Extended second random phase approximation applied to metallic clusters

D. Gambacurta[1,2,*] and F. Catara[1,2,†]
[1]*Dipartimento di Fisica e Astronomia dell'Universitá di Catania, Via S.Sofia 64, I-95123 Catania, Italy*
[2]*Istituto Nazionale di Fisica Nucleare, Sezione di Catania, Via S.Sofia 64, I-95123 Catania, Italy*
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The coupling to two particle-two hole configurations is at the basis of the calculations of the spreading width of the collective excitations that in random phase approximation (RPA) are described as superpositions of one particle-one hole elementary modes. Second RPA (SRPA) is just the extension of RPA including both kinds of configurations. In SRPA use is made of the quasiboson approximation (QBA) as in RPA. It has been found that this is at the origin of the fact that the coupling among them strongly lowers the RPA collective excitations, both in metallic clusters and in nuclei. When the coupling of the two particle-two hole configurations among themselves is also included, as it has to be done in order to be consistent, this effect is even enhanced. Thus the reasonably good description of the collective vibrational states obtained at the RPA level (as expected on physical grounds, for example, for the dipole modes) is completely spoiled in SRPA. In the present paper we show quantitatively, for metallic clusters, that this disturbing behavior is eliminated when no use is made of QBA and a better description of ground-state correlations is introduced.

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I. INTRODUCTION

The Hartree-Fock (HF) plus random phase approximation (RPA) (Refs. 1 and 2) is a largely used microscopic approach to study the properties of collective modes in many-body systems. The HF method allows us to obtain the self-consistent mean field in which the particles are assumed to move independently, their energies and wave functions, and thus the HF ground state. The solution of the RPA equations gives the energies and the wave functions of the collective excitations that are assumed to be superpositions of particle-hole and hole-particle configurations built on top of a correlated ground state whose explicit expression is unknown. It is well known that, on one hand, this correlated ground state is used in the formal derivation of the equations of the motion while, on the other hand, the HF state is used instead in calculating the expectation values appearing in these equations. This inconsistency is also known as quasiboson approximation (QBA) and it is the main limit of RPA.[2]

In nuclei vibrational states, both low lying and high lying [giant resonances, GR (Ref. 3)] are quite well described within RPA. The same is true in metallic clusters that show the dipole plasmon resonance[4,5] which is the analog of the nuclear dipole GR and is interpreted as due to the collective vibration of the electrons against the positive ions. However, some RPA limitations are well known. On one hand, RPA predicts a perfectly harmonic spectrum with regularly spaced multiphonon states. Anharmonicities are a well-established phenomenon in nuclei, both experimentally and theoretically.[3,6–11] 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.[12,13]

On the other hand, in several works[6,10,14,15] it has been underlined that the coupling of one particle-one hole configurations with more complex states has to be taken into account in order to describe some important effects, as, for example, the spreading width of the excitations. In such a direction, a natural extension of RPA is the so-called second RPA (SRPA) (Refs. 16 and 17) 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[18–20] that in this case its use is even more problematic than in RPA. This point has been analyzed in Ref. 21 within a schematic model and more recently in metallic clusters[22] where a complete SRPA scheme, including all kinds of coupling within the $1p$-$1h$ and $2p$-$2h$ configurations, is used. It has been found that within SRPA one gets very strong modifications of the RPA excitation spectrum. The coupling of $1p$-$1h$ configurations with the $2p$-$2h$ ones strongly pushes down the multipole strength distribution, also for the collective states whose description within RPA is rather good, as, for example, the dipole plasmon. A similar feature has been found also in nuclei.[23]

Some possible origins of these results have been discussed in the literature.[17–22] In particular, two aspects related to the need for a better treatment of short-range correlations have been discussed in a recent work.[22] On one hand the above cited use of QBA in calculating the expectation values appearing in the equations of motion of SRPA. Furthermore, the $2p$-$2h$ configurations included in SRPA introduce self-energy corrections[24–27] and their contributions are quite large when the uncorrelated HF reference state is used in calculating the SRPA matrices. Indeed, in Ref. 22 it has been shown in a qualitative, approximated way that they are appreciably reduced when a better description of ground-state correlations is used. In the present paper we confirm quantitatively this result by means of more complete calculations and show that this unpleasant behavior, which would spoil the quality of the RPA description of the collective plasmon excitation, is eliminated.

Our main concern here is on the search for a suitable approach to cure the above-mentioned limitations and to show how it works when applied to the study of a realistic many-body system. Therefore we have chosen the case of

metallic clusters within the uniform jellium approximation, thus neglecting, for the time being, the improvements over it that can be found in the literature.[28] We believe that the relatively simple scheme we have considered is representative of a realistic many-body system and is certainly a good laboratory for our analysis. On the other hand, our main purpose is not to make a realistic study of a metallic cluster but rather to compare different levels of approximations within the same model. We have also decided to make no use of an effective interaction, for example, derived from density energy functional, in order to avoid any possible source of double countings due to the use of the correlation terms in the energy functional. The interaction of the electrons among themselves and with the jellium is just the bare Coulomb one.[29]

The paper is organized as follows. In Sec. II we discuss the main motivations of the present work and the link and the differences with previous works are analyzed. In Sec. III the SRPA framework is reviewed and the extended SRPA (ESRPA) approach is presented and discussed. In Sec. IV we apply it to the study of the excitation spectrum of Na metallic clusters and a comparison with RPA and SRPA results is carried out. Finally, in Sec. V, we draw the main conclusions.

II. MOTIVATIONS AND LINK WITH PREVIOUS WORKS

SRPA studies in realistic systems are indeed really few. The number of $2p$-$2h$ configurations is usually very large making the calculations extremely heavy, and sometimes prohibitive, from a computational point of view. Current versions of SRPA are based on noninteracting $2p$-$2h$ configurations and only the interactions between $2p$-$2h$ and $1p$-$1h$ are explicitly taken into account. Such approximation allows us to reduce the SRPA to an energy-dependent RPA problem, overcoming thus the problem of the large dimensions of the $2p$-$2h$ configurations. The energy-dependent term is connected to the self-energy of the $1p$-$1h$ excitations and to their coupling with the $2p$-$2h$ configurations. It has been shown that the real part of this self-energy gives a shift of the RPA resonance energies while the imaginary part takes into account spreading width effects.[24–26] Several SRPA calculations have been done by neglecting the real part of the particle-hole self-energy (see, for example, Refs. 17 and 27) and considering only the spreading width. The justification of this choice is based on the fact that, when effective interactions are used, self-energy contributions are already taken into account in the single-particle energies. Thus, in order to avoid double counting, the real part of the particle-hole self-energy is omitted. However, since the real and imaginary parts of the self-energy obey a dispersion relation,[25] a consistent treatment of both contributions should be necessary. In Ref. 22, the real part of the self-energy acquired by the RPA collective dipole plasmon through its coupling to $2p$-$2h$ configurations has been calculated in two different cases: when the HF state is used as reference state and when ground-state correlations are included perturbatively. In the latter case an appreciable reduction in the energy shift has been found. This result suggests that the use of a correlated ground state in SRPA, overcoming thus the QBA, could be important. Indeed some SRPA calculations beyond QBA have been done[17,19,20] but including ground-state correlations only in the $1p$-$1h$ channel, i.e., in the RPA matrices. On the contrary, we believe that the effect of ground-state correlations is particularly important in calculating the coupling of $1p$-$1h$ and $2p$-$2h$ configurations. For this reason we carry out SRPA calculations including all kinds of coupling within the $1p$-$1h$ and $2p$-$2h$ configurations and where ground-state correlations are taken into account in all the SRPA block matrices. The effect of these correlations are studied in the case of metallic clusters, within the uniform jellium approximation[4,30] with bare Coulomb interaction both for the electrons with the jellium and for the electrons among themselves.[29]

III. FORMALISM

The excited stated $|\nu\rangle$ of a system can be described by means of a set of operators $Q_\nu^\dagger$ whose action on the ground state $|0\rangle$ is defined by

$$Q_\nu^\dagger|0\rangle = |\nu\rangle, \tag{1}$$

$$Q_\nu|0\rangle = 0. \tag{2}$$

It can be shown[2] that the following equations hold for an arbitrary operator δQ:

$$\langle 0|[\delta Q,[H,Q_\nu^\dagger]]|0\rangle = \omega_\nu \langle 0|[\delta Q,Q_\nu^\dagger]|0\rangle, \tag{3}$$

where ω_ν is the excitation energy of the $|\nu\rangle$ state.

Let $|\mathrm{HF}\rangle$ be the HF ground state of the system where the hole states below the Fermi energy are filled and the particle states above are empty. In the following, we use the indices m,n,p,q and i,j,k,l to indicate particle and hole states, respectively.

In the SRPA the $Q_\nu^\dagger$ operators have the following expression:

$$Q_\nu^\dagger = \sum_{pi} (X_{pi}^\nu a_p^\dagger a_i - Y_{pi}^\nu a_i^\dagger a_p) + \sum_{p<m,i<j} (X_{pimj}^\nu a_p^\dagger a_i a_m^\dagger a_j - Y_{pimj}^\nu a_i^\dagger a_p a_j^\dagger a_m) \tag{4}$$

containing thus, besides the usual $1p$-$1h$ excitations considered in RPA, also the $2p$-$2h$ ones.

The X''s and Y''s are solutions of the equations

$$\begin{pmatrix} \mathcal{A} & \mathcal{B} \\ -\mathcal{B}^* & -\mathcal{A}^* \end{pmatrix} \begin{pmatrix} \mathcal{X}^\nu \\ \mathcal{Y}^\nu \end{pmatrix} = \omega_\nu \begin{pmatrix} \mathcal{X}^\nu \\ \mathcal{Y}^\nu \end{pmatrix}, \tag{5}$$

where

$$\mathcal{A} = \begin{pmatrix} A_{mi,pk} & A_{mi,pqkl} \\ A_{pqkl,mi} & A_{mnij,pqkl} \end{pmatrix}, \quad \mathcal{B} = \begin{pmatrix} B_{mi,pk} & B_{mi,pqkl} \\ B_{pqkl,mi} & B_{mnij,pqkl} \end{pmatrix}$$

and

$$\mathcal{X}^\nu = \begin{pmatrix} X_{mi}^\nu \\ X_{minj}^\nu \end{pmatrix}, \quad \mathcal{Y}^\nu = \begin{pmatrix} Y_{mi}^\nu \\ Y_{minj}^\nu \end{pmatrix}.$$

The explicit form of the matrix blocks is given by

$$A_{pi,qj} = \langle 0|[a_i^\dagger a_p,[H,a_q^\dagger a_j]]|0\rangle, \tag{6}$$

$$A_{mi,pqkl} = \langle 0|[a_i^\dagger a_m,[H,a_p^\dagger a_q^\dagger a_l a_k]]|0\rangle, \tag{7}$$

$$A_{mnij,pqkl} = \langle 0|[a_i^\dagger a_j^\dagger a_n a_m,[H,a_p^\dagger a_q^\dagger a_l a_k]]|0\rangle, \tag{8}$$

The B matrix elements are easily obtained from the corresponding A elements, by substituting the first operator in double commutator with its adjoint and by changing the sign. For example, we have that

$$B_{pi,qj} = -\langle 0|[a_p^\dagger a_i,[H,a_q^\dagger a_j]]|0\rangle. \tag{9}$$

The matrix [Eq. (7)] describes the coupling of $1p$-$1h$ states to $2p$-$2h$ states while matrix [Eq. (8)] takes into account the coupling between $2p$-$2h$ states themselves. The dimension of these matrices, especially of the latter, can be very large. Standard SRPA equations are obtained by resorting to QBA, namely, by replacing in the evaluation of the previous matrix elements the state $|0\rangle$ with the HF ground state $|\mathrm{HF}\rangle$.

In the present work we present an ESRPA approach where no use of the QBA is made and the correlated state is used in place of the HF one. First of all, the double commutators appearing in Eqs. (6)–(8) and in the corresponding B matrices are calculated without any kind of approximation. After that, the two-body or higher terms coming out from the double commutators are contracted with respect to the reference state $|0\rangle$. In such a way, the A and B matrices are expressed in terms of the one-body density matrix (OBDM)

$$\rho(\alpha,\beta) \equiv \langle 0|a_\alpha^\dagger a_\beta|0\rangle, \tag{10}$$

which we assume to be diagonal

$$\rho(\alpha,\beta) = \delta_{\alpha\beta} n_\alpha, \tag{11}$$

where n_α is the occupation number of the α single-particle state. The explicit expression of the matrices used in the calculations is given in Appendix. For example, we have that the matrix [Eq. (6)] acquires the following expression:

$$A_{pi,qj} = \delta_{ij}\delta_{pq}(\epsilon_p - \epsilon_i)(n_i - n_p) + \bar{V}_{pjiq}(n_i - n_p)(n_j - n_q), \tag{12}$$

where the ϵ quantities are the HF single-particle energies and $\bar{V}$ indicates the antisymmetrized residual interaction. Similar expressions hold for the other matrix elements.

Standard SRPA equations can be derived by using the above described procedure and the HF state as reference state. This amounts to use as occupation numbers the HF ones, i.e., 0 and 1 for particle and hole states, respectively. For example, we note that, in this limit, the expression in Eq. (12) becomes the one of the standard RPA A matrix.

The final step of our procedure deals with the evaluation of the occupation numbers appearing in the ESRPA matrices. In this work we present the results by using two different choices.

In the first case we use as correlated ground state

$$|0\rangle = \mathcal{N}\left(1 + \frac{1}{2}\sum_{minj} C_{minj} a_m^\dagger a_i a_n^\dagger a_j\right)|\mathrm{HF}\rangle, \tag{13}$$

where $\mathcal{N}$ is a normalization factor and the C coefficients are evaluated in first-order Rayleigh-Schrödinger perturbation theory, i.e.,

$$C_{minj} = \frac{\langle \mathrm{HF}|\hat{V} a_m^\dagger a_i a_n^\dagger a_j|\mathrm{HF}\rangle}{E_{minj}}, \tag{14}$$

E_{minj} being the unperturbed energies of the $2p$-$2h$ configuration.

Although this choice allows us to go over the QBA, this procedure is not really self-consistent since the state [Eq. (13)] does not satisfy the vacuum condition [Eq. (2)]. In order to go one step further in this direction we proceed as follows. In Ref. 31, an extension of RPA (ERPA) based on a more consistent treatment of correlations was presented and applied to metallic clusters. By means of the method of linearization of equations of motion, a set of RPA-like equations was derived, depending only on the OBDM that is expressed in terms of the ERPA X and Y amplitudes by using the number operator method.[32] This set of nonlinear equations is solved self-consistently via an iterative procedure which allows to calculate the energies and the wave functions of the excited states together with the occupation numbers entering in the equations of motion. The extension of SRPA we propose is based on a similar procedure but using the ERPA ground state as reference state. Therefore, we first solve the ERPA procedure and then use the so obtained occupation numbers in the ESRPA equations. Of course, the best would be the use of the vacuum of the SRPA operators [Eq. (4)] as reference state. However, SRPA calculations are much more time expensive than the RPA ones and thus an iterative procedure involving SRPA matrices is not easily affordable.

IV. RESULTS

Let us now show the results for two sodium clusters, namely, the Na_{21}^+ and Na_{20}, within the RPA, SRPA, and the ESRPA. The Hamiltonian of the system is

$$H = \sum_i h_i + \sum_{i<j} v_{ij} \tag{15}$$

with

$$h_i = -\frac{\hbar^2}{2m}\nabla_i^2 + V(r_i); \quad v_{ij} = \frac{e^2}{4\pi}\frac{1}{|\vec{r}_i - \vec{r}_j|} \tag{16}$$

and

$$V(r) = \frac{Ne^2}{4\pi}\begin{cases} (1/2r_c)(r^2/r_c^2 - 3) & \text{for } r \le r_c \\ -1/r & \text{for } r \ge r_c, \end{cases} \tag{17}$$

where r_c is the radius of the jellium sphere, i.e., $r_c = r_s N^{1/3}$, r_s being the Wigner-Seitz radius and N the number of ions.

The single-particle basis in which all the subsequent calculations are carried out is fixed by solving the HF equations. The single-particle wave functions have been represented as

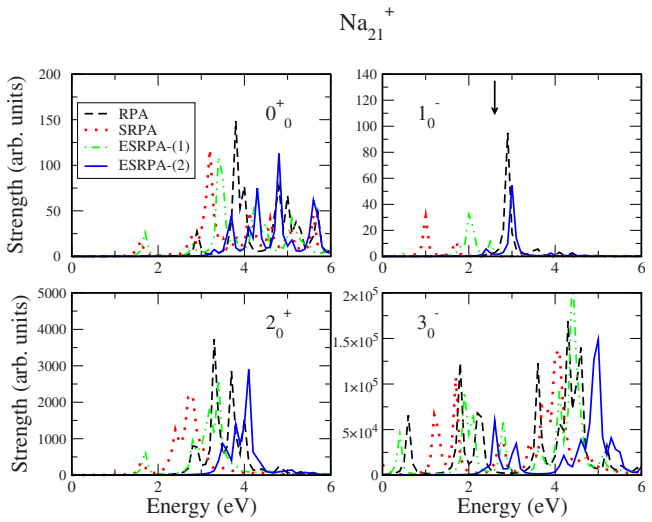

FIG. 1. (Color online) Natural parity $L=0$, 1 (upper panel) and $L=2$, 3 (lower panel) spin=0 multipole strength distributions for Na_{21}^{+} metallic cluster are shown. Dashed (black) lines refer to RPA calculations while the dotted (red) ones refer to SRPA results. The dot-dashed (green) lines and the solid (blue) lines show the ESRPA results where g.s. correlations are calculated perturbatively or within the ERPA procedure, respectively (see the text for more detail). The arrow roughly indicates the positions of the experimental dipole plasmon peak.

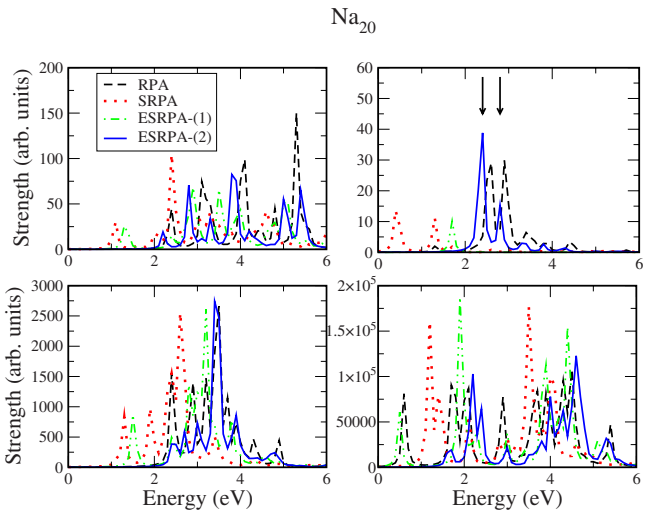

FIG. 2. (Color online) As in Fig. 1 but for the Na_{20} metallic cluster.

linear superposition of harmonic-oscillator ones, with principal quantum number ranging from $n=0$ to 10 for each orbital angular momentum l. The harmonic-oscillator parameter has been fixed in order to minimize the HF ground-state energy. The single-particle space in which we carry out our calculations has been truncated so that the Thouless theorem on the energy weighted sum rule[2] (EWSR) is satisfied in the RPA calculations better than 1% for all the multipolarities considered. We will focus our attention only on natural-parity states with multipolarity $L=0-3$ and spin $S=0$. In SRPA and ESRPA calculations we have included all the $2p$-$2h$ configurations with unperturbed energy lower than $E_{cut}=18$ eV. An accurate study of the stability of the results, in particular, of the strength distributions, by increasing E_{cut} has been done (see also Ref. 22).

In Fig. 1 we plot, for the Na_{21}^{+} metallic cluster, the strength distributions for the multipole operator

$$F^{(\lambda)}(r) = \begin{cases} r^{\lambda} Y_{\lambda 0} & \text{for } \lambda > 0 \\ r^{2} Y_{\lambda 0} & \text{for } \lambda = 0. \end{cases} \tag{18}$$

In order to make clearer the comparison, the discrete lines of RPA, SRPA, and ESRPA spectra are folded with a Lorentzian function. In all cases an artificial width $\Gamma=0.1$ eV has been used. In the upper and lower panels of the figure, we show, respectively, the $L=0$, 1 and $L=2$, 3 multipole strength distributions with spin $S=0$. Dashed (black) lines refer to RPA calculations while the dotted (red) ones refer to SRPA results. The dot-dashed (green) lines and the solid (blue) lines show the ESRPA results where g.s. correlations are calculated perturbatively [labeled by "ESRPA-(1)"] or within the above discussed ERPA procedure [labeled by "ESRPA-(2)"], respectively.

We see that going from RPA to SRPA the strength distributions are strongly shifted to lower energies and, at the same time, the height of the main peaks is reduced. In particular, we see that, in the dipole case, the plasmon energy is 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.[33] Moreover, very striking is the fact that the fraction of EWSR exhausted by this state goes from 79% to 10% while the total one (0.54733×10^{2} Å^2 eV) is very close to the RPA value (0.54167×10^{2} Å^2 eV), thus indicating a very strong fragmentation of the strength in SRPA. As far as the shift is concerned, we recall that no effective interaction is used in our calculations. Therefore, possible double countings induced by considering all couplings among all elementary configurations are not present.

Let us now examine the effect of the ground-state correlations in the ESRPA. When they are calculated perturbatively (dot-dashed green lines in the figure), we observe that the strength distributions are pushed toward higher energy, going close to the RPA results. However, in the dipole case, the strength distribution is still quite different from the RPA one and from the experimental peak whose position is roughly indicated by the arrow in the figure. Moreover, the height of the peak is practically unchanged from the SRPA case. When instead the ground-state correlations are taken into account by means of the ERPA procedure (solid blue lines), we see that the position of the peak is roughly the same as in RPA which is very close to the experimental value. The height of the peak is greater than the one obtained in SRPA and in the perturbatively ESRPA but still smaller than the RPA one. This is however related to the fragmentation of the strength due to the coupling of the $1p$-$1h$ configurations with the $2p$-$2h$ ones.

As shown in Fig. 2, qualitatively similar results have been found for Na_{20}. Also in this case we have a strong shift down going from the RPA to the SRPA, especially in the dipole case, where the two RPA peaks at about 2.5 and 3.0 eV are

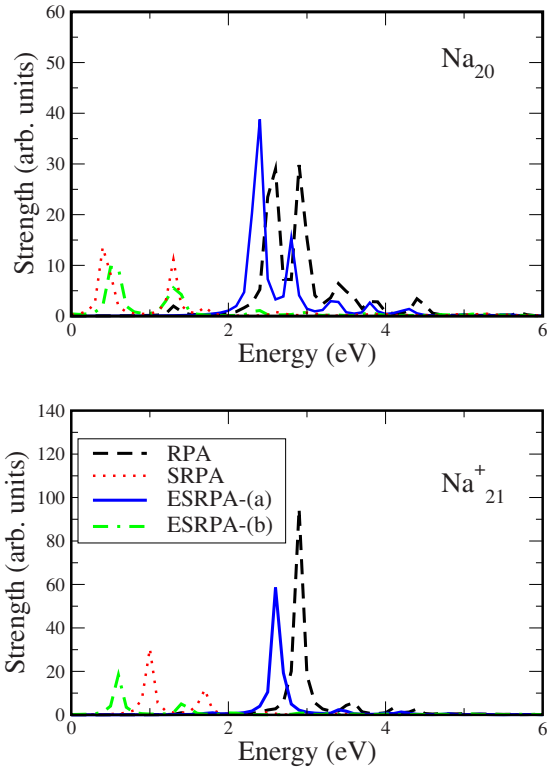

FIG. 3. (Color online) Comparison of the dipole strength distributions for the Na_{20} (upper panel) and Na_{21}^{+} (lower panel) obtained within different approximations: (black) dashed and red dotted lines refer to RPA and SRPA, respectively; (blue) solid lines [ESRPA-(a)] refer to the full ESRPA while the (green) dot-dashed ones [ESRPA-(b)] refer to an approximate ESRPA scheme in which ground-state correlations are taken into account only in the $1p$-$1h$ channel, i.e., in the RPA matrices. In both the ESRPA calculations the occupation numbers obtained by means of the ERPA procedure are used.

strongly pushed down in SRPA to 0.5 and 1.2 eV, respectively. In the dipole case the improvement within the ESRPA when ground-state correlations are calculated perturbatively is really poor while it is much better when the ERPA procedure is used. In particular, the two peaks are at 2.4 and 2.8 eV, very close to the RPA ones and to the experimental values (the arrows in the figure).

As discussed in Sec. II some SRPA calculations beyond QBA have been done[17,19,20] but including ground-state correlations only in the $1p$-$1h$ channel, i.e., in the RPA matrices. In Fig. 3 we compare, in the dipole case, this approximate ESRPA scheme, whose results are indicated in the figure with the label "ESRPA-(b)," with the complete calculation, i.e., when ground-state correlations are taken into account in the evaluation of all the SRPA matrices ["ESRPA-(a)"]. In both cases the occupation numbers obtained by means of the ERPA procedure are used. Both for the Na_{20} and Na_{21}^{+}, shown in the upper and lower panels of the figure, respectively, we see that the "ESRPA-(b)" results (green dot-dashed lines) are still very far from the RPA ones and are not very different from the ones obtained in standard SRPA (red dotted lines). These results clearly show that the effect of ground-state correlations is particularly important in calculating the coupling of $1p$-$1h$ and $2p$-$2h$ configurations and, at least in the metallic clusters, the approximate ESRPA is not adequate.

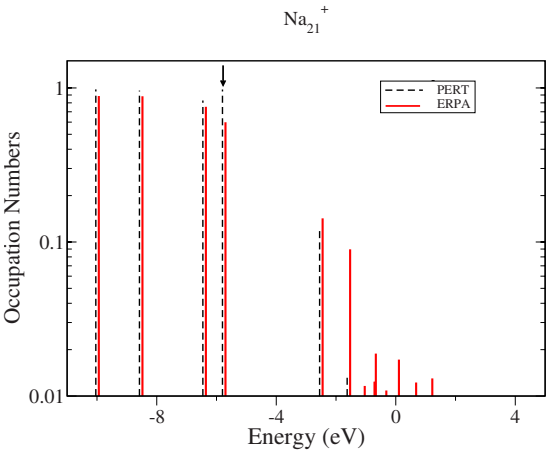

FIG. 4. (Color online) Occupation numbers for the Na_{21}^{+} metallic clusters calculated perturbatively (black) dashed lines and by using the ERPA procedure (red) solid lines. In the abscissa the HF single-particle energies are reported. The arrow indicates the last HF occupied orbital.

The differences between the two ESRPA calculations are related to the different description of the correlated ground state and, in particular, to the occupation numbers entering in the equations of motion. In Fig. 4, we show for the Na_{21}^{+} metallic cluster, the occupation numbers obtained when ground-state correlations are calculated perturbatively [Eq. (13)] or within the ERPA procedure, in dashed black lines and solid red lines, respectively. We see that, the differences from the HF values, namely, 0 (1) for particle (hole) states, are larger in the ERPA case than in the perturbative one. The same holds also for the Na_{20} metallic cluster, see Fig. 5. As discussed in Secs. I and II the strong differences between the RPA and SRPA results can be partially traced back to the use of the QBA and thus to the use of the HF state as reference state, which seems to be a more severe approximation in SRPA. In the present case, i.e., metallic clusters, we can thus suppose that the use of the state [Eq. (13)] is not adequate to describe ground-state correlations while the ERPA procedure

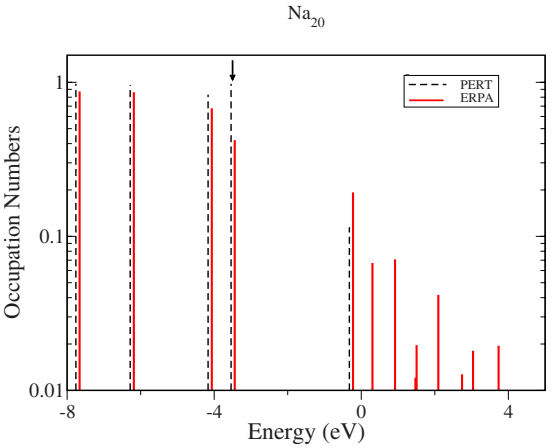

FIG. 5. (Color online) As in Fig. 4 but for the Na_{20} metallic clusters.

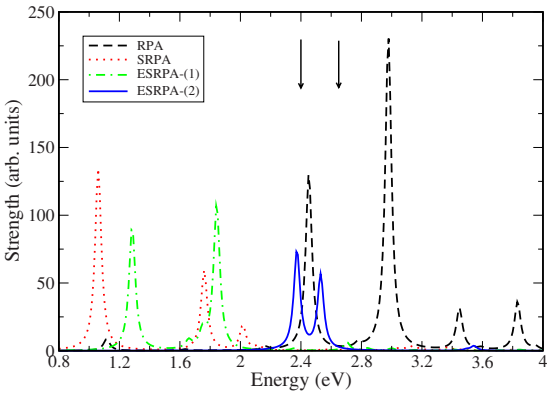

FIG. 6. (Color online) Dipole strength distribution for the Na_{40}. Dashed (black) lines refer to RPA calculations while the dotted (red) ones refer to SRPA results. The dot-dashed (green) lines and the solid (blue) lines show the ESRPA results where g.s. correlations are calculated perturbatively or within the ERPA procedure, respectively (see the text for more detail). The arrows roughly indicate the positions of the experimental dipole plasmon peaks (Ref. 5).

gives a better description (see also the end of Sec. IV of Ref. 31 for a more detailed discussion). Moreover, as mentioned before, the latter is of course a more consistent procedure in the fulfillment of the vacuum condition [Eq. (2)] used in the formal derivation of the equations of motion.

In order to have more informations about the limits and the merits of our extended SRPA approach, we have calculated the dipole strength of the Na_{40}, whose experimental distribution[5] exhibits two broad peaks at about 2.4 and 2.65 eV and the first one is higher than the second one. RPA predicts the first peak at an energy of about 2.45 eV, very close to the experimental value, and the second one much higher, at about 2.97 eV. Furthermore, the height of the second peak is about twice that of the first one. It is thus interesting to see whether in this case, in particular, as far as the issue of the relative intensities of the excitation peaks is concerned, the ESRPA approach gives a better description of the experimental results with respect to standard RPA. In Fig. 6, we plot the dipole strength distributions obtained in RPA, SRPA, and in the ESRPA approach.

We see that, going from RPA to SRPA, (black) dashed and (red) dotted lines, respectively, the height of the first peak is slightly increased while the second one is strongly reduced. However, as in the previous cases, the strength distribution is strongly shifted to lower energies and the quite good position of the energy peaks obtained in RPA is completely spoiled. When ground-state correlations are included perturbatively, dot-dashed (green) lines in the figure, the strength distribution is pushed toward higher energy but the change in the relative intensities is not in the right direction. When the ground-state correlations are taken into account by means of the ERPA procedure solid (blue) lines, we observe that: (i) the strength distribution is shifted toward higher energy with respect to the SRPA results and gets closer to the RPA one; (ii) the lower peak, that in RPA is slightly higher than the experimental value, is shifted toward lower energy, thus in the right direction; (iii) the second peak, that in RPA lies at 2.97 eV, is found now at 2.57 eV in a much better agreement with the experimental peak (2.65 eV); (iv) very stricking is also the fact that the relative intensities of the two peaks is now inverted with respect to the RPA description and it is very close to the experimental distribution.[5]

We can thus say that, the inclusion of the $2p$-$2h$ configurations in SRPA produces a fragmentation of the dipole strength distribution giving a better description of the relative intensities of the two experimental peaks. At the same time, the inclusion of the ground-state correlations is very important in order to have a good position of the two peaks. It is also confirmed that the procedure based on ERPA produces better results with respect to the perturbative one.

V. CONCLUSIONS

In this work we present an extended SRPA approach in which no use is made of QBA and ground-state correlations are taken into account. It is general and can be applied to the study of collective excitations of any realistic many-body system. In order to test how well the proposed approach works, we have applied it to the study of collective excitations in metallic clusters within the uniform jellium model. Different levels of approximation are compared. In particular, ground-state correlations are taken into account either in a perturbative way or by means of a more consistent procedure that allows us to obtain better results, especially in the dipole case. In SRPA and extended SRPA calculations all kinds of coupling within the $1p$-$1h$ and $2p$-$2h$ configurations are considered and ground-state correlations are taken into account in all the SRPA block matrices, differently from other works where they were included only in the RPA matrices. It has also been shown that the inclusion of ground-state correlations only at level of the $1p$-$1h$ channel, i.e., only in the RPA matrices, is not enough in order to describe dipole plasmon in an appropriate way.

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APPENDIX: EXTENDED SRPA MATRICES

In this appendix we give the explicit expression of the matrices appearing in the extended SRPA approach. We express these matrices in terms of the symmetrized double commutators defined as

$$[A,B,C] = \frac{1}{2}\{[A,[B,C]] + [[A,B],C]\} \tag{A1}$$

in order to preserve hermiticity.[34] As discussed in Sec. III all the double commutators are calculated without any kind of approximation and then the coming out two-body or higher terms are contracted with respect to the reference state $|0\rangle$. In

such a way, the A and B matrices are expressed in terms of the one body density matrix

$$\rho(\alpha,\beta) \equiv \langle 0|a_\alpha^\dagger a_\beta|0\rangle, \tag{A2}$$

which is assumed to be diagonal

$$\rho(\alpha,\beta) = \delta_{\alpha\beta} n_\alpha, \tag{A3}$$

where n_α is the occupation number of the α single-particle state.

In the $1p$-$1h$ channel we obtain

$$\begin{aligned} A_{ph,p'h'} &= \langle 0|[a_h^\dagger a_p, H, a_{p'}^\dagger a_{h'}]|0\rangle \\ &= \delta_{hh'}\delta_{pp'}(\epsilon_p - \epsilon_h)(n_h - n_p) \\ &\quad + \bar{V}_{ph'hp'}(n_h - n_p)(n_{h'} - n_{p'}), \\ B_{ph,p'h'} &= -\langle 0|[a_p^\dagger a_h, H, a_{p'}^\dagger a_{h'}]|0\rangle \\ &= \bar{V}_{pp'hh'}(n_h - n_p)(n_{h'} - n_{p'}), \end{aligned} \tag{A4}$$

where the ϵ's are the HF single-particle energies and $\hat{V}$ is the residual interaction. The coupling among the $1p$-$1h$ and $2p$-$2h$ configurations is given by the matrix

$$\begin{aligned} A_{ph,p_1p_2h_1h_2} &= \langle 0|[a_h^\dagger a_p, H, a_{p_1}^\dagger a_{p_2}^\dagger a_{h_2} a_{h_1}]|0\rangle \\ &= \mathcal{U}(h_1,h_2)\bar{V}_{h_1pp_1p_2}\delta_{hh_2}\chi^{(3p1h)} \\ &\quad - \mathcal{U}(p_1,p_2)\bar{V}_{h_1h_1p_1h}\delta_{pp_2}\chi^{(3h1p)}, \end{aligned} \tag{A5}$$

where $\mathcal{U}(ij)$ is the antisymmetrizer for the indices i and j, and the two factors χ are

$$\chi^{(3p1h)} = \frac{1}{2}[n_{h_1h}\bar{n}_{p_1p_2} + n_{p_1p_2}\bar{n}_{h_1h} + (n_h - n_p)(n_{h_1}\bar{n}_{p_1p_2} + n_{p_1p_2})],$$

$$\chi^{(3h1p)} = \frac{1}{2}[n_{h_1h_2}\bar{n}_{p_1p} + n_{p_1p}\bar{n}_{h_1h_2} + (n_h - n_p)(n_{p_1}\bar{n}_{h_1h_2} + n_{h_1h_2})] \tag{A6}$$

with the definition

$$n_{\alpha\beta} = n_\alpha n_\beta, \quad \bar{n}_{\alpha\beta} = 1 - n_\alpha - n_\beta. \tag{A7}$$

In the $2p$-$2h$ channel we have

$$\begin{aligned} A_{p_1h_1p_2h_2,p_1'h_1'p_2'h_2'} &= \langle 0|[a_{h_1}^\dagger a_{h_2}^\dagger a_{p_1} a_{p_2}, H, a_{p_2'}^\dagger a_{p_1'}^\dagger a_{h_2'} a_{h_1'}]|0\rangle \\ &= (\epsilon_{p_1} + \epsilon_{p_2} - \epsilon_{h_1} - \epsilon_{h_2})U(p_1,p_2)U(h_1,h_2) \\ &\quad \times \delta_{h_1h_1'}\delta_{p_1p_1'}\delta_{h_2h_2'}\delta_{p_2p_2'}\chi^{(0)} + U(h_1,h_2) \\ &\quad \times \bar{V}_{p_1p_2p_1'p_2'}\delta_{h_1h_1'}\delta_{h_2h_2'}\chi^{(4p)} + U(p_1,p_2) \\ &\quad \times \bar{V}_{h_1h_2h_1'h_2'}\delta_{p_1p_1'}\delta_{p_2p_2'}\chi^{(4h)} \\ &\quad + U(p_1,p_2)U(h_1,h_2)U(p_1',p_2')U(h_1',h_2') \\ &\quad \times \bar{V}_{p_1h_1'h_1p_1'}\delta_{h_2h_2'}\delta_{p_2p_2'}\chi^{(2p2h)}, \end{aligned} \tag{A8}$$

where

$$\chi^{(0)} = n_{h_1h_2}\bar{n}_{p_1p2} - n_{p_1p_2}\bar{n}_{h_1h2},$$

$$\chi^{(4p)} = \frac{1}{2}\chi^{(0)}(\bar{n}_{p_1p2} + \bar{n}_{p_1'p'2}),$$

$$\chi^{(4h)} = -\frac{1}{2}\chi^{(0)}(\bar{n}_{h_1h2} + \bar{n}_{h_1'h'2}),$$

$$\chi^{(2p2h)} = \frac{1}{2}\chi^{(0)}(n_{h_1} + n_{h_1'} - n_{p_1} - n_{p_1'}). \tag{A9}$$

Also the norm matrices acquire a new expression,

$$G_{ph,p'h'} = \langle 0|[a_h^\dagger a_p, a_{p'}^\dagger a_{h'}]|0\rangle = \delta_{hh'}\delta_{pp'}(n_h - n_p)$$

and

$$\begin{aligned} G_{p_1h_1p_2h_2,p_1'h_1'p_2'h_2'} &= \langle 0|[a_{h_1}^\dagger a_{h_2}^\dagger a_{p_1} a_{p_2}, a_{p_2'}^\dagger a_{p_1'}^\dagger a_{h_2'} a_{h_1'}]|0\rangle \\ &= U(p_1,p_2)U(h_1,h_2)\delta_{h_1h_1'}\delta_{p_1p_1'}\delta_{h_2h_2'}\delta_{p_2p_2'} \\ &\quad \times (n_{h_1h_2}\bar{n}_{p_1p2} - n_{p_1p_2}\bar{n}_{h_1h2}). \end{aligned}$$

The B matrices

$$B_{ph,p_1p_2h_1h_2} = \langle 0|[a_p^\dagger a_h, H, a_{p_1}^\dagger a_{p_2}^\dagger a_{h_2} a_{h_1}]|0\rangle,$$

$$B_{p_1h_1p_2h_2,p_1'h_1'p_2'h_2'} = \langle 0|[a_{p_1}^\dagger a_{p_2}^\dagger a_{h_1} a_{h_2}, H, a_{p_2'}^\dagger a_{p_1'}^\dagger a_{h_2'} a_{h_1'}]|0\rangle \tag{A10}$$

are found to be zero also in the extended SRPA. This is mainly related to the fact that we approximate two-body and higher density matrices in terms of the one-body density matrix, which is then assumed diagonal. For example, we have for the two-body density,

$$\begin{aligned} \langle 0|a_\alpha^\dagger a_\beta^\dagger a_{\beta'} a_{\alpha'}|0\rangle &= \rho(\alpha,\alpha')\rho(\beta,\beta') - \rho(\alpha,\beta')\rho(\beta,\alpha') \\ &= n_\alpha n_\beta[\delta(\alpha,\alpha')\delta(\beta,\beta') - \delta(\alpha,\beta')\delta(\beta,\alpha')]. \end{aligned}$$

Finally we stress that the standard SRPA matrices are easily obtained by using the HF occupation numbers, i.e., 0 and 1 for particle and hole states, respectively, in the above expressions.

*danilo.gambacurta@ct.infn.it
†francesco.catara@ct.infn.it

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