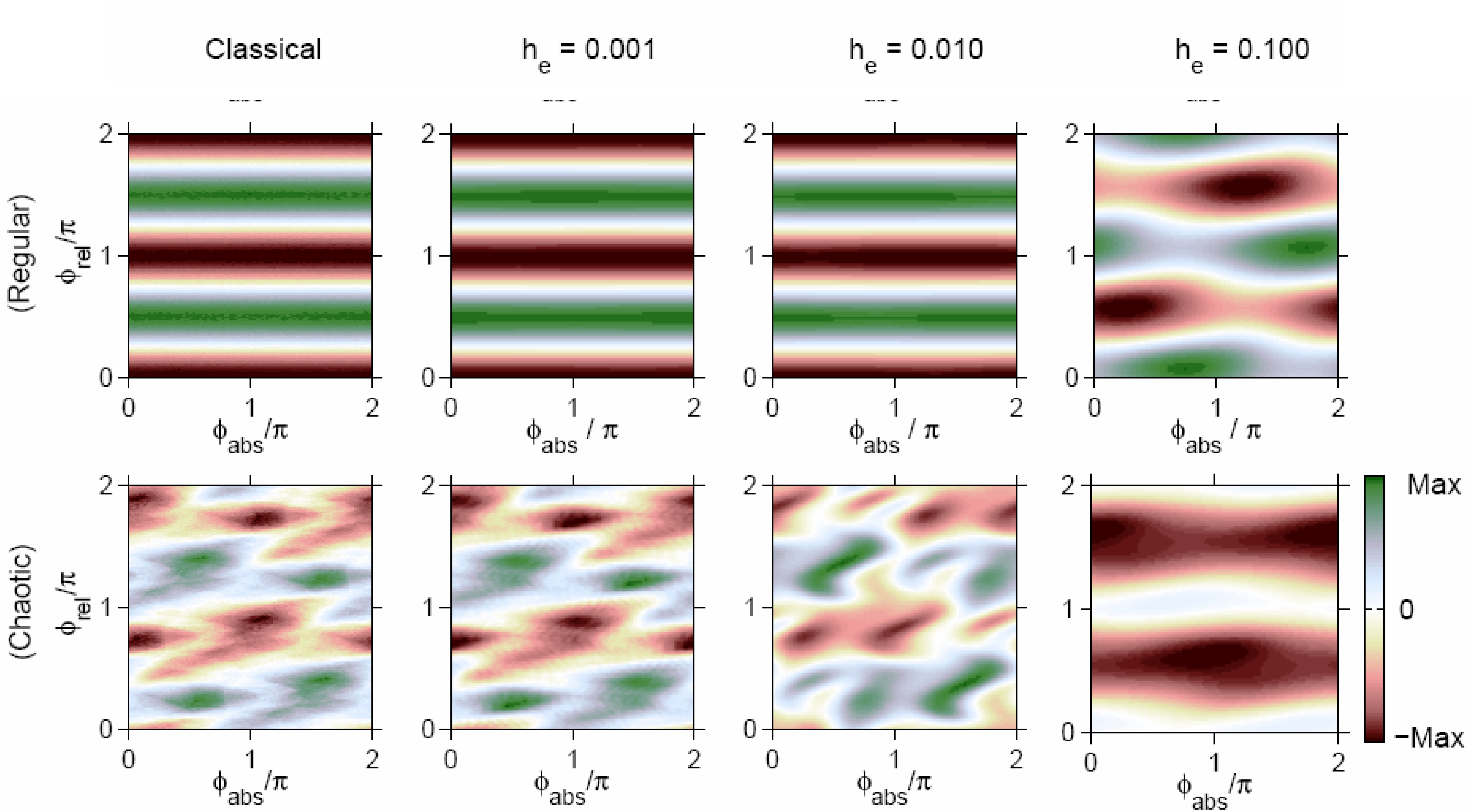
Focus on full calculation:



Evidently:

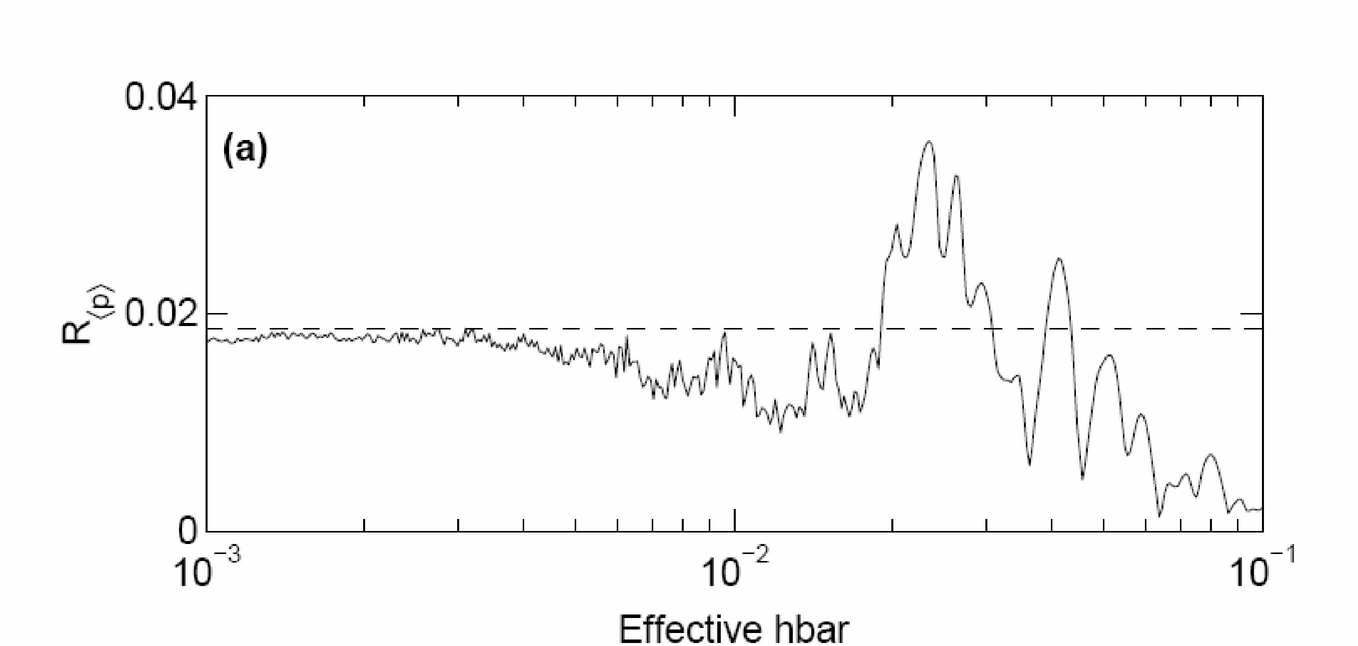
a. Quantum goes over to classical as h_e goes to zero --- i.e. the classical limit is indeed classical mechanics, which does show nice control.

b. The fully quantum shows no dependence on the absolute phase, unlike the small h_e and classical cases --- origin is in the chaotic region that is sensitive to the detailed initial conditions:



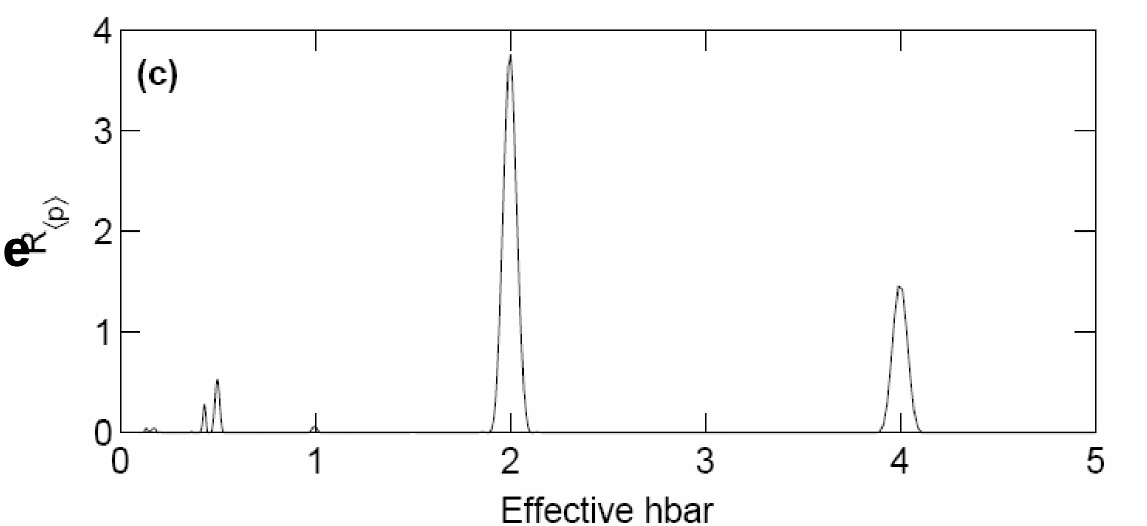
note dependence on Φ_{abs} arising entirely from the chaotic region, which eventually disappears in the quantum limit. Would be enlightening to see experimentally!

And range of control? $R_{\langle p \rangle} = \max\{\langle P_\theta \rangle)\} - \min\{\langle P_\theta \rangle)\}$



Solid is quantum;
Dashed is classical'
Essentially same
order of magnitude.

Although the situation
Can be quite different in
the strong quantum regime
due to highly resonant
contributions.



General Structure and Issues:

Theme B: Control in Open systems

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2. Coherent Control is based on the interference between pathways to the same final state. Such control is generally manifest via a dependence on features such as relative phases of incident laser fields.

3. But in open systems decoherence is expected to cause quantum → classical, and (quantum) control is lost.

But if classical control still exists then maybe control, as a tool, will not disappear due to decoherence.

4. Can we propose an experiment to examine the outcome of these open system considerations?

EXAMINE BOTH OF THESE THEMES USING ONE EXAMPLE: 1vs2 absorption

But this is another talk….

Summary:

Control can survive into the classical limit. It is qualitatively the same phenomenon, but can differ greatly quantitatively.

Control IS due to interference effects, but they can differ from the double slit paradigm insofar as they can be field driven. Such field driven interference terms may survive to the classical limit.

(Some control cases, e.g. collisional control scenarios based on entanglement will lose control in the classical limit – not driven)

Optical lattice experiment proposed to examine the quantum to classical transition.

Decoherence shows little effect on control for small hbar systems. Further studies on larger hbar in progress.

And thanks to NSERC as well as:

Ignacio Franco –
Univ of Toronto graduate student. Now postdoc at Northwestern.

and Michael Spanner –
postdoc at University of Toronto, now at NRC, Ottawa