POPULATION TRANSFER PROCESSES: FROM ATOMS TO CLUSTERS AND BOSE-EINSTEIN CONDENSATE

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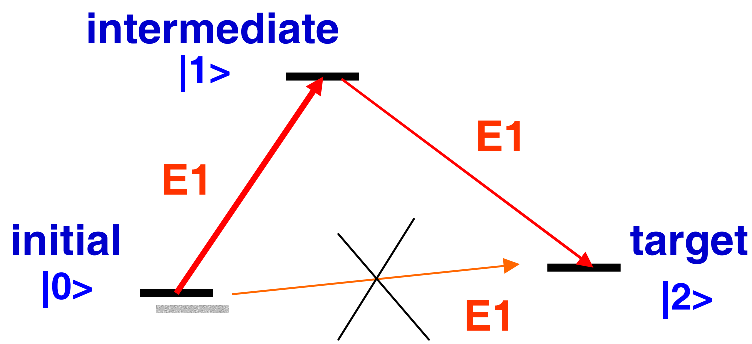
Typical problem of atomic/molecular spectroscopy:
how to provide the population transfer between the levels which cannot be directly related by dipole transition?



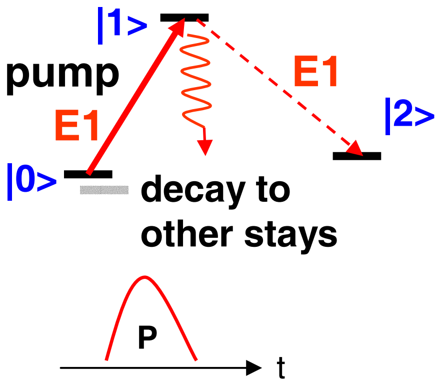
Modern quantum optics: various methods for two-photon population transfer in atoms and simple molecules:

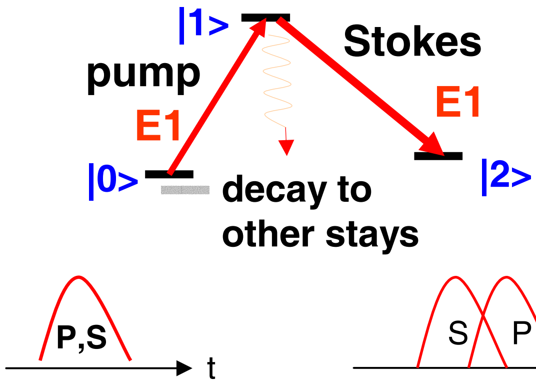
- Raman scattering,
- stimulated Raman,
- Rapid Adiabatic Passage (RAP)
- STIimulated Raman Adiabatic Passage (STIRAP),
- Stark-shift-Chirped Rapid Adiabatic Passage (SCRAP),
- ....

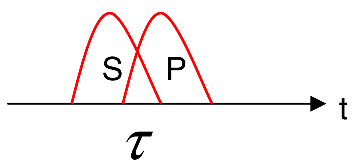
Is it possible to use these methods for other systems:
- **exploration of electronic of metal clusters**
- **transport of BEC**

?

Two-photon population transfer methods:







K. Bergman, et al,
Rev. Mod. Phys., 70, 1003 (1998)

- adiabatic process
- counterintuitive pulse order
- partial overlap
- dark state

Simple Raman scattering:
- only pump
- low transfer

Stimulated Raman scattering:
- pump + Stokes
- transfer up to 30%

STImulated Raman Adiabatic Passage (STIRAP):
- Stokes + pulse
- transfer up to **100% !!!**

Dressed states in STIRAP:

$$|\, a^{+} >= \sin\theta \sin\phi |\, 0 > + \cos\phi |\, 1 > + \cos\theta \sin\phi |\, 2 >$$

$$|\, a^{0} >= \cos\theta |\, 0 > + \sin\theta |\, 2 >$$ ← dark state: = |0> at t = 0; = |2> at t = ∞

$$|\, a^{-} >= \sin\theta \cos\phi |\, 0 > + \sin\phi |\, 1 > + \cos\theta \cos\phi |\, 2 >$$

$$\sin\theta = \frac{\Omega_P}{\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}}, \quad \cos\theta = \frac{\Omega_S}{\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}}$$

Is it possible to apply the fascinating methods of modern quantum optics to:

- exploration of electronic spectra in metal clusters,
- transport of BEC in multi-well traps or between BEC components

?

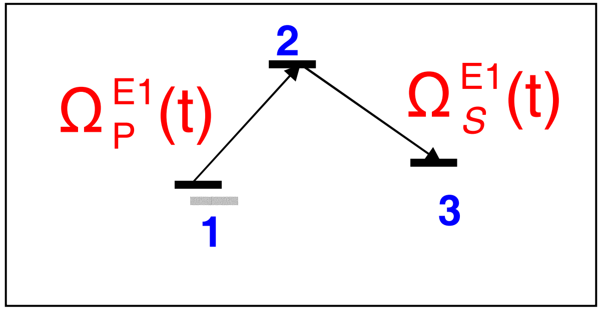
Atomic clusters: why not?

But the problems of:

- extremely short lifetimes (10-1000 fs),
- competition with plasmon mode,
- strong dynamical stark shifts from intense pulses

Transport of Bose-Einstein condensate: why yes?

Because TPP and BEC tunneling are similar physically and mathematically!



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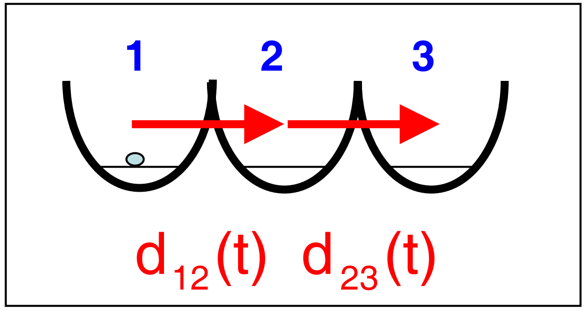
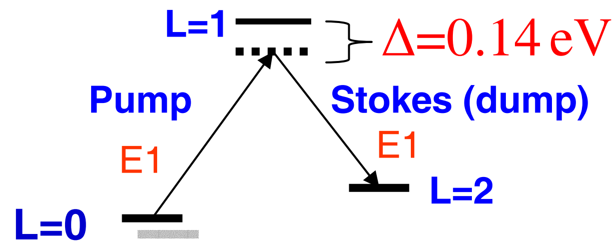


K. Eckert et al, PRA, 70, 023606 (2004)

But detrimental non-linear impact of interaction between BEC atoms!

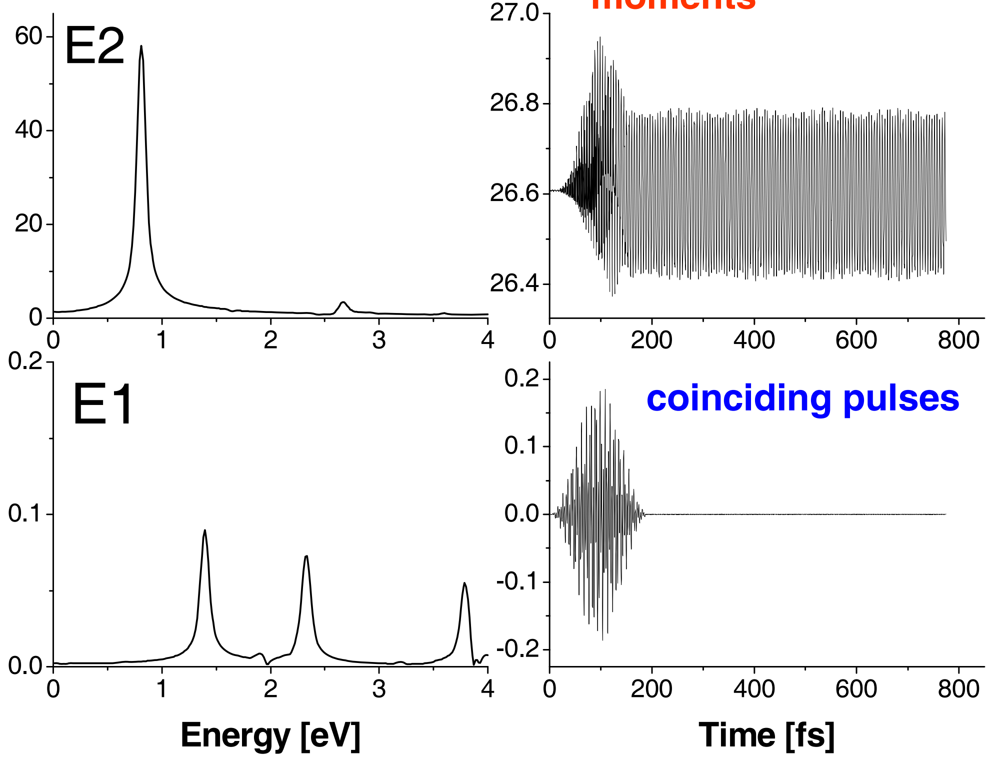
Atomic clusters: off-resonant stimulated Raman transfer to the quadrupole 1eh state at 0.8 eV

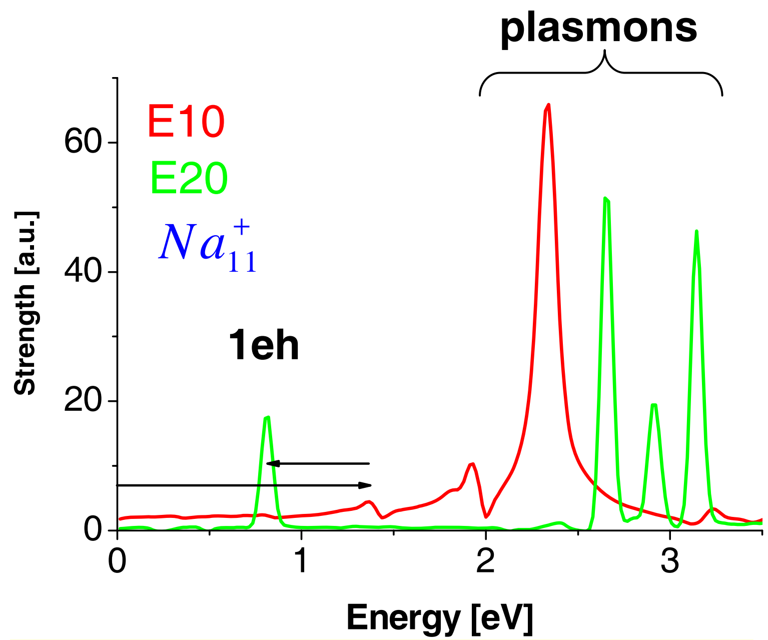
$e_{eh} = e_h - e_e \longrightarrow$ **Direct access to s-p spectra!**



E1/E2 strength

Oscillating E1/E2 moments





$\omega_{pump} = 1.25$ eV, $\omega_{stokes} = 0.45$ eV

$T_s = T_p = 200$ fs, $T_{shift} = 0$

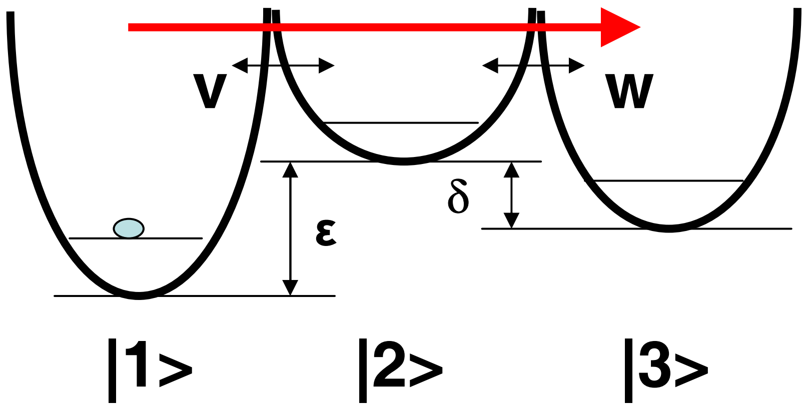
$I_s = 2.2 \times I_p = 2.2 \times 10^{10}$ W/cm^2

- **Time-dependent HF**
- **Kohn-Sham functional**
- **LDA Perdew-Wang xc**
- **jellium for ions**

ORSR works! But maybe low population?
STIRAP? SCRAP!

STIRAP transport of BEC between the wells

E.M. Graefe et al, PRA, 73, 013617 (2006)



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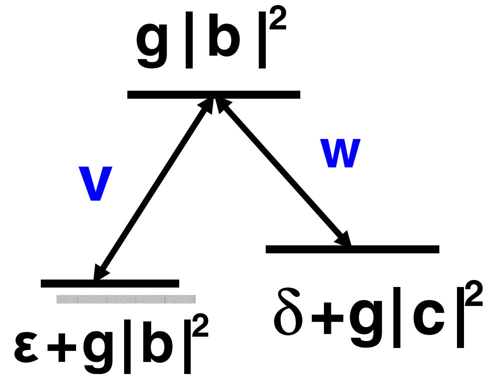


Analog of STIRAP!

$$\psi(t) = a(t)\,|1> + b(t)\,|2> + c(t)\,|3>$$

$$H(|a|^2,|b|^2,|c|^2)\begin{pmatrix} a \\ b \\ c \end{pmatrix} = ih\frac{d}{dt}\begin{pmatrix} a \\ b \\ c \end{pmatrix}$$

The parameters v, w, ε, δ can be varied by controlling the debts or separations of the wells.

$$H(|a|^2,|b|^2,|c|^2)=\begin{pmatrix} \varepsilon+g|a|^2 & v & 0 \\ v & g|b|^2 & w \\ 0 & w & \delta+g|c|^2 \end{pmatrix} \approx H_{STIRAP}=\begin{pmatrix} -2\Delta_P+S_1 & \Omega_P & 0 \\ \Omega_P & S_2 & \Omega_S \\ 0 & \Omega_S & -2\Delta_S+S_3 \end{pmatrix}$$

BEC/STIRAP model:

$\hat{\psi}_k^\dagger(\vec{r},t)$ -creates atom in component k at point r in time t

Equations for 3-component BEC:

$$i\hbar\frac{\partial}{\partial t}\hat{\psi}_k = [\hat{h}_k + \sum_{j=1}^{3} g_{kj}\hat{\psi}_j^\dagger\hat{\psi}_j]\,\hat{\psi}_k + \sum_{j=1}^{3}(1-\delta_{kj})\Omega_{kj}(t)\hat{\psi}_j$$

g_{kj} - interaction

$\Omega_{kj}(t)$ - coupling

Transfer to macroscopic order parameters $\psi_k(t)$: GPE

$$\hat{\psi}_k^\dagger(\vec{r},t) \simeq \psi_k(t)\Phi_k(\vec{r}), \quad \psi_k(t) = \sqrt{NN_k(t)}\exp\{-i\varphi_k(t)\}$$

$N_k(t)$ - normalized population

$\varphi_k(t)$ - phase

Equations for phases and populations:

$$\begin{cases} \dfrac{\partial}{\partial t}N_k = -\sum_{j=1}^{3}\bar{\Omega}_{kj}(t)\sqrt{N_j N_k}\,\sin(\varphi_k - \varphi_j) \\ \dfrac{\partial}{\partial t}\varphi_k = E_k + \sum_{j=1}^{3}\Lambda_{kj}N_j - \dfrac{1}{2}\sum_{j=1}^{3}\bar{\Omega}_{kj}(t)\sqrt{\dfrac{N_j}{N_k}}\cos(\varphi_k - \varphi_j) \end{cases}$$

$$\Omega_{kj}(t) = K\bar{\Omega}_{kj}(t)$$

$$\bar{\Omega}_{kj}(t) = \exp\{-(\frac{t_{kj}-t}{\Gamma})^2\}$$

The only parameter regulating Interaction-coupling ratio

$$\Lambda_{kj} = \frac{U_{kj}N}{2K}$$

$$\bar{E}_k = E_k/2K, \quad 2Kt \to t$$

$$U_{kj} \sim g_{kj}$$

Scaled dimensionless time

Classical Hamiltonian and canonical equations:

$$H_{CL} = \sum_{k=1}^{3} \bar{E}_k + \frac{1}{2}\sum_{k,j=1}^{3} \Lambda_{kj} N_j N_k - \frac{1}{2}\sum_{j=1}^{3} \bar{\Omega}_{kj}(t)\sqrt{N_j N_k}\cos(\varphi_k - \varphi_j)$$

$$\begin{cases} \dfrac{\partial}{\partial t} N_k = -\dfrac{\partial H_{cl}}{\partial \varphi_k} \\ \dfrac{\partial}{\partial t} \varphi_k = \dfrac{\partial H_{cl}}{\partial N_k} \end{cases}$$

Canonical transformation to new unknowns:

$$\begin{cases} z_k = \sum_{j=1}^{3} T_{kj} N_j & \text{- population imbalances} \\ \theta_k = \sum_{j=1}^{3} R_{kj} \varphi_j & \text{- phase differences} \end{cases}$$

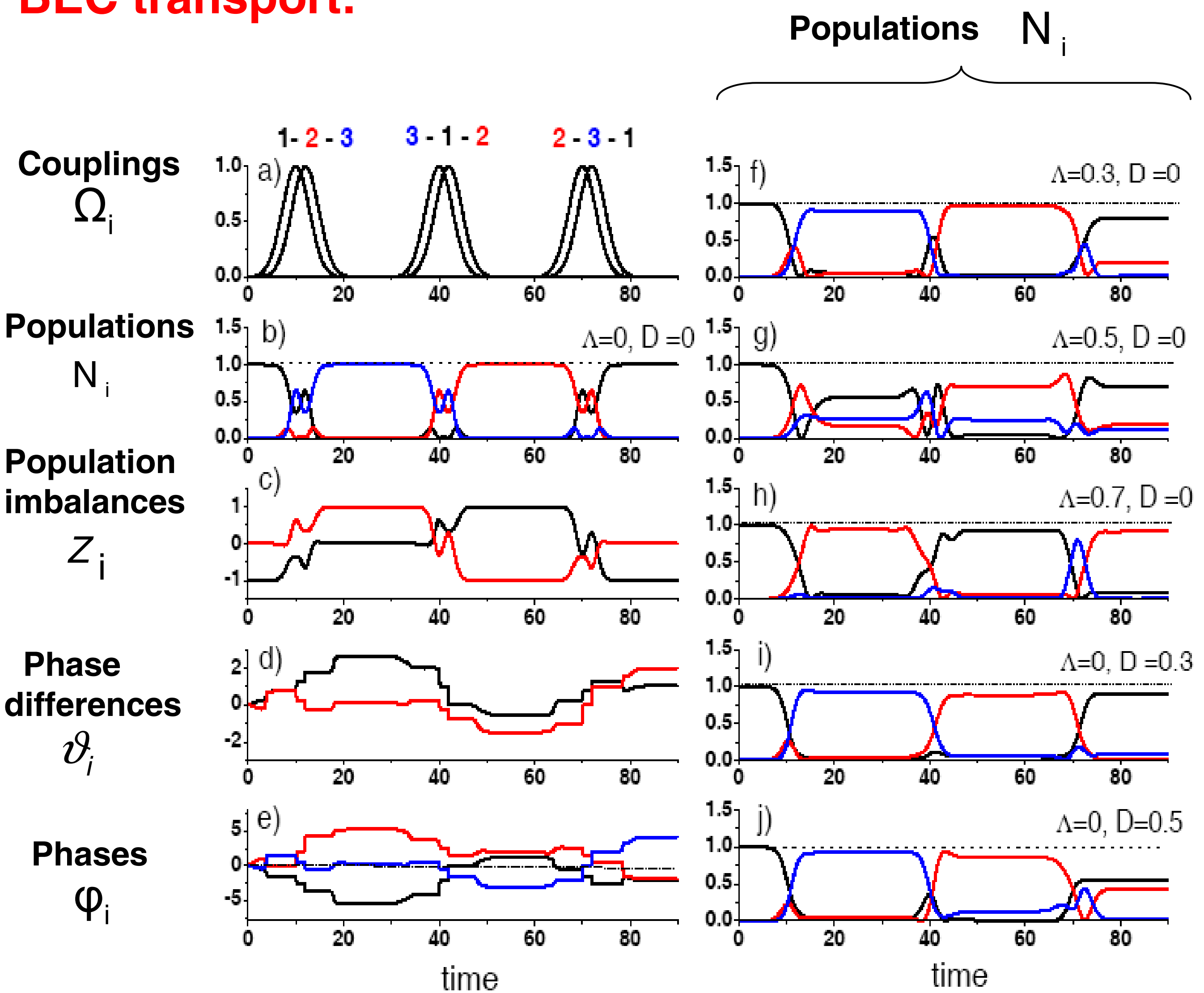
$$T = \begin{pmatrix} -1 & 1 & 0 \\ 0 & -1 & 1 \\ 1 & 1 & 1 \end{pmatrix}$$

$$R = \frac{1}{3}\begin{pmatrix} -2 & 1 & 1 \\ -1 & -1 & 2 \\ 1 & 1 & 1 \end{pmatrix}$$

so as to extract integrals of motion $N = \sum_{k=1}^{3} N_k(t)$ **and** $\Theta = \sum_{k=1}^{3} \varphi_k(t)$

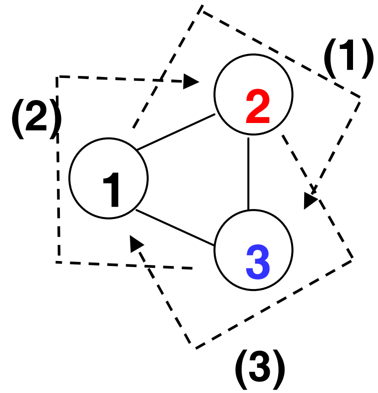
and to reduce 6 equations to 4 ones

BEC transport:



-- well 1
-- well 2
-- well 3

Circular well config.



STIRAP:

- complete at $\Lambda = D = 0$
- still survives at $\Lambda, D < 0.5$

$$\Lambda_{kj} = \frac{U_{kj} N}{2K}$$

STIRAP takes place even under (modest) interaction and so can be applied to realistic BEC!

Geometric phases!

Conclusions and Outlook

Particular two-photon population transfer methods can be applied to:

atomic clusters: ORSR, 1eh modes, s-p electron spectra

Single-particle (mean field) spectra
- **sensitive to cluster structure and thus deliver info on diverse cluster features,**
- **robust test for theory,**

STIRAP??
SCRAP!

BEC: STIRAP transport in multi-well traps

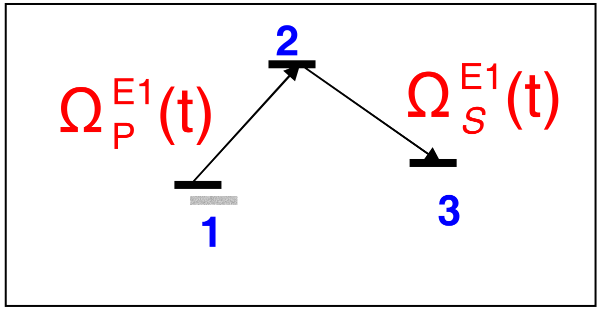
Perspectives:
- **geometric phases,**
- **quantum informatics (STIRAP of atoms),**
- **multi-component BEC, …**

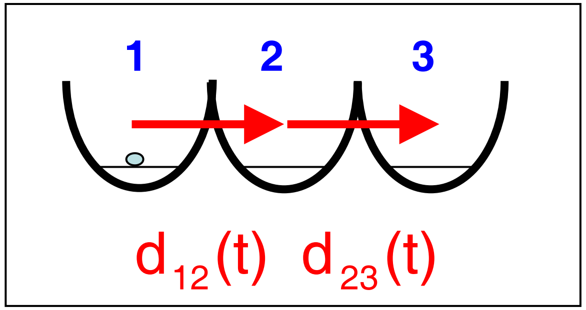
1-photon, 2-photon, multiphoton population transfer schemes
Thanks to similarity between multi-photon and tunneling schemes

Methods of modern quantum optics →

- **Spectroscopy of atomic clusters**
- **Transport of BEC, atoms, …**
- **…**

Equations describe two scenarios:

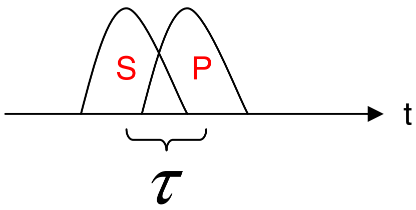




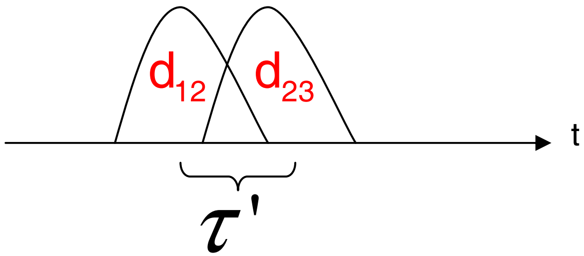
- Three-component BEC in single-well trap,
- coupling by pump and Stokes laser pulses,
- $U_{k\neq j} = U_{k=j} = U$

- One-component BEC in triple-well trap,
- coupling via barriers between traps,
- $U_{k\neq j} = 0, \quad U_{kk} = U$

Adiabatic condition:



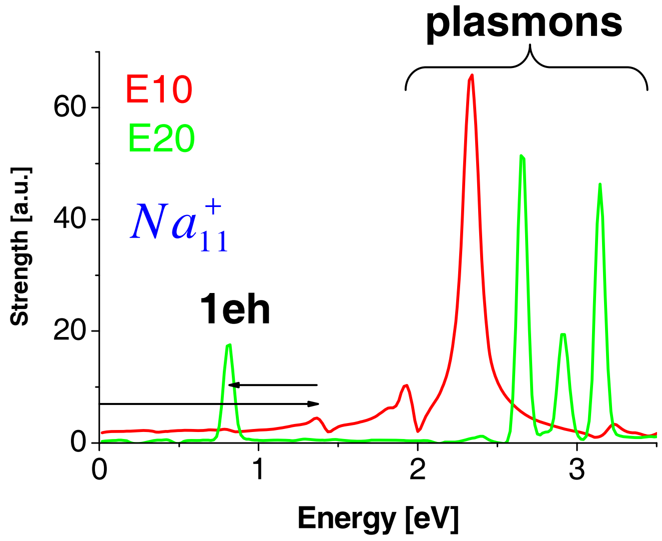
$$\Omega\tau > 10$$



$$2K\tau = \tau', \Omega\tau = K\tau = 0.5\tau' > 10,$$

$\tau' > 20$ - very simple form

Atomic clusters



Spectra of valence electrons:

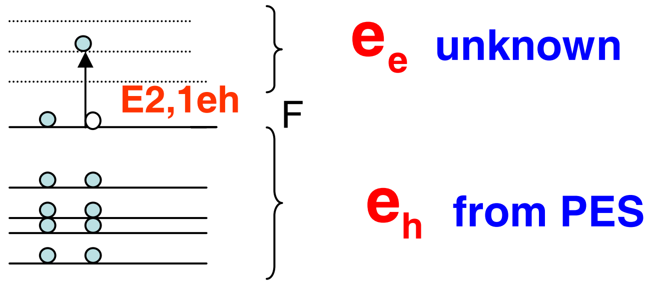
1) collective modes (plasmons)

2) infrared 1eh excitations

$$e_{eh} = e_h - e_e$$

3) Single-particle (mean field) spectra

- sensitive to cluster structure and thus deliver info on diverse cluster features,
- robust test for theory,
- still poorly studied, hot topic!



Infrared 1eh modes provide direct access to s-p spectrum above Fermi level

Problems:

- very short lifetimes (10-1000 fs) ⟶
- strong dynamical Stark shifts ⟵
- competition with plasmons

intense lasers with ultra-short (fs) pulses

Model:

-- Kohn-Sham functional, Perdew-Wang xc

-- Time Dependent Local Density Approximation (TDLDA)

-- propagation of single-electron wave function in time

-- including photoemission through absorption boundary

$$i\hbar \frac{\partial}{\partial t} \phi_i(\vec{r},t) = \hat{h}_{KS}(\vec{r},t)\,\phi_i(\vec{r},t)$$

-- expectation values of multipole moments $D(t) = \int d\vec{r}\, r^L Y_{L0}(\Omega)\rho(\vec{r},t)$

-- Fourier transformation into frequency domain $\tilde{D}(\omega) = \int dt\, e^{i\omega t} D(t)$

-- coherent (classical) laser field

E(t)cos(ωt), E(t) = sin^2(t/T), T = 100 - 500 fs

-- axially deformed cluster Na_{11}^{+}

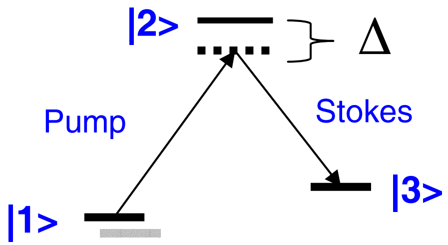
-- quadrupole (LM=20) infrared 1eh state at 0.75 eV

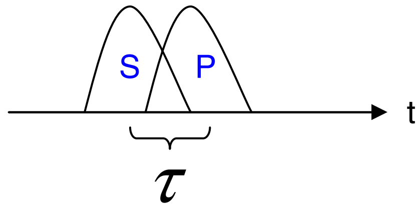
-- jellium approximation for ions

V.O. Nesterenko, W. Kleinig, P.-G. Reinhard, and D.S. Dolci, Phys. Rev. A70, 023205 (2004);
V.O. Nesterenko, P.-G. Reinhard, Th. Halfmann and L.I. Pavlov, Phys. Rev. A73, 02120 (2006);
V.O. Nesterenko, P.-G. Reinhard, Th. Halfmann and E. Suraud, J. Phys. B, 39, 3905 (2006).

STImulated Raman Adiabatic Passage (STIRAP): basic points

K. Bergman, et al,
Rev. Mod. Phys., 70, 1003 (1998)





STIRAP provides up to 100% of the population transfer!

Main requirements:

- **Two-photon resonance:** $\omega_P = \omega_2 - \omega_1 - \Delta \quad \omega_S = \omega_2 - \omega_3 - \Delta$ ⇨ $\omega_P - \omega_S = \omega_3 - \omega_1$
- **Overlapping pulses, counterintuitive order**
- **Adiabatic evolution:** $\Omega\tau > 10$
- **Dark state, no contribution from |2> at all !**

$$\Omega = \sqrt{\Omega_P^2 + \Omega_S^2}$$ - Rabi frequency

System of equations for **dressed** states |a >:

$$| a^+ >= \sin\theta \sin\phi | 1 > + \cos\phi | 2 > + \cos\theta \sin\phi | 3 >$$

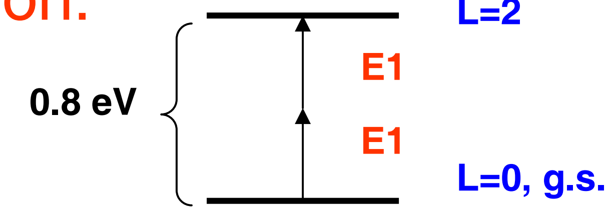
$$| a^0 >= \cos\theta | 1 > + \sin\theta | 3 >$$

$$| a^- >= \sin\theta \cos\phi | 1 > + \sin\phi | 2 > + \cos\theta \cos\phi | 3 >$$

STIRAP dark state. Maximal population at $\tau > 0$.

$$\sin\theta = \frac{\Omega_P}{\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}}, \quad \cos\theta = \frac{\Omega_S}{\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}}, \quad \omega^{\pm} = \Delta \pm \sqrt{\Delta^2 + \Omega_P^2 + \Omega_S^2}, \quad \omega^0 = 0$$

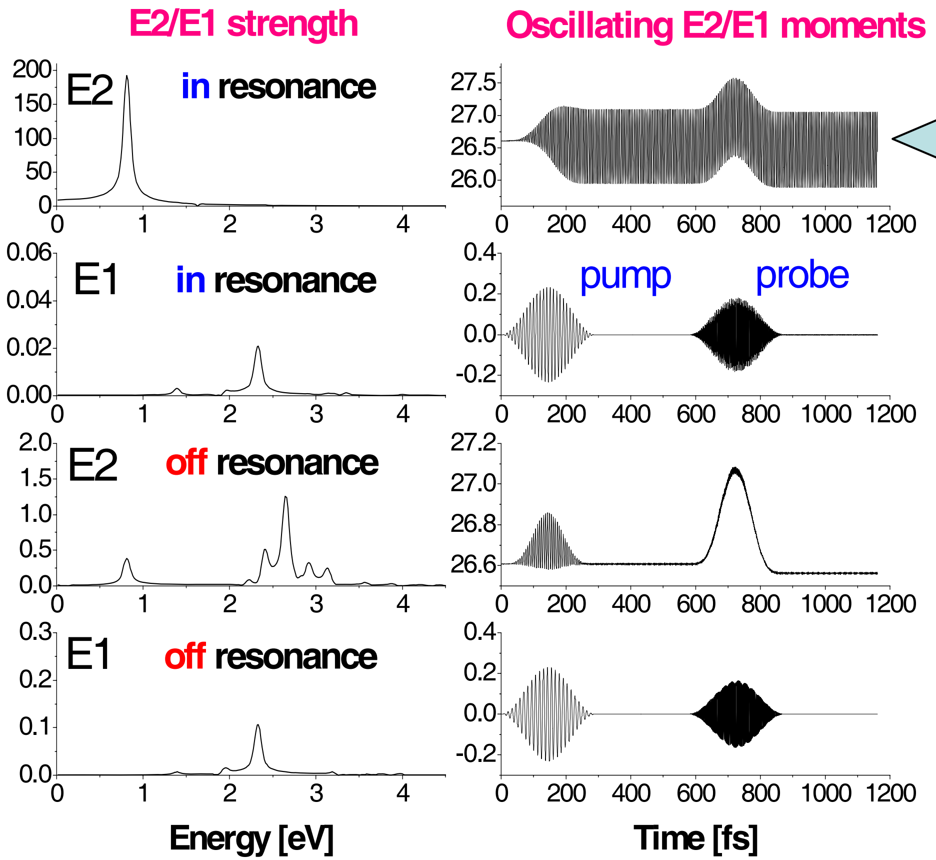
Direct two-photon population:



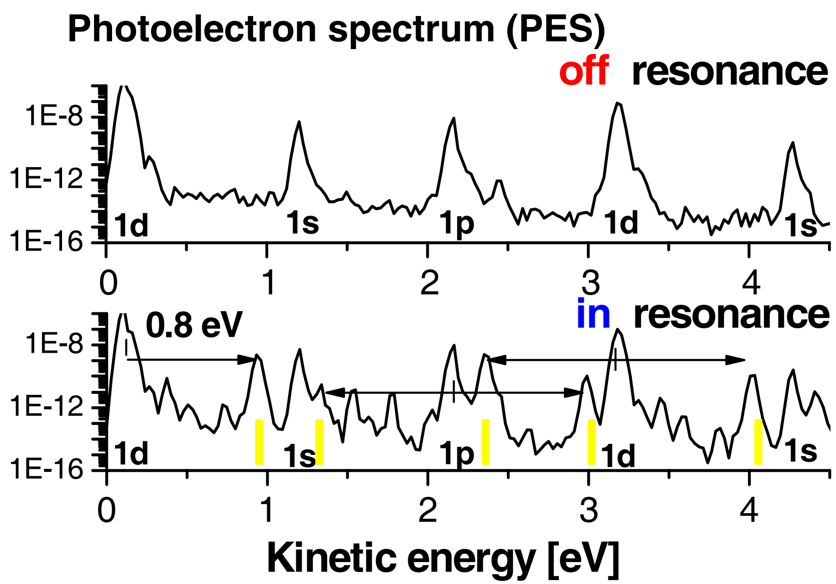
Na_{11}^{+}

jellium TDLDA

Measuring 1eh energy and lifetime



Endurant quadrupole oscillations result in PES satellites:

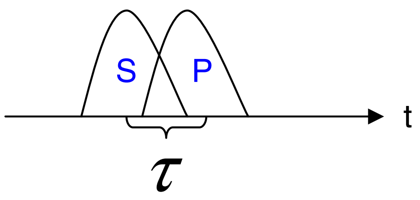


Probe pulse follows the pump pulse with a large delay 600 fs so as to detect only the endurant quadrupole mode

$I = 10^{11}\ \mathrm{W/cm^2},\ T = 300\ \mathrm{fs}$

$\omega_{pump} = 0.40\,\mathrm{eV}$ (in res.), $0.34\,\mathrm{eV}$ (off res.)

$\omega_{probe} = 3.1\,\mathrm{eV}$

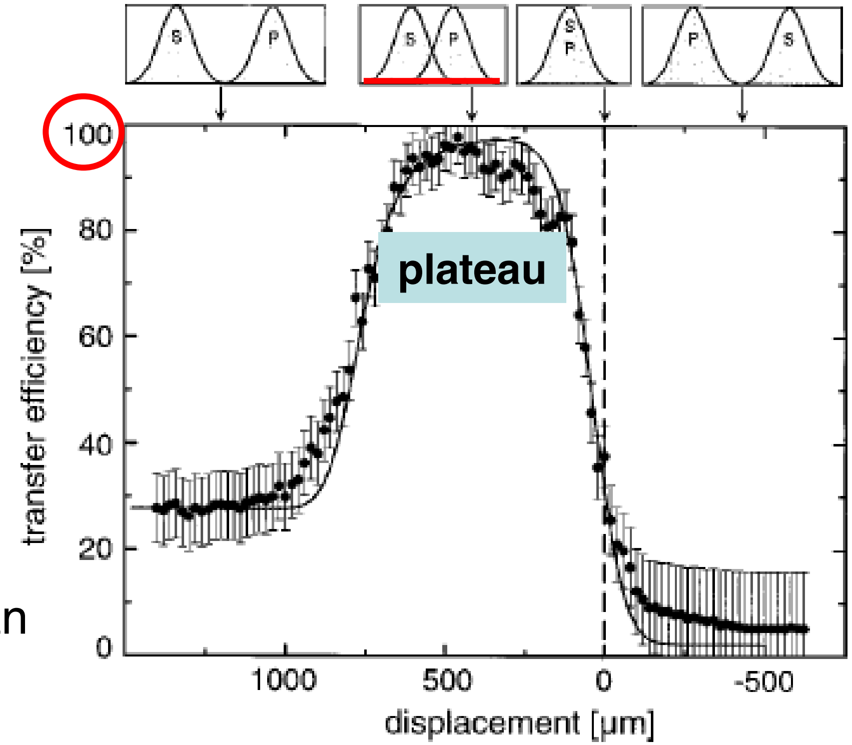


$$\Omega\tau > 10$$

Principle point:
slow **adiabatic** process

Overlapping time
or laser intensity
must be large enough!

Population transfer between $^3P_0$ and $^3P_2$ states in Ne atom



Maximal limit for stimulated Raman

Plateau:
The process is only sensitive slightly sensitive to variation of laser parameters.

- Principle signature of STIRAP: maximal population with plateau at counterintuitive order of pulses

K. Bergman, H. Theuer and B.W. Shore, Rev. Mod. Phys., 70, 1003 (1998)

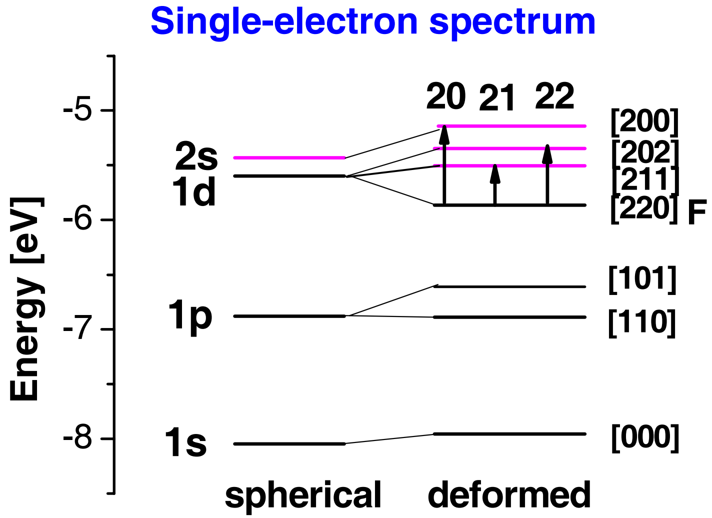
Advantages of light deformed clusters:

-- safe size selection, well known shape, routinely available beams

-- dilute infrared 1eh spectra,

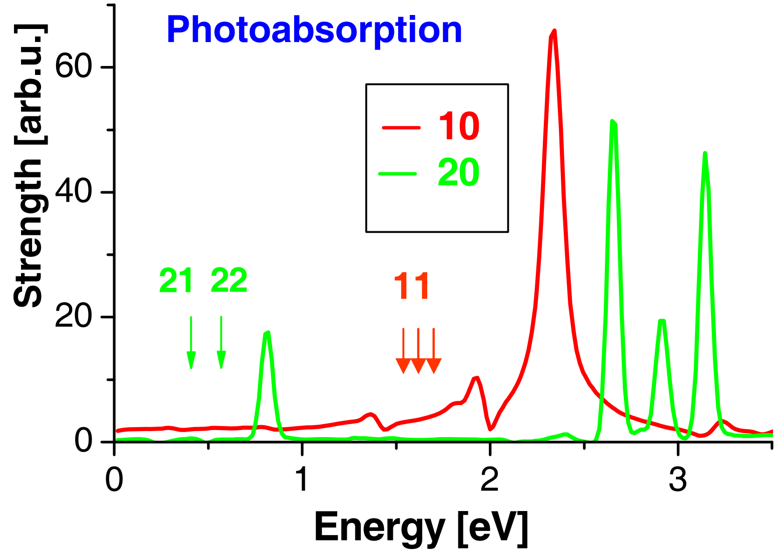
Na^{+}_{11}

prolate, axial



-- 21 and 22 1eh-modes are fully driven by cluster deformation

V.O. Nesterenko, P.-G. Reinhard, W. Kleinig, and D.S. Dolci, PRA, v.70, 023205 (2004).



Every infrared quadrupole mode is strictly dominated by one 1eh configuration:

E20: [220]-[200] 99.9%
E21: [220]-[211] 99.5%
E22: [220]-[202] 99.6%

Access to:

-- cluster mean field

-- deformation effects

τ_{1eh} **= 1-5 ps ⟹ fs intense lasers in TPP!**