Controlling Ultra-Cold Molecules with Collective Dissipation

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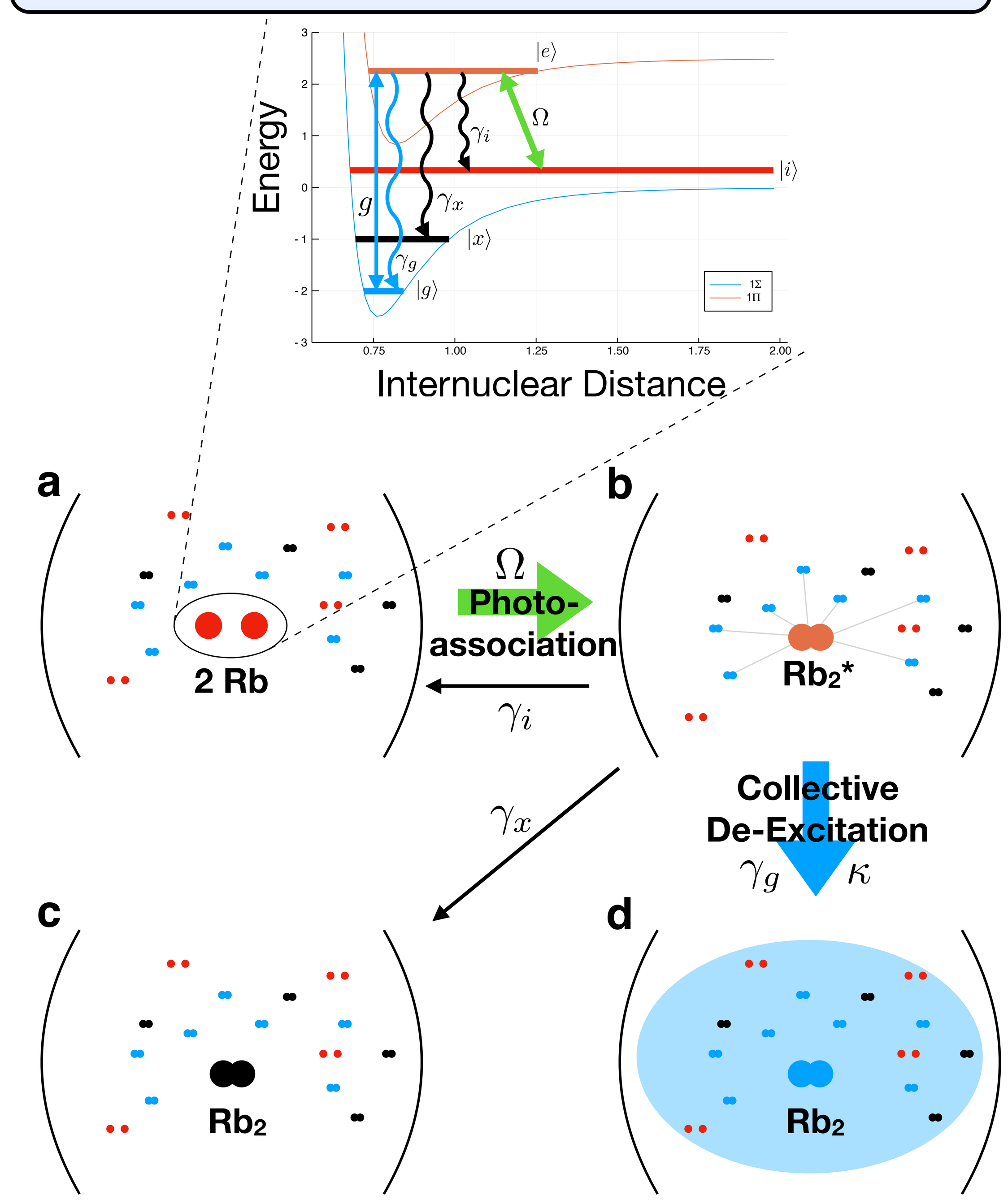
Abstract

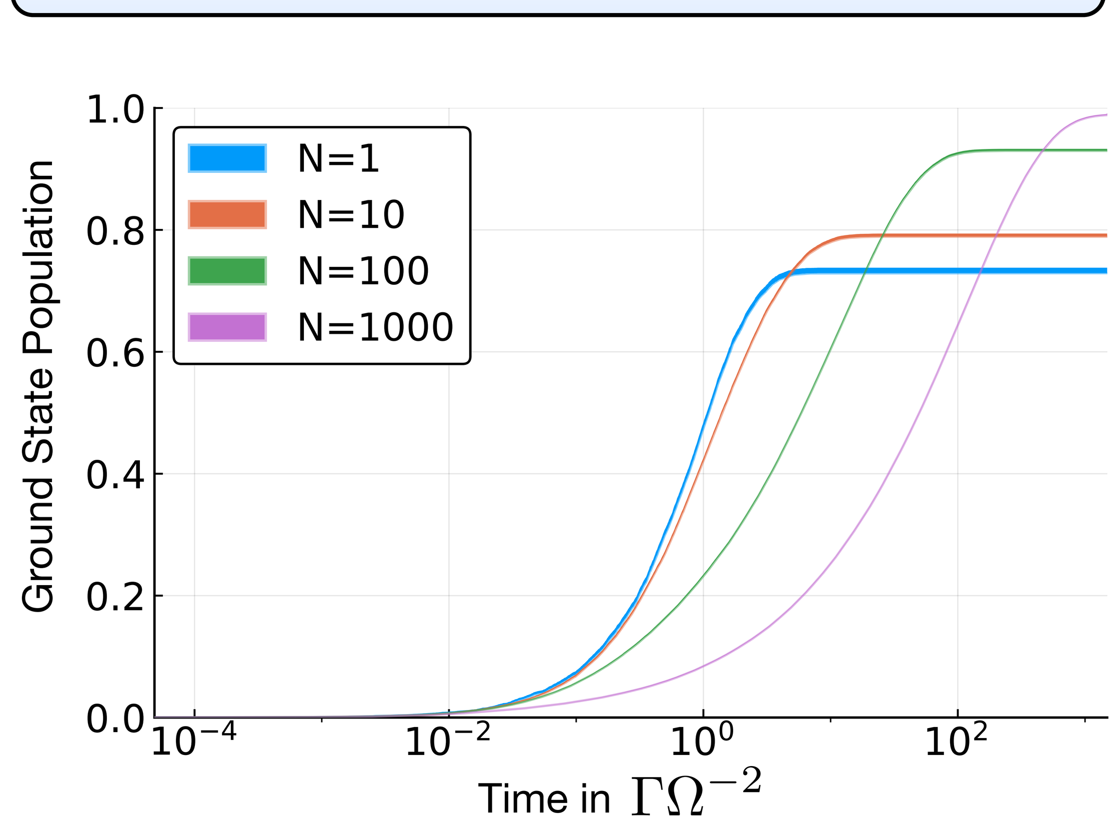
We show that the formation of a molecule from free atoms can be collectively assisted by coupling to a cavity. We consider a production process of photo-association followed by spontaneous emission, and demonstrate how collective dissipative mechanisms can drastically modify production rates and final molecular state fractions. Using both analytical and numerical tools, we analyze situations where super-radiant decay leads to a Zeno-modified formation rate for a cavity-coupled molecular state and propose scenarios where this can speed up the formation process. We show that final molecular fractions can be collectively enhanced almost to unity, even with very lossy cavities, solely by increasing the initial atom number.

Model



Two Rubidium atoms (see **a**) are pumped into an excited molecular state (see **b**). From there, it can decay — collectively or by spontaneous emission — into a vibrationally excited molecular state state (see **c**), into the molecular ground state (see **d**) or back into two separated atoms (see **a**).

Time Evolution



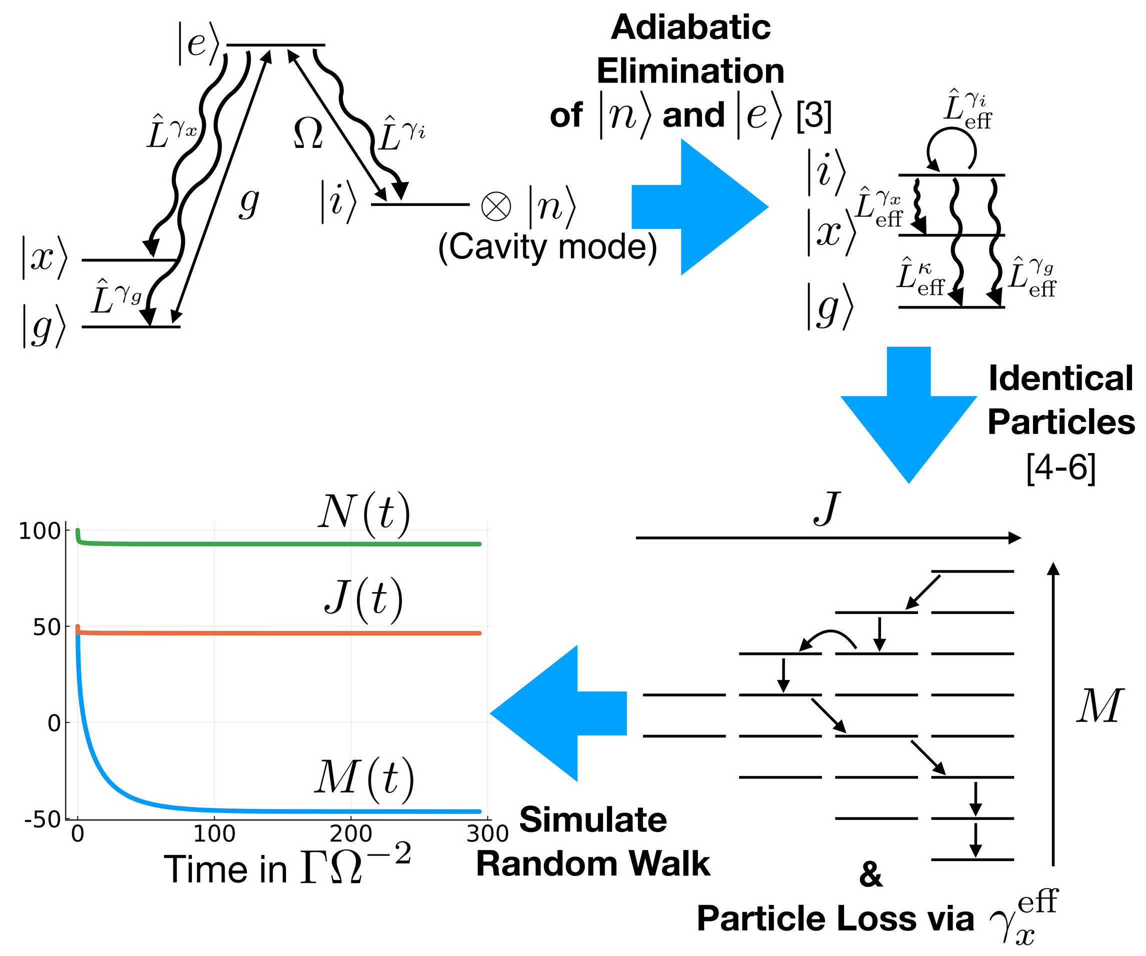
with $C = 0.5$ $\frac{\gamma_g}{\Gamma} = 0.37$ $\frac{\gamma_i}{\Gamma} = 0.32$ $\frac{\gamma_x}{\Gamma} = 0.31$

Effects of increasing the number of particles:

- Transfer is slowed down.
- Final ground state population increases.

Explanation:
The spontaneous emission of the excited state into the ground state is increased by superradiance, which slows down the transfer due to Zeno blocking, but at the same time increases the fraction brought into the ground state.

Master Equation and Simulation



Operators before adiabatic elimination

$$\hat H = \Omega\sum_n\left(\hat\sigma_{ie}^{(n)}+\hat\sigma_{ie}^{(n)\dagger}\right)+\Delta\sum_n\hat\sigma_{ee}^{(n)} + g\sum_n\left(\hat a^\dagger\hat\sigma_{ge}^{(n)}+\hat a\hat\sigma_{ge}^{(n)\dagger}\right)+\delta\hat a^\dagger\hat a$$

$$\hat L^{\kappa}=\sqrt{\kappa}\hat a \qquad \hat L^{\gamma_g^{(n)}}=\sqrt{\gamma_g}\hat\sigma_{ge}^{(n)}$$

$$\hat L^{\gamma_i^{(n)}}=\sqrt{\gamma_i}\hat\sigma_{ie}^{(n)} \qquad \hat L^{\gamma_x^{(n)}}=\sqrt{\gamma_x}\hat\sigma_{xe}^{(n)}$$

Effective operators for no detuning

$$\hat H_{\text{eff}}=0$$

$$\hat L^{\kappa}_{\text{eff}}=\frac{\Omega}{\sqrt{\Gamma}}\hat S^-_{gi}\frac{\sqrt{C}}{1+(\hat n_g+1)C}$$

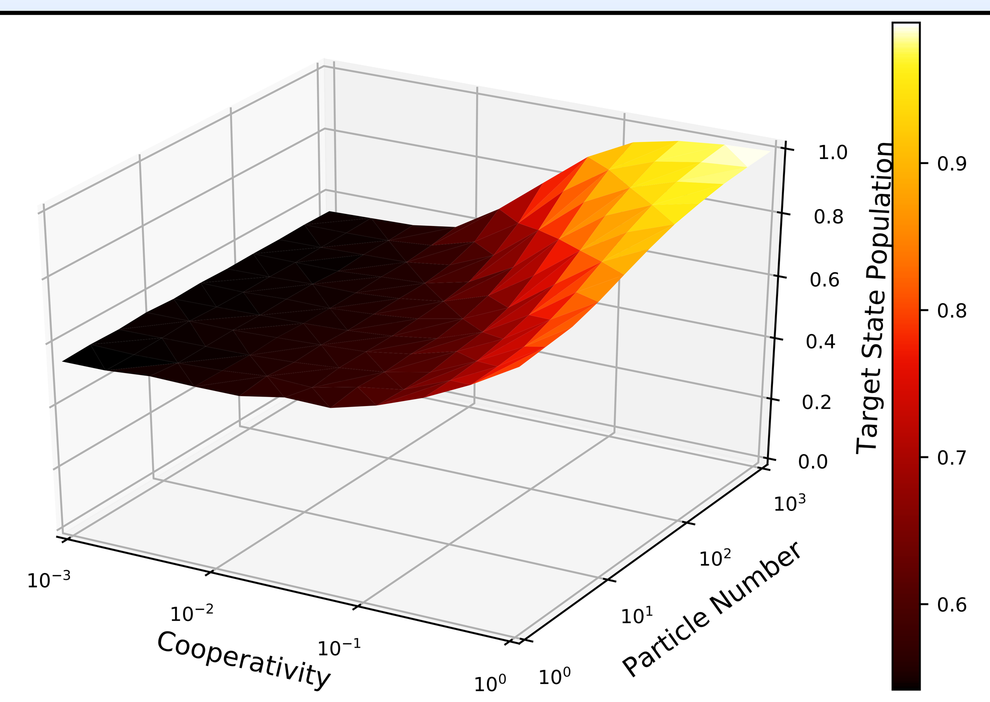
$$\hat L^{\gamma_g^{(n)}}_{\text{eff}}=\frac{\Omega\sqrt{\gamma_g}}{\Gamma}\left(\hat\sigma_{gi}^{(n)}-\hat\sigma_{gg}^{(n)}\hat S^-_{gi}\frac{C}{1+(\hat n_g+1)C}\right)$$

$$\hat L^{\gamma_i^{(n)}}_{\text{eff}}=\frac{\Omega\sqrt{\gamma_i}}{\Gamma}\left(\hat\sigma_{ii}^{(n)}-\hat\sigma_{ig}^{(n)}\hat S^-_{gi}\frac{C}{1+(\hat n_g+1)C}\right)$$

$$\hat L^{\gamma_x^{(n)}}_{\text{eff}}=\frac{\Omega\sqrt{\gamma_x}}{\Gamma}\left(\hat\sigma_{xi}^{(n)}-\hat\sigma_{xg}^{(n)}\hat S^-_{gi}\frac{C}{1+(\hat n_g+1)C}\right)$$

with $\Gamma=\gamma_g+\gamma_i+\gamma_x$ $\quad \hat\sigma_{ij}=|i\rangle\langle j|$ $\quad \hat S^-_{gi}=\sum_n\hat\sigma_{gi}^{(n)}$

Target Population Enhancement



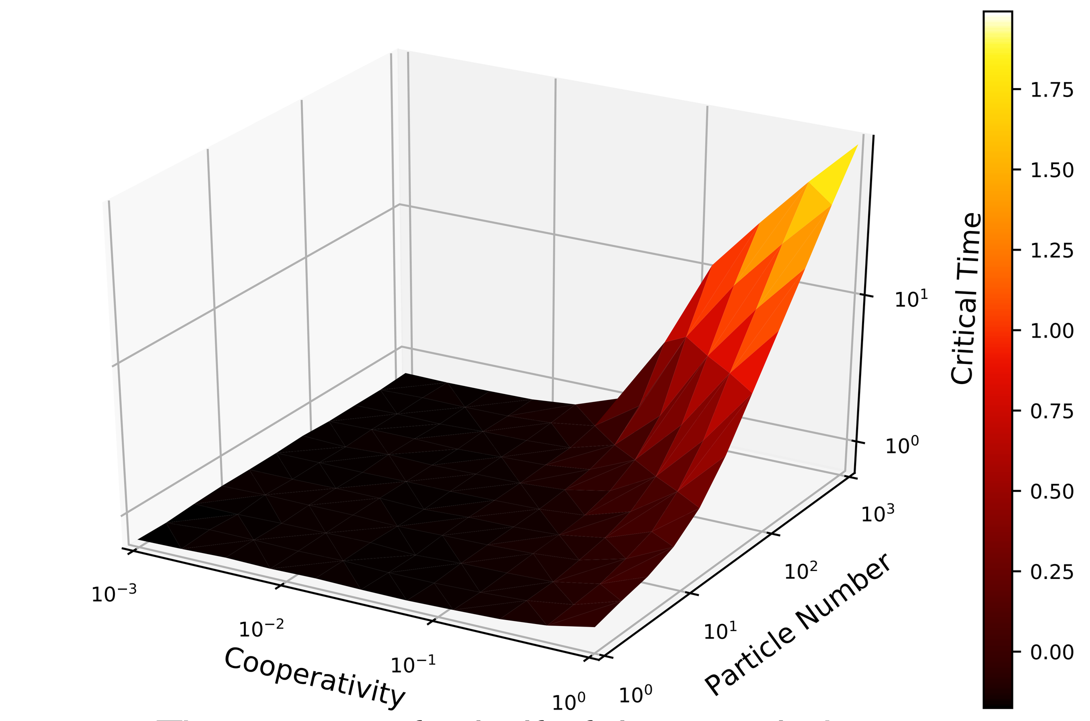
with $\frac{\gamma_g}{\Gamma}=0.37$ $\frac{\gamma_i}{\Gamma}=0.32$ $\frac{\gamma_x}{\Gamma}=0.31$

- Ground state population increases with particle number and cooperativity
- For small NC plateau at:
$$n_g^\infty=\frac{\gamma_g}{\gamma_x+\gamma_g}$$
- For large NC ground state population is given by
$$n_g^\infty\approx 1-\frac{\pi}{2}\frac{\gamma_x}{\Gamma}\frac{\ln(N)}{NC}$$

Explanation:
Decay into ground state is by a factor $\frac{\gamma_x}{\Gamma(n_g+1)C}$ faster than the decay into the $|x\rangle$ state. The additional factor comes from integrating until all the population is in the ground state $\ln N$.

Zeno Blocking (for $\gamma_x=0$)

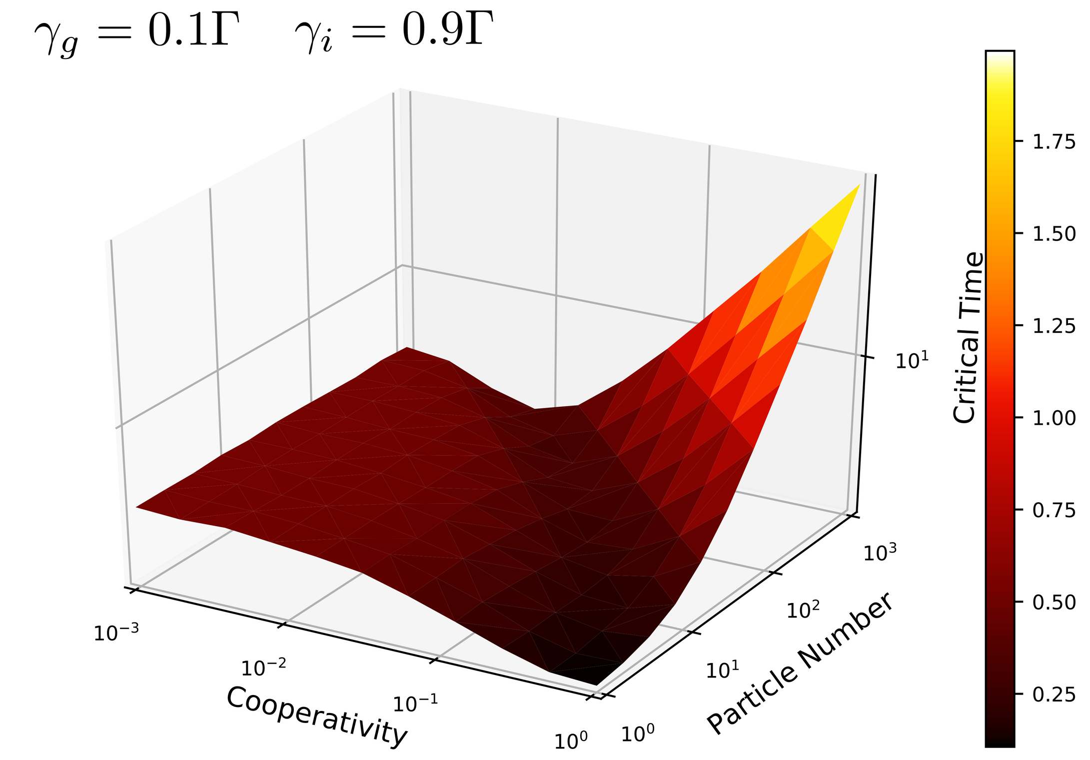
$\gamma_g=\gamma_i=0.5\Gamma$



Time to transfer half of the population

- For small NC the transfer time has a plateau.
- For large NC the transfer time is proportional to particle number and cooperativity.

$\gamma_g=0.1\Gamma$ $\quad\gamma_i=0.9\Gamma$



Time to transfer half of the population

- For small NC the transfer time has a plateau.
- For intermediate NC around 1 to 10 there is a speed-up.
- For large NC the transfer time is proportional to particle number and cooperativity.

Explanation:
Without the cavity the transfer rate is $\frac{\Omega^2\gamma_g}{(\gamma_g+\gamma_i)^2}$, which is maximal for $\gamma_g=\gamma_i$. Interpreting the superradiant emission into the ground state as a modification of γ_g^{eff} explains the speed up until $\gamma_g^{\text{eff}}\sim\gamma_i \Leftrightarrow NC\sim 1$ and a slow-down by Zeno blocking for large γ_g^{eff}.

Conclusion and Outlook

Conclusion

- In photo-association of molecules a cavity can collectively increase the spontaneous emission into a target state.
- Intermediate collective coupling: speed-up for low spontaneous emission into the target state.
- Strong collective coupling: Zeno blocking.
- The final population in the target state can be significantly enhanced.

Outlook

- Beyond adiabatic elimination: Target enhancement without slow-down?
- Applicable to room temperature chemistry experiments [2]?

References

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