Collective vibrational states with fast iterative QRPA method

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An iterative method we previously proposed to compute nuclear strength functions [1] is developed to allow it to accurately calculate properties of individual nuclear states. The approach is based on the quasi-particle-random-phase approximation (QRPA) and uses an iterative non-hermitian Arnoldi diagonalization method where the QRPA matrix does not have to be explicitly calculated and stored. The method gives substantial advantages over conventional QRPA calculations with regards to the computational cost. The method is used to calculate excitation energies and decay rates of the lowest lying $2^+$ and $3^-$ states in Pb, Sn, Ni and Ca isotopes using three different Skyrme interactions and a separable gaussian pairing force.

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I. INTRODUCTION

The goal of nuclear structure theory is to be able to predict and model the physics of the atomic nucleus. This involves the ground-state properties, as well as different modes of excitation and decay. One of the possible methods to compute excited states in nuclei is based on the quasi-particle-random-phase approximation (QRPA) [2]. This approach can be derived by considering the linear response of a nucleus when perturbed by an external field. From the response one can extract information about excited nuclear states and cross sections for nuclear reactions. The QRPA approach is particularly interesting in connection with nuclear density-functional theory (DFT) as the method can be applied also when starting from a density functional. In order for the QRPA method to be practical, it is very important to implement it in ways that have low computational costs. For phenomenological DFT approaches, a low computational cost would allow dynamical properties to be considered when fine tuning model parameters. A numerically efficient method is also essential for applications to deformed and heavy nuclei which are otherwise prohibited by the time and memory required to construct and diagonalize the large QRPA matrix.

Two recent solution methods address these issues. The Finite Amplitude Method (FAM) [3, 4] generates the response of a nucleus to an external field by solving the linear response equations iteratively for each requested external field frequency. FAM furthermore uses the same mean fields as in the Hartree-Fock-Bogoliubov (HFB) ground-state calculation and employs finite differences to linearize the equations of motion. In its current form FAM uses a smoothing method to improve stability and therefore one cannot easily extract the exact QRPA eigenamplitudes. The same is true for the iterative Arnoldi method [1] which is able to provide smoothened QRPA strength functions and their energy weighted moments, but does not generate accurate individual states. A common aspect of both methods is however their ability to generate partial solutions of the full QRPA problem with reduced computational effort.

The purpose of this paper is to generalize the Arnoldi method which we previously developed for iterative calculations of RPA strength functions. The generalization involves modifying the method so that it becomes possible to not only compute strength functions but also sets of individual excited states with high accuracy. As a first step the new method is applied to the calculation of excitation energies and decay rates of the lowest lying $2^+$ and $3^-$ states in several isotope and one isotone chain. Particular focus is given to the region around double-magic $^{208}$Pb where new experiments are currently planned [5].

This paper is organized as follows: in Sec. II the QRPA formalism is briefly reviewed and specific aspects of our formulation are discussed. In Sec III the computational cost and accuracy of the method is evaluated. In Sec. IV the method is applied to the calculation of energies and transition probabilities of the lowest $J^\pi = 2^+$ and $J^\pi = 3^-$ states in a selection of semi-magic even-even nuclei. Finally conclusions are given in section V.

II. QRPA IN TERMS OF FIELDS

The iterative method is based on the QRPA equations [2, 6–8] which can be derived by starting from time-dependent HFB theory. Here we present the main parts of the derivation, highlighting aspects relevant to the iterative formulation. In cases where the expressions are not fully defined we use notation consistent with Ref. [2]. The QRPA equations can be derived by considering a general time-dependent wavefunction which is oscillating between the ground state and an excited state with
excitation energy $\hbar \omega$

$$|\psi(t)\rangle = e^{-itE_{gs}/\hbar}C_{gs}|\psi_{g.s.}\rangle + e^{-it(E_{gs}+\hbar \omega)/\hbar}C_{exc}|\psi_{exc.}\rangle.$$

We limit the consideration to small-amplitude oscillations around the ground state so that the corresponding generalized density-matrix $R$ can be expanded to first order in $C_{exc}$

$$R(t) \simeq R_{gs} + e^{-i\omega t} \tilde{R} + e^{i\omega t} \tilde{R}^\dagger.$$

In order to make use of the time-dependent-Hartree-Fock-Bogoliubov (TDHFB) equations of motion, it is desired that the time-dependent density should be a HFB density at all times (i.e., it should be a projector $R^2 = R$). We further assume that $R_{gs}$ can be approximated with the ground-state HFB density. Then the most general approximation for the transition density which ensures that $R(t)$ a projector for small-amplitude vibrations involves both the forward $\tilde{Z}$ and the backward $\tilde{Z}'$ going amplitudes

$$U^\dagger \tilde{R} U = -C_{gs}^* C_{exc} \langle \psi_{g.s.} | \left( \frac{\alpha \alpha^\dagger}{\alpha^\dagger \alpha} \right) | \psi_{exc.} \rangle \quad (1)$$

In this expression we have made use of the matrix

$$U = \begin{pmatrix} U & V^\dagger \\ V & U^\dagger \end{pmatrix},$$

written in terms of the $U$ and $V$ pairing matrices related to the HFB ground state as well as the matrices of quasiparticle operators $[\alpha \alpha^\dagger]_{ij} = \alpha_i \alpha_j^\dagger$. It should be noted that if the wavefunctions in the beginning were taken as HFB vacuums one would not obtain any backward going amplitudes, as can be seen from Eq. (1) by inserting the HFB ground state. Thus it is the assumption of the transition density $R$ being as general as allowed by the $R^2 = R$ criteria, which allows for the existence of the implicitly defined correlated ground state.

Inserting the expression for $R(t)$ into the TDHFB equations of motion $i\hbar \frac{dR(t)}{dt} = [\mathcal{H}, R(t)]$ and taking the small-amplitude limit leads to the QRPA equation

$$\hbar \omega \tilde{R} \simeq [\mathcal{H}[R_{gs}], \tilde{R}] + [\mathcal{H}^1[\tilde{R}], R_{gs}].$$

In this expression the hermicity property $\mathcal{H}^1[\tilde{R}] = (\mathcal{H}^1[\tilde{R}])^\dagger$ of the effective interaction was assumed and the time-dependent fields $\mathcal{H}(t) = \partial \mathcal{E} / \partial R$ were expanded around the ground-state value

$$\mathcal{H}[R] \simeq \mathcal{H}[R_{gs}] + \mathcal{H}^1[R - R_{gs}].$$

This expansion is taken to first order in the transitional fields, which is enough for small-amplitude vibrations and leads to

$$\mathcal{H}^1[\tilde{R}] = \left( \frac{\hbar}{\Delta^\dagger} \frac{\Delta}{-\hbar^T} \right),$$

where

$$\Delta_{\mu \nu} = \sum_{\pi \lambda} \frac{\partial \hbar \mu \nu}{\partial \rho_{\pi \lambda}} \tilde{\rho}_{\pi \lambda},$$

$$\Delta'_{\mu \nu} = \sum_{kl} \nu^{pp}_{\mu \nu kl} \tilde{\rho}_{kl},$$

$$\Delta''_{\mu \nu} = \sum_{kl} \nu^{pp}_{\mu \nu kl} \tilde{\rho}_{kl}.$$

In our case with a density-independent pairing interaction it is only the $\hbar = \partial \mathcal{E} / \partial \rho$ field which is non-linear in the densities and becomes linearized. With a density-dependent pairing interaction the $\Delta, \Delta'$ fields would also have to be linearized and would give an additional contribution to the $\hbar$ field.

Inserting the expressions for fields and densities into the QRPA equation, and multiplying from the left with $U^\dagger$ and from the right with $U$, gives a system of equations for the unknown excitation energies $\hbar \omega$ and the $\tilde{Z}$ and $\tilde{Z}'$ amplitudes:

$$\hbar \omega \tilde{Z} = E \tilde{Z} + \tilde{Z} E + \tilde{W},$$

$$-\hbar \omega \tilde{Z}' = E \tilde{Z}' + \tilde{Z}' E + \tilde{W}' \tilde{\rho}.$$

In this equation, $E$ denotes a diagonal matrix of positive quasi-particle energies and the $W$ matrices depend on the linearized fields

$$\tilde{W} = U^\dagger \tilde{h} V^\dagger + U^\dagger \Delta U^\dagger + V^\dagger \Delta' V^\dagger - V^\dagger \tilde{h}^T U^\dagger,$$

$$\tilde{W}' = V^T \tilde{h} U + V^T \Delta V + U^T \Delta' U - U^T \tilde{h}^T V,$$

which can be expressed in terms of the transition densities

$$\tilde{\rho} = U \tilde{Z} V^T + V^\dagger \tilde{Z}' U^\dagger,$$

$$\tilde{\rho}' = U \tilde{Z} V^T + V^\dagger \tilde{Z}' U^\dagger,$$

$$\tilde{\rho}' = V \tilde{Z} V^T + U^\dagger \tilde{Z}' U^\dagger.$$
As an example to illustrate the iteration procedure, we neglect spin and isospin and consider a term in the energy of the form
\[ E [\rho] = \int \rho^{a+2} (\vec{r}) \, d\vec{r}, \]
which gives the linearized field
\[ \hat{h}_{\alpha m} = (\alpha + 2) (\alpha + 1) \int \phi^*_i (\vec{r}) (\rho_{g.s.} (\vec{r}) \hat{\rho} (\vec{r})) \phi_m (\vec{r}) \, d\vec{r}. \]

In this case, the action of the QRPA matrix on an eigenvector can be calculated in three steps.

The first step is to generate the densities \( \hat{\rho} \) according to Eq. (3) and expressing them in \( r \) space. For the next step, \( \hat{h} \) is calculated as above. Alternatively, in the case of a density-independent interaction, where fields are already linear in densities \( \mathcal{H}^I [\hat{\rho}] = \mathcal{H} [\hat{\rho}] \), this can be achieved using the HFB mean-field routines for calculating matrix elements. Finally multiplying the fields with \( U \) and \( V \) matrices as in Eq. (2) one obtains the \( W \) matrices.

The main advantage of expressing the equations in this form is that calculating and storing two-body matrix elements can be avoided and instead one can rely on the expressions for HFB fields. The price to pay is that the densities and integrals for the matrix elements of the fields are recalculated for each matrix vector product, in the same way as when performing the HFB iterations to find the ground state. Thus it is important to investigate how many iterations i.e. matrix-vector products are needed in order to obtain acceptable convergence, and whether the iteration procedure introduces numerical errors which could lead to instabilities.

III. ACCURACY AND EFFICIENCY OF THE METHOD

The iterative QRPA solver is implemented by extending the program HOSPHE (v1.02) [3] and will be included in the next published version of the program. This code uses a spherical harmonic oscillator basis and takes advantage of the Wigner-Eckart theorem in order to work with angular momentum reduced quantities. The use of reduced quantities keeps the HFB and QRPA dimensions small and makes the code a useful tool for testing different calculational methods.

In order to verify that the QRPA implementation is correct, a comparison is made with a recent QRPA implementation based on the HFTHO code [10]. This code is able to treat axially deformed nuclei and its QRPA implementation is based on the traditional diagonalization of a large QRPA matrix. Therefore, applications of this code are limited to cases where dimensions can be kept within manageable limits.

As a test case we consider the nucleus \( {}^{18}_{8}\text{O}_{10} \) and compare the ground-state energy and energies of the QRPA excitations obtained in both codes. In order to have a benchmark result that is useful for testing future QRPA codes we list values obtained from both codes in Table I. Several different recipes on how to truncate the pairing space and how to treat the Coulomb interaction exist in the literature, so in order to make the benchmark results as useful as possible, the results are obtained without any pairing truncation and without any Coulomb interaction. The remaining parameters are listed in the caption of Tab. I.

The implementation based on the axially deformed HFTHO code [10] allows us to test its accuracy by performing calculations for excitations with different angular momentum projections on a principal axis of the nucleus. Since the comparison is made for a spherical nucleus, these different calculations should ideally give the same result. In this way, for the QRPA implementation of [10], the precision of the \( 2^+ \) excitation was estimated to be roughly \( 10^{-4} \) both for the energy and for the \( B (E2) \) value. As seen from Table I, the lowest states calculated with both codes agree to about this precision. A similar accuracy test with the HOSPHE code is not possible as it works in spherical symmetry. But since HOSPHE uses the same mean-fields both in the QRPA and the HFB calculations we expect about the same accuracy for the QRPA excitations as for the groundstate energy. The full strength functions calculated with both codes were also compared and turned out to be indistinguishable by eye when the reduced transition probabilities are plotted as function of the energy of the excited states.

| Quantity       | HOSPHE | HFTHO+QRPA [10] |
|----------------|--------|-----------------|
| \( E_{\text{HFB}} \) | -131.677022532 | -131.677022519 |
| \( E_{\text{exc}} (0^+) \) | 20.49599056 | 20.4959974 |
| \( E_{\text{exc}} (1^+) \) | 14.02098704 | 14.02085 |
| \( E_{\text{exc}} (2^+) \) | 8.691200427 | 8.69120 |
| \( E_{\text{exc}} (3^+) \) | 12.91743893 | 12.91747 |
| \( E_{\text{exc}} (4^+) \) | 9.041422878 | 9.0414255 |

Table I: Comparison of HFB and QRPA calculations performed for the nucleus \( {}^{18}_{8}\text{O} \) without any pairing truncation and without Coulomb interaction. For the Skyrme interaction we use the SLy4 parametrization [11] with a delta (volume) pairing interaction [12] with strength \( V_0 = -200 \) MeV/(fm)\(^3\) and a one-body center of mass correction. The results are obtained with a harmonic oscillator basis where the maximum oscillator shell included has principal quantum number \( N_{\text{max}} = 5 \) and the oscillator constant is set to 0.865 (fm)\(^{-1}\). In this table, the transition strengths are calculated using the isoscalar transition operators of Ref. [13]. Energies have units of MeV and the \( B (EI) \) transitions are in units of \( e^2 \) (fm)\(^4\). For each multipolarity the state lowest in energy with an appreciable strength is compared. When both codes give the same decimals, they are printed in bold.
A. Iterative solutions

As described above, the product of the QRPA matrix acting on an arbitrary vector can be calculated without constructing the matrix explicitly. When this technique is used, traditional matrix diagonalisation routines, which need explicit information about the matrix elements, cannot be used. Instead, one must resort to indirect iterative methods, such as the Lanczos or Arnoldi methods. For non-Hermitian problems, such as the QRPA eigenvalue problem, the Implicitly Restarted Arnoldi method (IRA) [18, 19] is one of the most commonly used methods for finding accurate approximations for the eigenstates lowest in energy. IRA is a more advanced version of the original Arnoldi method, giving faster convergence and a reduction in computational effort. As with the iterative Arnoldi method of [1], the IRA method generates a set of basis vectors, usually called Ritz vectors, which span a vector space called the Krylov subspace, and uses these vectors to represent the QRPA eigenvectors. However, IRA’s use of restarting allows it to gradually improve the accuracy of a set of eigenstates during iteration, using a reasonably small number of Ritz vectors (typically a few hundred at most).

In the extended HOSPHE code, we use the numerical software ARPACK [20], which implements the IRA method. With this method, the number of matrix-vector products needed in order to reach convergence depends on the requested tolerance. Denoting the QRPA matrix $A$, the approximate eigenvector $x$ and the corresponding approximate eigenvalue $\lambda$, the iterations proceed until the accuracy measure $||Ax - \lambda x||$ is less than the requested tolerance.

Fig. 1 shows the convergence as a function of the basis size when applying the method for the calculation of the lowest $2^+$ state in $^{214}$Pb. The only truncation employed is the number of main oscillator shells used for the basis. As seen from this figure the accuracy measure is always lower than the requested tolerance. Denoting that the calculation with 33 oscillator shells is the wall clock time was 12 min on a desktop workstation (Intel Core i7-2600K, 3.4GHz). In all cases the Krylov subspace was constructed from 100 Ritz vectors which was estimated to give the fastest convergence with $N_{max} = 16$.

![Figure 1: Convergence of the excitation energy and reduced transition probability for the lowest $2^+$ state in $^{214}$Pb using the SLy4 interaction together with a separable Gaussian pairing force \cite{15, 16}. The convergence is shown as a function of the maximum oscillator shell $N_{max}$ included in the basis. The accuracy measure and the number of iterations needed to obtain convergence are also shown. The tolerance parameter which determines when to stop the Arnoldi iterations was set to 0.001. For $N_{max} = 16$, the wall clock time was 12 min on a desktop workstation (Intel Core i7-2600K, 3.4GHz). In all cases the Krylov subspace was constructed from 100 Ritz vectors which was estimated to give the fastest convergence with $N_{max} = 16$.](image)

Sets of a few lowest eigenvalues can also be extracted and requires about the same number of iterations. For example, in the case of $^{192}$Pb, the number of iterations needed to extract 10, 20 and 30 positive energy eigenstates becomes 1974, 2648 and 3427 respectively.

\[ \begin{align*}
10 & \quad 15 & \quad 20 & \quad 25 & \quad 30 \\
10^4 & \quad 10^5 & \quad 10^6 & \quad 10^7 & \quad 10^8 \\
|Ax - \lambda x| & \quad B(E2) \quad [\text{eIb}^2] & \quad \text{Iterations} \\
\end{align*} \]
In the equivalent spectra method the normal and ab-normal density-matrices are truncated during the HFB iterations [13]. However, in the subsequent QRPA calculation we used non-truncated wavefunctions without any energy cut for the residual particle-particle interaction. This way of using the equivalent spectra method is therefore slightly inconsistent and a better truncation recipe is desired. In the following we will only use the finite-range pairing interaction which does not need to be truncated and allows us to treat HFB and QRPA in a consistent way.

The effect of changing the strength of the Gaussian pairing interaction with ±5% is shown in Fig. 2 with dashed lines. Both the energies and the transitions are sensitive to such a change, i.e. a decrease of the pairing lowers the excitation energies and raises the $B\,(E2)$ values. The effect is seen to be largest for $N = 104$ which is just between two magic numbers. The schematic fits of the pairing strengths appear to give quite reasonable values for the lead isotopes with an average $E_{2^+}$ energy that agrees roughly with experiment.

In this work we consider three different Skyrme parameterizations: SkM*, SLy4 and SkX. SKM* is based on the SkM parameters [23] but has been adjusted further using results from fission barrier calculations [24]. The original SKM parameters were determined by considering both static ground state properties as well as some dynamical properties including monopole and quadrupole resonances [23]. SLy4 was adjusted with special care taken to model neutron matter in order to facilitate the description of neutron rich nuclei [11]. The accuracy of QRPA based on these interactions was recently studied and compared to calculations based on the generator-coordinate method GCM [25]. It was found that SkM* reproduced experimental $2^+$ states more accurately than SLy4 and the QRPA results were similar to results obtained with the GCM. In addition, we also consider the SkX interaction which has been tuned with special focus on reproducing single-particle states in double-magic nuclei [26].

Results for the lead isotopes using the three different Skyrme interactions are shown in the left hand panels of Fig. 3. For the double magic nucleus Pb\textsubscript{120}, SkX gives the correct $2^+$ energy while the other two forces overestimate this excitation energy. As the neutron number is reduced, the predictions show considerable differences. SLy4 gives zero energy solutions around $N = 112$, indicating a transition to a deformed ground state, while the other two interactions appear to be stiffer towards deformation and give more realistic results.

The ground-state energies of Pb\textsubscript{108} and Pb\textsubscript{112} as a function of quadrupole deformation are shown in Fig. 4. The energy curves are calculated using delta pairing instead of the Gaussian pairing which means that the curves are slightly inconsistent with the QRPA calcula-

### Table II: Strength parameters of the separable Gaussian pairing interaction in units of MeV(fm)$^3$.

| Interaction  | $G_n$  | $G_p$  |
|-------------|--------|--------|
| SLy4        | 655    | 600    |
| SkM*        | 610    | 550    |
| SkX         | 560    | 530    |

Figure 2: Excitation energies and reduced transition probabilities for Pb isotopes. Results are shown for different treatments of the pairing interaction. The dashed lines denote the result of changing the strength of the finite-range pairing with ±5%. Decreasing the pairing lowers the energies and raises the $B\,(E2)$ values. The strength parameters for the zero-range interaction were chosen as $V_n = -168$ and $V_p = -200$ (MeV(fm)$^3$) when the cutoff in the equivalent spectra was taken as 60 MeV and 20 MeV respectively.

### IV. NUMERICAL RESULTS

#### A. Influence of the pairing interaction

In order to study the influence of the pairing interaction on $2^+$ states we compare the use of a zero-range delta interaction [12] combined with a truncation in the equivalent spectra [13] to the use of a separable Gaussian pairing force [14]. This force has a finite range and therefore does not need to be truncated. In order to obtain reasonable pairing, the pairing strengths are tuned to get the lowest quasiparticle energies to agree with the experimental gaps extracted in Ref. [21] using a four-point formula. The resulting parameters obtained for the finite-range interaction are shown in Table II.

Results for the lead isotopes using the different pairing interactions are shown in Fig. 2. As seen in this figure there are fluctuations in the energies which depend on the choice of pairing force. Comparing the two pairing interactions, it appears that the finite-range interaction is slightly better in capturing the fluctuations of the experimental energies.

In the equivalent spectra method the normal and ab-
The spherical minimum obtained with SkM* is stiffer than the oblate one with quadrupole deformation. In this case our QRP A calculation is likely to give a zero energy solution as the assumed spherical ground state is no longer stable with respect to quadrupole deformations. The $2^+$ energies obtained with the SkM* interaction agree rather well with experiment and seem to favor the prediction of stiffer energy surfaces.

In the QRP A formalism, an expression for the operator

$$O_\alpha = \sum_{k<k'} Z_{kk'}^\alpha a_{k'}^\dagger a_k^\dagger - Z_{kk'}^\alpha^* a_k a_{k'}^\dagger,$$

which creates the excited states by acting on the QRP A ground state can be written as

$$\langle \alpha | O_\alpha | \beta \rangle = \sum_{m,m'} |Z_{\alpha jm,\alpha'j'm'}|^2 - |Z_{\alpha jm,\alpha'j'm'}^\alpha|^2.$$

Defined in this way, the largest components in the calculated $2^+_1$ states in the chain of lead isotopes are shown in Fig. 5. As seen in this figure, the largest proton and neutron components in Pb$^{126}$ involve particle-hole excitations across the $Z = 82$ and $N = 126$ gaps. In the other isotopes, where neutron pairing is active, the $2^+$ states mainly involve neutron excitations ($\sim 85\%$) with the dominating component being $15-30 \% \nu (i_{13/2})^2$ for $N = 100 - 116$. The calculated neutron single-particle levels are shown in Fig. 6 and as expected the dominating components in the $2^+$ states involve excitations among the shells close to the Fermi-level.

It should be noted that the transition strengths are calculated directly from the electromagnetic operators without any effective charges. Therefore it is the smaller
two double magic Sn isotopes, the excited states are calculated to be roughly even mixture of proton and neutron excitations, while the excited states in the semi-magic isotopes mainly involve neutron excitations. With SkX, the largest components are $\nu (d_{5/2})^2$ for Sn$_{52-54}$, $\nu (g_{7/2})^2$ for Sn$_{56-64}$ and $\nu (h_{11/2})^2$ for Sn$_{66-80}$. The positions of these shells (shown in Figure 4) are thus important in order to reproduce the details of the experimental data.

C. $J^\pi = 2^+$ states in Ni and Ca isotopes

The results for the Ni chain are shown in the left hand panels of Fig. 8. In Ni$_{28}$, the $2^+$ state is built from an almost equal mixture of proton and neutron excitations, while the $2^+$ states in the other nickel isotopes are dominated by neutron excitations. SkX predicts a smaller 28 gap than the other interactions and a $2^+$ state in Ni$_{28}$ which is lower than in experiment. SkX also predicts a smaller gap at $N=32$ and does not show the spike in excitation energy obtained with the other interactions for Ni$_{32}$.

QRPA calculations based on the relativistic-mean-field model for Ni$_{40}$ overpredicted the energy of the $2^+$ state by roughly three times the experimental energy. A suggested explanation was missing 2p-2h and higher order excitations among the neutrons. Since the neutron Fermi-level is located between opposite parity shells, this state will be overpredicted whenever the neutron pairing goes to zero. With SLy4 we also obtain an overprediction, although less severe, while the other interactions predict excitation energies close to the experimental value. It is interesting to note that with SkX the ground state is calculated to have an average gap of $\Delta_n = 1.38$ MeV and the $2^+$ state is obtained with both correct energy and transition strength. The state is built as a mixture of proton ($\sim 23\%$) and neutron ($\sim 77\%$) excitations, where the largest component is $\nu (g_{9/2})^2$.

Results for the Ca chain are shown in the right hand panel.

Figure 6: Theoretical (SkX) neutron single-particle levels. The left set of levels is for Ca$_{28}$, the middle set for Sn$_{52}$, and the right set is for Pb$_{126}$. Positive parity levels are shown with full lines and negative parity levels with dashed lines.

Figure 7: Same as in Fig. 3 but for $N=126$ isotones.
D. $J^e = 3^-$ states in Pb and Sn isotopes

The calculated $3^-$ states for lead isotopes are shown in the left hand panels of Fig. 9. The most striking feature of the data is the dip in excitation energy seen when going from $^{204}$Pb to $^{208}$Pb. In $^{208}$Pb the $3^-$ state is created by an roughly equal amount of neutron and proton excitations across the 82 and 126 gaps. When two more neutrons are added, negative parity states can be made by exciting particles between the neighboring positive parity $g_{9/2}$ shell and negative parity $j_{15/2}$ shell, located above the 126 gap (see Figure 9). Therefore the $3^-$ state in $^{208}$Pb can be built at a low cost mainly from neutron excitations, which explains the dip in the experimental energy. As seen in Fig. 9 SkX reproduces the experimental lowest $3^-$ energies and $B(E3)$ values almost perfectly except for the dip in the transition probability seen for $^{208}$Pb. The forces SkM* and SLy4 have a less perfect overall agreement, but SkM* agrees with experiment in the case of $^{208}$Pb.

In $^{208}$Pb with SkX, the excitation operator for the lowest $3^-$ state consists of 17 % proton excitations, where the largest component is only 6 % $\pi(d_{3/2})^1(h_{9/2})^1$ and the main neutron component is 62 % $\nu(f_{7/2})^1(i_{13/2})^1$. The $3^-$ states of the neutron deficient Pb isotopes have many small proton components contributing. When the neutron number increases, proton excitations become more dominant, but with fewer components contributing. At $N=124$, proton excitations constitute 48 % with one dominant component, 29 % $\pi(d_{3/2})^1(h_{9/2})^1$, while the largest neutron component is 13 % $\nu(p_{3/2})^1(g_{9/2})^1$. Thus, with QRPA, we see a transition from strong proton configuration mixing in $^{208}$Pb to strong neutron configuration mixing in $^{212}$Pb.

The lowest experimental and calculated $3^-$ states of tin isotopes are shown in the right hand panels of Fig. 9. The energies are reproduced almost perfectly in the region $N=60-72$, using SkX and SkM* forces. Closer to the $N=82$ shell closure, the predicted energies become too high. The experimental $B(E3)$ values are reproduced roughly by all three Skyrme forces, but none of the three forces gives a truly accurate description of the finer details. The excitations are dominated by the $\nu(d_{5/2})^1(h_{11/2})^1$ configuration which has a component of 70 % in $^{116}$Sn, and gradually goes down to 50 % in $^{120}$Sn. The second largest excitation is 3% $\nu(g_{9/2})^1(h_{11/2})^1$ in $^{120}$Sn, but starting from $^{116}$Sn the second largest excitation is $\nu(g_{7/2})^1(h_{11/2})^1$ and its amplitude grows steadily from 5% to 10% in $^{120}$Sn. Other neutron excitations between the $\nu h_{11/2}$ subshell and the $N_{osc} = 4$ orbitals are excluded because of angular momentum selection rules. The proton fraction of the transitions is an almost constant 10 % and is composed of many small-amplitude excitations. The lowest energies and the largest transition strengths are obtained for $N=66$ when the neutron Fermi-level is close to the $\nu h_{11/2}$ intruder shell and negative parity excitations correspond to a low energy cost.

E. $J^e = 3^-$ states in Ni and Ca isotopes

For the Ni isotopes shown in the left hand panels of Fig. 10 we also show experimental data for the second $3^-$ states in $^{58-62}$Ni taken from [30]. For some reason the calculations for $^{58-62}$Ni agree better with the second $3^-$ states. Especially the lowest $3^-$ state in $^{58}$Ni

Figure 9: Excitation energies and reduced transition probabilities ($B(E3; 0^+_g \rightarrow 3^-_i)$) for the lowest $3^-$ states in Pb and Sn isotopes. Results are shown for three different Skyrme parameterizations. The experimental data for the $3^-$ states are taken from [23].
is predicted about 3 MeV too high in energy. It should be noted that this state has not been clearly identified as $3^-$ in experiments [29], contrary to the $3^-$ states in the other Ni isotopes. One should also note that with Skyrme type interactions one usually neglects proton-neutron pairing and part of the isovector particle-hole interaction which may have an influence around the $N=Z$ line. With SkX, the lowest theoretical $3^-$ state in Ni$_{58}$ is calculated to be of isoscalar type. This state is built from smaller probability excitations to the orbitals of character.

For the semi-magic nickel isotopes with $N=30-48$, the $3^-$ states are dominated by neutron excitations, $\nu(d_{3/2})^1(g_{9/2})^1$ which decrease from 75% in $N=30$ to 16% in $N=48$ and $\nu(f_{5/2})^1(g_{9/2})^1$, which increase from 2% in $N=30$ to 74% in $N=48$. The proton components are small since it is more favorable to excite neutrons when the proton Fermi-level is just above $f_{7/2}$. The double magic Ni$_{50}$ has a different structure than the semi-magic isotopes, the dominating component being 36% $\nu(p_{1/2})^1(d_{5/2})^1$.

The results for calcium isotopes are shown in the right hand panels of Fig. [10] In this case, the lowest experimental $3^-$ energies are best reproduced by SkM* and SkX while SLy4 gives too high energies. However, in general the finer details of the $3^-$ energies between $N=20$ and $N=26$ are not reproduced. Especially for the $N=Z$ nucleus Ca$_{20}$, the energy calculated with SkM* and SkX becomes too low.

The leading wave function components of the calcium isotopes are shown in Fig. [10]. As seen in this figure, the $3^-$ state in $^{20}$Ca$_{20}$ is composed of a fairly even mixture of proton and neutron excitations from the $d_{3/2}$ and $s_{1/2}$ shells below the 20 gap to the $f_{7/2}$ shell just above the gap (see Figure 10). Going towards Ca$_{28}$ the neutron excitations become suppressed as the Fermi-level reaches the middle of the $N_{osc}=3$ shell and proton excitations start to dominate. When more neutrons are added, neutron excitations to the $g_{9/2}$ shell start to appear and become the largest components in Ca$_{40}$.

V. SUMMARY AND CONCLUSIONS

An iterative method for the solution of the QRPA equations which avoids the construction of the large QRPA matrix was employed for the calculation of low-lying vibrational states. The method uses the Implicitly Restarted Arnoldi approach for the solution of the non-hermitian eigenvalue problem. In this approach, only the action of the matrix on a Ritz vector is needed. As demonstrated, this can be expressed in terms of the fields generated by the transitional densities corresponding to the Ritz vector. Our study shows that the method is numerically stable and typically requires a few thousand iterations in order to produce well converged lowest even-states.

The new solution method was applied to the calculation of excitation energies and decay rates of the first $2^+$ and $3^-$ vibrational states in a set of spherical even-even nuclei. The calculations were performed using three different Skyrme interactions together with a finite-range pairing force. Overall a quite reasonable agreement with experimental data was obtained. The main difficulties seem to be in the description of $2^+$ states right between two magic neutron numbers where the different interactions tend to give different results and even zero energy solutions.

Difficulties were also observed for the Ca isotopes where all the interactions gave too little collectivity. These difficulties are probably related to the limitations of the QRPA method itself, as it only includes two-quasiparticle excitations. However, since our method is rather computationally inexpensive, it may become practical to consider extensions of QRPA, for example higher-order QRPA approaches or boson expansion methods, which allow to treat more complicated excitations that could improve the results.

Because of the low numerical cost and low memory requirements, the method appears promising for applications to deformed nuclei where the dimensions become substantially larger. Indeed, the methods of this work
are quite analogous to the ones used in the nuclear Shell Model community, where quite similar iterative methods have been used for large-dimensional hermitean eigenvalue problems. However, contrary to the Shell Model where the dimensions increase exponentially and multi-major shell calculations for heavy nuclei are almost impossible, the QRPA method stays tractable. Therefore, as a next step, we will implement the method in a code able to treat nuclei with deformed ground states.

For spherical nuclei, the speed of the iterative method opens the possibility to directly compute low-lying states and include them as part of the observables used when fitting the parameters of new improved Skyrme interactions. However, care must be taken to analyse the structure of the included states in order to ensure that a QRPA description is compatible with the experimental states.

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