COHERENT POPULATION TRANSFER BETWEEN ATOMIC AND MOLECULAR BECs

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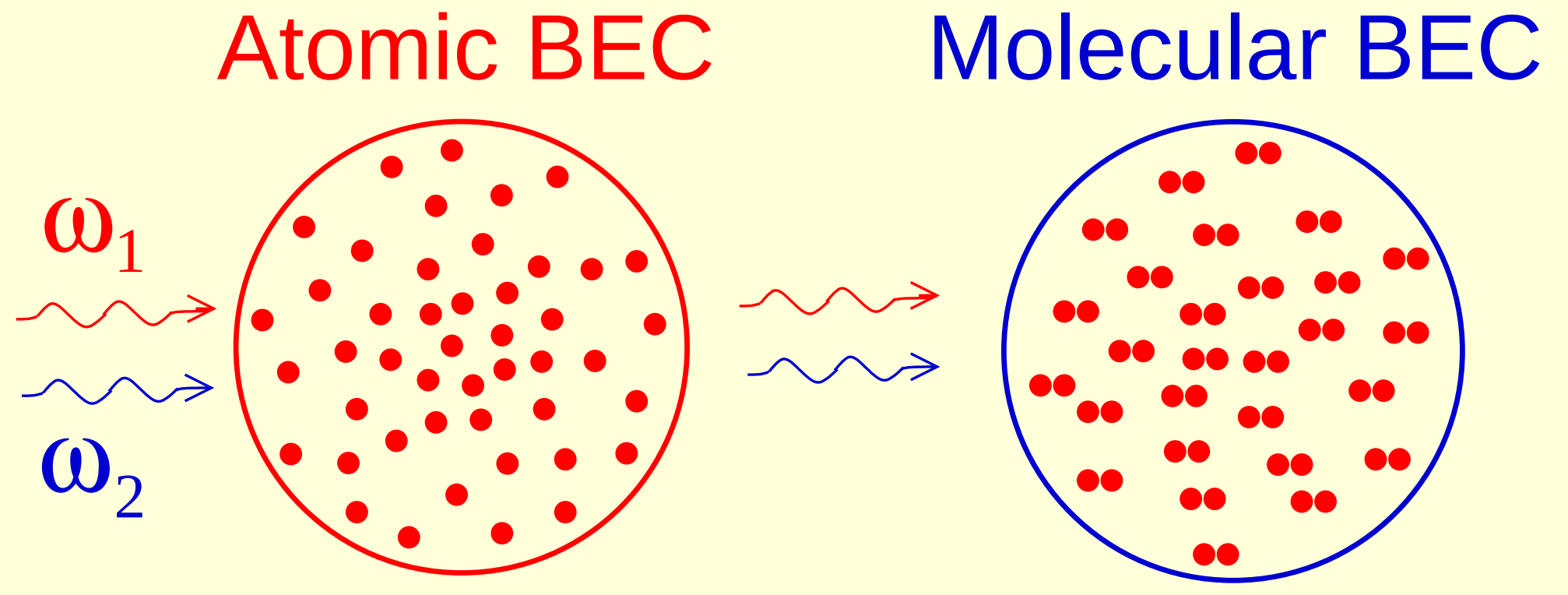
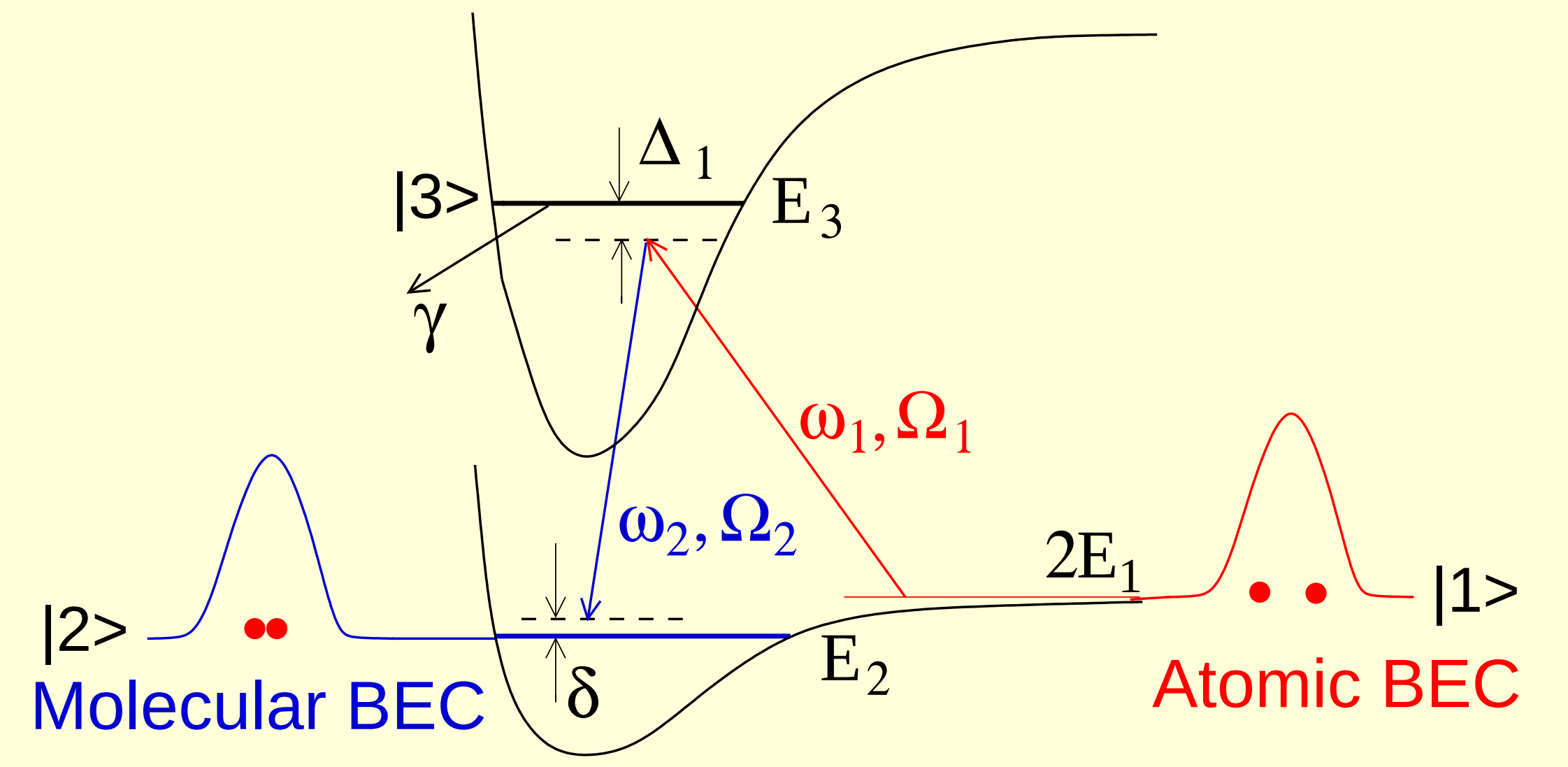
SUPERCHEMISTRY

- Superchemistry is a coherent stimulation of a chemical reaction involving BECs.
- Can a molecular BEC form coherently from an atomic BEC?
- What are the best strategies?
- We compare cw Raman photoassociation (PA) with Stimulated Raman Adiabatic Passage (STIRAP) using delayed pulses.

Raman PA in a BEC

We study photo-assisted conversion of pairs of atoms in a ^{87}Rb BEC to molecules, via stimulated Raman transitions.



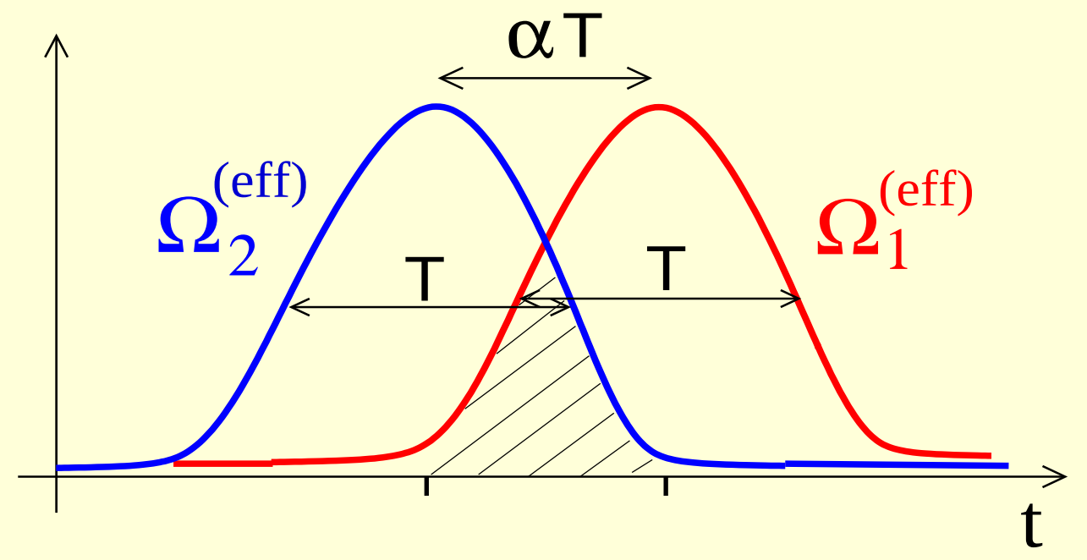


In off-resonant cw photoassociation, molecule production is limited by relatively small coupling and spontaneous emission losses **[D.Heinzen, R.Wynar, P.Drummond, K.Kheruntsyan, PRL 84, 5029 (2000)], [R.Wynar et al., Science 287, 1016 (2000)]**.

PA via STIRAP offers advantages **[J.Javanainen and M. Mackie, PRA 59, R3186 (1999)]**.

HOW STIRAP WORKS

– uses on-resonant counter-intuitive pulse sequence



The system is being prepared and follows the adiabatic eigenstate

$$|\psi(t)\rangle \propto \Omega_2^{(eff)}(t)\,|1\rangle - \Omega_1^{(eff)}(t)\,|2\rangle\,,$$

which is a "dark" superposition-state:

- does not involve "lossy" excited state $|3\rangle$!
- can provide coherent population transfer from $|1\rangle \to |2\rangle$, with up to 100% efficiency.

Adiabaticity criterion (global, simplified):

$$\boxed{\Omega_i^{(eff)}T \gg 1.}$$

Effective Rabi frequencies ($\Omega_i^{(el)} = |\bar{\mathbf{d}}^{(el)}\cdot\mathbf{E}_i|/\hbar$):

$$\Omega_1^{(eff,0)} = \Omega_1^{(el)}\sqrt{n_1}\ |\langle 1|3\rangle|^2\,,$$
$$\Omega_2^{(eff,0)} = \Omega_2^{(el)}\,|\langle 2|3\rangle|^2\,.$$

Difficulty

- **(1)** Typical values of the overlap integral $|\langle 1|3\rangle|$ are too small.

$\Rightarrow$ $\Omega_1^{(eff,0)}$ is limited from above; typically

$$\Omega_1^{(eff,0)} \lesssim 2\times 10^6 s^{-1};$$

- **(2)** There is a dephasing due to the mean-field energy shifts, occurring on time-scale $t_{ph} = [U_{11}n_1]^{-1} \simeq 5\times 10^{-5}$ s;

$\Rightarrow$ T has to be shorter than

$$T \lesssim 5\times 10^{-5}\ \text{s}.$$

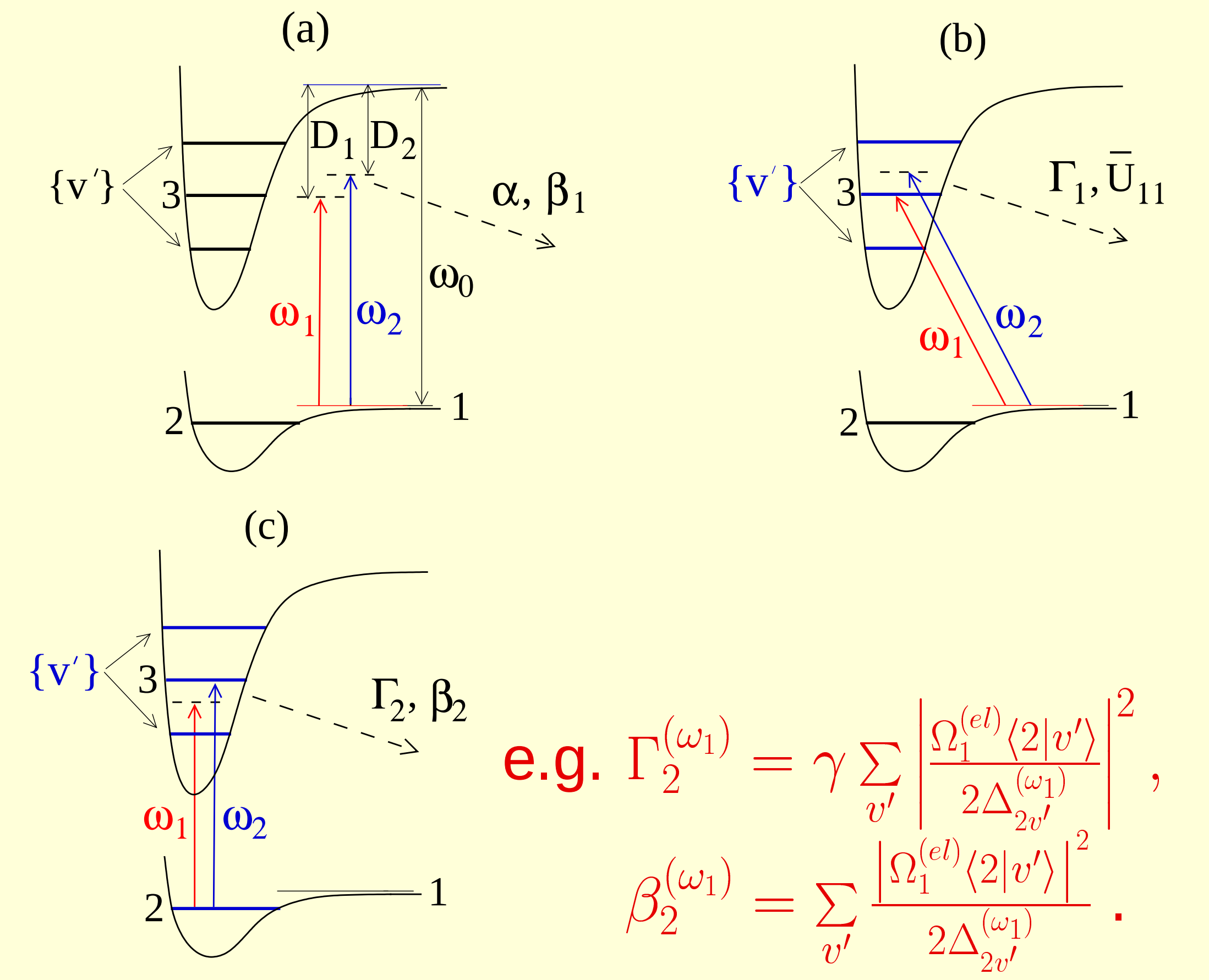
- **Combining these gives poor adiabaticity ($\Omega_1^{(eff)}T$ is not large enough), hence poor conversion efficiency ($\sim 10\%$).**

OPTIMIZATION

- Use asymmetric pulses: $\Omega_2^{(eff)} \gg \Omega_1^{(eff)}$;
- Compensate the dephasing due to mean-field energies by tuning δ;
- Optimize α and T (gives $T \simeq 2\times 10^{-4}$ s,);
- Can improve the conversion up to $\sim 50\%$.

Multi-level model

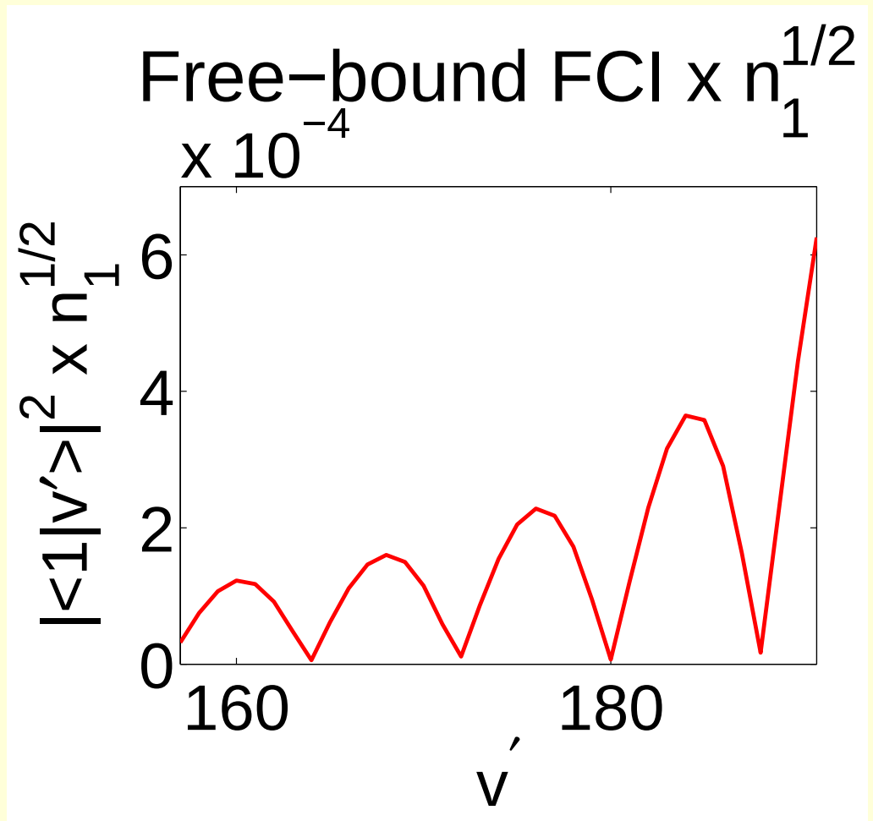
The following processes are NOT negligible:

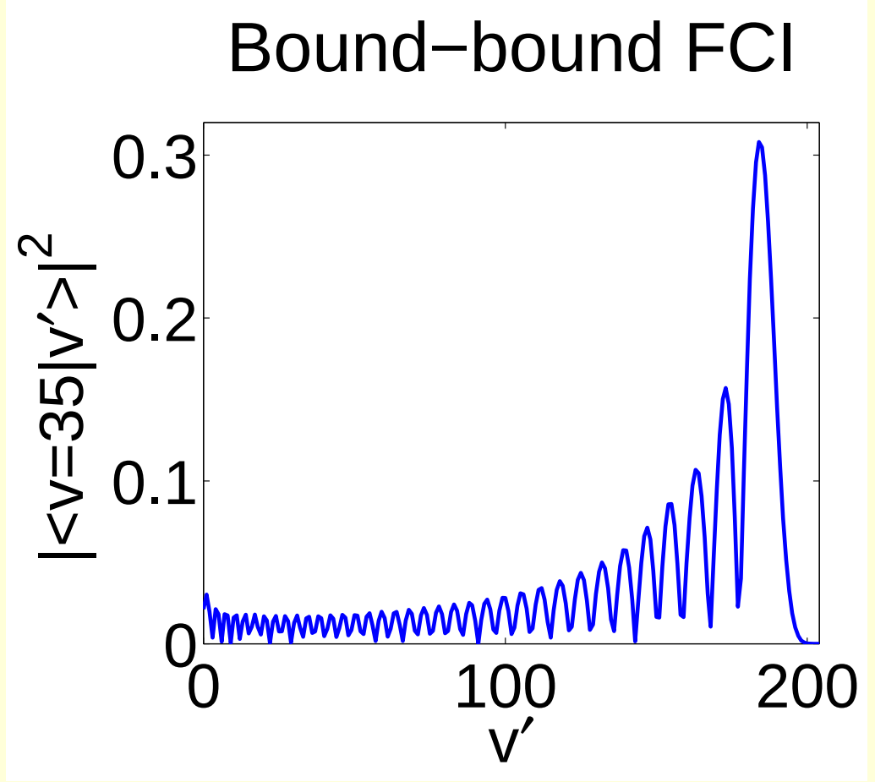


e.g. $\Gamma_2^{(\omega_1)} = \gamma\sum_{v'}\left|\frac{\Omega_1^{(el)}\langle 2|v'\rangle}{2\Delta_{2v'}^{(\omega_1)}}\right|^2$,

$$\beta_2^{(\omega_1)} = \sum_{v'}\frac{\left|\Omega_1^{(el)}\langle 2|v'\rangle\right|^2}{2\Delta_{2v'}^{(\omega_1)}}\,.$$

- The most disruptive loss coefficient is $\Gamma_2^{(\omega_1)}$; can easily have $[\Gamma_2^{(\omega_1)}]^{-1} < T \sim 2\times 10^{-4}$ s, and loose all the molecules, during T;
- Phase mismatch $\beta_2 - 2\beta_1$ due to Stark shifts is equally disruptive; with $[\beta_2 - 2\beta_1]^{-1} < T$, can easily lose the coherence;
- Need a detailed knowledge of the Franck-Condon overlap integrals $\langle 1|v'\rangle$ and $\langle 2|v'\rangle$.





SOLUTION

- Find and tune the transitions to appropriate levels that simultaneously (!) satisfy

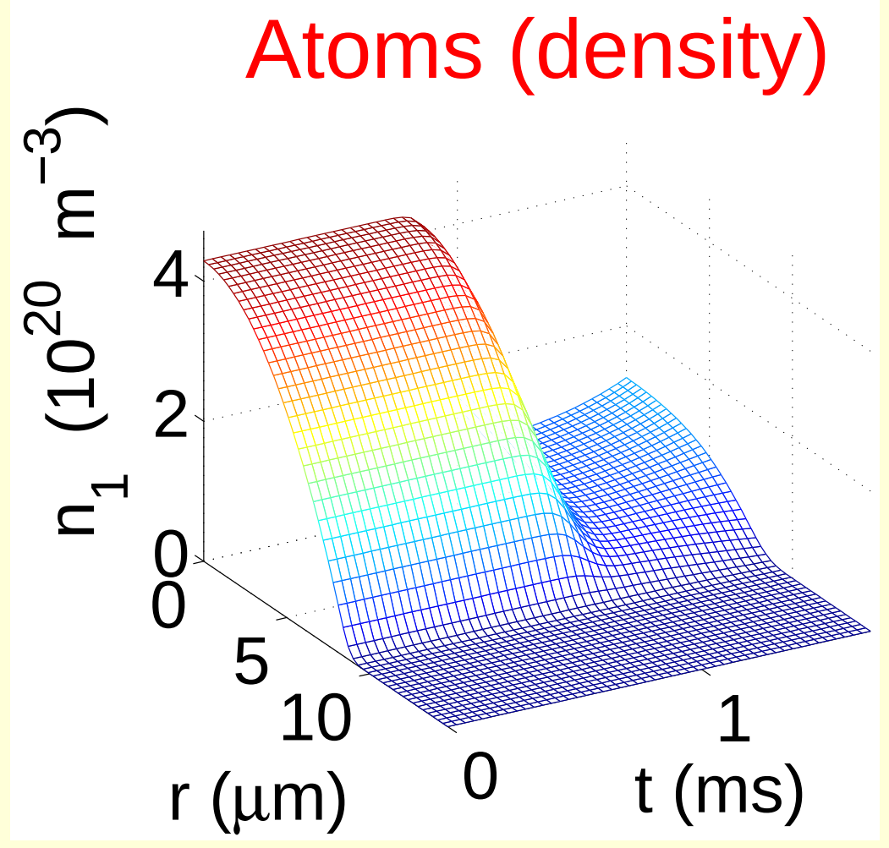
$$[\Gamma_2^{(\omega_1)}]^{-1} \gg T \quad \text{(need small } |\,\langle 2|3\rangle\,|\text{)},$$
$$[\beta_2 - 2\beta_1]^{-1} \gg T,$$

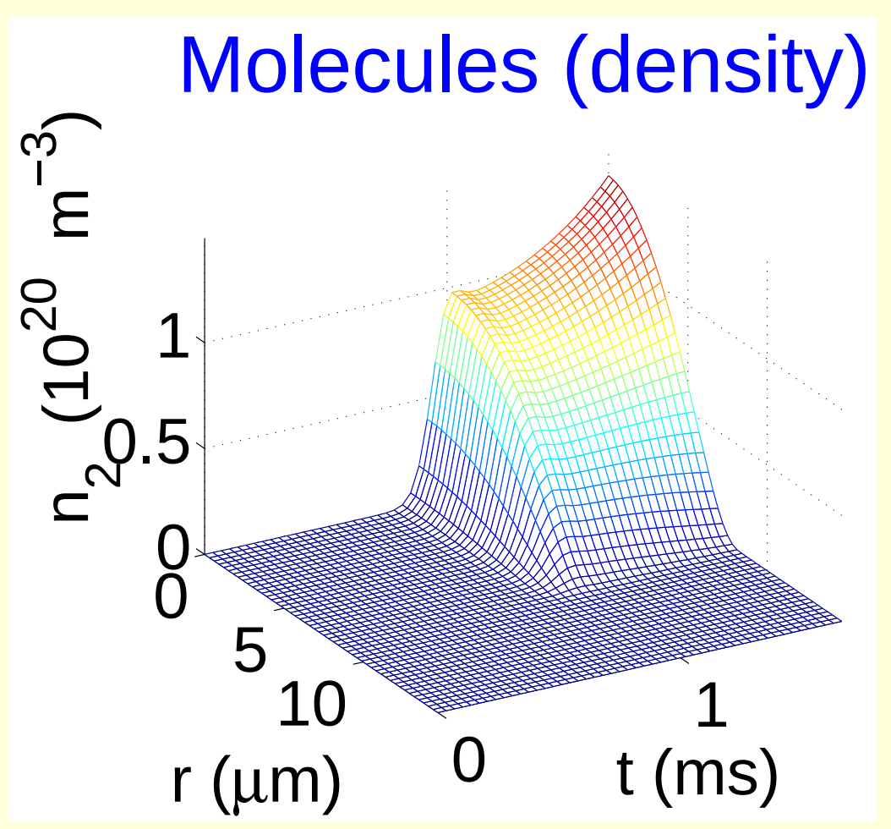
with $|\,\langle 1|3\rangle\,|$ staying large; a rare event!

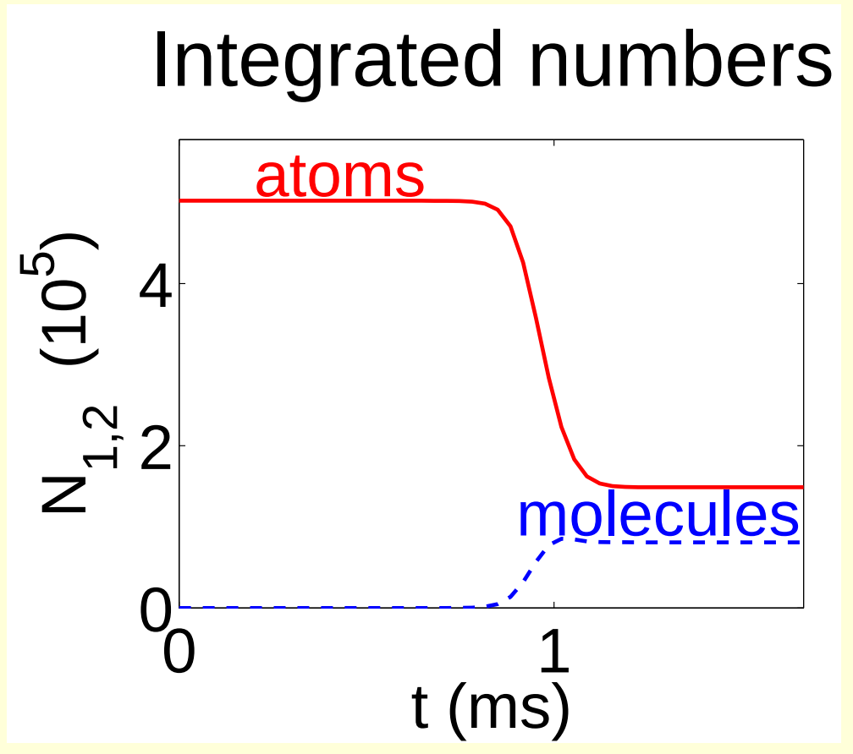
- Example: $|2\rangle \equiv |v = 35\rangle$, $|3\rangle \equiv |v' = 177\rangle$.

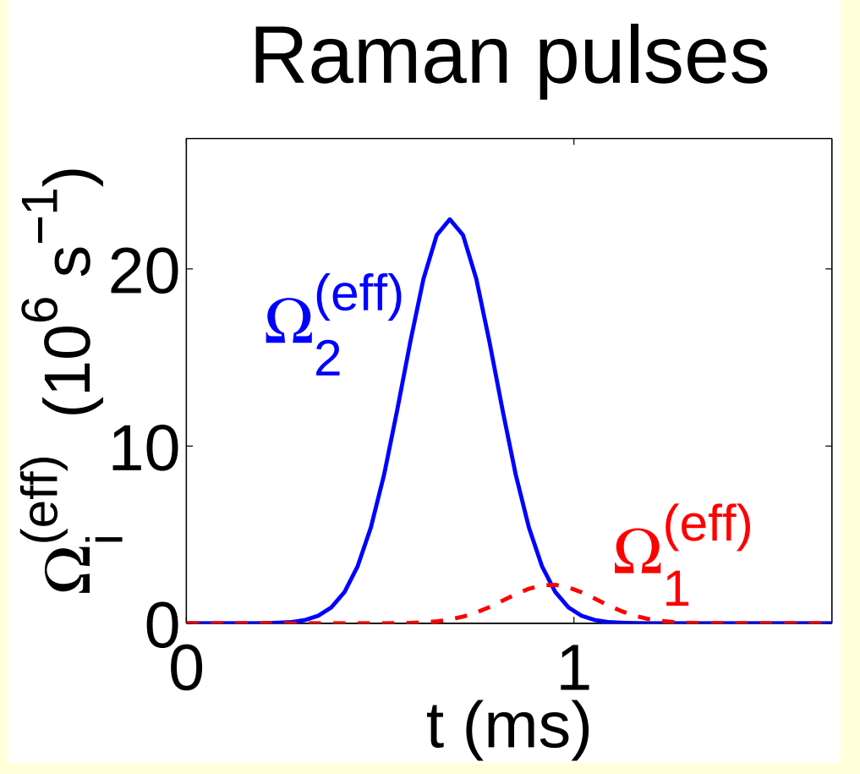
3D simulation results

We perform full multimode 3D simulations of the GP equations, including the trap potentials, kinetic energy terms, elastic collisions, induced radiative losses and the Stark shifts.









We get $\sim 32\%$ conversion efficiency; not bad!

CONCLUSIONS

- **STIRAP has advantages over cw PA: can reduce spontaneous emission losses.**
- **Limitations come from realistic laser powers, dephasing due to mean-field energies, induced radiative losses and Stark shifts.**

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