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Implementation and testing of Lanczos-based algorithms for Random-Phase Approximation eigenproblems

Myrta Grüning [a,e,*], Andrea Marini [b,c,d], Xavier Gonze [e]

[a] Centre for Computational Physics and Physics Department, University of Coimbra, Rua Larga, 3004-516 Coimbra, Portugal
[b] European Theoretical Spectroscopy Facility, Physics Department, University "Tor Vergata", I-00133 Rome, Italy
[c] Nano-Bio Spectroscopy Group, Universidad del País Vasco, E-20018 San Sebastián, Spain
[d] IKERBASQUE, Basque Foundation for Science, E-48011 Bilbao, Spain
[e] European Theoretical Spectroscopy Facility, NAPS/IMCN, Université Catholique de Louvain, B-1348 Louvain-la-Neuve, Belgium

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ABSTRACT

The treatment of the Random-Phase Approximation Hamiltonians, encountered in different frameworks, like time-dependent density functional theory or Bethe–Salpeter equation, is complicated by their non-Hermicity. Compared to their Hermitian Hamiltonian counterparts, computational methods for the treatment of non-Hermitian Hamiltonians are often less efficient and less stable, sometimes leading to the breakdown of the method. Recently [Grüning et al. Nano Lett. 8 (2009) 2820], we have identified that such Hamiltonians are usually pseudo-Hermitian. Exploiting this property, we have implemented an algorithm of the Lanczos type for Random-Phase Approximation Hamiltonians that benefits from the same stability and computational load as its Hermitian counterpart, and applied it to the study of the optical response of carbon nanotubes. We present here the related theoretical grounds and technical details, and study the performance of the algorithm for the calculation of the optical absorption of a molecule within the Bethe-Salpeter equation framework.

1. Introduction

The Random-Phase Approximation (RPA) Hamiltonian H^{RPA} appears in several areas of physics and theoretical chemistry, and describes strong collective excitations of a many-body system as the linear combination of particle–hole pairs $|\lambda\mu\rangle$ [1,2]. It has the form

$$H^{\mathrm{RPA}} = \begin{pmatrix} R & C \\ -C^* & -R^* \end{pmatrix}, \tag{1}$$

where the resonant R and anti-resonant $-R^*$ blocks are Hermitian matrices in the subspace generated by particle–hole pairs propagating respectively forward ($|\lambda\mu\rangle$) and backward ($|\mu\tilde{\lambda}\rangle$) in time (in what follows α, λ indicate particles and β, μ holes), and the C and $-C^*$ blocks are symmetric matrices coupling the particle–hole pairs propagating forward and backward in time. The excitation energies and strengths of the many-body system are the eigensolutions of Eq. (1). Note that the RPA Hamiltonian is not Hermitian, thus its eigenvalues are not necessarily real.

In quantum chemistry, condensed matter physics, nanoscience, or nuclear physics, the RPA Hamiltonian appears within the state-of-the-art approaches for calculating the excitations in an electronic system: the time-dependent density functional theory [3] (TD-DFT) and the Bethe–Salpeter [4] (BS) equation [5]. In the commonly used approximations to TD-DFT (e.g. real exchange-correlation kernel) and BS equation (static screening of the interaction), *all the eigenvalues are real*. TD-DFT is particularly successful for finite systems, namely molecules and molecular clusters, while the BS approach is mostly used for extended systems, like periodic bulk solids and, in general, for systems where excitonic effects play an important role [6]. Nowadays, the application of these approaches to the computation of the time-dependent responses of more and more complex systems, such as large bio-molecules or nanostructures, poses the problem of efficient solution of the eigenproblem for H^{RPA}. For large matrices, the direct diagonalization is usually not possible, and one has to resort to iterative algorithms, such as the Lanczos method. Such algorithms exist for both Hermitian and non-Hermitian Hamiltonian. However, compared to their Hermitian Hamiltonian counterparts, algorithms for the treatment of non-Hermitian Hamiltonians are often less efficient and less stable, sometimes leading to the breakdown of the method [7,8].

Within TDDFT, the very convenient Hermitian formulation of the eigenvalue problem proposed by Casida [9] exists. However its application is limited to finite systems and purely local effective potentials for which the H^{RPA} is real. The presence of e.g. spin-orbit coupling prevents the application of Casida's approach. In general a

* Corresponding author at: Centre for Computational Physics and Physics Department, University of Coimbra, Rua Larga, 3004-516 Coimbra, Portugal.
E-mail address: myrta@fis.uc.pt (M. Grüning).

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further approximation is introduced, the so-called Tamm–Dancoff approximation (TDA), that considers only particle–hole pairs propagating forward in time, so that the TDA Hamiltonian corresponds just to the resonant part, $H^{\mathrm{TDA}} = R$. The TDA is often sufficiently accurate, as in the case of optical absorption spectra of periodic bulk systems. On the other hand, the TDA becomes inaccurate or even unphysical in the case of electron-energy-loss spectra [10], reflectivity spectra [11], and also for the optical absorption of low-dimensional systems, e.g. nanosystems or π-conjugated molecules [12,13]. In a previous work [12] we have implemented an approach for the solution of the RPA Hamiltonian, that avoids the TDA, and still benefits of the efficiency and robustness of the algorithms for the Hermitian case. This approach has been already successfully applied to the calculation of the optical absorption and energy-loss spectra of a carbon nanotube. While our previous work focussed on the implications of the TDA for nanoscale systems, in this work the focus is on the theoretical grounds and some more technical aspects of that approach. We show here how the Lanczos algorithm for Hermitian eigenproblem (Section 2.1) can be used for the RPA Hamiltonian, that is pseudo-Hermitian with real eigenvalues (Section 2.3), by simply redefining the inner product (Section 3.1). We obtain (Section 3.2) the generalization to complex matrices of the scheme proposed by Van der Vorst [14]. The obtained algorithm is then further specialized (Section 3.3) to the calculation of the macroscopic dielectric function (from which the optical absorption and energy-loss spectra are derived) and finally applied to the calculation of the optical response of the trichloro-bezene isomers within the BS equation framework (Section 4), to show the algorithm accuracy (Section 4.2) and efficiency (Section 4.3).

2. Mathematical background

This section reviews briefly the two key "ingredients" of the presented approach: the Lanczos method for the solution of (non-)Hermitian eigenproblems, and the definition of pseudo-Hermitian matrix. The Lanczos method allows to calculate by recursion the eigenvalues, and eigenvectors, or directly the response spectrum, of large matrices. The pseudo-Hermicity is related to the reality of the eigenvalues of a matrix and with the possibility of transforming the matrix into a Hermitian matrix.

2.1. Lanczos method for Hermitian eigenproblems

The Lanczos recursion method [7] is a general algorithm for solving eigenproblems for a Hermitian operator H. This algorithm recursively builds an orthonormal basis $\{|q_i\rangle\}$ (Lanczos basis) in which H is represented as a real symmetric tridiagonal matrix,

$$T^k = \begin{pmatrix} a_1 & b_2 & 0 & \cdots & 0 \\ b_2 & a_2 & b_3 & & \vdots \\ 0 & \ddots & \ddots & \ddots & 0 \\ \vdots & & b_{k-1} & a_{k-1} & b_k \\ 0 & \cdots & 0 & b_k & a_k \end{pmatrix}. \tag{2}$$

The first vector $|q_1\rangle$ of the Lanczos basis is set equal to a (normalized) given vector $|u_0\rangle/\|u_0\|$. The next vectors are calculated from the three-term relation

$$|Q_{j+1}\rangle = H|q_j\rangle - a_j|q_j\rangle - b_j|q_{j-1}\rangle, \tag{3}$$

where

$$a_j = \langle q_j|H|q_j\rangle, \tag{4}$$

$$b_{j+1} = \|Q_{j+1}\|, \tag{5}$$

$$|q_{j+1}\rangle = |Q_{j+1}\rangle/b_{j+1}. \tag{6}$$

The algorithm is schematically described in Figs. 1 and 2. In steps (A)–(D) the variables are initialized before entering the conditional loop [steps (E)–(K)]. Here, at each iteration a new vector of the Lanczos basis is computed till the convergence criteria is met. The cost per iteration is given mainly by the matrix–vector multiplication at step (K), that is of $O(N^2)$ for non-sparse matrices, with N the size of H. In terms of memory and storage, if one is just interested in the eigenvalues, at each iteration only three vectors ($|q_{n-1}\rangle, |q_n\rangle, |q_{n+1}\rangle$) are needed, and only two reals (a_i, b_i) need to be stored. At the end of the process one gets the tridiagonal matrix of Eq. (2) of dimension $k \times k$, that can be diagonalized with a cost $\propto k$. Compared with the standard diagonalization, the advantages are the memory usage, and the computational cost $\propto kN^2$ (for diagonalization is $O(N^3)$) as soon as the number of iterations $k \ll N$. This is in practice always the case when we are interested only in a portion of the spectrum of H [15].

As first highlighted by Haydock [16,17], an additional advantage of Lanczos recursive approach is the possibility of calculating the resolvent $(\omega - H)^{-1}$ matrix elements, bypassing completely the diagonalization. In fact the resolvent for the state $|u_0\rangle$ takes the form of a continued fraction.

$$\langle u_0|(\omega - H)^{-1}|u_0\rangle = \|u_0\|^2 \cfrac{1}{(\omega - a_1) - \cfrac{b_2^2}{(\omega - a_2) - \cfrac{b_3^2}{\ddots}}}. \tag{7}$$

Other matrix elements can be then calculated by recursion (see Appendix A).

2.2. Lanczos method for non-Hermitian eigenproblems

The Lanczos recursive approach can be extended to the non-Hermitian case [7]. For a non-Hermitian matrix H, that we suppose diagonalizable, the action on a ket $|v\rangle$ differs from the action on a bra $\langle v|$: no orthogonal basis set exists, that could transform it into a diagonal form. The most straightforward extension of the Lanczos procedure illustrated in the previous subsection is the Arnoldi recursive approach that transforms H into an upper-Hessenberg matrix, instead of a tridiagonal one, and thus presents clear computational disadvantages with respect to the Hermitian case [18].

$b_0 \Leftarrow \|u_0\|$ (A)
$|s\rangle \Leftarrow |u_0\rangle/b_0$ (B)
$|t\rangle \Leftarrow H|s\rangle$ (C)
$b_1 \Leftarrow 0, |r\rangle \Leftarrow 0$ (D)
$i \Leftarrow 1$
until CONVERGENCE do
$a_i \Leftarrow \langle s|t\rangle$ (E)
$|t\rangle \Leftarrow |t\rangle - a_i|s\rangle - b_i|r\rangle$ (F)
$b_{i+1} \Leftarrow \|t\|$ (G)
$|r\rangle \Leftarrow |s\rangle$ (H)
$|s\rangle \Leftarrow |t\rangle/b_{i+1}$ (I)
$|t\rangle \Leftarrow H|s\rangle$ (K)
$i \Leftarrow i + 1$

Fig. 1. Hermitian Lanczos algorithm.