COHERENT ATOMIC-MOLECULAR SIMULTONS IN BEC

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PARAMETRIC FIELD THEORY

- Typical of parametric couplings, which convert one particle into two.
- In nonlinear optics, describes second harmonic generation.
- In nonlinear atom optics (BEC), describes coherent dimerization.

New physical effects resulting from this:

- **SIMULTONS** - coherent propagating atom-molecular solitons;
- **SUPERCHEMISTRY** - stimulated nonlinear coherent chemistry.
General parametric Hamiltonian

\[ H = H_0 + H_{int} + H_{self}, \]

\[ H_0 = \sum_i \int d^3 x \left[ \frac{\hbar^2}{2m_i} |\nabla \Psi_i(x)|^2 + V_i(x) \Psi_i^\dagger(x) \Psi_i(x) \right], \]

\[ H_{int} = \frac{\chi}{2} \int d^3 x \Psi_1^\dagger(x) \Psi_1^\dagger(x) \Psi_2(x) + H.c., \]

\[ H_{self} = \sum_{i,j} \frac{U_{ij}}{2} \int d^3 x \Psi_i^\dagger(x) \Psi_j^\dagger(x) \Psi_j(x) \Psi_i(x). \]
Definitions

- $\hat{\Psi}_1$ is the atomic field; $\hat{\Psi}_2$ is the molecular field.

- $m_1$ and $m_2 = 2m_1$ are the corresponding masses.

- $\chi$ is the molecular formation rate.

- $U_{11} = 4\pi\hbar^2 a / m_1$; $a$ is the atomic $S$-wave scattering length.

- $V_i(x)$ - trap potentials $[ V_i(x) = (m_i\omega_i^2 / 2)x^2 + V_i^{(0)} ]$.

- $\gamma_i(x)$ - linear loss rate; $\gamma_{ij}(x)$ - nonlinear loss rate.
APPROXIMATIONS

- The Hamiltonian is just an effective Hamiltonian.
- Replacement of the actual interatomic potential by a $\delta$-function pseudopotential is known to be only valid in combination with an ultraviolet momentum cutoff.
- If we could include the complete inter-atomic potential, we would have no need to treat the molecular field separately.
- The theory only treats a single molecular level.
- Atom-molecule and molecule-molecule scattering processes are neglected in the calculations, except for certain loss processes.
Mean field theory

Mean field theory ($\psi_i = \langle \hat{\Psi}_i \rangle$) or modified G-P equations are valid for large particle number:

\[
i\hbar \dot{\psi}_1 = -\frac{\hbar^2}{2m_1} \nabla^2 \psi_1 + V_1^c(x)\psi_1 + U_{11}^c|\psi_1|^2\psi_1 + \chi \psi_1^* \psi_2,
\]
\[
i\hbar \dot{\psi}_2 = -\frac{\hbar^2}{2m_2} \nabla^2 \psi_2 + V_2^c(x)\psi_2 + \frac{1}{2}\chi \psi_1^2.
\]

where $V_j^c = V_j - i\hbar \gamma_j$, $U^c$ are complex, to include loss processes.

- In the absence of the trap potentials and for $U_{11}^c = 0$, these coincide with coupled wave equations for frequency doubling in nonlinear optics ($\chi \rightarrow \chi^{(2)}$ -nonlinearity), which have stable classical 3D solitons.
QUANTUM SIMULTONS WITHOUT CUTOFF

We look for the energy eigenstates (or quantum simultons) ignoring losses. These must simultaneously be the eigenstates of

$$\hat{N} = \int d^3x \left[ |\hat{\Psi}_1|^2 + 2|\hat{\Psi}_2|^2 \right] = \hat{N}_1 + 2\hat{N}_2,$$

conserving the generalized particle number $N$ (total number of atoms if we count each molecule as two atoms).

Simplified version:

- in free space (no trap potentials);
- no momentum cutoff.
Two-particle quantum simultons

Simplest quantum soliton (simulton) is a di-boson bound state \((N = 2)\), which has the form of a superposition state:

\[
|\psi^{(2)}\rangle = \left[ \int d^3x \hat{\Psi}_2^\dagger(x) + \int \int d^3x d^3y \: g(x - y) \: \hat{\Psi}_1^\dagger(x) \hat{\Psi}_1^\dagger(y) \right] |0\rangle
\]

- a ‘dressed’ molecule which exists in a superposition with a pair of atoms.

Solutions of similar form were first discovered by T.D.Lee in meson physics (except using two fermion fields, and no S-wave repulsion)

• S. Schweber, Relativistic Quantum Field Theory.

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Properties of exact solutions

- Energy is bounded from below if $U_{11} > 0$, - unbounded if $U_{11} = 0$.

- Correlation function $g(r)$ has a point-like structure:

  $$ g(r) = 0 \text{ , if } |r| > 0 \text{ , } g(0) = -\chi/(2U_{11}) . $$

- **Exact** binding energy per atom: $E_g^{(2)} = \Delta V - \chi^2/(4U_{11})$.

- $\Delta V = V_2^{(0)}/2 - V_1^{(0)}$.

$N$-particle exact solution

Even $N$-particle eigenstates have the form of $N/2$ independent di-bosons:

$$|\psi^{(N)}\rangle = \left[ \int d^3 x \hat{\Psi}^\dagger_2(x) + \int \int d^3 x d^3 y \ g(x - y) \ \hat{\Psi}^\dagger_1(x) \hat{\Psi}^\dagger_1(y) \right]^{N/2} |0\rangle$$

Binding energy per atom is: $E^{(N)}_g = E^{(2)}_g = \Delta V - \chi^2/(4U_{11})$.

- In the limit $U_{11} \to 0$, we obtain singularity and energy collapse.
- This is in contrast to the known mean-field behavior of the classical energy, which is rigorously bounded from below.
QUANTUM SIMULTONS WITH CUTOFF

We introduce a momentum cutoff ( \( k_m \sim 2\pi/a \)):

\[
|\psi_Q^{(2)}\rangle = \left[ \hat{b}^\dagger (0) + \int_0^{k_m} d^3k \ G(k) \hat{a}^\dagger (k) \hat{a}^\dagger (-k) \right] |0\rangle,
\]

where \( \hat{a} \) and \( \hat{b} \) are Fourier components of \( \hat{\Psi}_1 \) and \( \hat{\Psi}_2 \).

- Quantum (di-boson) simulton acquires finite radius;

- the binding energy (for \( k_m \gg [\chi m_1/(2\pi\hbar)]^2 \)) is:

\[
E_Q^{(2)} = \Delta V - \chi^2/(5U_{11}).
\]
Approximate $N$-particle solutions: independent Q-ansatz

The total binding or ground state energy can be estimated using a variational approach:

\[
|\psi_{Q}^{(N)}\rangle = \left[\hat{b}^{\dagger}(0) + \int_{|k|=0}^{k_m} d^{3}k \ G(k)\hat{a}^{\dagger}(k)\hat{a}^{\dagger}(-k)\right]^{N/2} |0\rangle.
\]

- This corresponds to $N/2$ independent di-bosons or ‘dressed’ molecules;
- the energy upper bound is (for $k_m \gg [\chi m_1/(2\pi\hbar)]^2$):
  \[
  E^{(N)}_{Q} = E^{(2)}_{Q} = \Delta V - \chi^2/(5U_{11}).
  \]
Approximate $N$-particle solutions: Coherent ansatz

The above Q-ansatz, at finite $k_m$, does not give the lowest possible energy. To show this, we consider a coherent or mean-field theory (MFT) ansatz:

$$|\psi_C^{(N)}\rangle = \exp \left\{ \int d^3x \left[ \psi_1(x) \hat{\Psi}_1^\dagger(x) + \psi_2(x) \hat{\Psi}_2^\dagger(x) \right] \right\} |0\rangle .$$

- By varying $\psi_1(x)$ and $\psi_2(x)$, can get a lower energy than previously.
- The calculation makes use of the known result that the classical Hamiltonian is always bounded from below, and the bound is given by the classical soliton energy.
Energy estimate by Gaussian C-ansatz

- Chosing $\psi_j(x) = (-1)^{j-1} g_j N^2 [2/(\pi s_j)]^{3/4} \exp(-|x|^2 N^2 / s_j)$ results in:

$$E_C^{(N)} / \hbar = N^2 \left( \frac{3\hbar}{2m_1} \right) \left[ \frac{g_1^2}{s_1} + \frac{g_2^2}{2s_2} - \frac{\tilde{\chi} g_1^2 g_2 s_2^{3/4}}{(s_1 + 2s_2)^{3/2}} \right] + N^4 \tilde{U} \frac{g_1^4 s_1^{-3/2}}{s_1} + \rho g_2^2,$$

- where: $\tilde{\chi} = 2^{5/2} (2/\pi)^{3/4} m \chi / (3\hbar)$ and: $\tilde{U} = 2^{-5/2} (2/\pi)^{3/2} U_{11}$.

- Minimize $E_C^{(N)}$, under the constraint of a fixed $N$.

- P. D. Drummond, K. V. Kheruntsyan, and H. He, PRL 81, 3055 (1988).
Comparison of Q- vs C-ansatz

- The independent di-boson (Q-) and coherent (C-) ansatzs give different results; the coherent theory gives a better (lower) estimate for the ground state energy at $N \geq N_{cr}$, as $E_{C}^{(N)} \leq E_{Q}^{(N)}$.

- $N_{cr}$ depends on a combination of momentum cutoff and density effects!

- For $m_1 \sim 10^{-25}$ kg, $\chi/\hbar \sim 10^{-6}$ m$^{3/2}$/s, $a \sim 2\pi$ nm, so that $U_{11}/\hbar \sim 8 \times 10^{-17}$ m$^3$/s, and a cutoff at $k_m = 2\pi/a \sim 1$ nm$^{-1}$, we obtain:

$$N_{cr} \sim 1.4 \times 10^5.$$
GROUND STATE ENERGY ESTIMATES

\( \Delta V = 0 \)

\[
E_g^{(N)} = \frac{-1}{5} \chi^2 \frac{1}{U_{11}} \]

\[
E_g^{(N)} = \frac{-4}{5} \chi^2 \frac{1}{U_{11}}
\]

\( N_{cr} \approx 10^5 \)

Q-ansatz

C-ansatz

lower bound

exact ?
COUPLED BEC SOLITONS

Simulate MFT equations; assume that the parametric coupling is dominant (neglect $U_{11}$), and $\Delta V = 0$. 

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EXPERIMENTAL TECHNIQUES

Superchemistry BEC simultons

- Photo-association coupling of two atoms to one molecule.
- Include the effects of the trap potential.
Raman photoassociation

- Nonresonant coupling with two coherent lasers (D. Heinzen et al., QELS’99, QTuD3, QWH4).
How does it work?

- In **PHOTOASSOCIATION**: a pair of free atoms collide AND absorb a photon from a laser beam, giving an excited molecular state.

- In **RAMAN PHOTOASSOCIATION** two atoms collide, absorb a photon and are stimulated to emit one, giving a ground molecular state.

- Raman photoassociation can change the atomic scattering length.

- Less loss-prone than Feshbach resonance (Ketterle, Heinzen, Wieman).
Coherent dynamics of atom-molecular BEC

Direct simulation of the mean-field theory equations, in the presence of trap potentials:

\[
\begin{align*}
    i\hbar \dot{\psi}_1 &= -\frac{\hbar^2}{2m_1} \nabla^2 \psi_1 + V_1^c(x)\psi_1 + U_{11}^c|\psi_1|^2\psi_1 + \chi \psi_1^* \psi_2, \\
    i\hbar \dot{\psi}_2 &= -\frac{\hbar^2}{2m_2} \nabla^2 \psi_2 + V_2^c(x)\psi_2 + \frac{1}{2} \chi \psi_1^2, 
\end{align*}
\]

where parameters are calculated for the case of Rubidium dimerization using typical laser intensities and data from the Heinzen experiment.
SIMULATIONS

- The simulations are carried out in two stages.
- In the first stage, we assume that only atomic species are present.
- In the second stage we switch on the coupling $\chi$ for molecule formation.
- We include additional spontaneous decay terms.
- Giant oscillations appear in the occupation number of the molecules.
- Bose-enhanced nonlinear quantum dynamics replaces the usual chemical kinetics: SUPERCHEMISTRY.
‘SUPERCHEMISTRY’ OSCILLATIONS

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SUMMARY

- At low density, atoms couple to molecules in a particle-like way.

- For large momentum cutoff, these have a point-like structure.

- At large density, and large couplings $\chi$, the coherent coupling of two entire condensates is dominant - just as in nonlinear optics.

- Call this a type of superchemistry in BEC.

- Can form three-dimensional atom-molecular BEC simultons.

- Could stabilize atom laser outputs; coherent BEC nano-chemistry?