Description of Rabi oscillations in TDDFT: The effect of spin

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Outline

- Motivation
- Theory
- Results*
  - Exact calculation
  - TDDFT calculation
- Conclusions

Motivation

Rabi oscillation: Oscillation between the ground state and an excited state with laser frequency close to resonance

Large change in the density $\Rightarrow$ Problems with adiabatic approximations in TDDFT

Rabi frequency:

$$\Omega_0 = \langle \psi_g | \sum_j \hat{r}_j | \psi_e \rangle E_0$$
Rabi oscillation

Time-dependent many-body-wave function:

\[ |\psi(t)\rangle = a_g(t)|\psi_g\rangle + a_e(t)|\psi_e\rangle \]

Detuning from the resonance:

\[ \delta = \omega - \Delta \]

We should satisfy the conditions:

\[ \delta \ll \Delta, \quad \Omega_0 \ll \omega \]

Occupation number of the excited state:

\[ |a_e(t)|^2 = n_e(t) \]

Occupation number of the ground state:

\[ |a_g(t)|^2 = n_g(t) \]
Exact calculation

Two electron in one dimension

External potential (1D): 
\[ V_{1\text{ext}} = \frac{-2}{\sqrt{x^2 + 1}} \quad V_{2\text{ext}} = \frac{-4}{\sqrt{x^2 + 1}} \]

Electron-electron interaction: (soft Coulomb)
\[ V_{ee} = \frac{1}{\sqrt{(x_1 - x_2)^2 + 1}} \]

Equation for \( n_e(t) \):
\[ \partial_t^2 n_e(t) = - (\delta^2 + \Omega_0^2) n_e(t) + \frac{1}{2} \Omega_0^2 \]

\[ n_e^{\text{max}} = \frac{\Omega_0^2}{\Omega_0^2 + \delta^2} \]

J.I. Fuks et al., PRB 84, 75107 (2011)
D. Tannor, Introduction to Quantum Mechanics a time dependent perspective, pp. 479-482.
Equation for $n_e$ for singlet case

**EXX**

$$\partial_t^2 n_e^s(t) = -\left(\frac{\gamma^2}{2} n_e^s(t)^2 + \Omega_s^2\right) n_e^s(t) + \frac{1}{2} \Omega_s^2$$

**Resonant** $\omega = 0.549 \text{ Ha}$

**Dipole moment**
- $n_g$ (red)
- $n_e$ (blue)
Triplet
Exact calculation

\[ \Omega_0 = 0.0045 \text{ Ha} \]

\[ \Delta = 0.172\text{Ha} \]

\[ \delta = 0.02\Omega_0 \]

\[ \delta = 2.3\Omega_0 \]
Mitglied der Helmholtz-Gemeinschaft

T. Grabo et al., Orbital functionals in density functional theory: the optimized effective potential method.

\[ \varphi_2(x, t) = a_g(t)\varphi_2(x, 0) + a_e(t)\varphi_3(x, 0) \]

\[ \varphi_1(x, t) = e^{i\alpha(t)}\varphi_1(x, 0) \]

\[ \Omega_0 = 0.0049 \text{ Ha} \]

\[ \delta = 2.04\Omega_0 \quad \omega = 0.179 \text{ Ha} \]

\[ \omega = 0.169 \text{ Ha} \]
Triplet
TDDFT – EXX
Maximum $n_e(t)$

$\delta = 0.04 \Omega_0$

$\omega = 0.167 \text{ Ha}$
Triplet

TDDFT – ALDA

\[ V_{1ext} = \frac{-2}{\sqrt{x^2 + 1}} \]

\[ V_{2ext} = \frac{-4}{\sqrt{x^2 + 1}} \]
Triplet
TDDFT – ALDA
Maximum $n_e(t)$

ALDA

$\omega = 0.545$ Ha

$\omega_{res} = 0.556$ Ha

ALDA With SIC

$\omega = 0.530$ Ha

$\omega_{res} = 0.515$ Ha
Conclusion

Comparison between exact calculation and TDDFT for Rabi oscillations for triplet state.

ALDA  ➔ off resonance behaviour
       better behaviour with SIC

EXX  ➔ resonant behaviour with applying small detuning
       Numerically suggests detuning is constant

Outlook  ➔ Derive the detuning term and differential equation for \( n_e^s \)
Thank you for your attention!