Second Random Phase Approximation Studies in Metallic Clusters

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Outline

- Random Phase Approximation (RPA)
- RPA Limits
- Extension: Second Random Phase Approximation (SRPA)
  - Second Random Phase Approximation Studies in Metallic Clusters
- Results
- SRPA Problematic Aspects
- Conclusions and Outlook

\(^a\)D. Gambacurta and F. Catara, submitted to Phys. Rev. B
The Method of the Equations of Motion

Set of exact eigenstates of the Hamiltonian $H$:

$$H|\nu\rangle = E_\nu |\nu\rangle$$

where $|0\rangle$ is the ground state with energy $E_0$.

Let us introduce the operators $Q$’s:

$$Q^\dagger_\nu |0\rangle = |\nu\rangle$$

$$Q_\nu |0\rangle = 0.$$ 

Equations of Motion:

$$\langle 0 | [\delta Q, [H, Q^\dagger_\nu]] |0 \rangle = \omega_\nu \langle 0 | [\delta Q, Q^\dagger_\nu] |0 \rangle$$

where

$$\omega_\nu = E_\nu - E_0.$$
To obtain Random Phase Approximation (RPA) two approximations are made:

\[ Q^\dagger_\nu = \sum_{ph} X^{(\nu)}_{ph} a_p^\dagger a_h - \sum_{ph} Y^{(\nu)}_{ph} a_h^\dagger a_p \]

| 0 \rangle is not known: in Eqs. of Motion

\[ | 0 \rangle \sim | HF \rangle \Rightarrow \]

**Quasi-Boson Approximation (QBA)**

**RPA Limits**

- **Internal Inconsistency**: | 0 \rangle is used in the derivation of Eqs. of Motion
- | 0 \rangle (correlated) \rightarrow | HF \rangle (uncorrelated)
- QBA introduces a violation of the Pauli principle
- RPA predicts a harmonic spectrum and thus it is not suited to the study of the anharmonicities.
Second RPA

More general excitation operators: (Double Excitations)

\[ Q_\nu^\dagger = \sum_{p,h} \left( X_{ph}^{(\nu)} a_p^\dagger a_h - Y_{ph}^{(\nu)} a_h^\dagger a_p \right) \]

\[ + \sum_{p<p',h<h'} \left( X_{php'h'}^{(\nu)} a_p^\dagger a_h^\dagger a_{p'}^\dagger a_{h'} - Y_{php'h'}^{(\nu)} a_h^\dagger a_p a_{h'} a_{p'}^\dagger \right) \]

Several RPA properties hold in SRPA

**Thouless Theorem still holds**
SRPA preserves Energy Weighted Sum Rules (**No Spurious States**)

Equations of Motion (1\(\mapsto\) 1p-1h, 2\(\mapsto\) 2p-2h)

\[
\begin{pmatrix}
A_{11} & A_{12} & B_{11} & B_{12} \\
A_{21} & A_{22} & B_{21} & B_{22} \\
-B_{11}^* & -B_{12}^* & -A_{11}^* & -A_{12}^* \\
-B_{21}^* & -B_{22}^* & -A_{21}^* & -A_{22}^*
\end{pmatrix}
\begin{pmatrix}
\chi_1^{\nu} \\
\chi_2^{\nu} \\
\gamma_1^{\nu} \\
\gamma_2^{\nu}
\end{pmatrix}
= \omega_{\nu}
\begin{pmatrix}
\chi_1^{\nu} \\
\chi_2^{\nu} \\
\gamma_1^{\nu} \\
\gamma_2^{\nu}
\end{pmatrix}
The Method of the Equations of Motion

RPA and SRPA
Metallic Clusters
Calculations
Results
Conclusions and Outlook

Matrices

RPA Matrices (1p-1h configurations)

\[ A_{1,1'} = \langle HF | [a_h^\dagger a_p, [H, a_p^\dagger a_{h'}]] | HF \rangle \]
\[ B_{1,1'} = -\langle HF | [a_h^\dagger a_p, [H, a_{h'} a_p^\dagger]] | HF \rangle. \]

SRPA Matrices (1p-1h and 2p-2h configurations)

\[ A_{1,2} = A_{2,1}^* = \langle HF | [a_h^\dagger a_p, [H, a_{p_1}^\dagger a_{p_2}^\dagger a_{h_1} a_{h_2}]] | HF \rangle \]
\[ A_{2',2} = \langle HF | [a_{h_2'}^\dagger a_{h_1'}^\dagger a_{p_2}^\dagger a_{p_1}^\dagger, [H, a_{p_1}^\dagger a_{p_2}^\dagger a_{h_1} a_{h_2}]] | HF \rangle \]
\[ B_{1,2} = B_{2,1}^* = -\langle HF | [a_i^\dagger a_m, [H, a_k^\dagger a_l^\dagger a_q a_p]] | HF \rangle = 0 \]
\[ B_{2',2} = -\langle HF | [a_{p_1}^\dagger a_{p_2}^\dagger a_{h_1} a_{h_2'}, [H, a_{p_1}^\dagger a_{p_2}^\dagger a_{h_1} a_{h_2}]] | HF \rangle = 0 \]

QBA is still used
Diagonal Approximation

- The dimension of the $A_{2,2}$ matrix can be very large.
- The SRPA problem can be reduced to an RPA eigenvalue problem but with $A_{1,1}$ depending on the Energy $\omega$

$$\tilde{A}_{1,1'} = A_{1,1'} + \sum_{2,2'} A_{1,2}(\omega + i\eta - A_{2,2'})^{-1}A_{2',1'}$$

- The matrix inversion is very expensive.
- If we neglect the residual interaction among the $2p - 2h$ states

$$A_{2,2} \simeq U(ij)U(mn)\delta_{ik}\delta_{jl}\delta_{mp}\delta_{nq}(\epsilon_m + \epsilon_n - \epsilon_i - \epsilon_j)$$

the inversion is algebraic, (diagonal approximation)
QBA in SRPA

QBA is still used and it has been suggested that it is an even more severe approximation than in RPA:


In a previous work (Phys. Rev. C73, 024319 (2006))

- We presented an approach to go beyond QBA and applied it to the 3 level Lipkin model
- The inclusion of g.s. correlations improves agreement with exact results
- However the model is very schematic and contains several parameters (coupling among 1p – 1h and 2p – 2h configurations)

Metallic Clusters

Study of the vibrational spectrum of a realistic many-body system within SRPA
Metallic Clusters

- Metallic Clusters are aggregates of atoms with a well defined size.
- At lowest order the valence electrons can be approximately treated as independent \( \rightarrow \) HF.

Anallogies between metallic clusters and nuclei:

- Shell Structure and Magic Numbers
- Collective Excitations (Only Dipole Plasmon observed experimentally)

Differences:

- Ionic Background
  Jellium approximation: *uniform positive charge distribution (ions) which interacts with valence electrons (also interacting among themselves) via the bare Coulomb interaction*
- No adjustable parameters
  (clearer comparison between different approximations)
As a first step, we have fixed the single particle (s.p.) basis by solving the HF equations.

The s.p. wavefunctions are represented as linear superposition of harmonic oscillator ones.

We have solved the RPA and SRPA equations for natural parity states, with multipolarities ranging from $L = 0$ to $L = 3$.

SRPA including all kinds of couplings among all $1p-1h$ and $2p-2h$ configurations.

In the following we focus our attention only on spin $S = 0$ states.

However, both spin $S = 0$ and $1$ ($1p - 1h$) states are considered in the construction of the $2p - 2h$ configurations:

$$
\left[ B_{L_1 S_1}^{\dagger} (p_1 h_1) B_{L_2 S_2}^{\dagger} (p_2 h_2) \right]_{00}^L S, \quad (L_1, L_2 = 0 - 3, S_1, S_2 = 0, 1)
$$

RPA and SRPA strength distributions for the multipole operator $F(r)^{\lambda} = r^{\lambda} Y_{\lambda 0}$ are compared.
Strength Distributions

Possible reasons for such a behavior
QBA
Self-Energy

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Second Random Phase Approximation Studies in Metallic Clusters
Strength Distributions

Possible reasons for such a behavior
QBA
Self-Energy

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Possible reasons for such a behavior

- QBA
- **Self-Energy** of particle-hole state
As above discussed, both in RPA and SRPA, QBA is used. QBA is justified only if ground state correlations are not too strong. In RPA the $Y_{ph}^\nu$ are a measure of these correlations and thus, looking at them could be useful about the adequacy of QBA (see next slide).

RPA occupation numbers can be explicitly obtained

$$n_h = 1 - \frac{1}{2} \sum_{p,\nu} | Y_{ph}^\nu |^2,$$
$$n_p = \frac{1}{2} \sum_{h,\nu} | Y_{ph}^\nu |^2.$$

To our knowledge, the corresponding SRPA expressions have not been obtained.

However, it could be instructive to compare these quantities when the $Y_{ph}^\nu$ obtained in RPA and SRPA are used.

In this sense we will refer in the following to SRPA occupation numbers.
Analysis of $Y_{ph}^\nu$ amplitudes

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<td>Y_{ph}^\nu</td>
<td>^2$</td>
<td>RPA</td>
<td>SRPA-A22D</td>
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<tr>
<td>$\sum_{ph,\nu}</td>
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<td>^2$</td>
<td>RPA</td>
<td>SRPA-A22D</td>
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Occupation Numbers

RPA

\[ n_p \]

\[ (n_h - 1) \]

SRPA

\[ n_p \]

\[ (n_h - 1) \]

Energy (eV)

-12 -8 -4 0
As above mentioned, the SRPA problem can be reduced to an energy-dependent RPA problem

\[ \tilde{A}_{1,1'} = A_{1,1'} + \Sigma_{1,1'}(\omega) \]

\( \Sigma_{1,1'}(\omega) \) is the p-h Self-Energy

- Its real part gives a shift of the (RPA) resonance energies \(^a\)
- While its imaginary part takes into account spreading width effects

In order to have a quantitative evaluation we have calculated the real part of the RPA Self-Energy due to coupling to 2p-2h configurations \(^b\)

\[ \Sigma^{RPA}_\nu(\omega) = \sum_{2p2h} \frac{|<\nu|V|2p2h>|^2}{\omega - \epsilon_{2p2h} + i\eta} = \sum_{2p2h} K(\omega, 2p2h) \]


\(^b\) D. Lacroix et al., Progress in Part. and Nucl. Phys. 52 (2004) 497
Self Energy (Dipole Plasmon in Na$_9^+$)

![Graphs showing self-energy and strength distributions](image)
Conclusions and Outlook

Analysis of Excitation Spectrum of Metallic Clusters within SRPA

- Comparison with RPA description shows that
  - Large shift down of the RPA energies
  - Strong modifications of the Strength distributions (Dipole Case)
- We discussed some possible origins of these unpleasant results
  - QBA (more severe than in RPA)
  - Real part of p-h Self-Energy gives rise to large shifts

Outlook

- Short range correlations are neglected. This limitation could be overcome:
  - Better treatment of g.s. correlations (avoiding QBA) by using extended RPA and SRPA approaches\(^a\)^\(^b\)
  - By using an effective interaction derived from density functional including such short range correlations
- It is also interesting to analyze how much such strong modifications depend on the studied system.
  - We plan to apply SRPA in nuclei with effective Skyrme-type interactions.

The valence electrons motion is determined by

\[ H = \sum_i h_i + \sum_{i<j} v_{ij}, \]

\[ h_i = -\frac{\hbar^2}{2m} \nabla_i^2 + V(r_i); \quad v_{ij} = \frac{e^2}{4\pi\varepsilon_0} \frac{1}{|\vec{r}_i - \vec{r}_j|}, \]

\[ V(r) = \begin{cases} 
\frac{Ne^2}{4\pi\varepsilon_0} \left( \frac{1}{2r_c} \right) \left( \frac{r^2}{r_c^2} - 3 \right) & \text{for } r \leq r_c \\
-\frac{1}{r} & \text{for } r \geq r_c 
\end{cases}, \]

\[ r_c = r_s N_e^{1/3} \] is the radius of the jellium sphere

\[ r_s \] the Wigner-Seitz radius

\[ N \] is the number of ions
RP A and SRPA preserve EWSR

**Energy Weighted Sum Rule (EWSR)**

If \( \vert 0 \rangle \) and \( \vert \nu \rangle \) are a complete set of exact eigenstates with eigenvalues \( E_0 \) and \( E_\nu \) and \( F \) a transition operator, the following identity holds:

\[
\sum_\nu (E_\nu - E_0) \vert \langle \nu | F | 0 \rangle \vert^2 = \frac{1}{2} \langle 0 | [F^\dagger, [H, F]] | 0 \rangle.
\]

Violated when \( \vert 0 \rangle \) and \( \vert \nu \rangle \) are calculated with some approximation.

It is a measure of the adequacy of the approximation.

**Thouless theorem**

*the above equality is satisfied if one evaluates the l.h.s. within RPA and the mean value of the double commutator in the r.h.s. in the \( |HF \rangle \) state*

- The same holds in SRPA
- The first moment \( m_1 = \sum_\nu \omega_\nu \vert \langle \nu | F | 0 \rangle \vert^2 \) is the same in RPA and SRPA
- Thouless theorem guarantees that spurious excitations separate out and are orthogonal to the physical states.
Strength Distributions

- The SRPA strength distributions are shifted to lower energies and the height of the main peaks is reduced.
- However the first moment $m_1$ is the same in RPA and SRPA (numerically verified).
- Overall contribution of the high lying SRPA states to $m_1$ is quite big.
- For example, in the dipole case, the states lying above 10 eV exhaust 40% of $m_1$. 

![Graph showing strength distributions](image-url)
SRPA with density dependent forces

In the case of density dependent forces the RPA Equations are obtained in the Time Dependent HF (small amplitudes limit)

- One Body Density \( \rho_{kl}(t) = \langle \psi(t) | a_l^\dagger a_k | \psi(t) \rangle \)

- External Perturbation \( f(t) \)

- Eqs. of Motion: \( i\hbar \dot{\rho} = [h + f(t), \rho] \) (Linearization)

- Residual interaction defined as the second derivative of g.s. energy with respect to the density \( v_{psqr} = \frac{\partial^2 E}{\partial \rho_{qp} \partial \rho_{rs}} \)

SRPA Generalization

- Two-Body Density \( \rho_{klpq}^{(2)}(t) = \langle \psi(t) | a_p^\dagger a_q^\dagger a_l a_k | \psi(t) \rangle \)

- Eqs. of Motion: \( i\hbar \dot{\rho}^{(2)} = [h + f(t), \rho^{(2)}] \)

- Linearization \( \mapsto \)

- More general definition of residual interaction
P. Papakonstantinou and R. Roth (Arxiv.0805.4086, (2008))
Self-Energy(1)

D. Lacroix et al., Progress in Part. and Nucl. Phys. 52 (2004) 497
Screening effects

Self-Energy$(2)$

\[ \Sigma_{\nu}(\omega) = \Delta_{\nu}(\omega) - i\frac{\Gamma_{\nu}(\omega)}{2}. \]

\[ \Delta_{\nu}(\omega) = \frac{\mathcal{P}}{2\pi} \int_{-\infty}^{\infty} d\omega' \frac{\frac{\Gamma_{\nu}(\omega')}{\omega - \omega'}}{\omega - \omega'} \]

We use the **bare** Coulomb interaction. Possible screening effects are introduced by performing some calculations by with a Yukawa type interaction

\[ V_0 \frac{e^{-\mu r}}{r} \]

However, large shifts still remain.