Is there an exact Kohn-Sham system in Current Density Functional Theory ?

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- Density Functional Theory (DFT): $E_0[\, n(\mathbf{r}) \,]$
  [Hohenberg and Kohn (1964)]

$$\begin{tikzcd} v^{ext}(\mathbf{r}) & \Psi_0 \arrow[l, "\mathcal{C}"'] \arrow[r, shift left, "\mathcal{D}"] & n_0(\mathbf{r}) \arrow[l, shift left, "\mathcal{D}^{-1}"] \end{tikzcd}$$

$\Rightarrow$ Existence of a KS system for the ground states follows from these mappings.

- Current Density Functional Theory (CDFT): $E_0[\, n(\mathbf{r}), \mathbf{j}^p(\mathbf{r}) \,]$ (semi-relativistic)
  [Vignale and Rasolt (1987), Capelle and Vignale (2002)]

$$\begin{tikzcd} \mathcal{V}_1^{ext}(\mathbf{r}) \arrow[dr] & & \\ \mathcal{V}_2^{ext}(\mathbf{r}) \arrow[r] & \Psi_0 \arrow[r, shift left] & \mathcal{N}_0(\mathbf{r}) \arrow[l, shift left] \\ \cdots \arrow[ur] & & \end{tikzcd}$$

where: $\mathcal{V}^{ext}(\mathbf{r}) = [v^{ext}(\mathbf{r}), \mathbf{A}^{ext}(\mathbf{r})]$ and $\mathcal{N}(\mathbf{r}) = [n(\mathbf{r}), \mathbf{j}^p(\mathbf{r})]$

$\Rightarrow$ Existence of a KS system is **neither** guaranteed **nor** excluded.

Q: If we cannot prove the existence, can we reject it by providing a counter-example using an exactly solvable system ?

- Densities in terms of KS wave functions (WFs)

  (atomic units $\hbar = m = e = 1$ are used throughout the talk)

  KS WFs: $\varphi_k(\mathbf{r}) = R_k(\mathbf{r})\, e^{i\zeta_k(\mathbf{r})}$

  densities of a *non-interacting* system :

$$
\begin{aligned}
n(\mathbf{r}) &= \sum_k^{occ} \varphi_k^*(\mathbf{r})\, \varphi_k(\mathbf{r}) \\
&= \sum_k^{occ} R_k^2(\mathbf{r}) \\
\mathbf{j}^p(\mathbf{r}) &= \frac{1}{2i} \sum_k^{occ} \left[\varphi_k^*(\mathbf{r})\, \boldsymbol{\nabla}\varphi_k(\mathbf{r}) - \varphi_k(\mathbf{r})\, \boldsymbol{\nabla}\varphi_k^*(\mathbf{r})\right] = \\
&= \sum_k^{occ} R_k^2(\mathbf{r})\, \boldsymbol{\nabla}\zeta_k(\mathbf{r})
\end{aligned}
$$

  Q: Is it possible to express both exact densities of our solvable system in terms of the same set of KS-WFs ?

- standard CDFT [Vignale and Rasolt (1987)]:

  these expressions are used *ad hoc* as an *ansatz* for deriving the KS equations in terms of the XC energy functional $E_{xc}[\, n(\mathbf{r}), \mathbf{j}^p(\mathbf{r})\,]$.

- paramagnetic current density $\mathbf{j}^p(\mathbf{r})$ is gauge dependent

  ⇒ use the gauge-invariant vorticity instead

$$
\boldsymbol{\gamma}(\mathbf{r}) = \boldsymbol{\nabla} \times \frac{\mathbf{j}^p(\mathbf{r})}{n(\mathbf{r})}
$$

- observe:

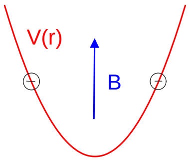
  for each individual KS state the vorticity vinishes

$$
\boldsymbol{\gamma}_k(\mathbf{r}) = \boldsymbol{\nabla} \times \frac{\mathbf{j}_k^p(\mathbf{r})}{n_k(\mathbf{r})} = \boldsymbol{\nabla} \times \boldsymbol{\nabla}\, \zeta_k(\mathbf{r}) = 0
$$

  (independent of the form of the WF)

1. Exactly Solvable Model in 2D

two electron harmonic quantum dot in **B**



1.1 Exact Decoupling

- Hamiltonian

$$H = \sum_{i=1}^{2} \left\{ \frac{1}{2} \left[\mathbf{p}_i + \frac{1}{2c} \mathbf{B} \times \mathbf{r}_i \right]^2 + \frac{1}{2} \omega_0^2 \, r_i^2 \right\} + \frac{1}{|\mathbf{r}_2 - \mathbf{r}_1|}$$

- Relative and Center–of–Mass Coordinates

$$\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1 \quad ; \quad \mathbf{R} = \frac{1}{2}(\mathbf{r}_1 + \mathbf{r}_2)$$

- Decoupled Hamiltonian

$$\begin{aligned} H &= 2\, H_r + \frac{1}{2}\, H_R \\ H_r &= \frac{1}{2} \left[\mathbf{p} + \frac{1}{4c} \mathbf{B} \times \mathbf{r} \right]^2 + \frac{1}{8} \omega_0^2 \, \mathbf{r}^2 + \frac{1}{2\, r} \\ H_R &= \frac{1}{2} \left[\mathbf{P} + \frac{1}{c} \mathbf{B} \times \mathbf{R} \right]^2 + 2\, \omega_0^2 \, \mathbf{R}^2 \end{aligned}$$

- Eigenfunctions and Eigenvalues

$$\Psi = \varphi(\mathbf{r}) \cdot \xi(\mathbf{R}) \cdot \chi(s_1, s_2)$$

$$E = 2\, \varepsilon_r + \frac{1}{2}\, \eta_R$$

where ε_r, $\varphi(\mathbf{r})$ and η_R, $\xi(\mathbf{R})$ are the eigenvalues and eigenfunctions of the operators H_r and H_R, respectively.

1.2 Center–of–Mass Motion

- Eigenfunctions in polar coordinates $\mathbf{R} = (R, \mathcal{A})$

$$\xi(\mathbf{R}) = \frac{e^{iM\mathcal{A}}}{\sqrt{2\pi}} \; \frac{U(R)}{R^{1/2}} \qquad ; \qquad M = 0, \pm 1, \pm 2, \ldots$$

M= c.m. angular momentum quantum number

- Radial Part

$$U(R) \propto R^{|M|+(1/2)} \; L_N^{|M|}(2\,\widetilde{\omega} R^2)\; e^{-\widetilde{\omega} R^2} \quad ; \quad N = 0, 1, 2, ...$$

$\widetilde{\omega} = \sqrt{\omega_0^2 + (\omega_c/2)^2} \quad ; \quad \omega_c = \frac{B}{c}$
$L_N^{|M|}(x)$= associated Laguerre polynomial
N= number of nodes

- Eigenvalues

$$\eta_{M,N} = (2N + |M| + 1)\; 2\widetilde{\omega} + M\, \omega_c$$

1.3 Relative Motion

- Eigenfunctions in polar coordinates $\mathbf{r} = (r, \alpha)$

$$\varphi(\mathbf{r}) = \frac{e^{im\alpha}}{\sqrt{2\pi}} \; \frac{u(r)}{r^{1/2}} \qquad ; \qquad m = 0, \pm 1, \pm 2, \ldots$$

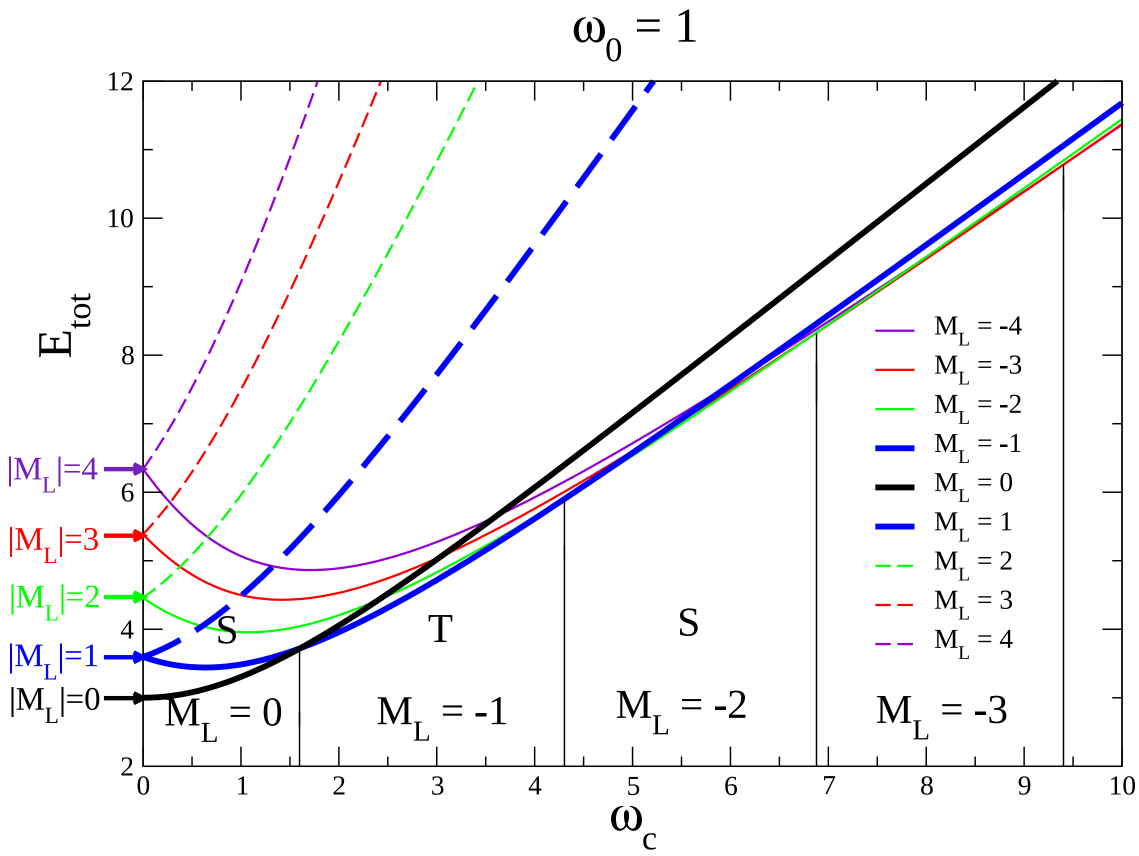
m= relative angular momentum quantum number

- Radial Schrödinger equation

$$\left\{ -\frac{1}{2}\,\frac{d^2}{dr^2} + \frac{1}{2}\left(m^2 - \frac{1}{4}\right)\frac{1}{r^2} + \frac{1}{8}\,\widetilde{\omega}^2\, r^2 + \frac{1}{2r} \right\} u(r) = \tilde{\varepsilon}_r\; u(r)$$

eigenvalues: $\tilde{\varepsilon}_r = \varepsilon_r - \frac{1}{4}\, m\, \omega_c$
normalization: $\int_0^\infty dr |u(r)|^2 = 1$
This equation can be solved analytically for a discrete, but infinite set of values for $\widetilde{\omega} = \sqrt{\omega_0^2 + (\omega_c/2)^2}$ [Taut(1994)]

- Eigenvalues for $M = 0$ (c.m. system in ground state)



⇒ angular momentum of the ground state decreases monotously with increasing magnetic field

1.4 Exact densities

- density and paramagnetic current density

$$\begin{aligned} n(\mathbf{r}) &= 2\int d\mathbf{r}'\, |\Phi(\mathbf{r},\mathbf{r}')|^2 \\ \mathbf{j}^p(\mathbf{r}) &= -i\int d\mathbf{r}'\, [\Phi^*(\mathbf{r},\mathbf{r}')\boldsymbol{\nabla}\Phi(\mathbf{r},\mathbf{r}') - \Phi(\mathbf{r},\mathbf{r}')\boldsymbol{\nabla}\Phi^*(\mathbf{r},\mathbf{r}')] \end{aligned}$$

- for M=0 in terms of radial WF $u_m(r)$

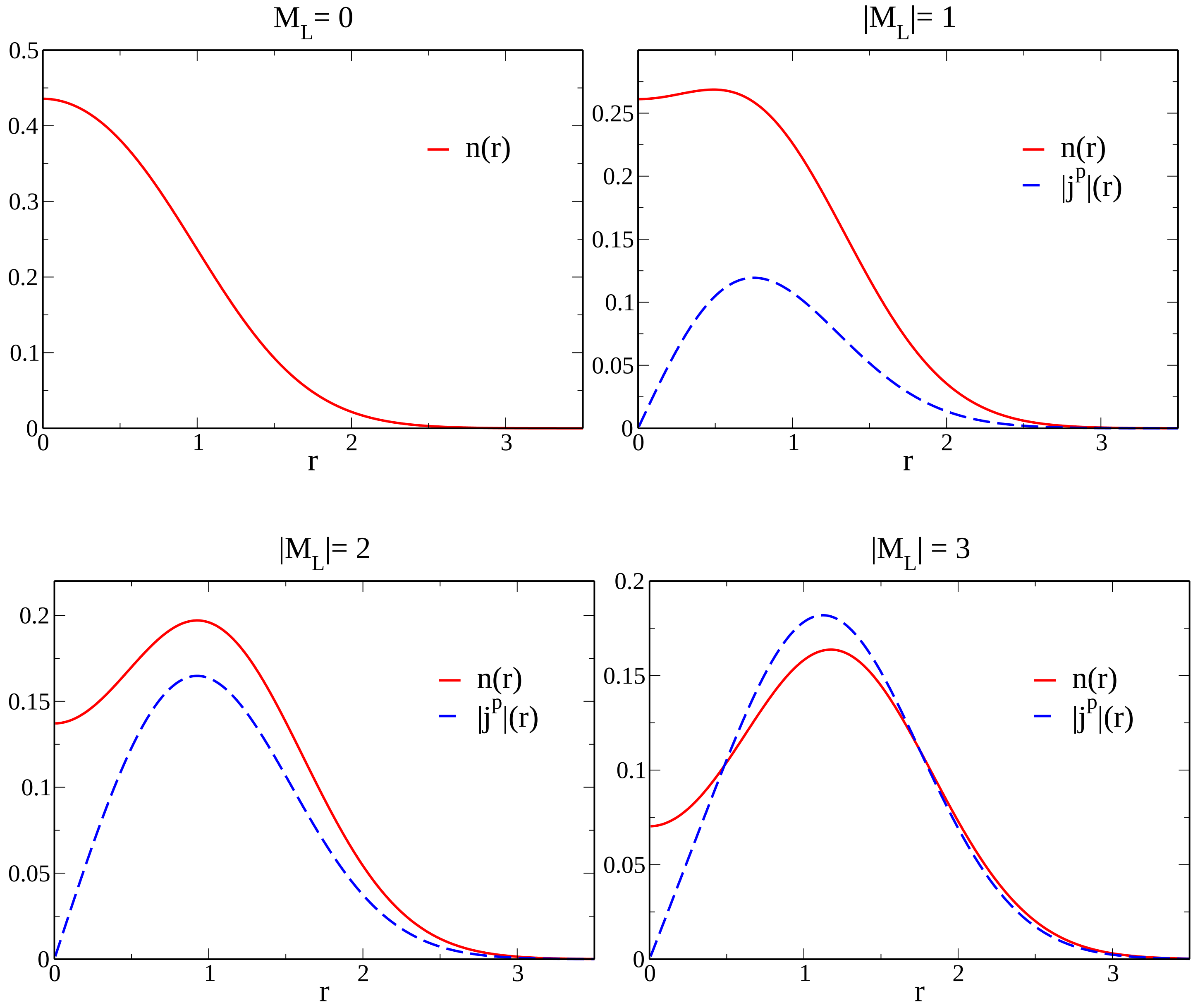
$$\begin{aligned} n(r) &= \frac{4\widetilde{\omega}}{\pi} e^{-2\widetilde{\omega}\,r^2} \int_0^\infty dr'\, e^{-(\widetilde{\omega}/2)r'^2}\, I_0(2\widetilde{\omega}rr')\, [u_m(r')]^2 \\ \mathbf{j}^p(\mathbf{r}) &= \mathbf{e}_\alpha\, m\, \frac{4\widetilde{\omega}}{\pi} e^{-2\widetilde{\omega}\,r^2} \int_0^\infty dr'\, e^{-(\widetilde{\omega}/2)r'^2}\, \frac{I_1(2\widetilde{\omega}rr')}{r'}\, [u_m(r')]^2 \end{aligned}$$

$I_n(x)$ = modified Bessel function

- power series for $r \longrightarrow 0$

$$\begin{aligned} n(r) &\longrightarrow \frac{4\,\widetilde{\omega}}{\pi} e^{-2\,\widetilde{\omega}\,r^2}\, [A_0 + A_2\,\widetilde{\omega}^2\, r^2 + \cdots] \\ j^p(r) &\longrightarrow M_L \frac{4\,\widetilde{\omega}^2}{\pi} e^{-2\,\widetilde{\omega}\,r^2}\, r\, [A_0 + \frac{1}{2} A_2\,\widetilde{\omega}^2\, r^2 + \cdots] \\ \gamma(r) &\longrightarrow M_L\, 2\,\widetilde{\omega}\, [1 - \widetilde{\omega}^2\, \frac{A_2}{A_0}\, r^2 + \cdots] \end{aligned}$$

where:
M_L= total orbital angular momentum
$\widetilde{\omega} = \sqrt{\omega_0^2 + (\omega_c/2)^2}$ = eff. confinement frequency
$A_k = \int_0^\infty dr\, r^k\, e^{-(\widetilde{\omega}/2)r^2}\, [u_m(r)]^2$ = pos. definite coefficients



Exact densities and paramagnetic current densities for $\tilde{\omega} = 1$ and the orbital angular momenta given in the titles of the figures. The sign of $j^p(r)$ agrees with the sign of M_L.

2. Matching density and vorticity

2.1 Singlet state (M_L even)

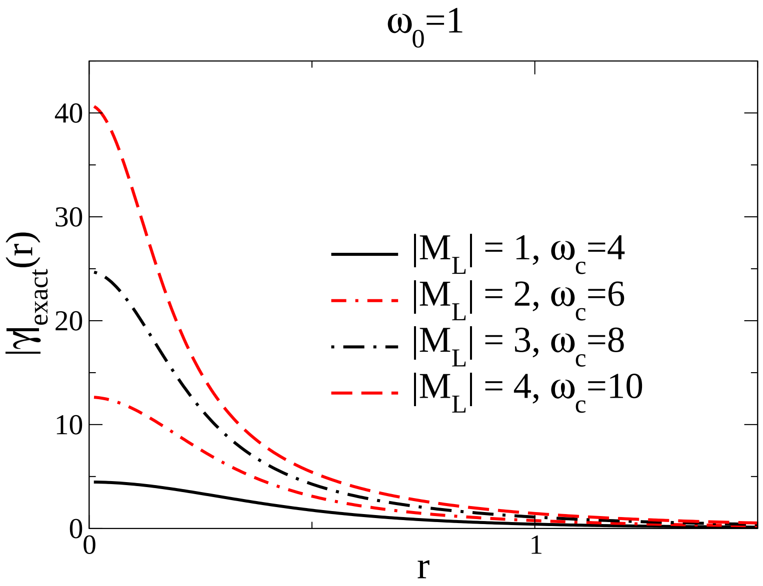
two electrons in the same orbital state $\varphi_k(\mathbf{r}) = R_k(\mathbf{r})\, e^{i\zeta_k(\mathbf{r})}$

$$n_{exact}(r) \overset{!}{=} n_{KS}(\mathbf{r}) = 2\,[R_k(\mathbf{r})]^2$$
$$\boldsymbol{\gamma}_{exact}(\mathbf{r}) \overset{!}{=} \boldsymbol{\gamma}_{KS}(\mathbf{r}) = \boldsymbol{\gamma}_k(\mathbf{r}) = 0$$

- $\Rightarrow R_k(r) = \sqrt{\frac{1}{2}\, n_{exact}(r)}$
- Matching of vorticities only for $\boldsymbol{\gamma}_{exact}(\mathbf{r}) = 0$ possible.

  limit $r \rightarrow 0$: $\gamma(r) \longrightarrow M_L\, 2\, \widetilde{\omega}\, [1 - \widetilde{\omega}^2\, \frac{A_2}{A_0}\, r^2 + \cdots]$

  general:



  $\Rightarrow$ Condition is (trivially) fulfilled only for $M_L = 0$, because $\boldsymbol{\gamma}_{exact}(\mathbf{r}) \propto M_L$

- 'order of violation': $|\gamma_{exact}(0)| = 2\,\sqrt{\omega_0^2 + (\omega_c/2)^2}\,|M_L|$
- The same result is obtained if we match $\mathbf{j}^p(\mathbf{r})$ instead of $\boldsymbol{\gamma}(\mathbf{r})$.

2.2 Triplet state (M_L odd) in the limit $r \longrightarrow 0$

two electrons in the orbital states $\varphi_1(\mathbf{r})$ and $\varphi_2(\mathbf{r})$

- Even for non-spherical systems the solutions of a KS equation with 'non-pathological' effective potentials have in the limit $r \longrightarrow 0$ the form

$$\varphi_k(\mathbf{r}) \longrightarrow R_k(r)\, e^{i\widetilde{m}_k\alpha} \quad ; \quad \widetilde{m}_k = 0, 1, 2, \cdots$$

$$\boxed{\begin{aligned} n_{exact}(r) &\overset{!}{=} n_{KS}(r) = [R_1(r)]^2 + [R_2(r)]^2 \\ \boldsymbol{\gamma}_{exact}(\mathbf{r}) &\overset{!}{=} \boldsymbol{\gamma}_{KS}(\mathbf{r}) = \frac{1}{r}\frac{d}{dr}\left[r\, \frac{j^p_{KS}(r)}{n_{KS}(r)}\right] \mathbf{e}_z \end{aligned}}$$

where: $j^p_{KS}(r) = \widetilde{m}_1 \frac{[R_1(r)]^2}{r} + \widetilde{m}_2 \frac{[R_2(r)]^2}{r}$

- Power expansion of the radial part in the limit $r \longrightarrow 0$ $R_k(r) \longrightarrow c_k\, r^{\widetilde{m}_k}$; 'small-r-exponents' $\widetilde{m}_k = 0, 1, 2, \cdots$

$$\boxed{\begin{aligned} n_0 + n_2\, r^2 + \cdots &\overset{!}{=} c_1^2\, r^{2\widetilde{m}_1} + c_2^2\, r^{2\widetilde{m}_2} + \cdots \\ \gamma_0\, M_L + \cdots &\overset{!}{=} \frac{2\, c_1^2\, c_2^2\, (\widetilde{m}_1 - \widetilde{m}_2)^2\, r^{2(\widetilde{m}_1+\widetilde{m}_2-1)}}{(n_0 + n_2\, r^2)^2} + \cdots \end{aligned}}$$

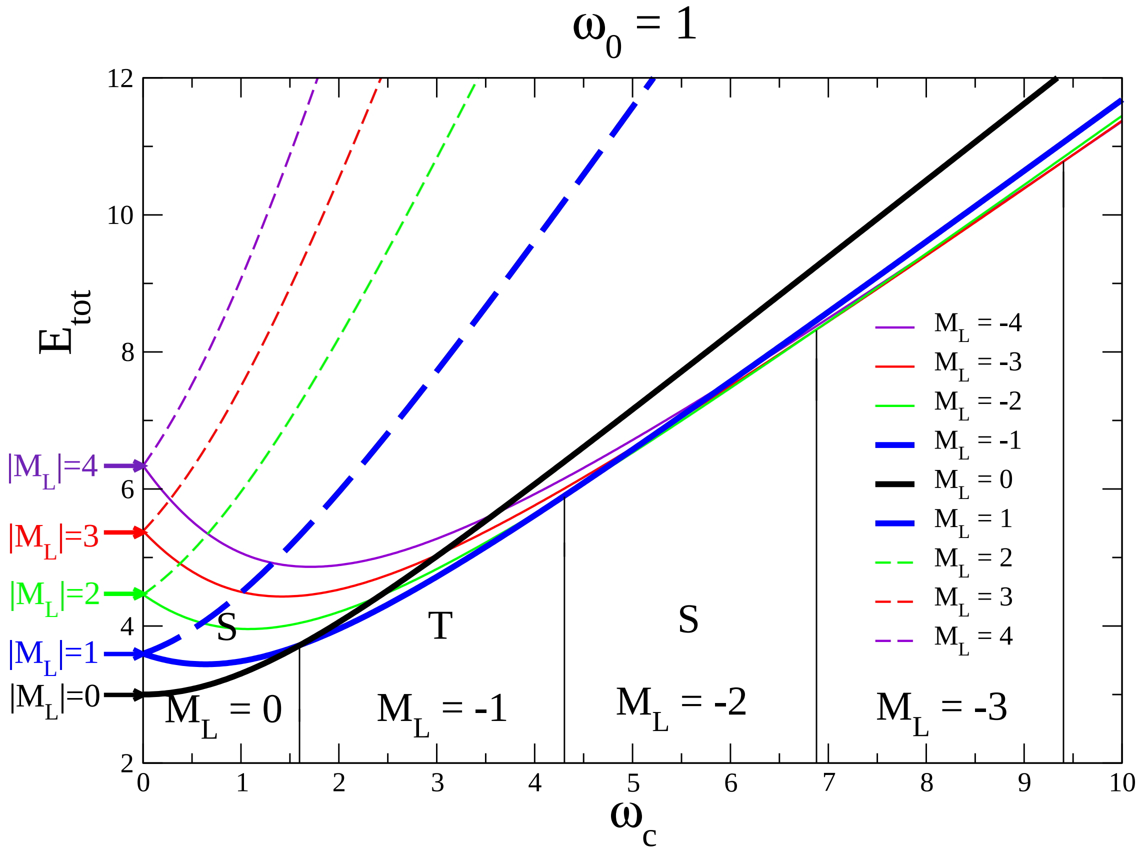
$\Rightarrow$ only compatible for $\widetilde{m}_1 = 0$; $\widetilde{m}_2 = 1$

- **Assume axial symmetry for KS system:**

$\Rightarrow \widetilde{m}_k \longrightarrow |m_k|$ (angular momentum) $\Rightarrow |M_L| = 1$

Summary

- we considered the *exact solutions* of a *two-electron* system in a *parabolic confinement* and a *homogeneous magnetic field*:
- exact Kohn-Sham (KS) system can exist only for:
  $M_L = 0$ (spin singlet)
  $|M_L| = 1$ (spin triplet)
- In other words:
  KS system can exist only for those states, which are continuously connected to the singlet and triplet ground states at $B = 0$.
  (see thick lines in figure below).



- existence of Kohn-Sham WFs in CDFT is **neither** guaranteed for **nor** restricted to ground states.

References

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