Collective excitations in metallic clusters within the second random phase approximation

D Gambacurta\textsuperscript{1,2} and F Catara\textsuperscript{2,3}

\textsuperscript{1}Consortium COMETA,
\textsuperscript{2}Dipartimento di Fisica e Astronomia dell'Università di Catania
\textsuperscript{3}Istituto Nazionale di Fisica Nucleare, Sezione di Catania, Via S.Sofia 64, I-95123 Catania, Italy
E-mail: Danilo.Gambacurta@ct.infn.it

Abstract. The second random-phase approximation (SRPA) is a natural extension of the RPA. It essentially consists in enlarging the space of the elementary modes introduced to describe the collective states by adding 2 particle - 2 hole excitations to the 1 particle - 1 hole ones of the RPA. However SRPA calculations are really few and almost all are done by resorting to strong approximations. We apply the SRPA to the study of collective states in metallic clusters. All kinds of couplings among all 1 particle - 1 hole and 2 particle - 2 hole configurations are taken into account. The coupling of 1 particle - 1 hole configurations with the 2 particle - 2 hole ones strongly pushes down the multipole strength distribution, especially for the dipole case, with respect to RPA. The inclusion of the coupling of the 2 particle - 2 hole among themselves enhances this behaviour also for the collective states whose description within RPA is widely accepted as satisfactory. Some possible origins of these unpleasant results are discussed. In particular, the need for a better treatment of short range correlations is underlined.

1. Introduction

Collective excitations are one of the most common and studied features of many-body systems. Of particular interest are the collective modes which can be interpreted in terms of vibrations. In nuclei such vibrations, both low-lying and high-lying (Giant Resonances, GR), have been known for a long time [1]. Metallic clusters show the dipole Plasmon resonance [2] which is the analogue of the nuclear dipole GR and is interpreted as due to the collective vibration of the electrons against the positive ions. The Random Phase Approximation (RPA) [3] has been extensively used as a microscopic theory to study the basic properties of these collective excitations. In both systems, RPA is able to describe the basic properties of these vibrations. However, some limitations are well known. On one hand, in the evaluation of the matrix elements appearing in the RPA equations of motion one has to resort to the Quasi Boson Approximation (QBA) introducing a clear loss of self-consistency and violations of the Pauli principle. Another limitation is that RPA predicts a perfectly harmonic spectrum with regularly spaced multiphonon states. Anharmonicities are a well established phenomenon in nuclei, both experimentally and theoretically [1, 4]. In metallic clusters no experimental evidence has been found until now for the existence of states corresponding to the double excitation of the dipole Plasmon. Theoretically such states have been predicted at energies deviating by about 10% from the double of the Plasmon energy [5].
A natural extension of RPA, aimed to study double excitations, is the so called Second RPA (SRPA) \([6, 7]\) where two particle-two hole \((2p-2h)\) excitations, in addition to the one particle-one hole \((1p-1h)\) ones, are introduced. The derivation of SRPA is also based on QBA and it has been argued \([8, 9]\) that in this case its use is even more problematic than in RPA. This point has been recently analyzed in \([10]\), where an approach to go beyond QBA was presented and applied to a 3 level Lipkin model. However, such analysis is limited by the fact that the model is very schematic and contains several parameters. Aim of the present paper is to analyze merits and limitations of SRPA for the study of the vibrational spectrum of a realistic many-body system. As a test laboratory we have chosen the case of metallic clusters, within the uniform jellium approximation \([2]\) with bare Coulomb interaction both for the electrons with the jellium and for the electrons among themselves. The reason for choosing such an admittedly simplified model is that it contains many characteristics of a generic realistic many body system and the interaction does not contain any adjustable parameter.

The Paper is organized as follows. In section 2 the SRPA framework is briefly presented. In section 3 we apply it to the study of the excitation spectrum of metallic clusters and a comparison with RPA results is carried out. Finally, in section 4, we draw the main conclusions.

2. Formalism

In SRPA, the excited states \(|\nu\rangle\) of the system are described as superpositions of \(1p-1h\) and \(2p-2h\) configurations built on top of the ground state \(|0\rangle\). One thus introduces the operators

\[
Q^\nu_1 = \sum_{pa} (X^\nu_{pa} a_p^\dagger a_i - Y^\nu_{pa} a_i^\dagger a_p) + \sum_{pqkl} (X^\nu_{pqkl} a_p a_i^\dagger a_m a_j - Y^\nu_{pqkl} a_i^\dagger a_p a_j a_m).
\]

such that

\[
Q^\nu_1 |0\rangle = |\nu\rangle, \quad Q^\nu_1 |0\rangle = 0.
\]

The \(X\)'s and \(Y\)'s are solutions of the equations

\[
\begin{pmatrix}
A & B \\
-B^* & -A^*
\end{pmatrix}
\begin{pmatrix}
X^\nu \\
Y^\nu
\end{pmatrix} = \omega^\nu
\begin{pmatrix}
X^\nu \\
Y^\nu
\end{pmatrix},
\]

where

\[
A = \begin{pmatrix}
A_{mi, pk} & A_{mi, pqkl} \\
A_{pqkl, mi} & A_{mnij, pqkl}
\end{pmatrix}, \quad B = \begin{pmatrix}
B_{mi, pk} & B_{mi, pqkl} \\
B_{pqkl, mi} & B_{mnij, pqkl}
\end{pmatrix}, \quad X^\nu = \begin{pmatrix}
X^\nu_{mi} \\
X^\nu_{mnij}
\end{pmatrix}, \quad Y^\nu = \begin{pmatrix}
Y^\nu_{mi} \\
Y^\nu_{mnij}
\end{pmatrix}.
\]

The elements \(A_{mi, pk}\) and \(B_{mi, pk}\) of \(A\) and \(B\) are the standard RPA matrices

\[
A_{pi,qj} = \langle HF | [a_i^\dagger a_p, [H, a_j^\dagger a_q]] | HF \rangle, \quad B_{pi,qj} = -\langle HF | [a_i^\dagger a_p, [H, a_j^\dagger a_q]] | HF \rangle.
\]

while the others are

\[
A_{mi,pqkl} = \langle HF | [a_i^\dagger a_m, [H, a_p^\dagger a_q a_k]] | HF \rangle, \quad A_{pqkl, mi} = A^*_{mi, pqkl},
\]

\[
A_{mnij,pqkl} = \langle HF | [a_i^\dagger a_j^\dagger a_m a_n, [H, a_p^\dagger a_q a_k]] | HF \rangle,
\]

\[
B_{mi,pqkl} = -\langle HF | [a_i^\dagger a_m, [H, a_k^\dagger a_p a_q]] | HF \rangle, \quad B_{pqkl, mi} = B^*_{mi, pqkl},
\]

\[
B_{mnij,pqkl} = -\langle HF | [a_i^\dagger a_j^\dagger a_m a_n, [H, a_k^\dagger a_p a_q]] | HF \rangle.
\]

Both in RPA and SRPA, since the exact ground state \(|0\rangle\) is not known, the HF ground state \(|HF\rangle\) is used in the evaluation of the matrix elements appearing in the equations of motion.
This replacement is also called Quasi Boson Approximation (QBA). As a consequence of the use of the $|HF\rangle$ state in the evaluation of the SRPA matrices we obtain, in particular

$$B_{mi,pqkl} = B_{pqkl,mi} = B_{mnij,pqkl} = 0. \quad (9)$$

The matrix (5) describes the coupling of $1p - 1h$ states to $2p - 2h$ states, while matrix (6) takes into account the coupling between $2p - 2h$ states themselves. The dimension of these matrices, especially of the latter, can be very large. If we neglect the residual interaction among the $2p - 2h$ states, the matrix (6) acquires a simple form,

$$A_{mnij,pqkl} = U(ij)U(mn)\delta_{ik}\delta_{jl}\delta_{mp}\delta_{nq}(\epsilon_m + \epsilon_n - \epsilon_i - \epsilon_j) \quad (10)$$

where $U(ij)$ is the antisymmetrizer for the indices $i, j$ and the $\epsilon$ quantities are the HF single particle energies. In this case, the SRPA problem can be reduced to an RPA eigenvalue problem, (whose dimensions are determined by the $1p - 1h$ space), but where the $A$ matrix (4) depends now on the excitation energies $\omega$ [7].

3. Results

In the following, we discuss some results obtained by applying the SRPA to the study of collective excitations in metallic clusters. The ionic background is described within the jellium approximation [2]. The reason for this choice is that the interaction of the electrons among themselves and with the ionic background is completely fixed as the bare Coulomb interaction and the model does not contain any adjustable parameter. This allows to compare in a clear way different degrees of approximation.

As a first step, we have fixed, by solving the HF equations, the single particle basis in which all the subsequent calculations are carried out. After that, we have solved the RPA and SRPA equations for natural parity states, with multipolarities ranging from $L = 0$ to $L = 3$. The single particle space has been truncated so that the Thouless theorem, and thus the preservation of EWSR, is satisfied in the RPA calculations better than 1 % for all the multipolarities. We focus our attention only on spin $S = 0$ states. However we remark that, for what concerns the SRPA calculations, all the $1p - 1h$ states with $L = 0 - 3$, both spin $S = 0$ and 1, are considered in the construction of the $2p - 2h$ configurations with good total spin and angular momentum.

We show the results for two sodium clusters, namely the $Na_9^+$ and $Na_{11}^+$. In the latter case an energy cutoff $E_{cut}$ has been used and only the $2p - 2h$ configurations with unperturbed energy lower than $E_{cut} = 15eV$ are included. In order to check that the configuration space is large enough, we have verified that the EWSR are numerically satisfied, (better than 1 % in the present calculations), for all the considered multipolarities. In figure 1 we plot the strength distributions for the multipole operator $F^{(\lambda)}(r) = r^{\lambda}Y_{00}$ for the $Na_{9}^+$ metallic cluster. In order to make clearer the comparison, the discrete lines of RPA and SRPA spectra are folded with a Lorentzian function. In both cases an artificial width $\Gamma = 0.1eV$ has been used. In the upper and lower panel of figure 1, we show, respectively, the $L = 0, 1$ and $L = 2, 3$ multipole strength distributions with spin $S = 0$ for $Na_{9}^+$ metallic cluster. Solid (red) lines refer to SRPA results, while the dashed (black) ones refer to those obtained within RPA. With the dotted (blue) lines we indicate the SRPA results when the diagonal approximation (10) is used. In the following figures we use the label “SRPA-A22D” for the results obtained when this diagonal approximation is used. We see that going from RPA to SRPA the strength distributions are shifted to lower energies, especially in the dipole case. The results shown in figure 1 refer to the lower part of the excitation spectrum, which is mainly composed by $1p - 1h$ configurations. However, even for such states the inclusion of the coupling among $2p - 2h$ configurations leads to a further shift down. Let us look in some detail to the collective dipole $S = 0$ state, which in RPA is located at 2.98 eV, and that experimentally is found at about 2.60 eV [11]. Within the approximated
Figure 1. (Color online) Natural parity \( L = 0,1 \) (upper panel) and \( L = 2,3 \) (lower panel) spin\(=0 \) multipole strength distributions for \( Na_9^+ \) metallic cluster are shown. Solid (red) lines refer to SRPA results, while the dashed (black) ones refer to RPA calculations. SRPA-A22D dotted (blue) lines show the SRPA results when the approximation (10) is used. The arrow roughly indicates the positions of the experimental dipole plasmon peak.

SRPA, that is when only the coupling of \( 1p - 1h \) to \( 2p - 2h \) configurations is taken into account, the energy moves to 1.88 eV, while in the full SRPA one gets 1.47 eV. As shown in figure 2, qualitatively similar results have been found for \( Na_{21}^+ \). Also in this case we have a strong shift down, especially for the dipole excitations, the plasmon energy being lowered from the RPA value of 2.90 eV to the full SRPA result of 1.00 eV very far from the experimental peak at about 2.65 eV [11]. Also in the nuclear [12] case one finds that the SRPA excitations spectrum is very much different from the RPA one, even for excitations like the Giant Resonances whose description within RPA is widely accepted as satisfactory. Probably in all cases, the problems arise from the fact that the self-energy-like terms present in SRPA take contributions from very high energy "incoherent" particle \( - \) hole configurations. In addition to that, we stress again that the derivation of the SRPA equations of motion is based on QBA and this might be more doubtful than in RPA. Indeed, a better treatment of ground state correlations has been found to improve the results, within a 3 level Lipkin model [10]. The replacement of the correlated ground state with the HF one is justified only if ground state correlations are not too strong. In RPA the \( Y_{\nu ph}^\mu \) amplitudes are a measure of these correlations [3] and thus, looking at their behaviour going from RPA to SRPA could be useful in order to have more information about the adequacy of QBA. In table 1 we report, in the case of \( Na_9^+ \) and for different multipolarities, the sum of the squares of the \( Y_{\nu ph}^\mu \) amplitudes in RPA (first column), in SRPA (third column) and in SRPA when the diagonal approximation (10) is used (second column). We see that going from RPA to SRPA an increase of these quantities is observed and the inclusion of the residual interaction coupling the \( 2p - 2h \) configurations among themselves amplifies this effect. Similar results have been found in the case of \( Na_{21}^+ \).
Figure 2. (Color online) As in figure 1 but for Na_{21}^+ metallic cluster.

Table 1. The sum of the squares of the $Y_{\nu ph}$ amplitudes in the case of Na_{0}^+ and for different multipolarities, in RPA (first column), in SRPA (third column) and in SRPA when the diagonal approximation (10) is used (second column).

| $\sum_{p,h,\nu} |Y_{\nu ph}^\nu|^2$ | RPA     | SRPA-A22D | SRPA     |
|-------------------------------|---------|------------|----------|
| L=0, S=0                      | 0.0458  | 0.0512     | 0.0532   |
| L=1, S=0                      | 0.1601  | 0.2962     | 0.4170   |
| L=2, S=0                      | 0.0256  | 0.0316     | 0.0347   |
| L=3, S=0                      | 0.0198  | 0.0282     | 0.0388   |

The RPA occupation numbers can be calculated by using, for example, the number operator method [13] and one gets:

$$n_h = 1 - \frac{1}{2} \sum_{p,\nu} |Y_{\nu ph}^\nu|^2, \quad n_p = \frac{1}{2} \sum_{h,\nu} |Y_{\nu ph}^\nu|^2.$$  \hspace{1cm} (11)

To our knowledge, the explicit form of the SRPA occupation numbers. Nevertheless, it could be instructive to compare the values of quantities (11) when the $Y_{\nu ph}^\nu$ obtained in RPA and SRPA are used. In this sense we will refer in the following to SRPA occupation numbers. In figure 3 we show, in the case of the Na_{21}^+ metallic cluster, the occupation numbers $n_p$ for particle states and the opposite of the depletion numbers $n_h - 1$ for hole states obtained in SRPA and in RPA,
in the lower and upper panel, respectively. We note that the deviations from the HF limit, i.e. $n_h = 1$ and $n_p = 0$, are greater in SRPA than in RPA. Since, as mentioned above, the use of QBA is justified only when the HF state does not differ very much from the correlated one, the larger deviations found in SRPA suggest that this approximation could be more severe than in RPA.

As discussed at end of section 2, the SRPA problem can be reduced to an energy-dependent RPA problem. In this case, the new RPA $A$ matrix is

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

where $A$ is the usual RPA matrix, $\Sigma$ is the energy dependent term

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

and the indices 1 and 2 are a short-hand notation for the $1p - 1h$ and $2p - 2h$ configurations, respectively. The energy dependent term is connected to the self-energy of the $1p - 1h$ excitation and it is due to the coupling with the $2p - 2h$ configurations. It has been shown that its real part gives a shift of the RPA resonance energies while the imaginary part takes into account spreading width effects [14, 15]. Several SRPA calculations have been done by neglecting the real part of the particle-hole self-energy (see for example [7, 16]) and considering only the spreading width. So it is interesting to study the energy shift due to the real part of the self energy in this case. In order to have a quantitative, though approximate, evaluation of this effect we consider the self-energy acquired by a RPA phonon as a consequence of the coupling with the $2p - 2h$ configurations [15]

$$\Sigma^{RPA}(\omega_{\nu}) = \sum_{2p2h} |<\nu|V|2p2h>|^2$$

$$\omega_{\nu} - \epsilon_{2p2h} + i\eta$$

Figure 3. Occupation numbers $n_p$ for particle states and the opposite of depletion numbers $n_h - 1$ for hole states for $Na^{2}_{21}$ metallic cluster. RPA and SRPA results are reported in the upper and lower panel, respectively. Only Spin S=0 states are included in the calculations. In the abscissa the single-particle energies in eV are indicated.
where $\omega_\nu$ is the excitation energy of the RPA phonon $|\nu>$, $V$ is the residual interaction and $\epsilon_{2p2h}$ is the unperturbed energy of the $2p-2h$ configuration $|2p2h> = a_{p2}^\dagger a_{h2} a_{p1}^\dagger a_{h1} | HF >$. We have calculated the real part of this quantity for the dipole plasmon in the $Na^9$ case. The $\eta$ parameter used in the calculation is 0.2 eV. As mentioned above, the RPA dipole plasmon $\omega_\nu$ is located at 2.98 eV. Since the unperturbed $2p-2h$ energies $\epsilon_{2p2h}$ are larger than this value, the denominator in Eq. (14) is always negative and thus the resonance peak is shifted downwards with respect to $\omega_\nu$. In the upper panel of figure 4 we plot the absolute value of the real part of the $k$ and $K$ quantities defined in Eqs. (15) and (16), respectively. The abscissa of both the figures shows the unperturbed energies of the $2p-2h$ configurations.

In order to put in evidence in a clearer way this fact, we have divided the unperturbed $2p-2h$ energy range in bins of 0.5 eV and we have summed all the terms defined in Eq. (15) lying in each interval identified by the index I. In the lower panel of figure 4, where we plot the the absolute value of the real part of the quantities

$$k(\epsilon_{2p2h}) = \frac{|\langle \nu | V | 2p2h >|^2}{\omega_\nu - \epsilon_{2p2h} + i\eta}$$

i.e. the contribution to the self-energy of each $2p-2h$ configuration identified by its unperturbed energy $\epsilon_{2p2h}$. We see that, although the largest contributions are given by the lower $2p-2h$ configurations, also the ones with higher energy contribute and the overall effect is quite large. In order to put in evidence in a clearer way this fact, we have divided the unperturbed $2p-2h$ energy range in bins of 0.5 eV and we have summed all the terms defined in Eq. (15) lying in each interval identified by the index I. In the lower panel of figure 4, where we plot the the absolute value of the real part of the quantities...
\[ K(I) = \sum_{\epsilon_{2p2h} \in I} k(\epsilon_{2p2h}) \]  

we see that the total contributions of high energy configurations, arising from a coherent sum of many small terms, become comparable with the ones of the lowest energies. Similar results are obtained in the case of Na\textsuperscript{+} \textsubscript{21}. By the above shown analysis of the \( Y \) amplitudes and of the occupation numbers, we can conclude that a better treatment of these correlations should be necessary in SRPA.

4. Conclusions

In the present paper we have applied the SRPA to the study of collective excitations in metallic clusters. We have found strong modifications of the RPA excitation spectrum, the energy being pushed down. When the coupling of 2\( p \) – 2\( h \) configurations among themselves is included, a further lowering is obtained even for those collective excitations whose description within RPA is widely assumed as satisfactory. Such strong modifications could be traced back to the use of QBA in calculating the expectation values appearing in the equations of motion of SRPA, although the same approximation is used in RPA, it seems more severe in SRPA.

These results suggest that a better treatment of ground state correlations along the path of extended RPA and SRPA approaches, as done for example in [10, 17], is necessary. Work in this direction is in progress. It is also interesting to analyze how much such strong modifications with respect to RPA description depend on the studied system. With this respect, in the near future, we plan to apply SRPA in nuclear systems by using effective Skyrme-type interactions.

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References

[1] Bohr A and Mottelson B R 1975 Nuclear Structure vol II (Benjamin: New York)
[2] Brack M 1993 Rev. Mod. Phys. 65 677
[3] Ring P and Schuck P 1980 The Nuclear Many-Body Problem (Springer-Verlag: Berlin)
[4] Chomaz P and Frascaria N 1995 Phys. Rep. 252 275
[5] Gerchikov L G et al. 2002 Phys. Rev. A 66 053202; Catara F et al. Phys. Lett. 2006 A 349 345
[6] Yannouleas C 1987 Phys. Rev. C 35 1159
[7] Drozdz G, Nishizaki S, Speth J and Wambach J 1990 Phys. Rep. 197 1
[8] Takayanagi K, Shimizu K and Arima A 1988 Nucl. Phys. A 477 205
[9] Lauritsch G and Reinhard P G 1990 Nucl. Phys. A 509 287
[10] Gambacurta D, Grasso M, Catara F and Sambataro M 2006 Phys. Rev. C 73 024319
[11] Borggreen J et al. 1993 Phys. Rev. B 48 17507
[12] Papakonstantinou P and Roth R Preprint arXiv:0805.4086 [nucl-th]
[13] Rowe D J 1968 Phys. Rev. 175 1283
[14] Wambach J 1988 Rep. Prog. Phys. 51 989
[15] Lacroix D, Ayik S and Chomaz P 2004 Prog. Part. Nucl. Phys. 52 497
[16] Ait-Tahar S and Brink D M 1993 Nucl. Phys. A 560 765
[17] Gambacurta D and Catara F 2008 Phys. Rev. B 77 205434 and 2009 Phys. Rev. B 79, 085403