# Coherent Control: from concept to reality

General objectives: Control of future events.

Tools: Use controlled quantum interference between material waves. We access the same final state using more than one pathway. Lacking the "which way" information these pathways interfere.

But interference is not enough. In order to achieve control we need to "tune" this interference, and this is done with a photon.

A possible objective: to control the outcome of chemical reactions.

$$\text{H-O + D} \xleftarrow{\hspace{2cm}} \text{H}\overset{\text{O}}{\diagup\diagdown}\text{D} \xrightarrow{\hspace{2cm}} \text{H + O-D}$$

Cleave the **O-D** bond   Cleave the **O-H** bond
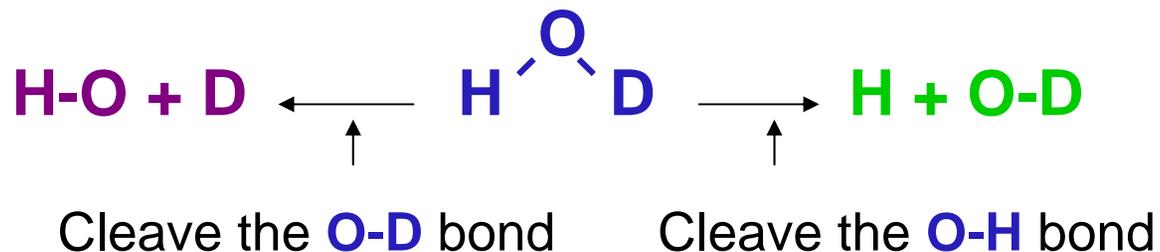
Or in greater generality:

$$\text{A-B + C} \xleftarrow{\hspace{2cm}} \text{A}\overset{\text{B}}{\diagup\diagdown}\text{C} \xrightarrow{\hspace{2cm}} \text{A + B-C}$$

Cleave the **B-C** bond   Cleave the **A-B** bond
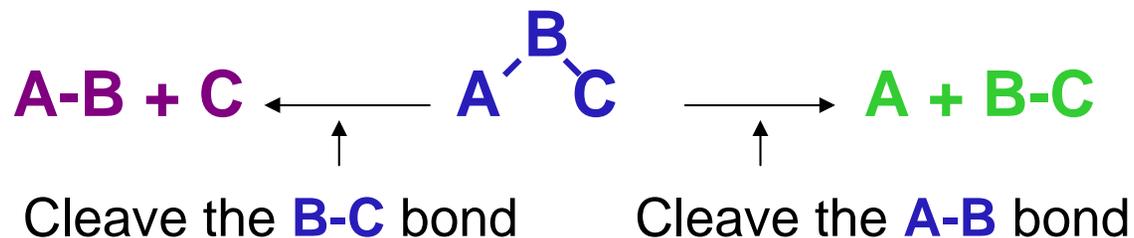
where **A**, **B**, or **C** can be any atom or any group of atoms.

photodissociation
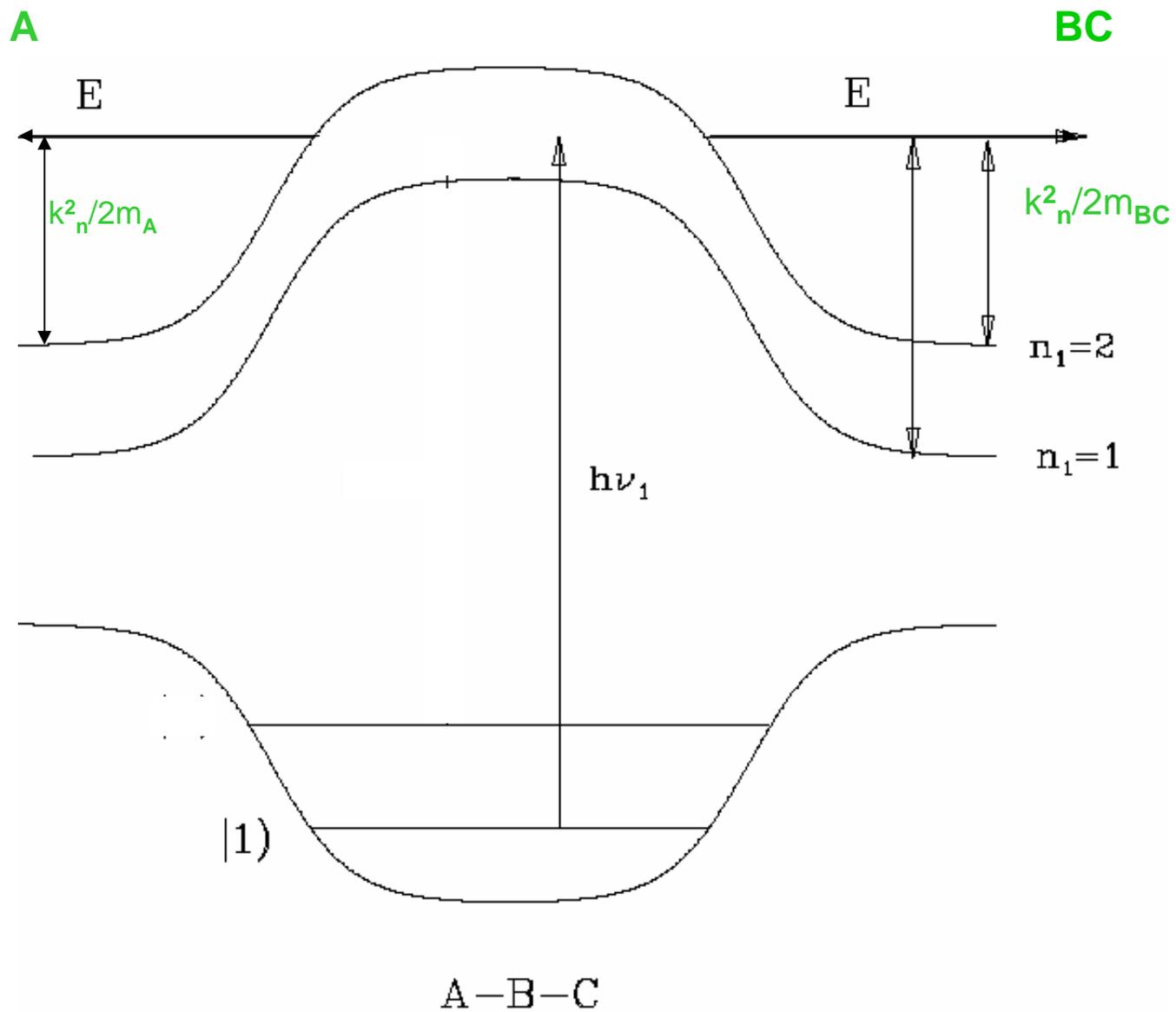
Photo-exciting a Molecule with a Pulse of Light

The matter-radiation Schrödinger equation

$$i\hbar \frac{d\Psi(t)}{dt} = H(t)\Psi(t),$$

where $H(t) = H_M + H_{MR}$ is the total Hamiltonian, H_M is the material Hamiltonian, and H_{MR} is the matter-radiation interaction.

in the dipole approximation,

$$H_{MR} = -\mathrm{d} \cdot E_R(z,t)$$

where $\quad \mathrm{d} \equiv \sum_j q_j r_j$.

$E_R(r,t)$ is **a Pulse of Light**

$$E_R(z,t) = \hat{\epsilon} \int_{-\infty}^{\infty} d\omega \epsilon(\omega) \exp(-i\omega\tau)$$

where $\quad \tau \equiv t - z/c \quad$ is the "retarded time",

$$\epsilon(\omega) = |\epsilon(\omega)|e^{i\phi(\omega)},$$

where $\phi(\omega)$ are frequency dependent

phases. Because of the reality of the field

$$\epsilon(-\omega) = \epsilon^*(\omega) .$$

Hence

$$\phi(-\omega) = -\phi(\omega)$$

$$|\epsilon(-\omega)| = |\epsilon(\omega)| .$$

We can solve the Schrödinger equation

$$i\hbar \frac{d\Psi(t)}{dt} = H\Psi(t)$$

using the eigenstates of the material

Hamiltonian, $\quad H_M|\,E_n\,\rangle = E_n|\,E_n\,\rangle$

as a basis set, by expanding $\Psi(t)$ as,

$$\Psi(t) = \sum_n b_n(t)|\,E_n\,\rangle e^{-iE_n t/\hbar}.$$

We obtain that

$$\frac{db_m(t)}{dt} = \frac{-i}{\hbar} \sum_n b_n(t) e^{i\omega_{m,n} t} \langle E_m | H_{MR} | E_n \rangle$$

where $\omega_{m,n} \equiv (E_m - E_n)/\hbar.$

The molecule is initially $(t = -\infty)$ in a

single state $| E_1 \rangle$, i.e.,

$b_1(t = -\infty) = 1,$ and $b_{k \neq 1}(t = -\infty) = 0$

If $H_{MR}(t)$ is weak, namely,

$$\int_{-\infty}^{\infty} dt |\langle E_i | H_{MR}(t) | E_j \rangle| / \hbar \ll 1,$$

then,

$$b_m(t) = \frac{i d_{m,1}}{\hbar} \int_{-\infty}^{t} dt' e^{i\omega_{m,1}t'} \varepsilon(z,t') =$$

$$\frac{i d_{m,1}}{\hbar} \int_{-\infty}^{\infty} d\omega \bar{\epsilon}(\omega) \int_{-\infty}^{t} dt' e^{i(\omega_{m,1}-\omega)t'},$$

where
$$\mathrm{d}_{m,1} \equiv \langle E_m | \mathrm{d} \cdot \hat{\boldsymbol{\epsilon}} | E_1 \rangle,$$

$$\varepsilon(z,t) = \int_{-\infty}^{\infty} d\omega \, \bar{\epsilon}(\omega) e^{-i\omega t}$$

is the length of the $E_R(z,t) = \varepsilon(z,t)\hat{\epsilon}$

vector and

$$\bar{\epsilon}(\omega) \equiv |\epsilon(\omega)| e^{i(\phi(\omega) + \omega z/c)}.$$

$\Psi(t)$ as $t \longrightarrow +\infty$ is obtained using

$$\int_{-\infty}^{\infty} dt' e^{i(\omega_{m,1}-\omega)t'} = 2\pi\delta(\omega_{m,1} - \omega),$$

to yield

$$b_m(+\infty) = \frac{2\pi i}{\hbar}d_{m,1}\bar{\epsilon}(\omega_{m,1}) =$$

$$\frac{2\pi i}{\hbar}d_{m,1}|\epsilon(\omega_{m,1})|e^{i(\phi(\omega_{m,1})+\omega_{m,1}z/c)}.$$

A material energy state $|E_m\rangle$ only absorbs

or emits modes, at infinite time, for which

$$\omega = \omega_{m,1} \quad \text{or} \quad \omega' = -\omega_{m,1}.$$

State Preparation During the Pulse

At finite times, while the pulse is on,

$$b_m(t) = \langle E, m \,|\Psi(t)\rangle =$$

$$(i/\hbar)\mathrm{d}_{m,1} \int_{-\infty}^{t} dt'\varepsilon(t')e^{i\omega_{m,1}t'}$$

The probability of observing state $|E, m\rangle$

is, $\quad P_m(t) = |\langle E, m | \Psi(t) \rangle|^2 = |b_m(t)|^2$

$$b_m(t) = (i/\hbar)\mathrm{d}_{m,1}\bar{\varepsilon}(\omega_{m,1})c_m(t),$$

where

$$c_m(t) \equiv \frac{1}{\bar{\varepsilon}(\omega_{m,1})} \int_{-\infty}^{\infty} d\omega \epsilon(\omega) \frac{e^{i(\omega_{m,1}-\omega)t}}{i(\omega_{m,1}-\omega)} \ .$$

For a Lorentzian profile,

$$\epsilon(\omega) = (1/2\pi)\frac{\epsilon_a\Gamma}{(\omega - \omega_a)^2 + \Gamma^2/4},$$

the contribution of the "rotating wave" term is given as,

$$c_m^+(t) = 2\pi\left\{\theta(t) - \left[\frac{i(\omega_{m,1} - \omega_a)}{\Gamma} + \frac{sgn(t)}{2}\right]\cdot\right.$$

$$\left.\exp[-\Gamma|t|/2 + i(\omega_{m,1} - \omega_a)t]\right\},$$

where

$$sgn(t) = 1 \quad for \quad t \geq 0, \quad = -1 \quad for \quad t < 0,$$

and
$$\theta(t) = [1 + sgn(t)]/2.$$

A Lorentzian frequency profile is equivalent

to choosing

$$\epsilon(t) = \epsilon_a \exp(-\Gamma|t|/2)\exp(-i\omega_a t) + c.c.,$$

which is unrealistic because of the cusp at

For a Gaussian pulse,

$$\epsilon(t) = \epsilon_a e^{-4\ln 2(\Gamma t)^2}\left[e^{-i\omega_a t} + c.c.\right],$$

we have that

$$\epsilon(\omega) = \pi^{-1/2}\epsilon_a \alpha e^{-\alpha^2(\omega-\omega_a)^2},$$

where $\alpha \equiv 1/(\Gamma 4\ln^{1/2} 2)$.

$$c_m(t) = sgn(t')2\pi\left\{\theta(t) - \frac{e^{\beta^2}}{2}W[sgn(t)\beta]\right\}$$

where, $\beta \equiv \alpha(\omega_{m,1} - \omega_a) + i\dfrac{t}{2\alpha}.$

$W[z]$ is the complex error function.

$$c_m(t) \rightarrow 0, \text{ for } t \ll -1/\Gamma,$$

and

$$c_m(t) \rightarrow 2\pi, \quad \text{for } t \gg 1/\Gamma \quad.$$

Pulse preparation coefficients

a)

E=0.

E=24.

E=48.

E=72.

E=96.

E=120.

$|c'_m(t)|$

t(psec)

b)

E=0.

E=24.

E=48.

E=72.

E=96.

E=120.
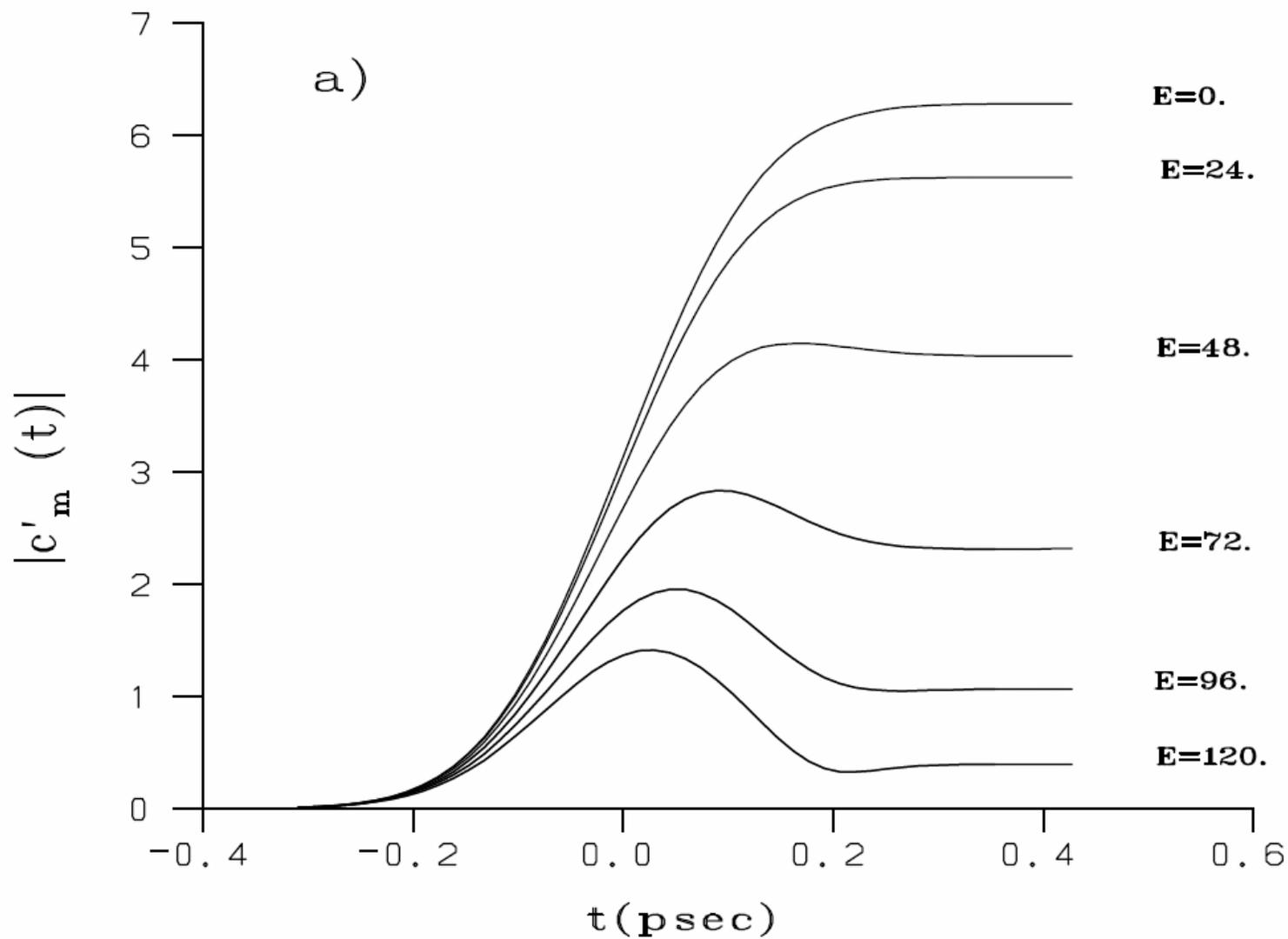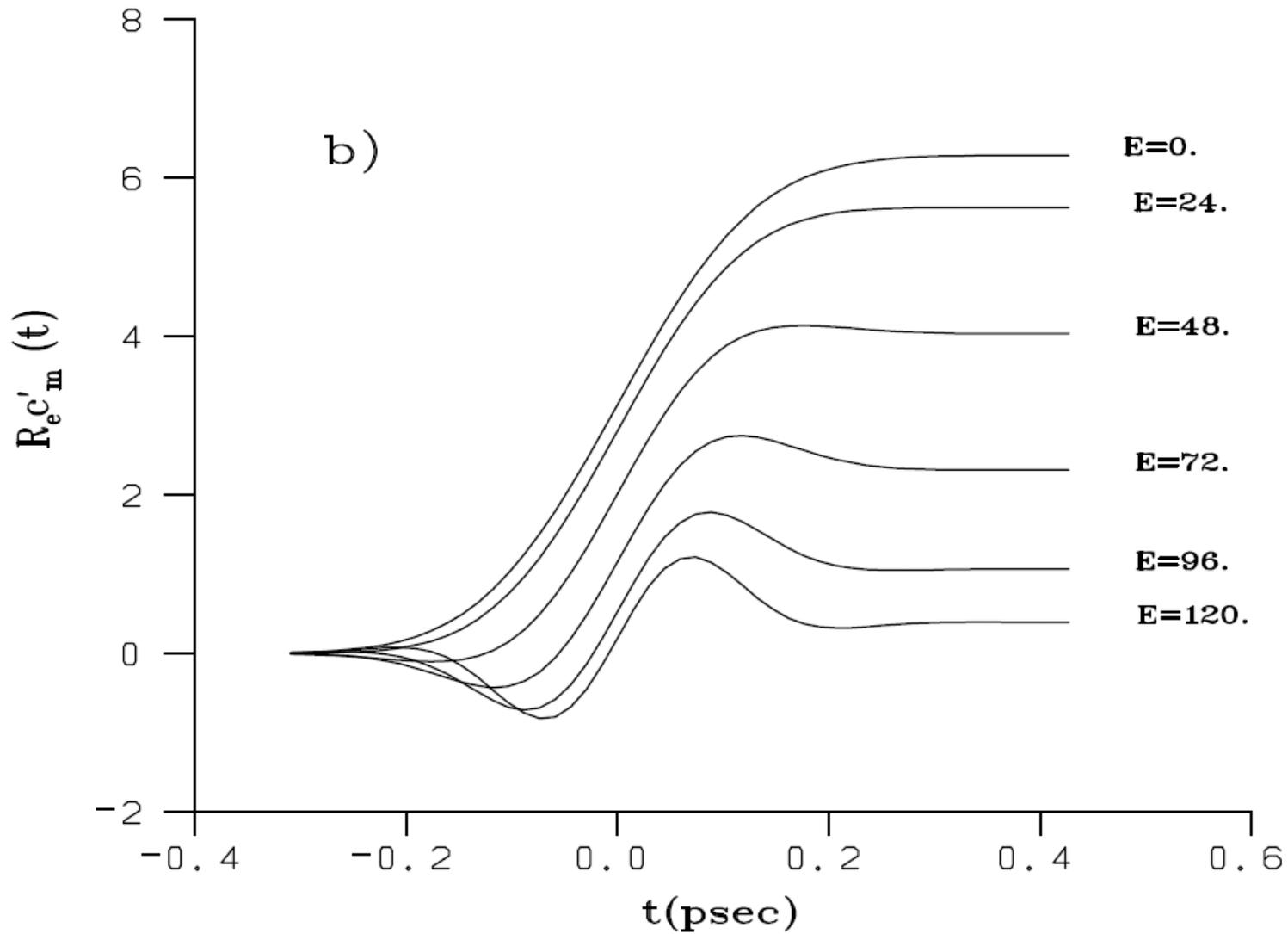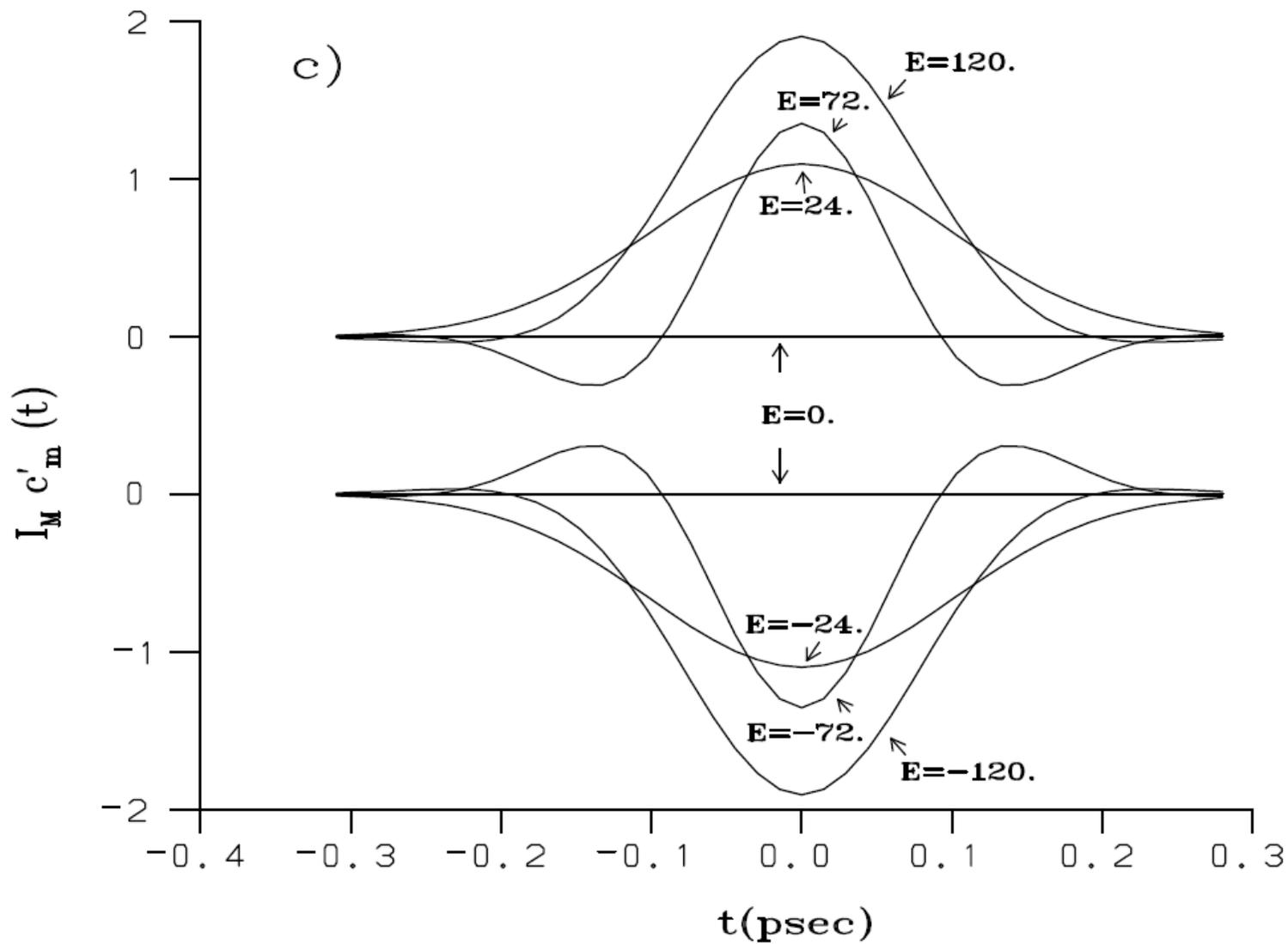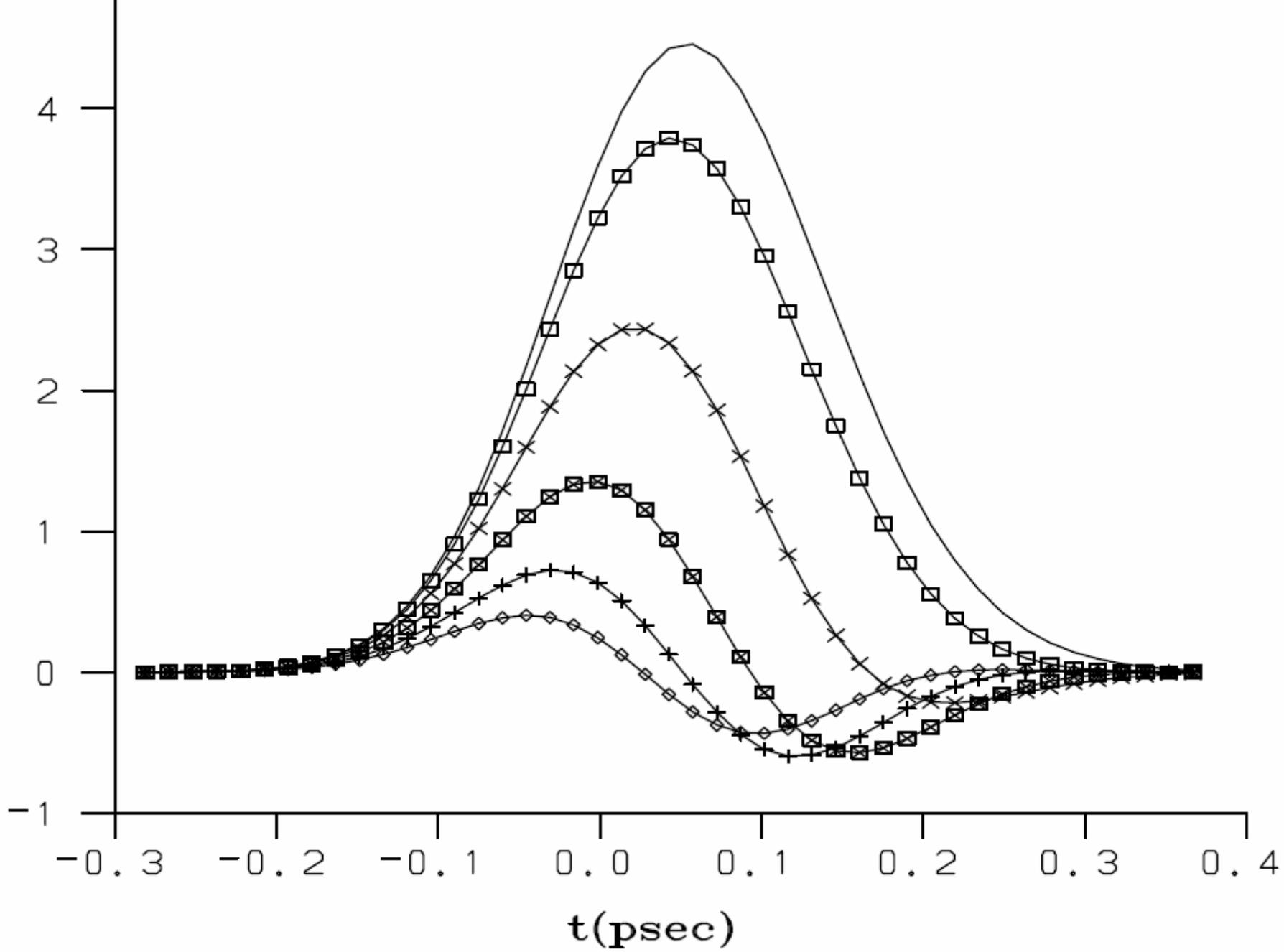
During the excitation pulse

$$\Psi(t) = |\, E_1 \,\rangle e^{-iE_1 t/\hbar} +$$

$$(i/\hbar) \sum_m c_m(t) \bar{\varepsilon}(\omega_{m,1}) \mathrm{d}_{m,1} |\, E_m \,\rangle e^{-iE_m t/\hbar}.$$

At the end of the pulse, for an absorption

process $(E_m > E_1)$,

$$\Psi(t \gg 1/\Gamma) = |\, E_1 \,\rangle e^{-iE_1 t/\hbar} +$$

$$(2\pi i/\hbar) \sum_m \bar{\varepsilon}(\omega_{m,1}) \mathrm{d}_{m,1} |\, E_m \,\rangle e^{-iE_m t/\hbar}.$$

The absorption of a photon has created a wavepacket in which the coefficients of preparation are proportional to the field amplitude at the $\omega_{m,1}$ frequency.

The Outcome of a State

The amplitude of creating a discrete

excited state $|E_m\rangle$ after the pulse is

$$b_m = \langle E_m | \Psi(t \gg 1/\Gamma)\rangle =$$

$$\frac{2\pi i}{\hbar} \langle E_m |\mathrm{d}| E_1 \rangle \bar{\varepsilon}(\omega_{m,1})$$

where $\quad \bar{\varepsilon}(\omega) \equiv \dfrac{1}{2\pi} \displaystyle\int_{-\infty}^{\infty} dt\varepsilon(t)e^{i\omega t}.$

The excited portion of the wavepacket

$$\Psi(t \gg 1/\Gamma) = (2\pi i/\hbar)$$

$$\sum_m \bar{\bar{\varepsilon}}(\omega_{m,1})\langle\, E_m\,|\mathrm{d}|\, E_1\,\rangle|\, E_m\,\rangle e^{-iE_m t/\hbar}.$$

If the spectrum is continuous

$$[E - H_M]|\, E, m\,\rangle = 0,$$

with m designating any additional quantum number.

In that case

$$\Psi(t) = \frac{2\pi i}{\hbar} \sum_n \int dE \bar{\varepsilon}(\omega_{E,1}) \cdot$$

$$\langle E, n \left| \mathrm{d} \right| E_1 \rangle \left| E, n \right\rangle e^{-iEt/\hbar}.$$

normalization

$$\langle E', m \left| E, n \right\rangle = \delta(E - E') \delta_{m,n}.$$

$\psi_n(E, R) \equiv \langle R \left| E, n \right\rangle$ has dimensions

of $\quad [L]^{-1/2}[E]^{-1/2}.$

Consider a triatomic molecule - ABC, which breaks apart to yield, the $A + BC$ products or the $B + AC$ products,

$$(q = 1) \ A + BC \leftarrow ABC \rightarrow B + AC \ (q = 2)$$

H_M is now composed of three parts,

$$H_M = K_R + K_r + W(R, r).$$

where $\quad K_R = \dfrac{-\hbar^2}{2\mu}\nabla_R^2, \quad K_r = \dfrac{-\hbar^2}{2m}\nabla_r^2,$

μ and m being the reduced masses,

$$\mu = m_A(m_B + m_C)/(m_A + m_B + m_C) ,$$

$$m = m_B m_C/(m_B + m_C).$$

$v(r)$ is the asymptotic limit of $W(R, r)$ as

A departs from $B - C$,

$$v(r) = \lim_{R \to \infty} W(R, r) .$$

The $A-BC$ interaction potential, defined as,

$$V(R, r) \equiv W(R, r) - v(r)$$

vanishes as $R \to \infty$,

$$\lim_{R \to \infty} V(R, r) = 0.$$

Defining the $B - C$ Hamiltonian as,

$$h_r \equiv K_r + v(r)$$

$$H_M = K_R + K_r + W(R, r) =$$

$$K_R + h_r + V(R, r).$$

The interaction potential $V(R, r)$ couples the motion of the A atom to the motion of the BC diatomic.

$$H_0 \equiv K_R + h_r$$

The eigenstates, $|E, m; 0\rangle$, of the free Hamiltonian

$$H_0 \equiv K_R + h_r$$

satisfying,

$$[E - H_0]| E, m; 0\rangle = 0,$$

are given as products

$$|E, m; 0\rangle = |e_m\rangle| E - e_m\rangle$$

where

$$[e_m - h_r]| e_m\rangle = 0,$$

e_m is the internal energy of $B - C$.

$|E - e_m\rangle$ describe free motion of A

relative to BC. They satisfy the equation

$$[E - e_m + \frac{\hbar^2}{2\mu}\nabla_R^2]\langle R| E - e_m\rangle = 0$$

whose solutions are plane waves of kinetic

energy $E - e_m$, $\quad \langle R| E - e_m\rangle = e^{ik_m \cdot R}$,

where $\quad k_m = \{2\mu(E - e_m)\}^{1/2}/\hbar$,

They are normalized as,

$$\langle E', m; 0 | E, n; 0 \rangle = \delta(E - E')\delta_{m,n}.$$

Relating the free states to the fully interacting states: The Schrödinger equation, $[E - H_0]| E, n \rangle = V| E, n \rangle,$

can be solved formally as a sum of

$$| E, n \rangle = [E - H_0]^{-1} V | E, n \rangle$$

and any solution $| E, n; 0 \rangle$ of the

homogeneous part that equation,

$$| E, n \rangle = | E, n; 0 \rangle + [E - H_0]^{-1} V | E, n \rangle .$$

Using the spectral resolution,

$$[E - H_0]^{-1} = \int dE' \frac{| E', n; 0 \rangle \langle E', n; 0 |}{E - E'},$$

we have that,

$$| E, n \rangle = | E, n; 0 \rangle +$$

$$\int dE' \frac{| E', n; 0 \rangle \langle E', n; 0 | V | E, n \rangle}{E - E'}$$

The $\int dE'$ integral is ill defined in the Riemann sense. In the Cauchy sense we consider the limit of a series of well defined Riemann integrals.

We add a small $i\epsilon$ imaginary part to E, calculate the integrals and then let $\epsilon \longrightarrow 0$. The limiting value thus obtained depends on the sign of ϵ.

We consider adding $i\epsilon$ or subtracting $i\epsilon$, with $\epsilon > 0$.

$$| E, n^\pm \rangle = | E, n; 0 \rangle +$$

$$\lim_{\epsilon \to 0}[E \pm i\epsilon - H_0]^{-1}V| E, n^\pm \rangle.$$

This is the Lippmann-Schwinger eq.

The $\pm$ solutions are not independent of (and definitely not orthogonal to) one another, so we can use either one or the other.

$$\phi_{n2}e^{ik.R}$$

$$\psi^{+}{}_{n2}(E)$$

$$\phi_{n_1} e^{ik.R}$$
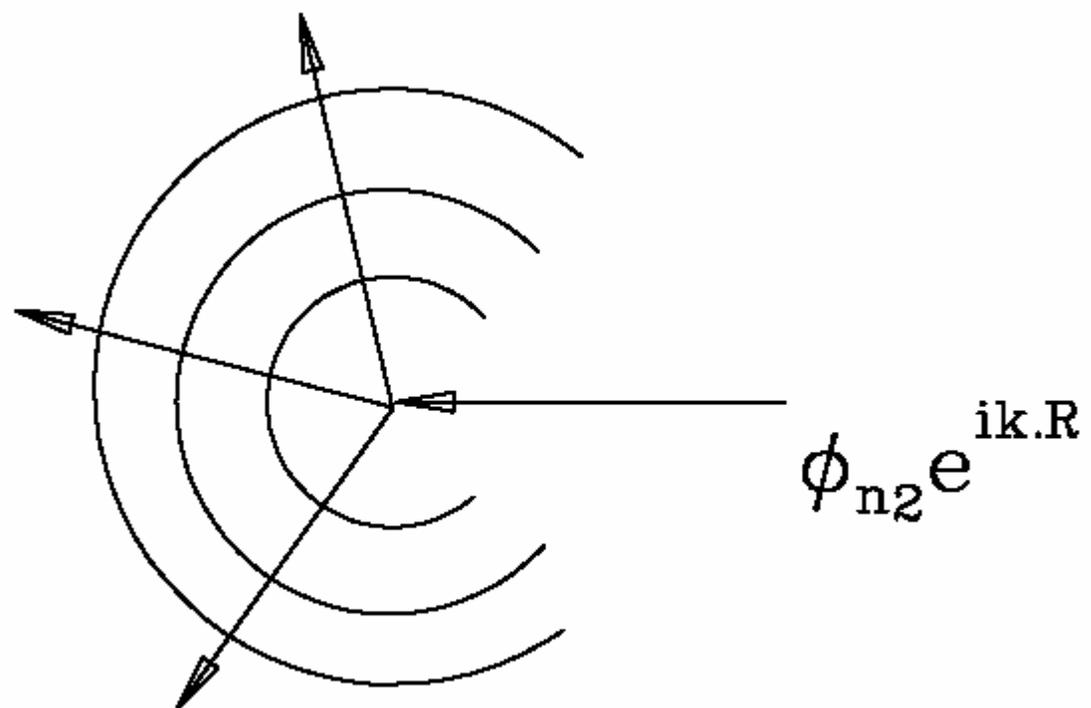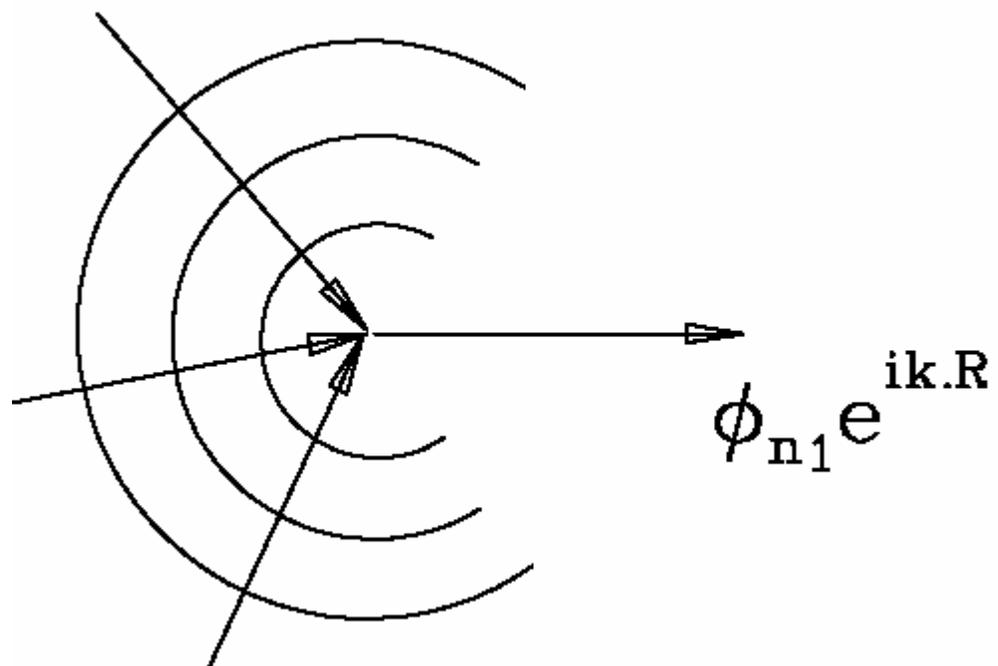
$$\psi^-_{n_1}(E)$$

The long-time behaviour of $\Psi(t)$ is obtained as,

$$\Psi(t) = \frac{2\pi i}{\hbar} \sum_n \int dE e^{-iEt/\hbar}.$$

$$\bar{\varepsilon}(\omega_{E,1}) \langle E, n^{\pm} | \mathrm{d} | E_1 \rangle \cdot$$

$$\left\{ | E, n; 0 \rangle + [E \pm i\epsilon - H_0]^{-1} V | E, n^{\pm} \rangle \right\}.$$

Using the spectral resolution of

$[E \pm i\epsilon - H_0]^{-1}$ the probability of finding a

free state $|E', m; 0\rangle$ at time t is given as

$$\langle E', m; 0 | \Psi(t) = \frac{2\pi i}{\hbar} \sum_n \int dE e^{-iEt/\hbar}.$$

$$\bar{\varepsilon}(\omega_{E,1}) \langle E, n^{\pm} | \mathrm{d} | E_1 \rangle \Big\{ \langle E'm; 0| E, n; 0\rangle +$$

$$[E \pm i\epsilon - E']^{-1} \langle E'm; 0 | V | E, n^{\pm}\rangle \Big\}.$$

$$\langle E', m; 0 | \Psi(t) = \frac{2\pi i}{\hbar} \, e^{-iE't/\hbar} \cdot$$

$$\left\{ \bar{\varepsilon}(\omega_{E',1}) \langle E', m^{\pm} | \mathrm{d} | E_1 \rangle + \right.$$

$$\sum_n \int dE e^{-iEt/\hbar} \bar{\varepsilon}(\omega_{E,1}) \langle E, n^{\pm} | \mathrm{d} | E_1 \rangle \cdot$$

$$\left. [E \pm i\epsilon - E']^{-1} \langle E'm; 0 | V | E, n^{\pm} \rangle \right\}.$$

In the $t \to \infty$ limit the integration over

E can be performed analytically by contour

integration.
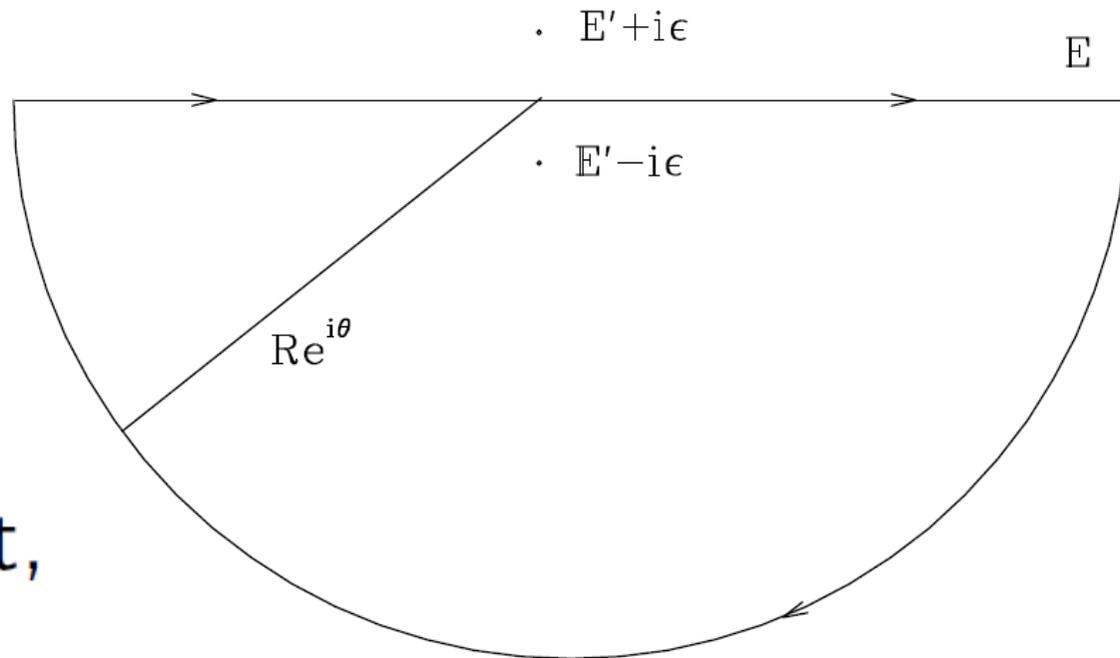
Noting that

in that limit,

when $E = Re^{i\theta}$, with $\theta < 0$,

$\cdot \ \mathbb{E}'+i\epsilon$

$\mathbb{E}$

$\cdot \ \mathbb{E}'-i\epsilon$

$Re^{i\theta}$

$$e^{-iEt/\hbar} = e^{-iRe^{i\theta}t/\hbar} =$$

$$e^{-iR\cos\theta t/\hbar} \cdot e^{R\sin\theta t/\hbar} \xrightarrow{t\to\infty} 0 \, .$$

$$\uparrow$$
$$\theta < 0$$

The real E integration remains unchanged

by supplementing it with integration along

the above large semi-circle in the lower

half E-plane.

Since in the $-i\epsilon$ case, the integrand has

a pole at $E = E' + i\epsilon$ which is outside
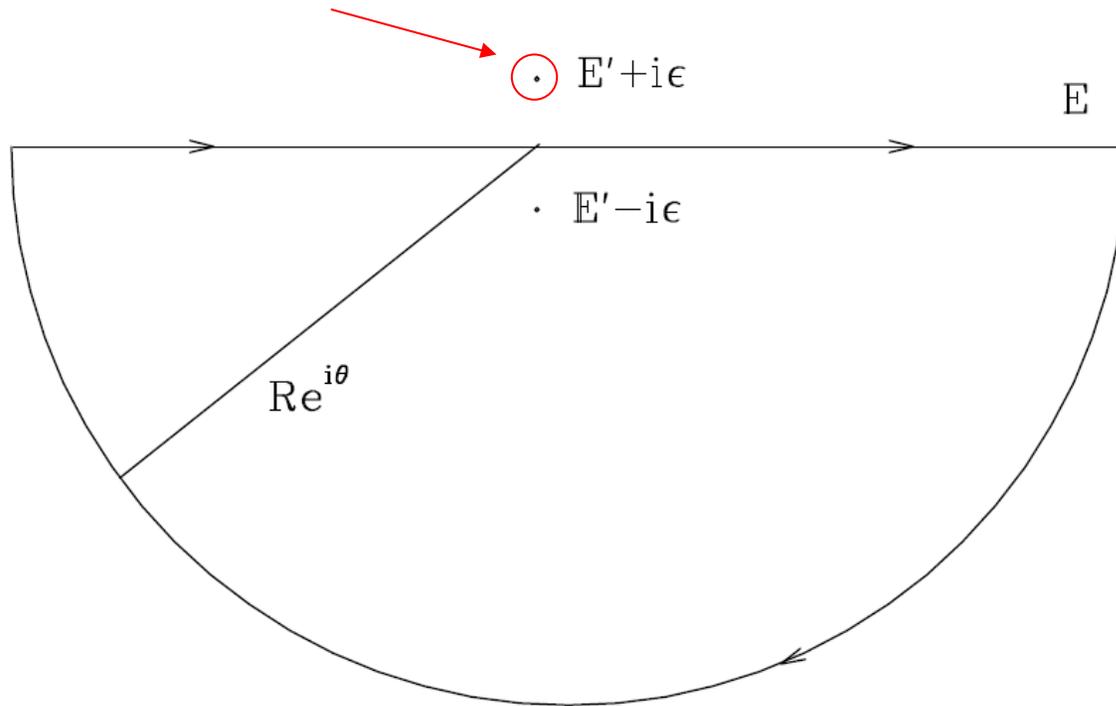
the closed

contour, the whole integral is zero.

Thus,

$$\lim_{t \to \infty} \langle\, E', m; 0\, | \Psi(t) =$$

$$\frac{2\pi i}{\hbar} \bar{\varepsilon}(\omega_{E',1})\, e^{-iE't/\hbar} \langle\, E', m^-\, |\mathrm{d}|\, E_1\, \rangle.$$

Hence the coefficients of expansion of the full wavepacket in terms of the $|\, E, m^-\, \rangle$ states, tell us immediately what is the probability of observing states $|\, E, m; 0\, \rangle$ in the distant future.

If instead of the incoming states we use

the outgoing states, the closed contour

integration encircles a pole at $E = E' - i\epsilon$,
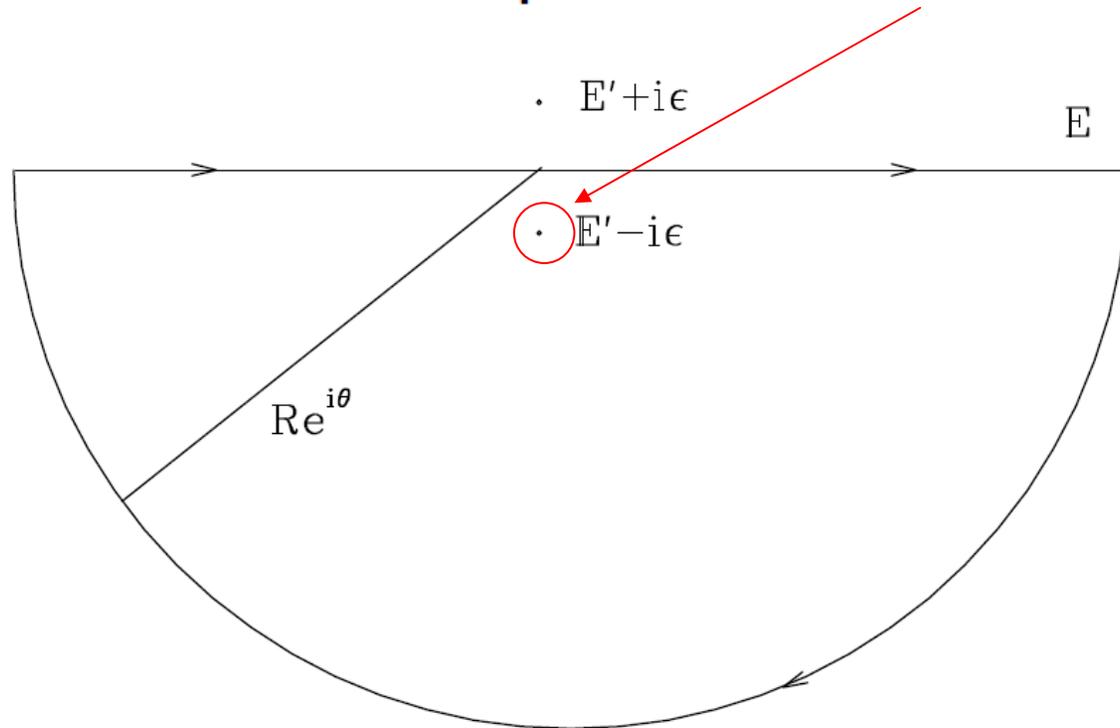
hence the integration yields

$$\lim_{t \to \infty} \langle\, E', m; 0 \,|\Psi(t) = \frac{2\pi i}{\hbar} e^{-iE't/\hbar} \bar{\varepsilon}(\omega_{E',1})\cdot$$

$$\sum_n S_{n,m}(E') \langle\, E', n^+ \,|\mathrm{d}|\, E_1 \,\rangle.$$

where the $S_{n,m}(E')$ matrix,

$$S_{n,m}(E') \equiv \delta_{n,m} - 2\pi i \langle\, E'm; 0 \,|V|\, E', n^+ \,\rangle$$

is the S-matrix or Scattering matrix.

In contrast, in the $t \to -\infty$ limit,

$$\lim_{t \to -\infty} \langle E', m; 0 | \Psi(t) =$$

$$\frac{2\pi i}{\hbar} \bar{\varepsilon}(\omega_{E',1}) \, e^{-iE't/\hbar} \langle E', m^+ | \mathrm{d} | E_1 \rangle.$$

while if we use the $-$ solutions

$$\lim_{t \to -\infty} \langle E', m; 0 | \Psi(t) =$$

$$\frac{2\pi i}{\hbar} e^{-iE't/\hbar} \bar{\varepsilon}(\omega_{E',1}) \sum_n S_{n,m}^-(E') \langle E', n^- | \mathrm{d} | E_1 \rangle.$$

where the $S_{n,m}^-(E')$ matrix is defined as,

$$S_{n,m}^-(E') \equiv \delta_{n,m} + 2\pi i \langle E'm; 0 | V | E', n^- \rangle$$

Direct Photo-Dissociation

by a Strong Laser Field

We now present a non-perturbative solution of the pulsed photo-dissociation problem.

Writing the pulse electric field as,

$$\mathbf{E}(t) = \mathcal{R}_e \hat{\boldsymbol{\epsilon}} \mathcal{E}(t) e^{-i\omega_a t},$$

where ω_a, called "carrier" frequency, represents the average frequency in the spectrum of the pulse $\bar{\epsilon}(\omega)$. $\mathcal{E}(t)$ is called the "envelope" of the pulse.

As before we expand the full time-dependent wavefunction as,

$$\Psi(t) = b_1(t)|\, E_1\,\rangle e^{-iE_1 t/\hbar} +$$

$$\sum_n \int dE b_{E,n}(t)|\, E, n^-\,\rangle e^{-iEt/\hbar},$$

Substituting into the time-dependent Schrödinger equation we obtain as before

$$\frac{db_1}{dt} = \frac{i}{\hbar} \sum_n \int dE \langle E_1 |\mathrm{d}| E, n^- \rangle \mathcal{E}^*(t) b_{E,n}(t) \cdot$$

$$e^{-i(\omega_{E,1} - \omega_a)t} \quad ,$$

$$\frac{db_{E,n}}{dt} = \frac{i}{\hbar} \langle E, n^- |\mathrm{d}| E_1 \rangle \mathcal{E}(t) b_1(t) e^{i(\omega_{E,1} - \omega_a)t}$$

where $\omega_{E,1} \equiv (E - E_1)/\hbar$, and we have

invoked the rotating wave approximation.

Substituting the formal solution

$$b_{E,n}(t) =$$

$$\frac{i\langle E, n^- |\mathrm{d}| E_1 \rangle}{\hbar} \int_{-\infty}^{t} dt' \mathcal{E}(t') b_1(t') e^{i(\omega_{E,1} - \omega_a)t'},$$

we obtain that,

$$\frac{db_1}{dt} = -\frac{1}{\hbar^2} \int dE \sum_n |\langle E_1 |\mathrm{d}| E, n^- \rangle|^2 \mathcal{E}^*(t) \cdot$$

$$\int_{-\infty}^{t} dt' \mathcal{E}(t') e^{-i(\omega_{E,1} - \omega_a)(t-t')} b_1(t').$$

This is an *integro-differential* equation.

According to it the change in b_1 at time t

depends on its history at all preceding times

$t' < t$. The $\mathcal{E}(t')e^{-i(\omega_{E,1}-\omega_a)(t-t')}$ factor is

sometimes called the "memory-Kernel" of

the integral equation.

Defining the the spectrum,

$$A(E) \equiv \sum_n |\langle E, n^- |\mathrm{d}| E_1 \rangle|^2$$

and the "spectral autocorrelation function", as its Fourier transform,

$$F(t - t') \equiv \int dE A(E) e^{-i\omega_{E,1}(t-t')},$$

we have that

$$\frac{db_1}{dt} =$$

$$-\frac{\mathcal{E}^*(t)}{\hbar^2} \int_{-\infty}^{t} dt' e^{i\omega_a(t-t')} \mathcal{E}(t') F(t-t') b_1(t').$$

If $A(E)$ is slowly varying we can replace it by its value $A(E_a)$ at the center energy of the pulse ($E_a = E_1 + \hbar\omega_a$).

The above equation transforms into,

$$F(t - t') \approx A(E_a) \int dE e^{-i\omega_{E,1}(t-t')} =$$

$$2\pi\hbar A(E_a)\delta(t - t').$$

Using this equation and remembering that

we integrate t' from $-\infty$ to the singularity

point t, which introduces a factor of $\frac{1}{2}$, we

obtain that

$$\frac{db_1}{dt} = -\frac{\pi A(E_a)}{\hbar}|\mathcal{E}(t)|^2 b_1(t),$$

whose solution is

$$b_1(t) =$$

$$b_1(-\infty)\exp\left[-\frac{\pi A(E_a)}{\hbar}\int_{-\infty}^{t} dt'|\mathcal{E}(t')|^2(t')\right].$$

The above is a statement of what may be termed the "slowly varying continuum approximation" (SVCA).

We see that in this approximation $b_1(t)$ decreases monotonically with time, though not necessarily as an exponential function.

The SVCA need not be assumed for the continuum coefficients themselves, because once we know $b_1(t)$ the $b_{E,n}$ coefficients are given as,

$$b_{E,n}(t) = \frac{i \langle E, n^- | \mathrm{d} | E_1 \rangle}{\hbar} \int_{-\infty}^{t} dt' \mathcal{E}(t') \cdot$$

$$e^{\left[i(\omega_{E,1} - \omega_a)t' - \frac{\pi A(Ea)}{\hbar} \int_{-\infty}^{t'} |\mathcal{E}(t'')|^2 dt'' \right]} .$$

Coherent Control

We now wish to see how the laser field can be made to modify the outcome of the photodissociation process. As we saw the probability of populating a "free" state $|e_m\rangle|\mathbf{k}_m\rangle$ at any given time is given as

$$P_m(E)(t) = |\langle\, e_m\,|\langle\, \mathbf{k}_m\,|\Psi(t)\rangle|^2.$$

Using the expansion of the wavepacket

$\Psi(t)$ we have that,

$$\langle\, e_m\,|\langle\, \mathbf{k}_m\,|\Psi(t)\rangle = b_1(t)\langle\, e_m\,|\langle\, \mathbf{k}_m\,|E_1\rangle e^{-iE_1 t/\hbar}$$

$$+\sum_n \int dE b_{E,n}(t)\langle\, e_m\,|\langle\, \mathbf{k}_m\,|E, n^-\rangle e^{-iEt/\hbar}.$$

Assuming that $\langle\, e_m\,|E_1\rangle = 0$, (e.g., they

belong to different electronic states),

it follows that in the long-time limit

$$P_m(E) = |\langle e_m | \langle \mathbf{k}_m | \Psi(t \to \infty) \rangle|^2 =$$

$$|b_{E,m}(t \to \infty)|^2.$$

Since

$$b_{E,n}(t \to \infty) =$$

$$\frac{i}{\hbar} \langle E, n^- | \mathrm{d} | E_1 \rangle \int_\infty^\infty dt' \varepsilon(t') e^{-i\omega_{E,1}t} b_1(t')$$

it follows that,

$$\frac{P_n(E)}{P_m(E)} = \left| \frac{b_{E,n}(\infty)}{b_{E,m}(\infty)} \right|^2 = \left| \frac{\langle E, n^- |\mathrm{d}| E_1 \rangle}{\langle E, m^- |\mathrm{d}| E_1 \rangle} \right|^2 .$$

We see that the relative probabilities of populating different asymptotic states at a fixed energy E are *independent of the laser power and pulse shape.*

This result which coincides with that of perturbation theory, holds true provided there is only *one* initial ("precursor") state $|E_1\rangle$ coupled to the continuum.

In order to affect the long time outcome we must therefore move away from the single initial (precursor) state situation.

Bichromatic photodissociation

A+BC

C+AB

E

E

$h\nu_2$

$h\nu_1$

$n_1=2$

$n_1=1$

$|2)$

$|1)$

$A-B-C$
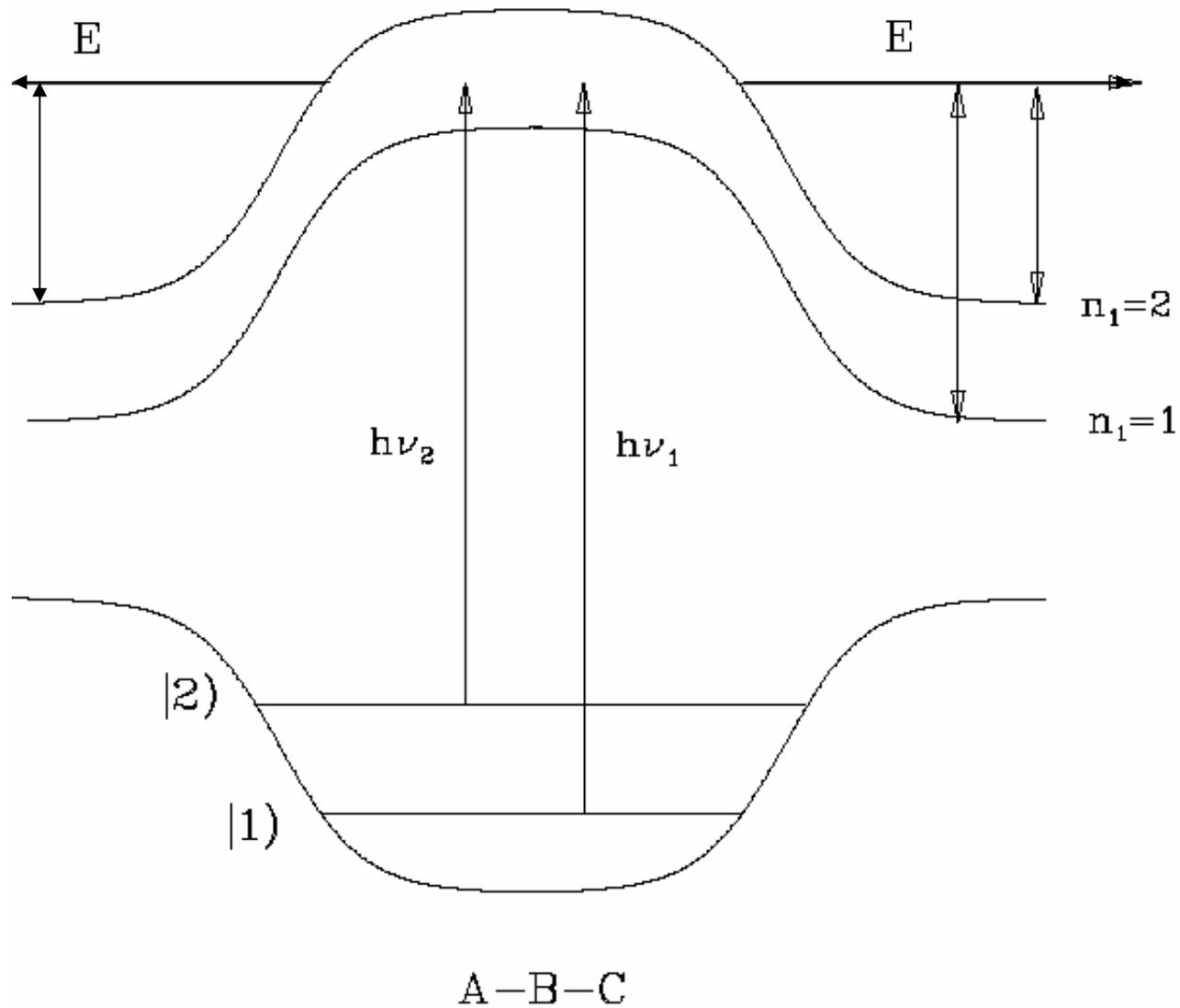
Starting from a linear superposition of two initial states

$$\Phi(t) = b_1 |E_1\rangle e^{-iE_1 t/\hbar} + b_2 |E_2\rangle e^{-iE_2 t/\hbar}$$

we have that

$$b_{E,n}(t \to \infty) =$$

$$\frac{i}{\hbar} \left\{ \langle E, n^- |\mathrm{d}| E_1\rangle \int_{-\infty}^{\infty} dt' \varepsilon(t') e^{-i\omega_{E,1} t'} b_1(t') \right.$$

$$\left. + \langle E, n^- |\mathrm{d}| E_2\rangle \int_{-\infty}^{\infty} dt' \varepsilon(t') e^{-i\omega_{E,2} t'} b_2(t') \right\}.$$

In first order perturbation theory $b_1(t)$ and $b_2(t)$ are constant, hence in the weak field regime,

$$b_{E,n}(t \to \infty) \approx \frac{2\pi i}{\hbar}.$$

$$\{\langle E, n^- | \mathrm{d} | E_1 \rangle \bar{\epsilon}(\omega_{E,1}) b_1 +$$

$$\langle E, n^- | \mathrm{d} | E_2 \rangle \bar{\epsilon}(\omega_{E,2}) b_2 \}$$

where

$$\bar{\epsilon}(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \varepsilon(t) e^{-i\omega t}.$$

Recognizing as we did in previous lectures

that $\bar{\epsilon}(\omega)$ has a phase, we can write

$$\bar{\epsilon}(\omega_{E,i}) = |\bar{\epsilon}(\omega_{E,i})|e^{-i\theta(\omega_{E,i})}, \qquad i = 1, 2,$$

and obtain, $\qquad b_{E,n}(\infty) =$

$$\frac{2\pi i}{\hbar}\Big\{\langle E, n^- |\mathrm{d}| E_1\rangle|\bar{\epsilon}(\omega_{E,1})|e^{-i\theta(\omega_{E,1})}b_1 +$$

$$\langle E, n^- |\mathrm{d}| E_2\rangle|\bar{\epsilon}(\omega_{E,2})|e^{-i\theta(\omega_{E,2})}b_2\Big\}.$$

The probability of seeing product state n at infinite time is,

$$P_n(E) = \frac{4\pi^2}{\hbar^2} \cdot$$

$$\left| \langle E, n^- | \mathrm{d} | E_1 \rangle |\bar{\epsilon}(\omega_{E,1})| e^{-i\theta(\omega_{E,1})} b_1 \right.$$

$$\left. + \langle E, n^- | \mathrm{d} | E_2 \rangle |\bar{\epsilon}(\omega_{E,2})| e^{-i\theta(\omega_{E,2})} b_2 \right|^2.$$

We see that the pulse attributes have been "entangled" into the material matrix elements.

The properties we wish to control in Chemistry are often the branching ratios to different chemical products. Realizing that any Chemical process such as

$$A + BC(n) \overset{q=1}{\longleftarrow} ABC \overset{q=2}{\longrightarrow} AB(n) + C,$$

involves both the chemical products (q) of interest, and the multitude of internal fragment states $(|e_n\rangle)$ in each chemical channel, which are now of no interest. We calculate the total probability to produce products in one of the q channels as,

$$P^{(q)}(E) = \sum_n P_n^{(q)}(E) = \sum_n \frac{4\pi^2}{\hbar^2}.$$

$$\left| \langle E, q, n^- | \mathrm{d} | E_1 \rangle | \bar{\epsilon}(\omega_{E,1}) | e^{-i\theta(\omega_{E,1})} b_1 + \right.$$

$$\left. \langle E, q, n^- | \mathrm{d} | E_2 \rangle | \bar{\epsilon}(\omega_{E,2}) | e^{-i\theta(\omega_{E,2})} b_2 \right|^2.$$

Hence

$$P^{(q)}(E) = P_{11}^{(q)}(E) + P_{22}^{(q)}(E) + P_{12}^{(q)}(E)$$

where

$$P_{11}^{(q)}(E) = \frac{4\pi^2}{\hbar^2} \mu_{11}^{(q)} |\bar{\epsilon}(\omega_{E,1})|^2 b_1^2$$

$$P_{22}^{(q)}(E) = \frac{4\pi^2}{\hbar^2} \mu_{22}^{(q)} |\bar{\epsilon}(\omega_{E,2})|^2 b_2^2$$

$$P_{12}^{(q)}(E) = \frac{4\pi^2}{\hbar^2} |\bar{\epsilon}(\omega_{E,1})| |\bar{\epsilon}(\omega_{E,2})| \cdot$$

$$2\mathcal{R}_e\{\mu_{12}^{(q)} e^{-i\theta_{12}} b_1^* b_2\},$$

where

$$\mu_{11}^{(q)} \equiv \sum_n |\langle E, q, n^- |\mathrm{d}| E_1 \rangle|^2,$$

$$\mu_{22}^{(q)} \equiv \sum_n |\langle E, q, n^- |\mathrm{d}| E_2 \rangle|^2,$$

$$\mu_{12}^{(q)} = \sum_n \langle E_1 |\mathrm{d}| E, q, n^- \rangle \langle E, q, n^- |\mathrm{d}| E_2 \rangle,$$

and $\theta_{12} \equiv \theta(\omega_{E,2}) - \theta(\omega_{E,1})$.

The $P_{11}^{(q)}(E)$ and $P_{22}^{(q)}(E)$ terms are the individual probabilities of photodissociating level $|E_1\rangle$ or $|E_2\rangle$ respectively.

$P_{12}^{(q)}(E)$ is the interference terms. It is the only term influenced by the relative phase θ_{12} between the $\bar{\epsilon}(\omega_{E,1})$ and $\bar{\epsilon}(\omega_{E,2})$.

In order to make the structure of the probability expression clearer, we write the complex amplitude $\mu_{12}^{(q)}$ as,

$$\mu_{12}^{(q)} = |\mu_{12}^{(q)}|e^{i\phi_{12}^{(q)}},$$

where $\phi_{12}^{(q)}$ is the so-called "molecular" or "material" phase. In addition we write,

$$b_1^* b_2 = |b_1 b_2|e^{-i\alpha_{12}}.$$

The interference term is now,

$$P_{12}^{(q)}(E) = \frac{8\pi^2}{\hbar^2} \left| \bar{\epsilon}(\omega_{E,1}) \bar{\epsilon}(\omega_{E,2}) \mu_{12}^{(q)} b_1 b_2 \right| \cdot$$

$$\cos \left[\phi_{12}^{(q)} - \alpha_{12} - \theta_{12} \right],$$

This term can be positive ("constructive interference") or negative ("destructive interference") with respect to one q channel and the opposite with respect to the other.

By tuning the external phases α_{12} or θ_{12} we can make the sign of this term be negative with respect to one q chemical channel and positive with respect to another. In this way by changing an external phase factor that is indifferent to the final channels we attain selectivity (discrimination) between the final channels.

The two slit perspective: the importance of the relative phase

Screen

a

b

+

-

+

-

+

-

+

-

0

0

Interference
pattern
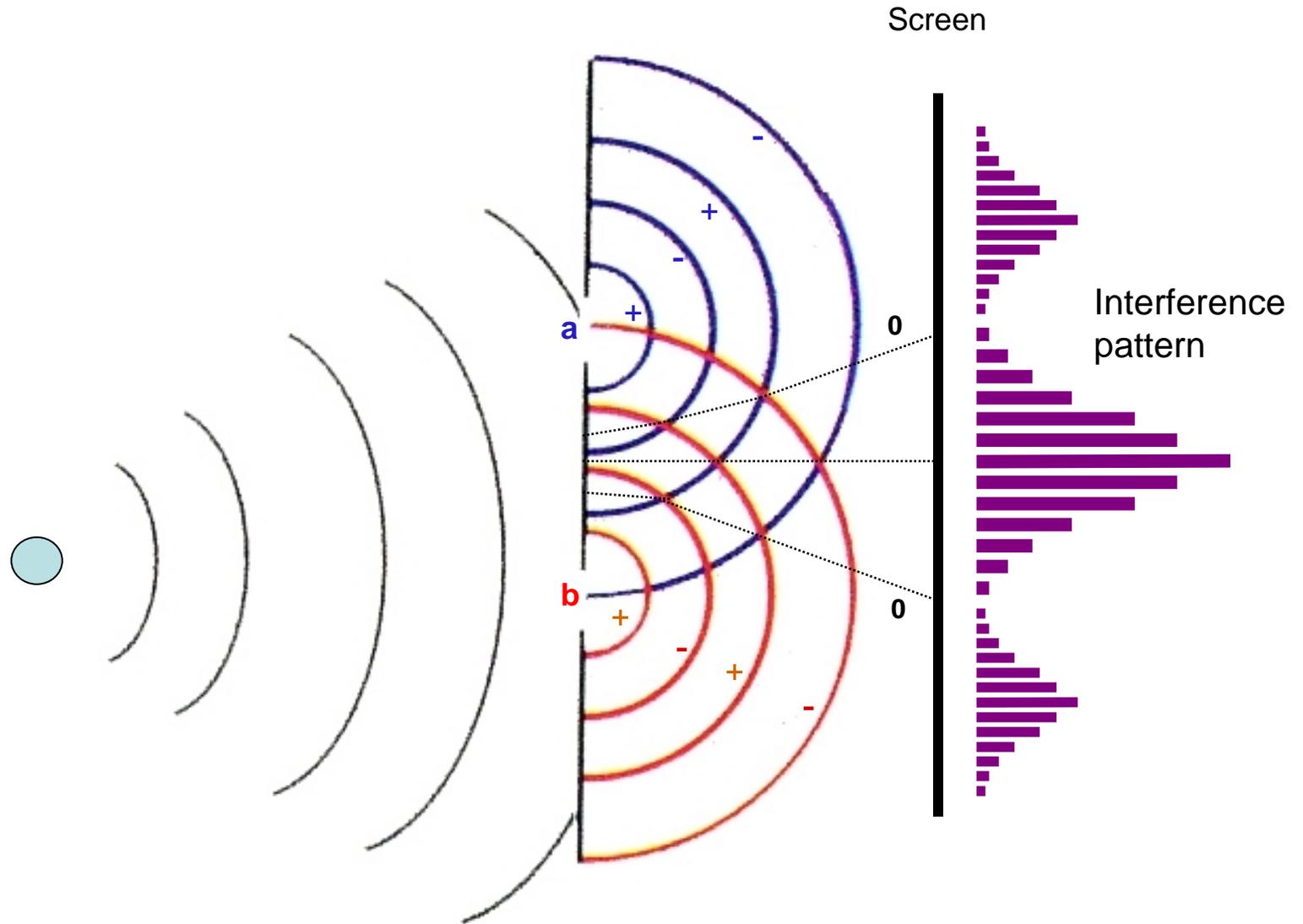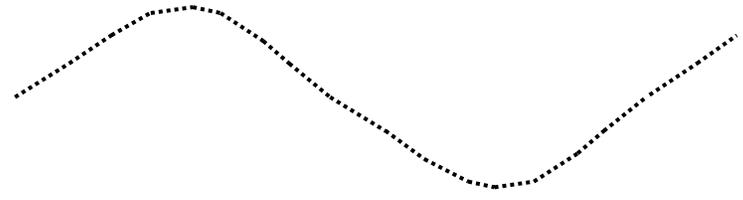
light wave a
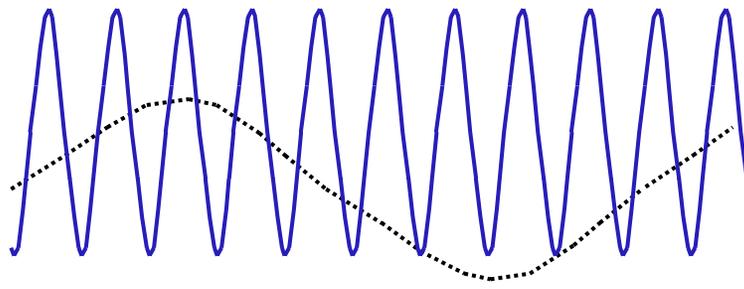
light wave a

final matter state

light wave a
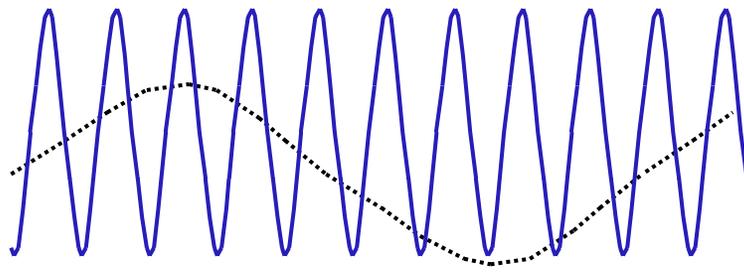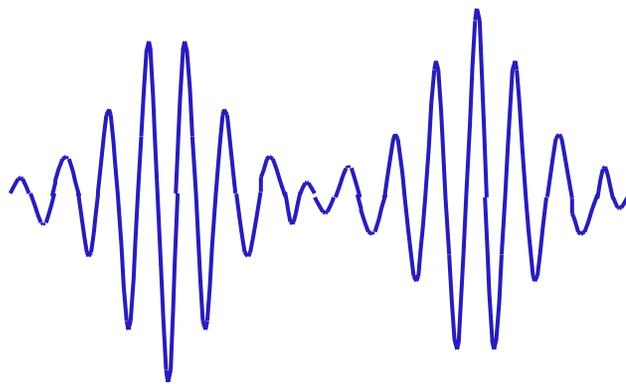
amplitude for
absorbing
light wave a

phase
shift

light wave a

amplitude for
absorbing
light wave a
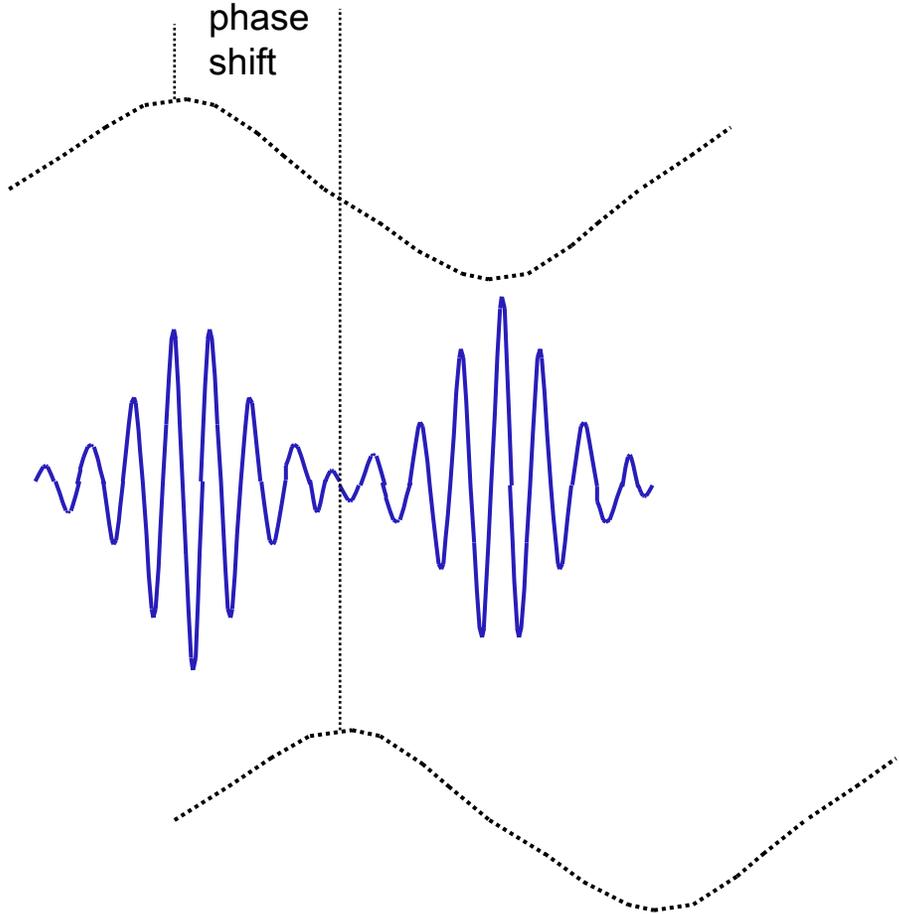
light wave b

phase shift
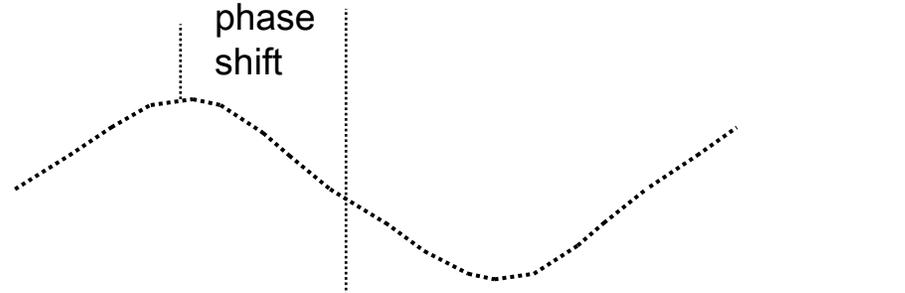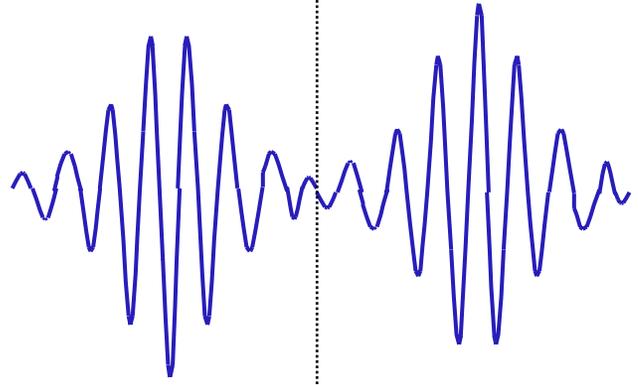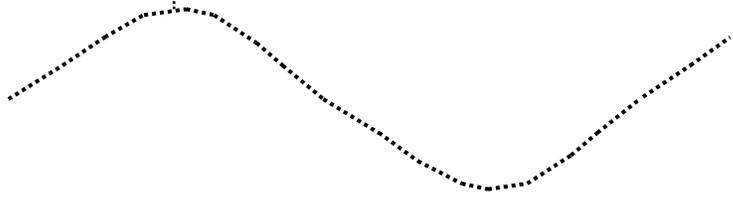
light wave a

amplitude for absorbing light wave a

light wave b

amplitude for absorbing light wave b
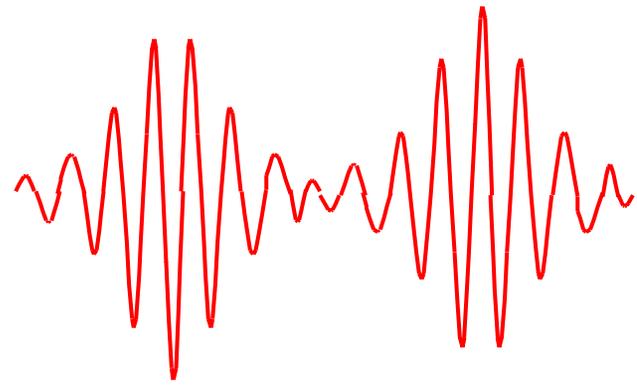
phase
shift

light wave a

amplitude for
absorbing
light wave a

interfere

light wave b

amplitude for
absorbing
light wave b
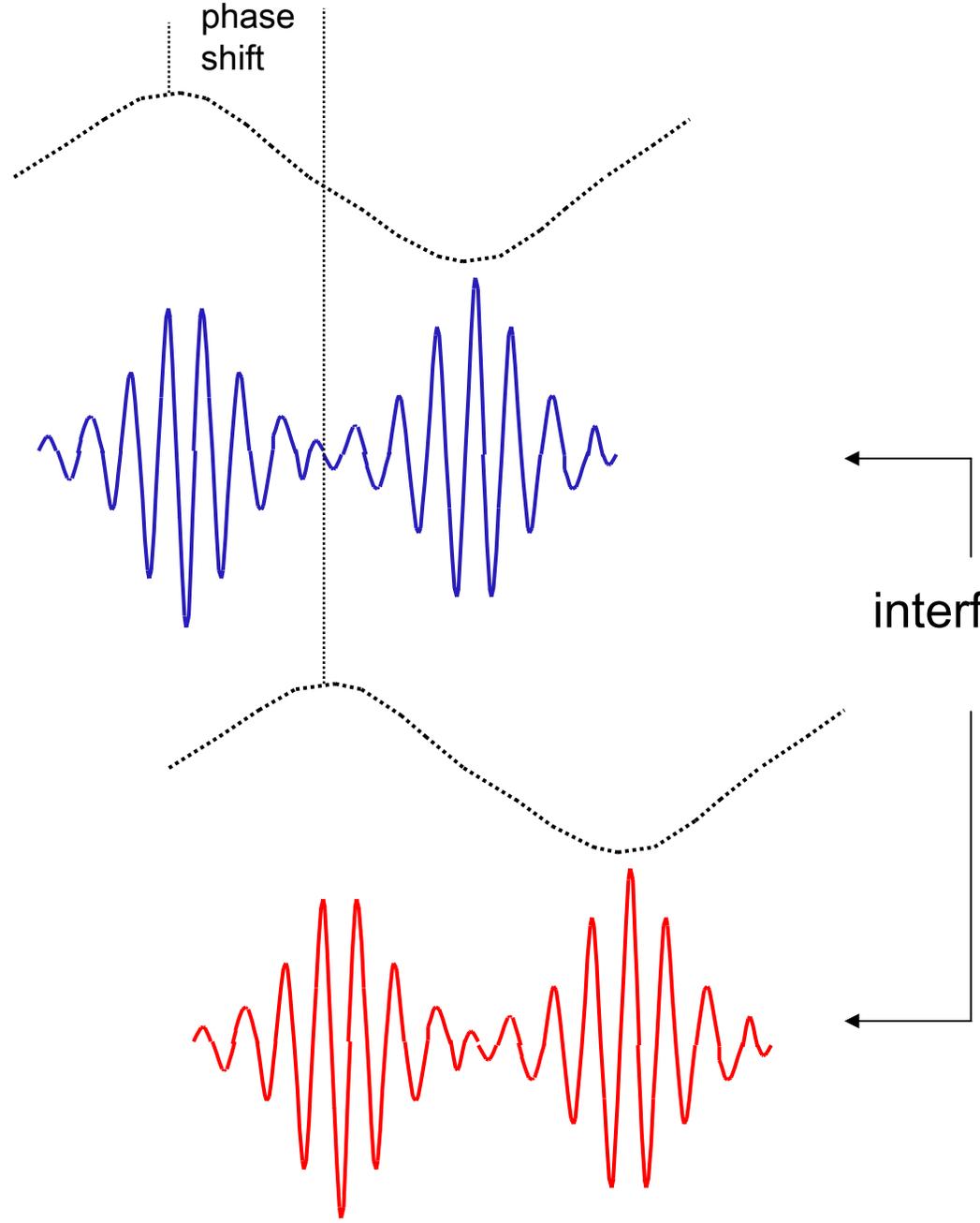
The **key** to control is that the interference patterns of different outcomes be shifted in phase.

A-B + C

the "screen" of
relative phases

A + B-C

hʋ

■ is favored

A-B + C

A + B-C

$h\nu$

■ is favored

A-B + C

A + B-C

$h\nu$

■ is favored

A-B + C

A + B-C

$h\upsilon$
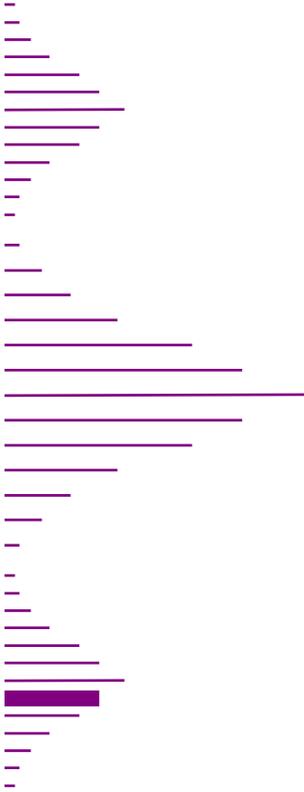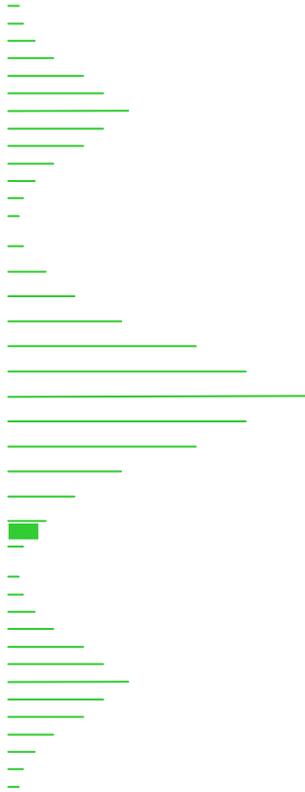
■ is favored
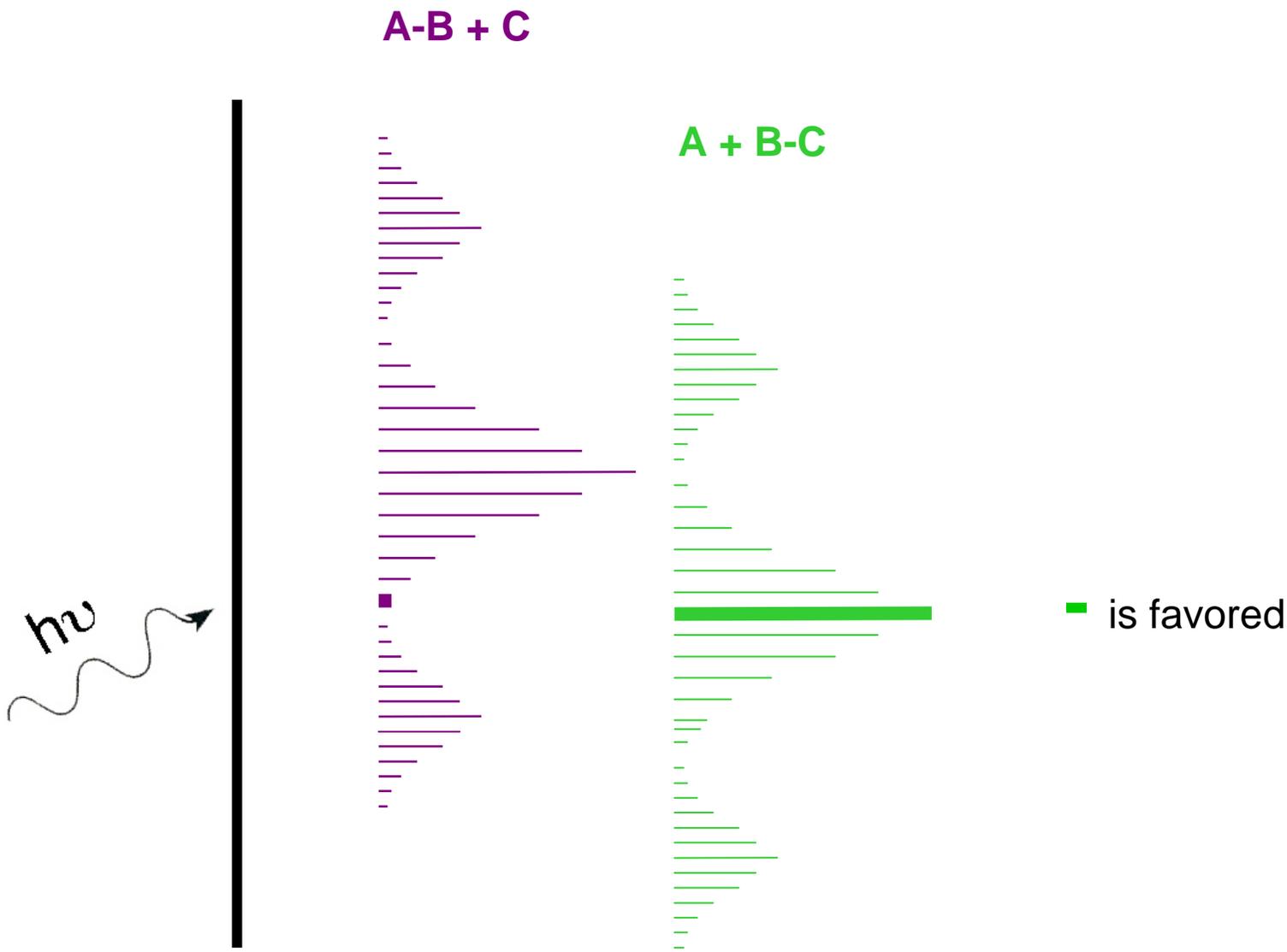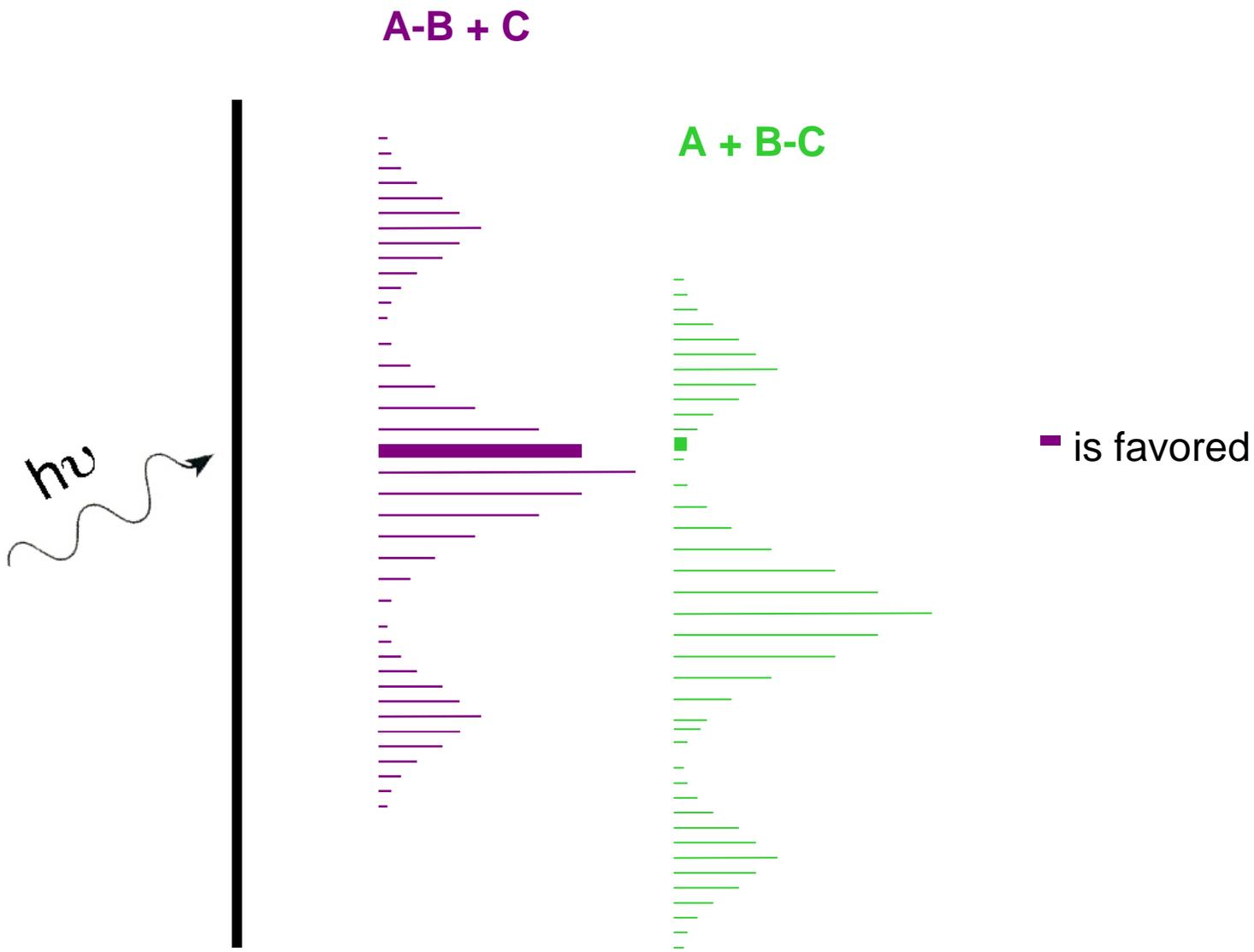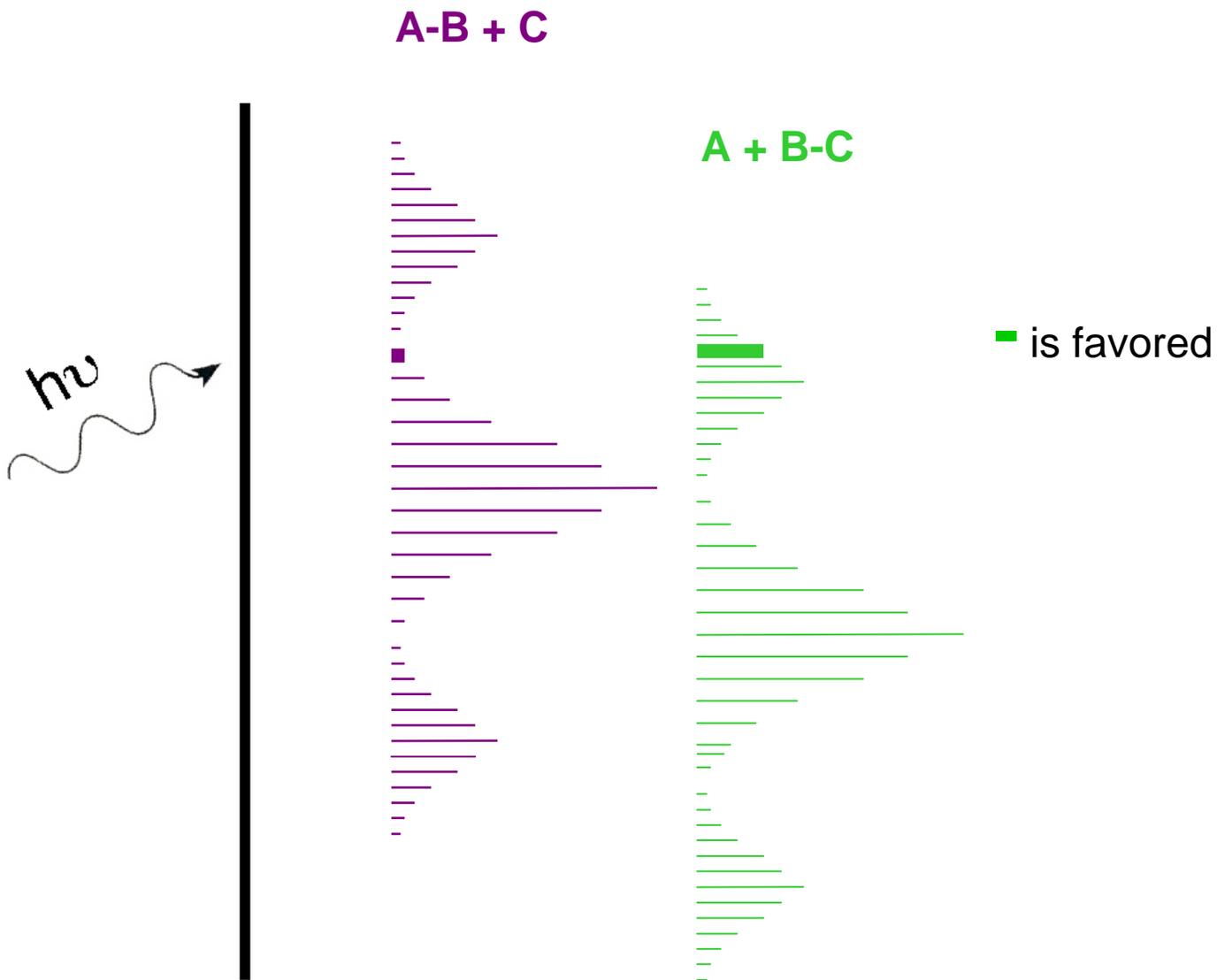
The effect can be enhanced by varying

$$x \equiv \frac{|\bar{\epsilon}(\omega_{E,1})b_1|}{|\bar{\epsilon}(\omega_{E,2})b_2|}$$

The method of controlling the final

outcome of a process in this way is called

"Coherent Control" (CC).

In order to see the structure more clearly we write the CC equations as,

$$P^{(q)}(E) = \frac{4\pi^2}{\hbar^2}|\bar{\epsilon}(\omega_{E,1})b_1|^2 \cdot$$

$$\{\mu_{11}^{(q)} + x^2\mu_{22}^{(q)} + 2x|\mu_{12}^{(q)}|\cos[\phi_{12}^{(q)} - \alpha_{12} - \theta_{12}]\}.$$

In order to attain maximum control we want to set $P^{(q)}(E) = 0$ for one of the q channels. This will automatically maximize the probability in the other q' channel(s).

Since $P^{(q)}(E)$ is always positive and since $\mu_{11}^{(q)} + x^2 \mu_{22}^{(q)}$ is also positive, the only way we can minimize $P^{(q)}(E)$ is by making the interference term $2x|\mu_{12}^{(q)}| \cos[\phi_{12}^{(q)} - \alpha_{12} - \theta_{12}]$ as negative as possible.

Thus, we must set the external phases $\alpha_{12} + \theta_{12}$ such that, $\alpha_{12} + \theta_{12} = \pi - \phi_{12}^{(q)}$.

When this is done $\cos[\phi_{12}^{(q)} - \alpha_{12} - \theta_{12}] = -1$

and we obtain that,

$$P^{(q)}(E) =$$

$$\frac{4\pi^2}{\hbar^2} |\bar{\epsilon}(\omega_{E,1})b_1|^2 \{\mu_{11}^{(q)} + x^2 \mu_{22}^{(q)} - 2x|\mu_{12}^{(q)}|\} :$$

We obtain that $P^{(q)}(E) = 0$ when this

quadratic equation has a solution.

There is a solution

$$x = |\mu_{12}^{(q)}|/\mu_{22}^{(q)} \quad \text{iff} \quad |\mu_{12}^{(q)}|^2 = \mu_{11}^{(q)}\mu_{22}^{(q)},$$

i.e., when the discriminant vanishes.

We now write,

$$\mu_{11}^{(q)} = \sum_n \langle E_1 |\mathrm{d}| E, q, n^- \rangle \langle E, q, n^- |\mathrm{d}| E_1 \rangle$$

$$= \langle E_1 |\mathrm{d} \sum_n | E, q, n^- \rangle \langle E, q, n^- |\mathrm{d}| E_1 \rangle$$

$$= \langle E_1 \mathrm{d}P | P \mathrm{d} E_1 \rangle,$$

$$\mu_{22}^{(q)} = \sum_n \langle\, E_2\, |\mathrm{d}|\, E, q, n^- \,\rangle\langle\, E, q, n^-\, |\mathrm{d}|\, E_2 \,\rangle$$

$$= \langle E_2 \mathrm{d}P | P\mathrm{d}E_2 \rangle,$$

$$\mu_{12}^{(q)} = \sum_n \langle\, E_1\, |\mathrm{d}|\, E, q, n^- \,\rangle\langle\, E, q, n^-\, |\mathrm{d}|\, E_2 \,\rangle$$

$$= \langle E_1 \mathrm{d}P | P\mathrm{d}E_2 \rangle,$$

where

$$P \equiv \sum_n |\, E, q, n^- \,\rangle\langle\, E, q, n^-\, |$$

Hence the $\mu_{11}^{(q)}$, $\mu_{22}^{(q)}$ and $\mu_{12}^{(q)}$ matrix elements are related as scalar products of the $|E_1 \mathrm{d}P\rangle$ and $|P\mathrm{d}E_2\rangle$ vectors. By the Schwarz inequality,

$$|\langle E_1 \mathrm{d}P | P\mathrm{d}E_2\rangle|^2 \leq \langle E_1 \mathrm{d}P | P\mathrm{d}E_1\rangle \langle E_2 \mathrm{d}P | P\mathrm{d}E_2\rangle$$

with the equality holding only when the $|E_1 \mathrm{d}P\rangle$ and $|P\mathrm{d}E_2\rangle$ vectors are parallel.

If P is a projection onto a single state i.e.,

no n summation, this in fact is the case,

because by definition

$$|\langle E_1 \mathrm{d}|\, E, q^- \rangle\langle E, q^-\, |\mathrm{d}E_2\rangle|^2 =$$

$$\langle E_1 \mathrm{d}|\, E, q^- \rangle\langle E, q^-\, |\mathrm{d}E_1\rangle \cdot$$

$$\langle E_2 \mathrm{d}|\, E, q^- \rangle\langle E, q^-\, |\mathrm{d}E_2\rangle$$

In all other cases due to the existence of many n internal states the strict inequality holds and complete control can never be realized. Nevertheless one can minimize $P^{(q)}(E)$ as much as possible. In that case the solution of the CC equation yields the "optimal" solution.

Bichromatic "coherent control" (Chem. Phys. Lett. 126, 541 (1986))

B + A-C   A + B-C

E

ω_{E2}

ω_{E1}

E_2

pathway a   pathway b

E_1

ω_{2g}

ω_{1g}

E_g

A-B-C
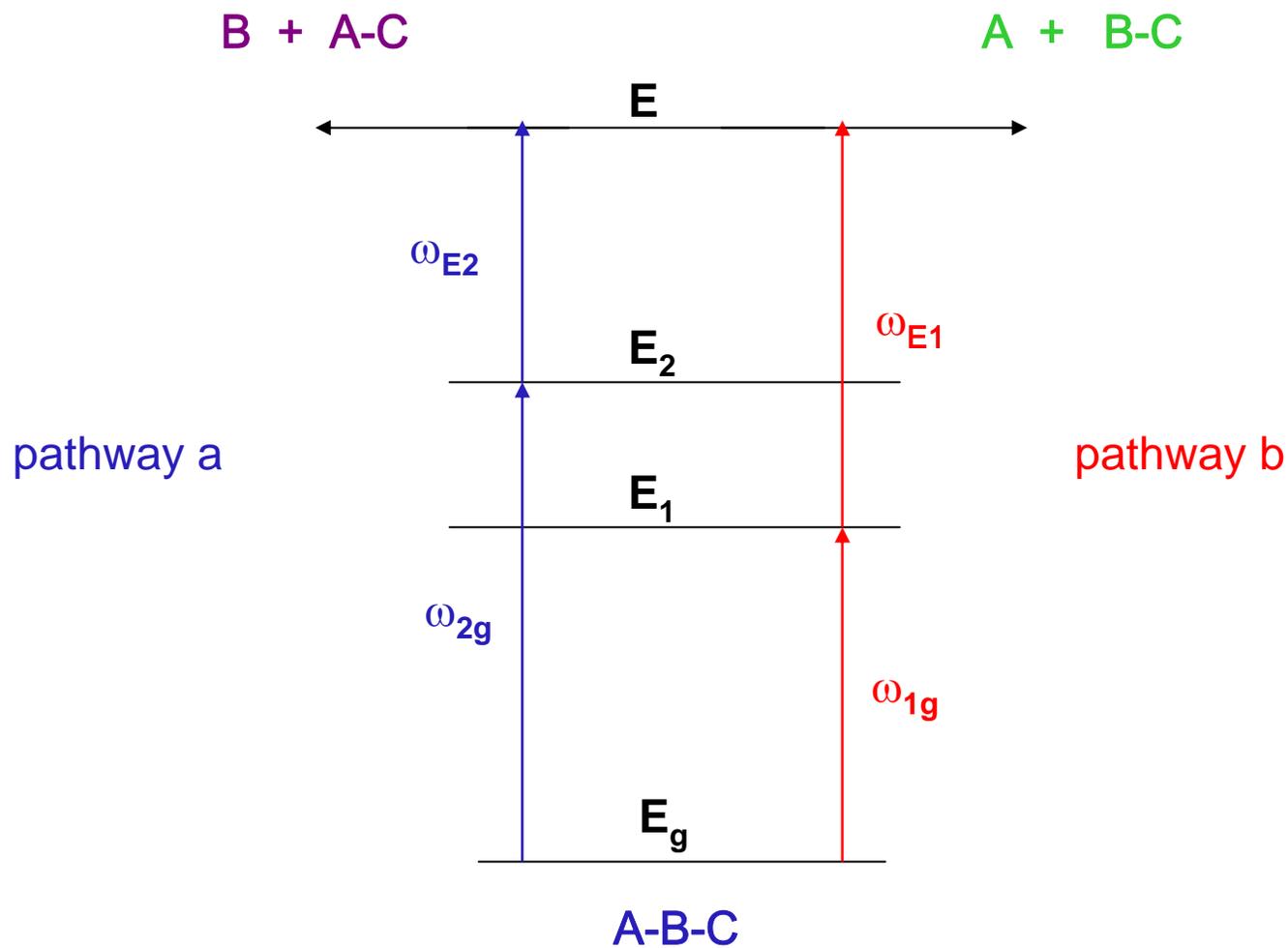
A second objective: to control of the direction of electronic motion.
The generation of current without voltage!

(G. Kurizki, M. Shapiro, P. Brumer, Phys. Rev. B, **39**, 3435 (1989)).

pathway a

1-photon path 5.3μm

pathway a

pathway b

1-photon path 5.3μm

Nonresonant 2-photon path 10.6μm

IE,A>

IE,S>

Energy/meV

300

200

100

0

-40 -20   0   20   40

distance/nm
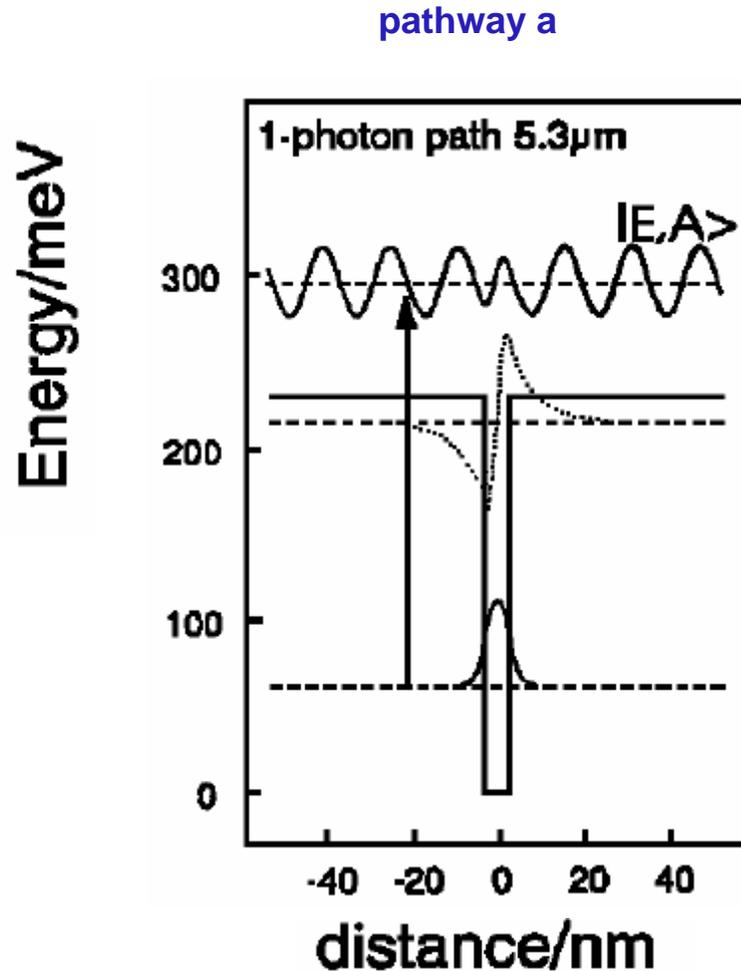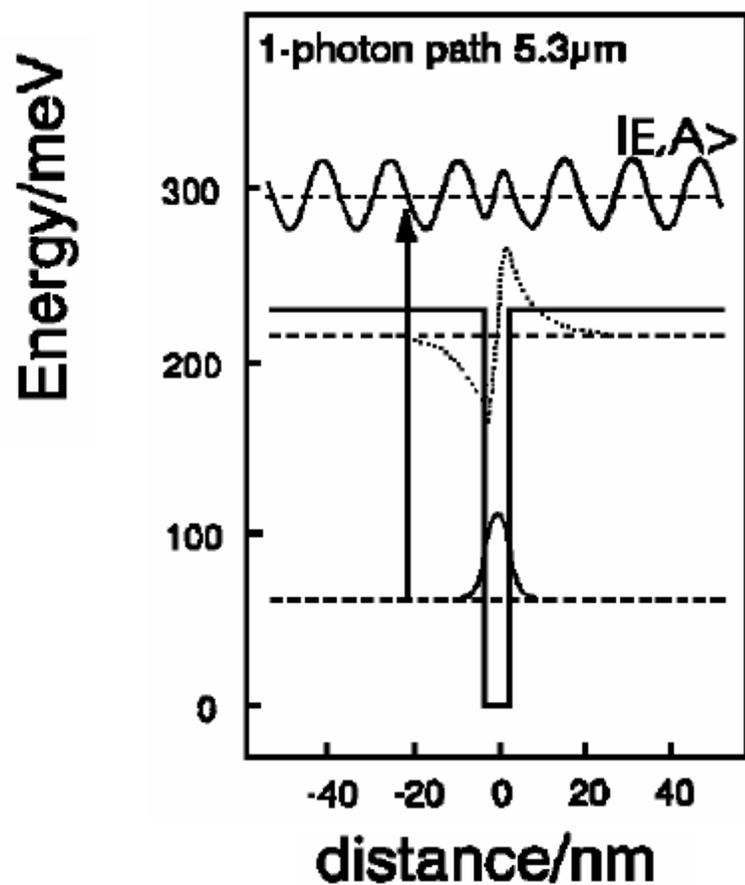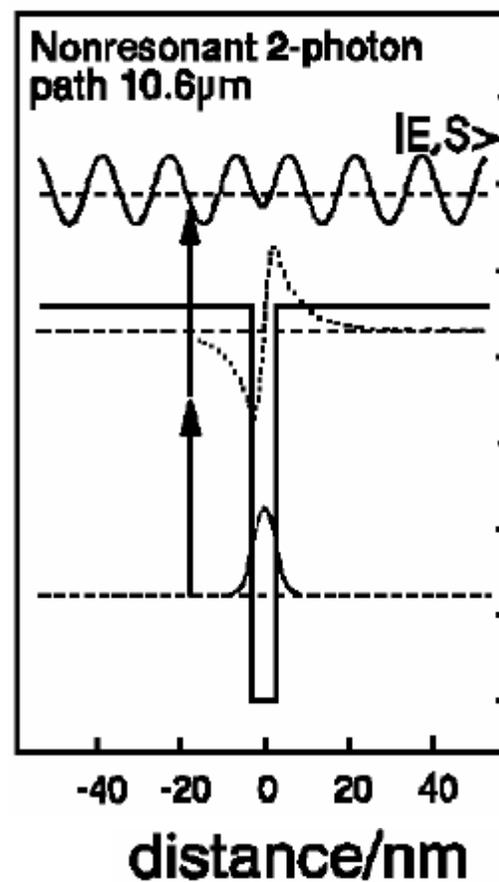
We can break the symmetry by irradiating the system with two fields,

$$A_1 \, e^{-i\omega_1 t - i\phi_1}$$ leading to a 1 photon transition, and

$$A_2 \, e^{-i\omega_2 t - i\phi_2}$$ where $\omega_1 = 2\,\omega_2$ leading to a 2 photon transition.

The initial state is $|k_v\rangle$ and the final state is $|k_c\rangle$

The probability of populating conduction state $|k_c\rangle$ is given as,

$$P_{k_c} = \left| A_1 p_{cv} e^{-i\phi_1} + A_2^2 \sum_i \frac{p_{c,i} p_{i,v}}{\hbar\omega_2 - E_i} e^{-2i\phi_2} \right|^2$$

where $\quad p_{cv} = \langle \mathbf{k}_c | \mathrm{p} | \mathbf{k}_v \rangle \quad p_{ci} = \langle \mathbf{k}_c | \mathrm{p} | \mathbf{k}_i \rangle$

$$p_{iv} = \langle \mathbf{k}_i | \mathrm{p} | \mathbf{k}_v \rangle$$

Opening the square we obtain that

$$P_{k_c} = P_{k_c}^{1,1} + P_{k_c}^{2,2} + P_{k_c}^{1,2}$$

where

$$P_{k_c}^{1,1} = \left| A_1 p_{cv} \right|^2 \quad , \quad P_{k_c}^{2,2} = \left| A_2^2 \sum_i \frac{p_{c,i} p_{i,v}}{\hbar\omega_2 - E_i} \right|^2$$

$$P_{k_c}^{1,2} = B_{k_c} \exp(i\delta_{k_c}) \exp(i\phi_1 - 2i\phi_2) + c.c.$$

where

$$B_{k_c} = \left| A_1 p_{cv} \cdot A_2^2 \sum_i \frac{p_{c,i} p_{i,v}}{\hbar\omega_2 - E_i} \right|$$

so that

$$P_{k_c}^{1,2} = B_{k_c} \cos(\delta_{k_c} + \phi_1 - 2\phi_2)$$

In particular

$$\delta_{-k_c} = \pi + \delta_{k_c}$$

E. Dupont, P.B. Corkum, H.C. Liu, M. Buchanan, and Z.R. Wasilewski, Phys. Rev. Lett. **74**, 3596 (1995)
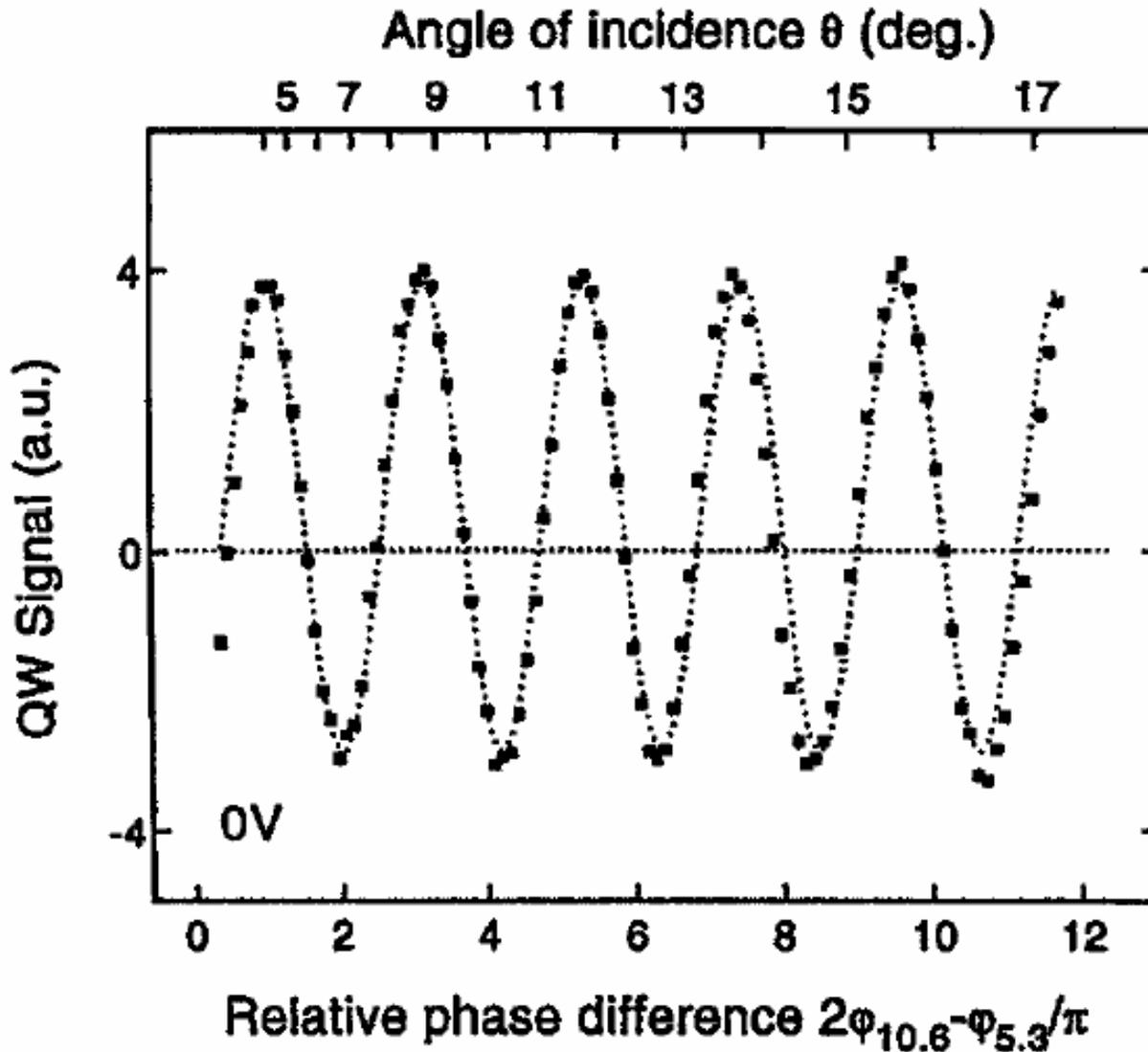
A pictorial representation

Anti-symmetric

Symmetric
(s wave)

1- photon absorption

p wave

2- photon absorption

or

s wave

d wave

Symmetric

(forward current)

j

$\phi_1 - 2\phi_2 = 0$

pathway a

$+$
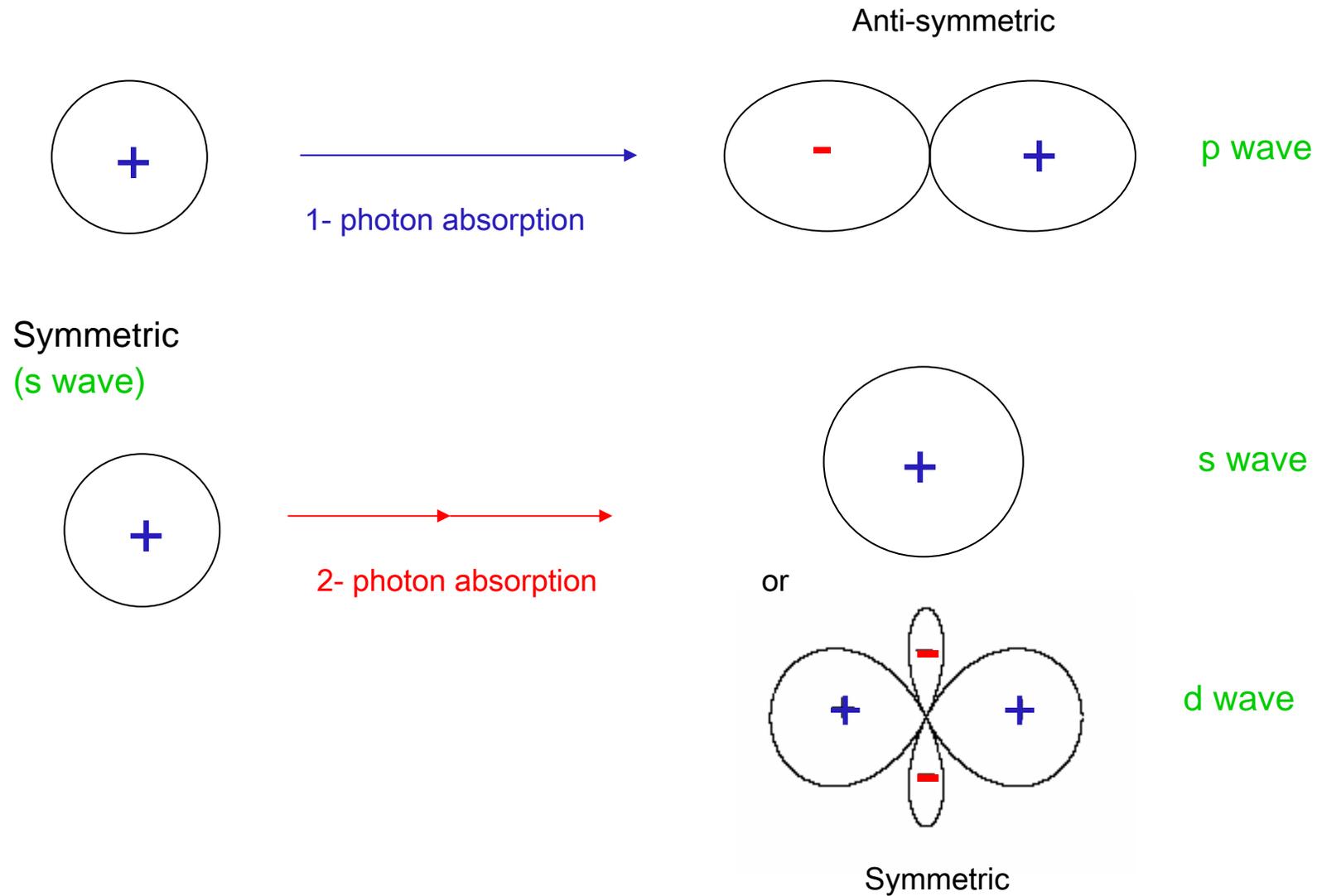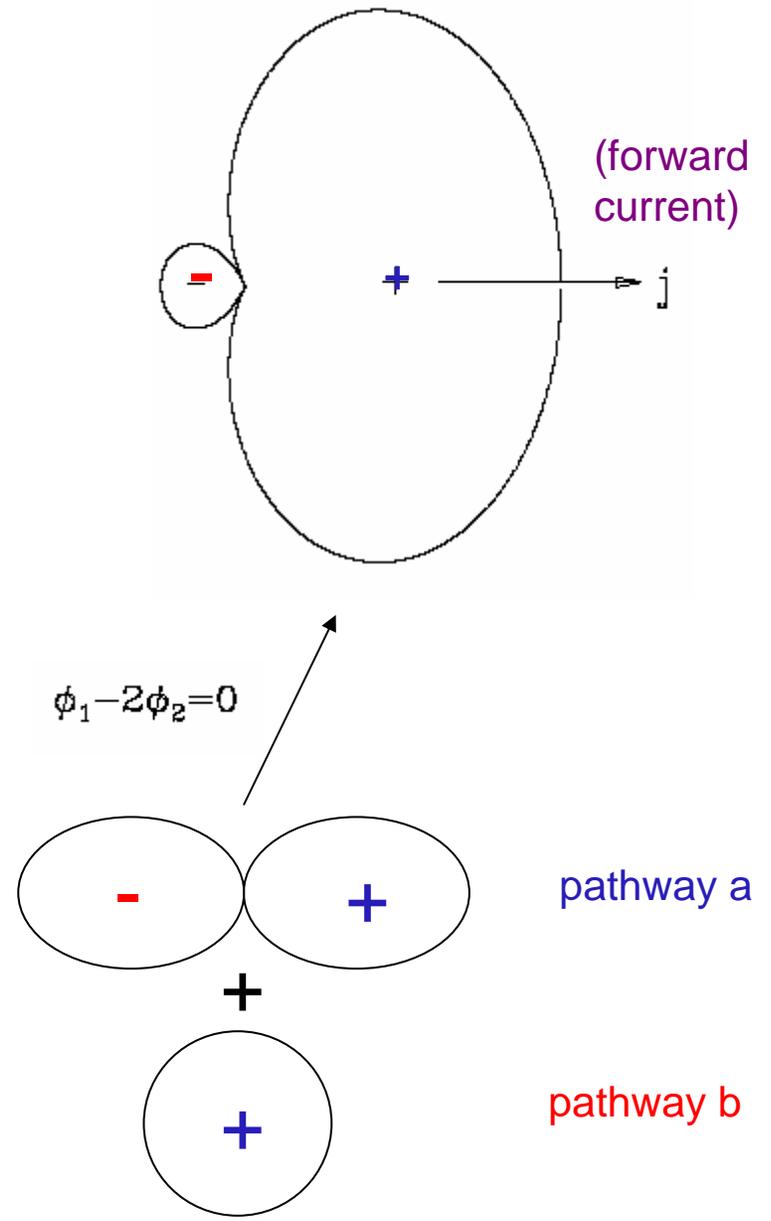
pathway b

(backward
current)

(forward
current)

j ⟵

⟶ j
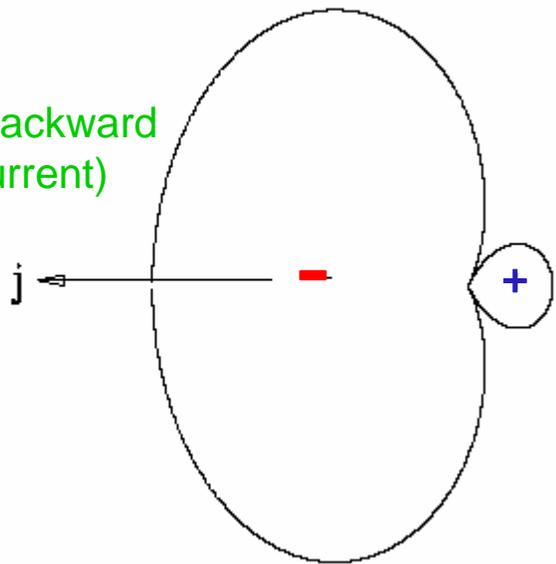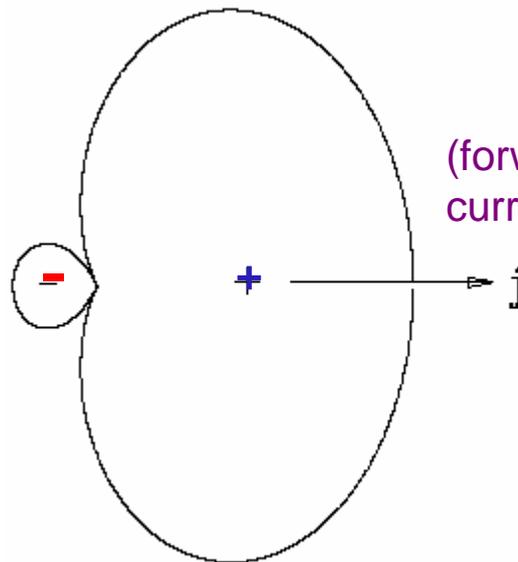
$\phi_1 - 2\phi_2 = \pi$

$\phi_1 - 2\phi_2 = 0$

−   +

+

−

−   +

+

+
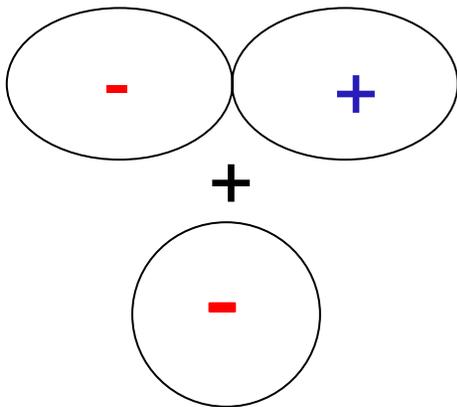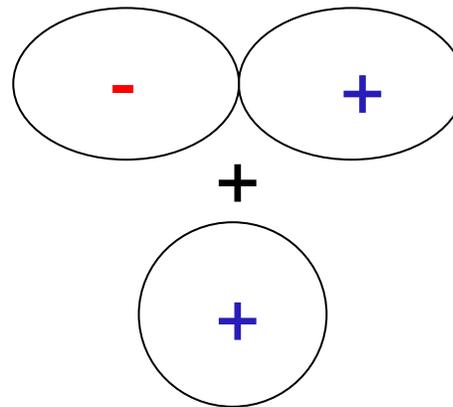
pathway a

pathway b

Generation of DC current in a molecular "wire" suspended between two leads

$\phi = 0$

$\phi = \pi / 2$

LL

RR
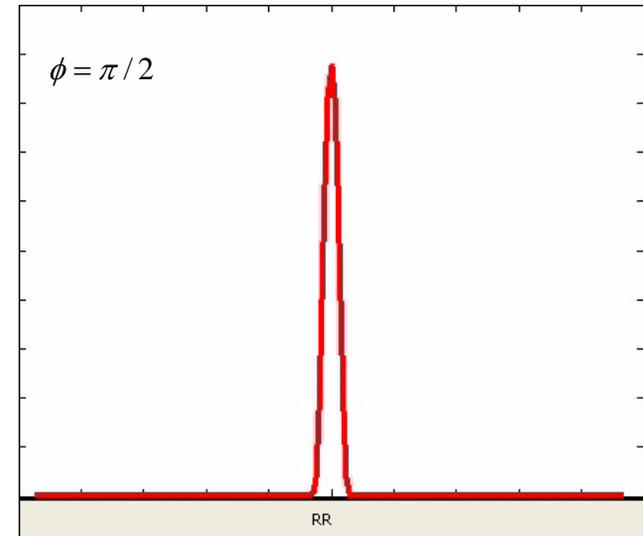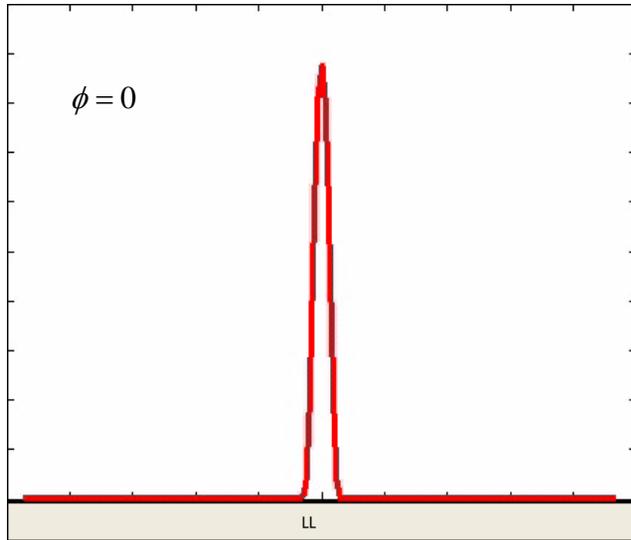
$$\dot{\psi} = -iH\psi$$

$$H = -\frac{\hbar^2}{2m}\frac{d^2}{dx^2} + V(x) + U(x,t)$$

$$V(x) = \begin{cases} 0 & |x| > 0.1 \text{ nm} \\ -0.5 \text{ eV} & |x| < 0.1 \text{ nm} \end{cases}$$

$$U(x,t) = x\left[\sin(\omega t) + \sin(2\omega t + \varphi)\right]f(t)$$

$$f(t) = e^{-(t/\lambda)^2} \quad \text{a short pulse}$$

$\omega=0.1$; $\Delta t= 3 \ 10^{-2}$ [au]
$\lambda=150$
nstep=100; N_T=2000;
$\Psi(x,t=0)=\phi_0(x)$

V(x) [eV]

x [A]
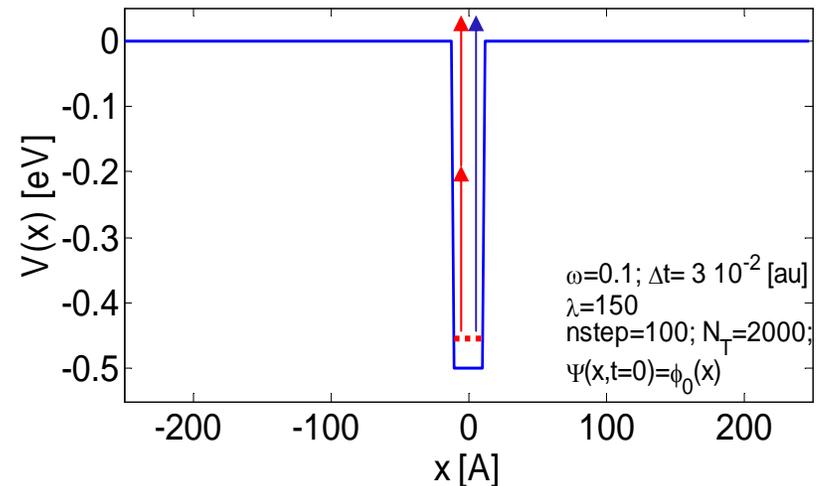
Control of Electron Localization in Molecular Dissociation

M. F. Kling,[1] Ch. Siedschlag,[1] A. J. Verhoef,[2] J. I. Khan,[1] M. Schultze,[2] Th. Uphues,[3] Y. Ni,[1] M. Uiberacker,[4] M. Drescher,[3,5] F. Krausz,[2,4] M. J. J. Vrakking[1]

A

D_2^{2+}

RCI

SI

$2p\sigma_u^+$

D_2^+

EI

$D^+ + D^+$

RCE

SE

$1s\sigma_g^+$

BS

$D^+ + D$

$D + D$

D_2

e^-

energy

bond distance

B

LP 8.0 fs

LP 6.5 fs

LP 5.0 fs

CP 5.0 fs

signal / a.u.

0   2   4   6   8   10   12

energy / eV
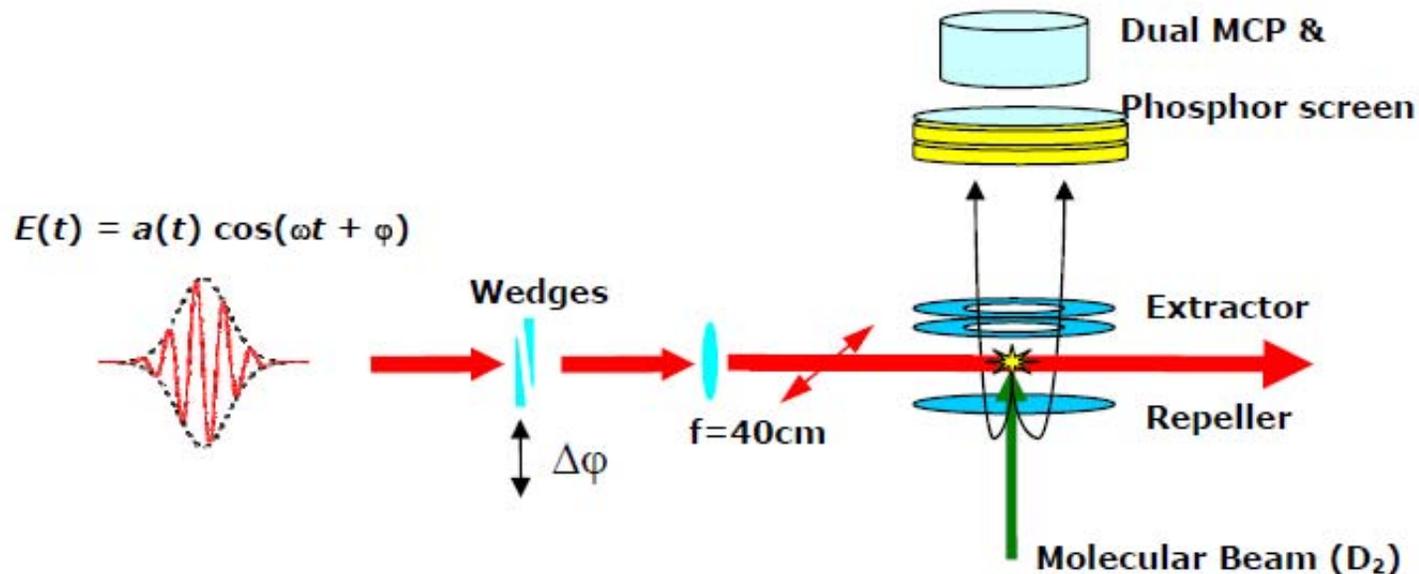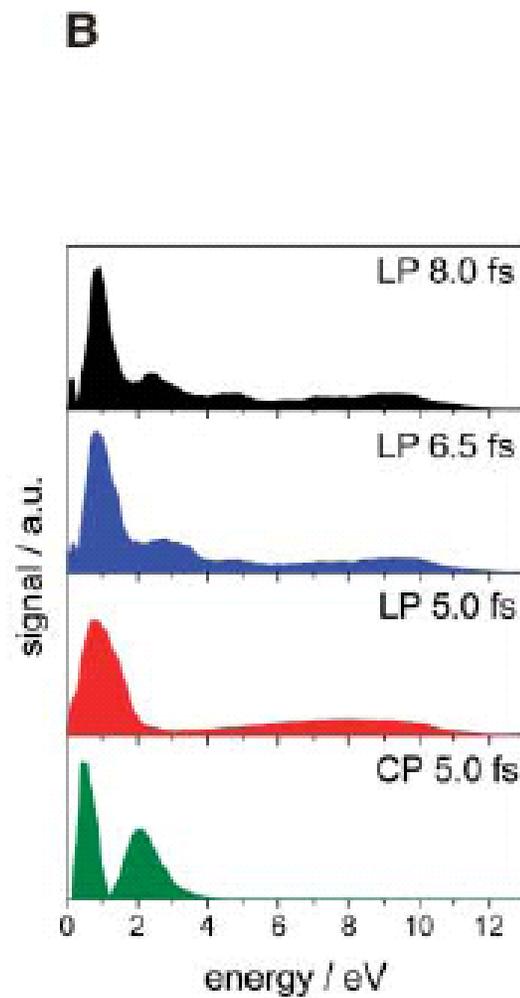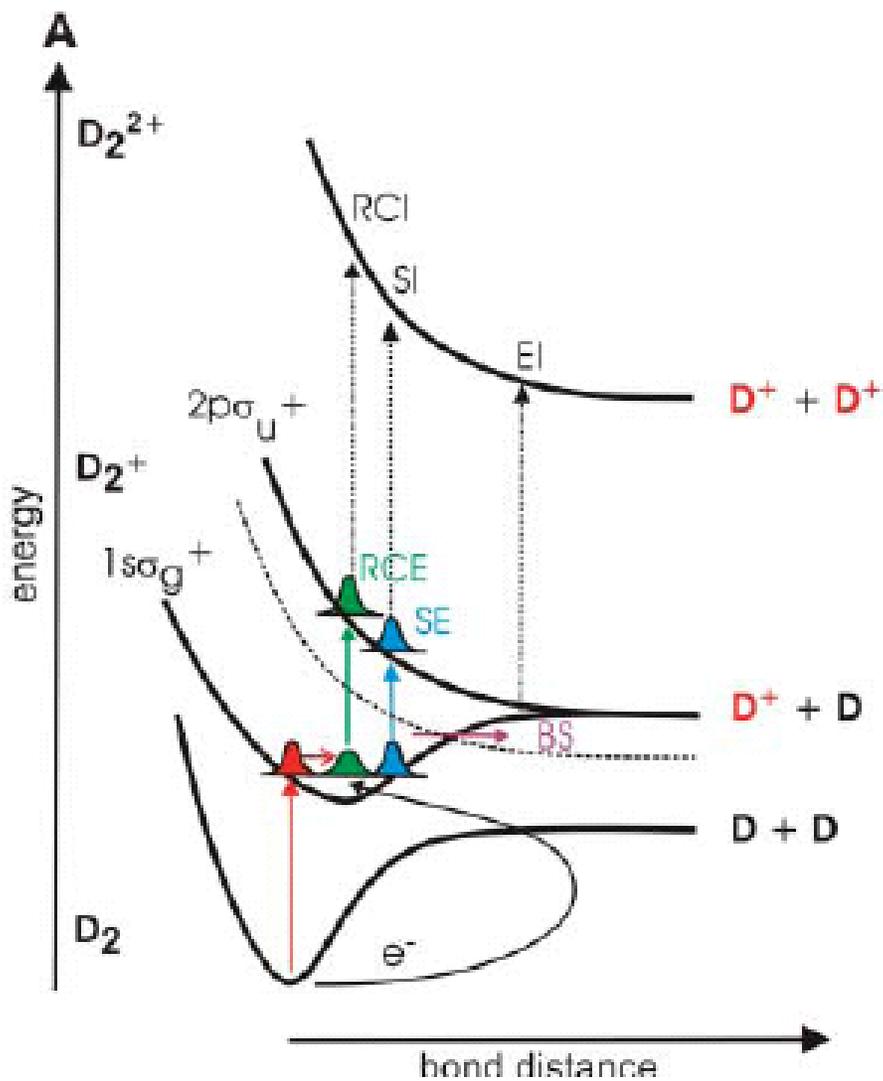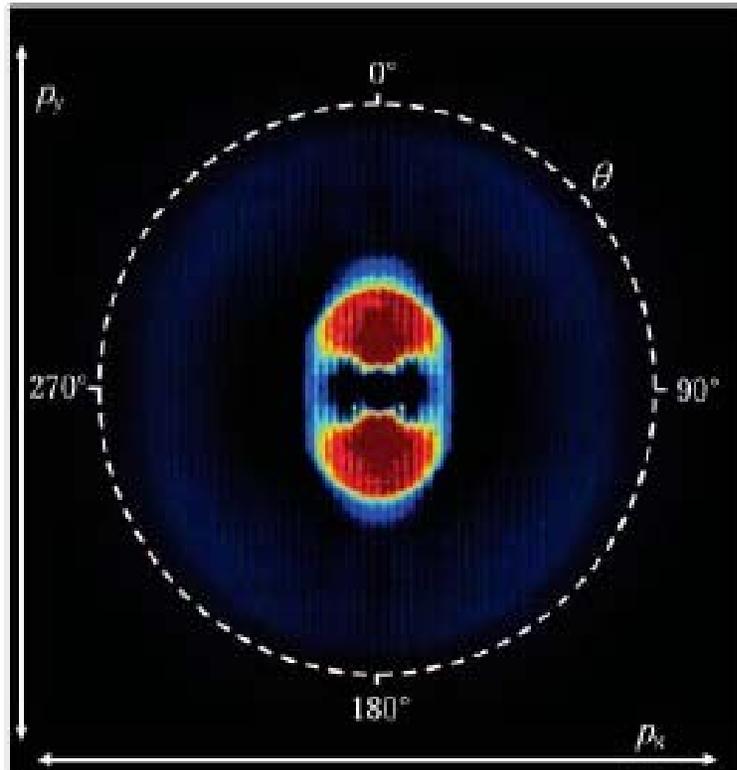
Fig. 2. Two-dimensional D^+ momentum image for D_2 dissociation in a 5-fs, 1×10^{14} W cm^{-2} laser field without phase stabilization.

signal

A

B

C

0 - 3 eV   3 - 8 eV
> 8 eV

energy / eV

asymmetry

phase / π

J

$$C_2H_3Cl + \omega_3, 3\omega_1 \rightarrow C_2H_3Cl*$$

$$C_2H_3Cl* \rightarrow C_2H_3 + Cl$$

$$C_2H_3Cl* \rightarrow C_2H_2 + HCl$$

$$C_2H_3Cl* + 2\omega_1 \rightarrow C_2H_3Cl^+.$$
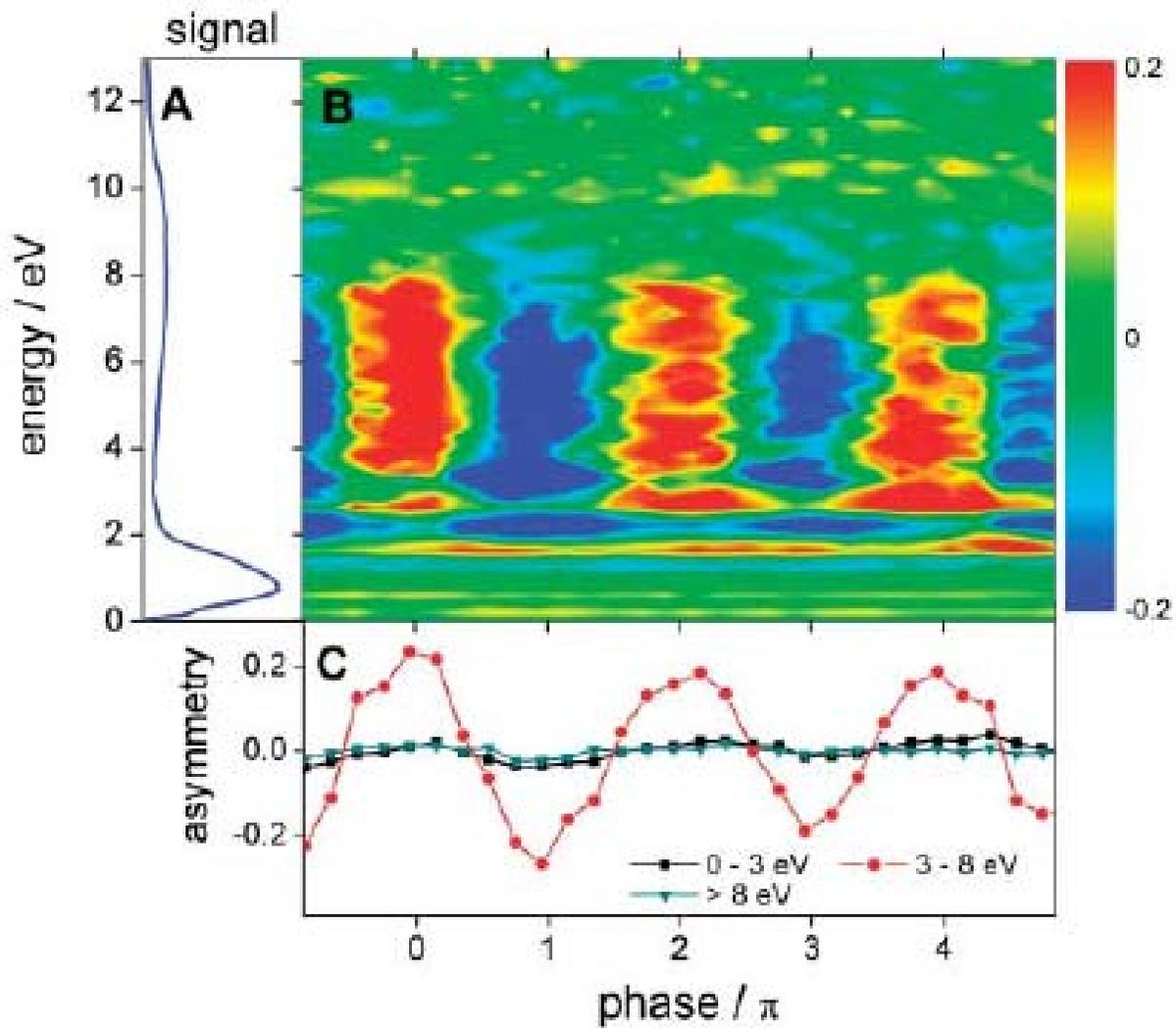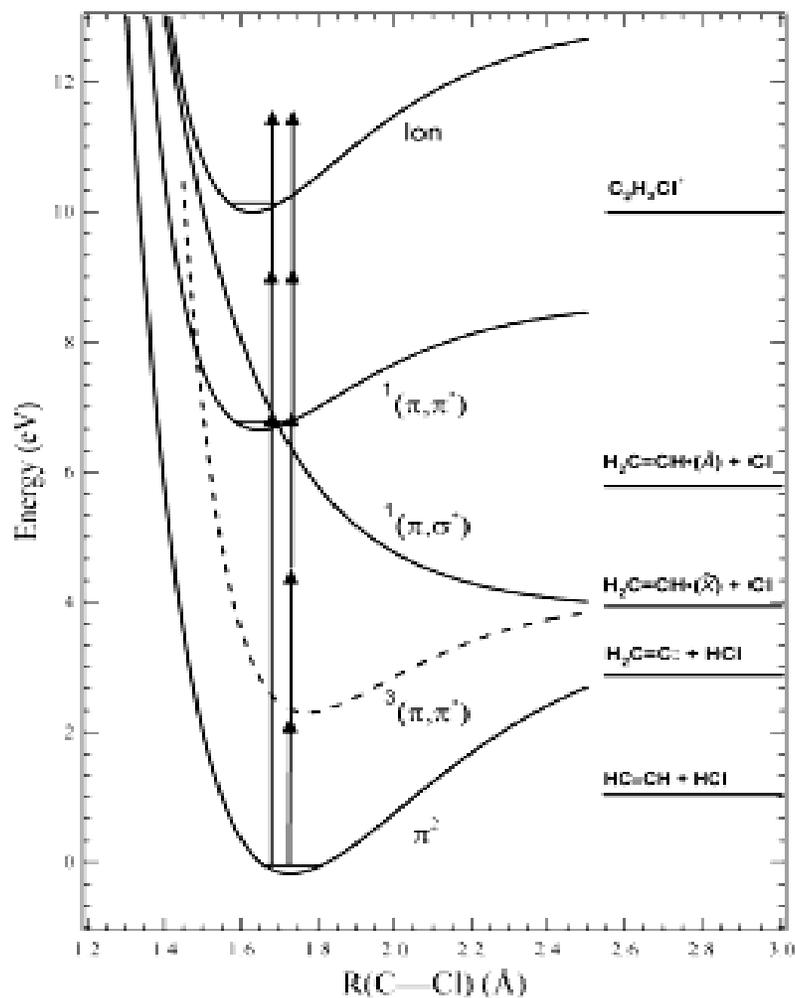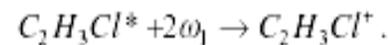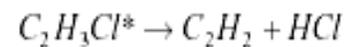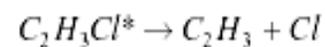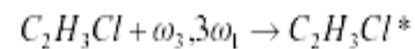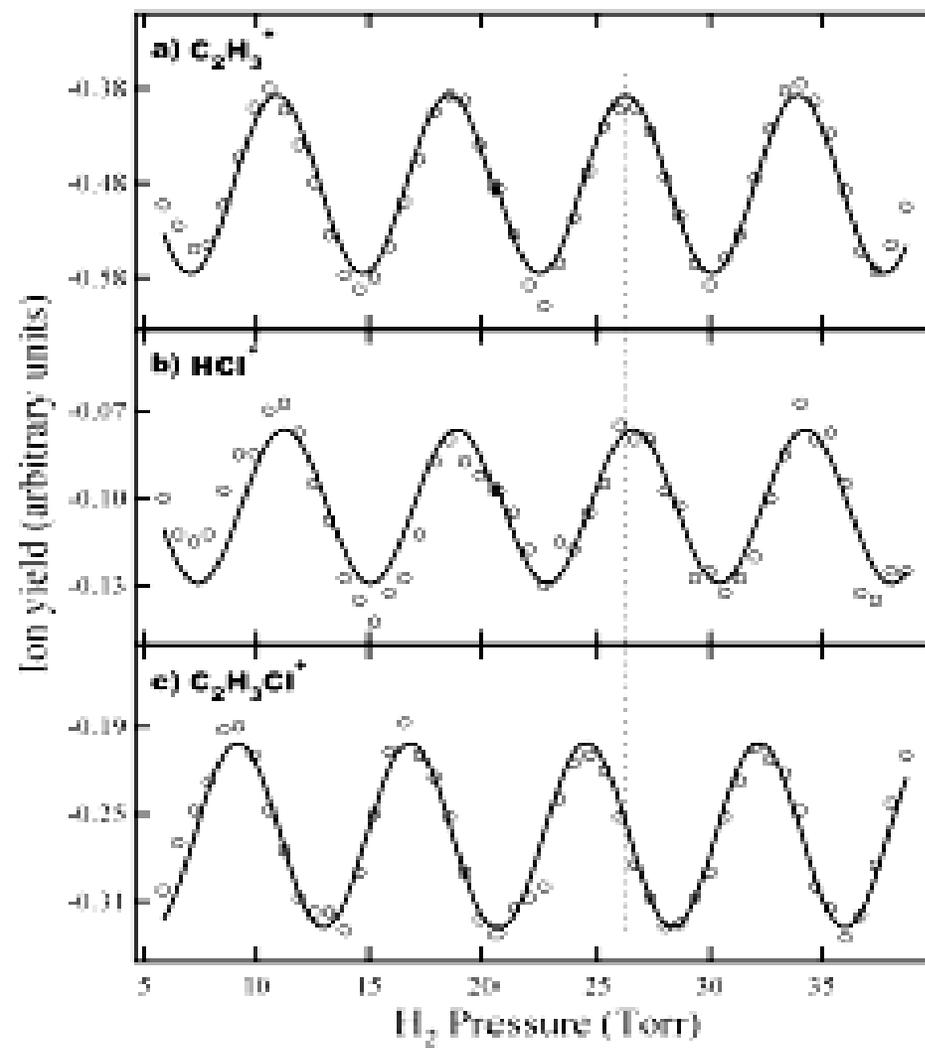
Milestones in the development of coherent control
## blue – coherent control (theory)
red - experiments

- **1985/1986:** PB +MS (trying to understand why control in IR multiphoton dissociation failed) suggest using quantum interference for control. Introduction of bi-chromatic control.
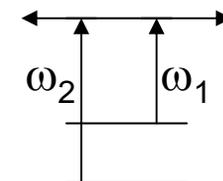

- **1987**: First theoretical results on the control of realistic systems: The control of electronic branching in the $CH_3 + I^* \leftarrow CH_3I \rightarrow CH_3 + I$, reaction

- **1987:** Coining the phrase "Coherent Control"

- **1988:** (with Asaro) Elliptic polarization control of differential cross-sections

1988: (with Hepburn) Introduction of 1 photon vs. 3 photon interference as a means of control

B+AC   A+BC

ABC
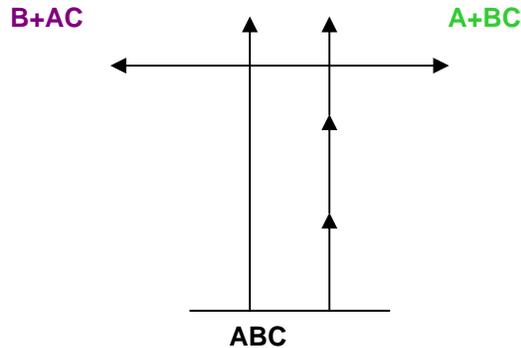
1989: (with Kurizki) Introduction of 1 photon vs. 2 photon as a means of symmetry breaking and differential control: prediction of creation of DC photo-current in semi-conductors with *no bias voltage*.

conduction band

backward   forward

e⁻
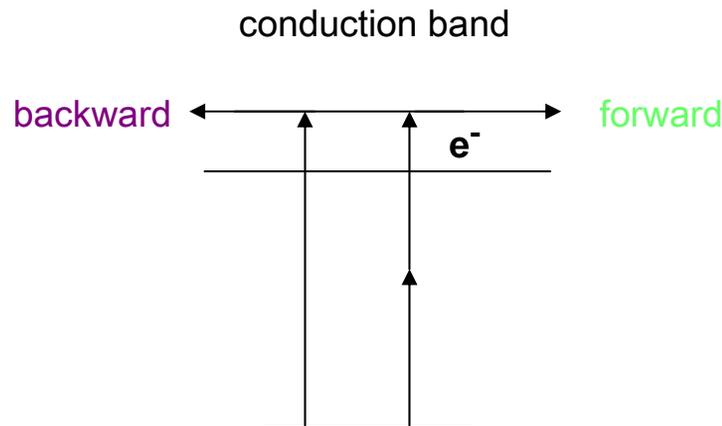
1990: First experimental verification of the 1 vs. 2 coherent control scenario. Control over photo-current directionality in semiconductors (Zeldovitch)

1990/1991: First experimental verifications of the 1 vs. 3 coherent control scenario: The control of photoionization yields in atoms (Elliott, Bucksbaum) and molecules (Gordon, Bersohn)

1991: Coherent control of molecular chirality introduced

1991: First experimental realization of the pump-dump control scheme in Na $+$ Na $\longleftarrow$ Na$_2$ $\longrightarrow$ Na$_2^+$ **(**Baumert +Gerber)

1994/1995: Interference Control with Incoherent light introduced

B+AC   A+BC

$-\phi_2$ ϕ_2

ϕ_1
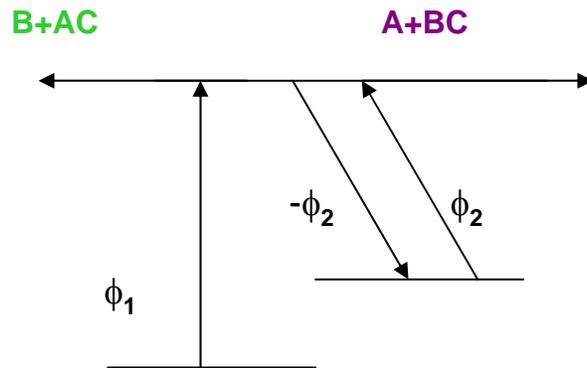
1995: Experimental demonstration of coherent control of photo-current generation in quantum wells by the 1 vs. 2 photon scenario (Corkum)

1-photon path 5.3μm   IE,A>

Nonresonant 2-photon path 10.6μm   IE,S>
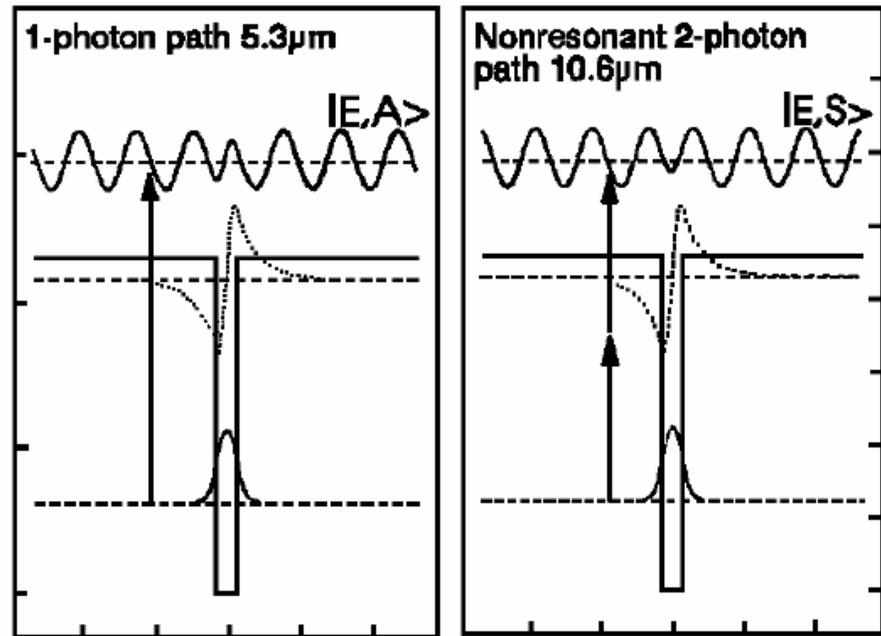
1995: Experimental demonstration of control of electronic hopping in a dissociating molecule, in
1 vs. 2 photon $H^+ + D \longleftarrow HD^+ \longrightarrow H + D^+$ (DiMauro)

1995: (Gordon) First experimental demonstration of control over a branching process: control over the ionization vs. dissociation in 1 vs. 3 photon $HI^+ \longleftarrow HI \longrightarrow H + I$

**1996:** First experimental demonstration of control over electronic branching ratios: interference control with incoherent light in
$Na + Na(3p) \longleftarrow Na_2 \longrightarrow Na + Na(3d)$ (Shnitman+Yogev+,Shapiro)

1997: (with Vardi) Photoassociation of ultracold atoms to form ultracold molecules via Coherent Raman Process suggested

1997: First experiments on automated feedback control via pulse shapings (Wilson, Silberberg, Gerber)

1998: Experimental demonstration of coherent control via phase modulation of two photon absorption of atoms (Silberberg)

1998: (Gerber) First experimental demonstration of adaptive feedback control of a branching photochemical reaction: Control over the photodissociation of $Fe(CO)_5$ and

$$Cp + 2CO + FeCl \longleftarrow CpFe(CO)_2Cl \longrightarrow CpFeCOCl + CO$$

2000: Coherent Control of refractive indices introduced

2000: (with Frishman) Enantiomeric purification of chiral racemic mixtures by coherent control techniques

- **2000:** Theory of nanoscale deposition on surfaces

- **2000:** (with Frishman) Coherent Suppression of spontaneous emission using overlapping resonances

2001: Adaptive feedback control of the photodissociation of $C_6H_5CO + CH_3 \longleftarrow C_6H_5CH_3CO \longrightarrow C_6H_5 + CH_3CO$ in the high field regime (Levis)
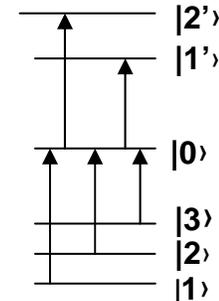
2002: (with Kral): An exact analytical solution of the non-degenerate quantum control problem

$|2'\rangle$
$|1'\rangle$
$|0\rangle$
$|3\rangle$
$|2\rangle$
$|1\rangle$

2002: Adaptive feedback control of internal conversion channel in a light harvesting antenna complex of photosynthetic purple bacterium (Motzkus)

2003: (with Kral and Thanopoulos): An analytic solution for the degenerate quantum control problem

2003: (with Zhang and Keil) Experimental demonstration of phase locking between two 2-photon processes in 2-photon vs. 2-photon coherent control

2003-2004: Experimental control of high harmonic generation (Murnane, Gerber)

2004: Coherent Control techniques used in the "streak camera" phase measurement attosecond pulses (Krausz, Corkum)
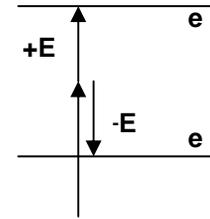
- **2005:** Control of radiationless transitions by interference between overlapping resonances

2005: (with Thanopoulos) Automatic repair of mutations caused by dihydrogenic tunneling between two nucleotides using coherent control

2005: (with Kral) Coherent control with non-classical light

2005: Experimental demonstration of coherent photoassociation of Rb + Rb BEC to form Rb_2 molecular BEC using the Vardi+Shapiro scheme (Grimm)

2005: Experimental demonstration of bond breaking selectivity in the CH_2=CH. + Cl ⟵ CH_2=CHCl ⟶ CH= CH + HCl (Gordon)

2005: (with Segal) Suggestion of quantum computation using electron trapped in an

quadrupoles of carbon nanotubes