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)
- STImulated 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:

Dressed states in STIRAP:

\[
|a^+\rangle = \sin \theta \sin \phi |0\rangle + \cos \phi |1\rangle + \cos \theta \sin \phi |2\rangle
\]

\[
|a^0\rangle = \cos \theta |0\rangle + \sin \theta |2\rangle
\]

\[
|a^-\rangle = \sin \theta \cos \phi |0\rangle + \sin \phi |1\rangle + \cos \theta \cos \phi |2\rangle
\]

\[
sin \theta = \frac{\Omega_p}{\sqrt{\Omega_p^2(t) + \Omega_s^2(t)}}, \quad \cos \theta = \frac{\Omega_s}{\sqrt{\Omega_p^2(t) + \Omega_s^2(t)}}
\]


- 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%

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

Decay to other stays

- \(|0\rangle\) at \(t = 0\)
- \(|2\rangle\) at \(t = \infty\)
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?
Transport of Bose-Einstein condensate: why yes?
Because TPP and BEC tunneling are similar physically and mathematically!

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

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

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

E1/E2 strength

Oscillating E1/E2 moments

coinciding pulses

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

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

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

- 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)

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

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

$H(|a|^2, |b|^2, |c|^2) = \begin{pmatrix} \varepsilon + g|a|^2 & v & 0 \\ v & g|b|^2 & w \\ 0 & w & \delta + g|c|^2 \end{pmatrix}$

$\approx H_{\text{STIRAP}} = \begin{pmatrix} -2\Delta_P + S_1 & \Omega_P & 0 \\ \Omega_P & S_2 & \Omega_S \\ 0 & \Omega_S & -2\Delta_S + S_3 \end{pmatrix}$

Analog of STIRAP!

The parameters $v, w, \varepsilon, \delta$ can be varied by controlling the debts or separations of the wells.
BEC/STIRAP model:

Equations for 3-component BEC:

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

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

\[ \hat{\psi}_k^\dagger(\vec{r},t) = \psi_k(t) \Phi_k(\vec{r}), \quad \psi_k(t) = \sqrt{N N_k(t)} \exp\{-i\varphi_k(t)\} \]

Equations for phases and populations:

\[
\begin{align*}
\frac{\partial}{\partial t} N_k &= -\sum_{j=1}^{3} \overline{\Omega}_{kj}(t) \sqrt{N_j N_k} \sin(\varphi_k - \varphi_j) \\
\frac{\partial}{\partial t} \varphi_k &= E_k + \sum_{j=1}^{3} \Lambda_{kj} N_j - \frac{1}{2} \sum_{j=1}^{3} \overline{\Omega}_{kj}(t) \sqrt{\frac{N_j}{N_k}} \cos(\varphi_k - \varphi_j)
\end{align*}
\]

The only parameter regulating Interaction-coupling ratio

\[ \Lambda_{kj} = \frac{U_{kj} N}{2K} \]

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

\( g_{kj} \) - interaction

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

\( N_k(t) \) - normalized population

\( \varphi_k(t) \) - phase

\( \overline{\Omega}_{kj}(t) = K \overline{\Omega}_{kj}(t) \)

\( \overline{\Omega}_{kj}(t) = \exp\{-\left(\frac{t_{kj} - t}{\Gamma}\right)^2\} \)

\( E_k = E_k / 2K, \quad 2Kt \to t \)

\( U_{kj} \sim g_{kj} \)
Canonical Hamiltonian and canonical equations:

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

$$\begin{align*}
\frac{\partial}{\partial t} N_k &= -\frac{\partial H_{cl}}{\partial \varphi_k} \\
\frac{\partial}{\partial t} \varphi_k &= \frac{\partial H_{cl}}{\partial N_k}
\end{align*}$$

Canonical transformation to new unknowns:

$$\begin{align*}
Z_k &= \sum_{j=1}^{3} T_{kj} N_j \\
\Theta_k &= \sum_{j=1}^{3} R_{kj} \varphi_j
\end{align*}$$

- population imbalances

- phase differences

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,

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:

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

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

**Adiabatic condition:**

$$\Omega \tau > 10$$

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

$$\tau' > 20$$

- 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

-- expectation values of multipole moments

\[ D(t) = \int d\mathbf{r} r^L Y_{L0}(\Omega) \rho(\mathbf{r}, t) \]

-- Fourier transformation into frequency domain

\[ \tilde{D}(\omega) = \int dt e^{i\omega t} D(t) \]

-- coherent (classical) laser field

\[ E(t) \cos(\omega t), \quad E(t) = \sin^2(t/T), \quad T = 100 - 500 \text{ fs} \]

-- axially deformed cluster \( Na_{11}^+ \)
-- quadrupole (LM=20) infrared 1eh state at 0.75 eV
-- jellium approximation for ions

STImulated Raman Adiabatic Passage (STIRAP): basic points


STIRAP provides up to 100% of the population transfer!

Main requirements:

• Two-photon resonance: \( \omega_P = \omega_2 - \omega - \Delta \), \( \omega_S = \omega_3 - \omega - \Delta \) \( \rightarrow \omega_P - \omega_S = \omega_3 - \omega_1 \)

• Overlapping pulses, counterintuitive order

• Adiabatic evolution: \( \Omega \tau > 10 \)

• Dark state, no contribution from \( |2\rangle \) at all!

System of equations for dressed states \( |a\rangle \):

\[
\begin{align*}
|a^+\rangle &= \sin \theta \sin \phi |1\rangle + \cos \phi |2\rangle + \cos \theta \sin \phi |3\rangle \\
|a^0\rangle &= \cos \theta |1\rangle + \sin \theta |3\rangle \\
|a^-\rangle &= \sin \theta \cos \phi |1\rangle + \sin \phi |2\rangle + \cos \theta \cos \phi |3\rangle \\
\end{align*}
\]

\[
\begin{align*}
\sin \theta &= \frac{\Omega_P}{\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}}, \quad \cos \theta = \frac{\Omega_S}{\sqrt{\Omega_P^2(t) + \Omega_S^2(t)}}, \quad \omega^z = \Delta \pm \sqrt{\Delta^2 + \Omega_P^2(t) + \Omega_S^2(t)}, \quad \omega^0 = 0
\end{align*}
\]
Direct two-photon population:

Measuring 1eh energy and lifetime

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

\[ I = 10^{11} \text{ W/cm}^2, \; T = 300 \text{ fs} \]

\[ \omega_{\text{pump}} = 0.40 \text{ eV (in res.), } 0.34 \text{ eV (off res.)} \]

\[ \omega_{\text{probe}} = 3.1 \text{ eV} \]
 Principle signature of STIRAP: maximal population with plateau at counterintuitive order of pulses


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

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

Advantages of light deformed clusters:
-- safe size selection, well known shape, routinely available beams
-- dilute infrared 1eh spectra,

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%

$\tau_{1eh} = 1-5$ ps $\rightarrow$ fs intense lasers in TPP!