Minimization Principle for Linear Response Eigenvalue Problem II: Computation

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Abstract

In Part I of this paper we presented a trace minimization principle and related theoretical results for the linear response eigenvalue problem. Here we develop best approximations of the few smallest eigenvalues with the positive sign via a structure-preserving subspace projection. Then we present a four-dimensional subspace search conjugate gradient-like algorithm for simultaneously computing these eigenvalues and their associated eigenvectors. Finally, we present numerical examples to illustrate convergence behaviors of the proposed methods with and without preconditioning.

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1 Introduction

This is the second paper of ours in a sequel of two. Building upon the theoretical results in [2], here we will focus on the numerical aspect of the LR eigenvalue problem:

$$Hz \equiv \begin{pmatrix} 0 & K \\ M & 0 \end{pmatrix} \begin{pmatrix} y \\ x \end{pmatrix} = \lambda \begin{pmatrix} y \\ x \end{pmatrix} \equiv \lambda z, \tag{1.1}$$

where K and M are $n \times n$ symmetric positive semi-definite matrices and one of them is definite. It is an equivalent problem obtained from the original LR (a.k.a. Random Phase Approximation (RPA)) eigenvalue problem:

$$\begin{pmatrix} A & B \\ -B & -A \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} = \lambda \begin{pmatrix} u \\ v \end{pmatrix} \tag{1.2}$$

by an orthogonal similarity transformation to give $K = A - B$ and $M = A + B$ [2], where A and B are $n \times n$ real symmetric matrices such that the symmetric matrix $\begin{pmatrix} A & B \\ B & A \end{pmatrix}$ is positive definite [35, 41]. Note that if obtained from the original LR eigenvalue problem, K and M are both definite but here we relax this condition to one of them being definite.

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As noted in [3], the eigenvalue problem (1.1) is equivalent to any one of the following product eigenvalue problems

$$KMy = \lambda^2 y, \tag{1.3a}$$

$$MKx = \lambda^2 x. \tag{1.3b}$$

Their equivalences have led to solving (1.1) through solving one of the eigenvalue problems in (1.3). They also imply that the eigenvalues of H come in $\pm\lambda$ pairs. As in [2], we will denote the positive eigenvalues[1] of H by λ_i $(1 \le i \le n)$ and

$$-\lambda_n \le \cdots \le -\lambda_2 \le -\lambda_1 \le 0 \le \lambda_1 \le \lambda_2 \le \cdots \le \lambda_n.$$

The eigenvalue problem (1.2) has the same eigenvalues $\pm\lambda_i$.

An important minimization principle of Thouless [40] is

$$\lambda_1 = \min_{u,v} \varrho(u, v), \tag{1.4}$$

where $\varrho(u, v)$ is defined by

$$\varrho(u, v) = \frac{\begin{pmatrix} u \\ v \end{pmatrix}^{\mathrm{T}} \begin{pmatrix} A & B \\ B & A \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix}}{|u^{\mathrm{T}}u - v^{\mathrm{T}}v|}, \tag{1.5}$$

and the minimization is taken among all vectors u, v such that $u^{\mathrm{T}}u - v^{\mathrm{T}}v \neq 0$. For H, this minimization principle translates into

$$\lambda_1 = \min_{x,y} \rho(x, y), \tag{1.6}$$

where

$$\varrho(u, v) \equiv \rho(x, y) \stackrel{\text{def}}{=} \frac{x^{\mathrm{T}}Kx + y^{\mathrm{T}}My}{2|x^{\mathrm{T}}y|}, \tag{1.7}$$

where the minimization is taken among all x and y such that either $x^{\mathrm{T}}y \neq 0$ or $x^{\mathrm{T}}y = 0$ but $x^{\mathrm{T}}Kx + y^{\mathrm{T}}My > 0$. This removes those x and y that annihilate both the numerator and the denominator from the domain. In particular $x = y = 0$ is excluded.

Thouless' minimization principle (1.4) and consequently the induced (1.6) for H were proved under the condition that both $A \pm B$ (thus K and M, too) are symmetric positive definite. In [2], they were extended to include the case when one of K and M is definite.

Since the RPA and linear response theory was proposed by Bohm and Pines for studying the collective motion of many particles in the early 1950's [7], the development of numerical methods for solving the eigenvalue problem (1.2) and equivalently (1.1) has been an active research subject in computational (quantum) physics and chemistry for over four decades. In [9], it was suggested to solve the equivalent product eigenvalue problem (1.3a) instead by converting it to the symmetric eigenvalue problem of $R^{\mathrm{T}}KR$ using the Cholesky decomposition of $M = R^{\mathrm{T}}R$. In [29, 38], Davidson's algorithm for the symmetric eigenvalue problem was extended to the large scale eigenvalue problem (1.2). In [14, 15, 43, 44, 45], Lanczos-like algorithms were proposed. Given the minimization principle (1.4) or equivalently (1.6), conjugate gradient (CG) methods become nature choices for finding the smallest positive eigenvalue and indeed they have (see for example [25, 27]).

[1] Note our convention of assigning the positive sign to half of the eigenvalues 0 and the negative sign to the other half in [2].

Meanwhile, the eigenvalue problems in the forms of (1.1), (1.3), and (1.2) have also attracted a great deal of attention in numerical analysis community over the past four decades, too. As early as in 1960's, Wilkinson discussed the product eigenvalue problems (1.3) arising from theoretical physics, and proposed the method of transforming them to the standard symmetric eigenvalue problems by using the Cholesky decomposition [48, p.35, p.337]. Wilkinson's method is implemented as LAPACK's subroutine `xSYGVD` [1], where the product eigenvalue problems in (1.3) are classified as *the types 2 and 3* of the generalized symmetric definite eigenvalue problems, respectively. Alternatively the structure-preserving GR algorithm, a generalization of the well-known QR algorithm, can also be used for small to medium size problems [47, Chapter 8]. For large and sparse cases, the Lanczos algorithm, Krylov-Schur algorithm and Jacobi-Davidson algorithm all have been generalized to the product eigenvalue problems (see [17, 21, 46]). On the other hand, since the RPA eigenvalue problem (1.2) is a special case of the *Hamiltonian matrix eigenvalue problem*, an extension of the QR algorithm made for Hamiltonian matrix eigenvalue problems can be used to solve the problems of small to medium sizes [6, 8, 12, 47]. In particular, the work [12] treated a more general linear response eigenvalue problem. Algorithms for large scale Hamiltonian eigenvalue problems can be found in [4, 5] and references therein. An RPA test case is given in [4] to illustrate the computational efficiency of a Hamiltonian Krylov-Schur-type algorithm.

A recent survey study [42] compared four numerical methods (namely Lanczos, Arnoldi, Davidson, and CG) and discussed the limitations of each of these methods for developing an efficient linear-scaling eigensolver for the RPA eigenvalue problem (1.2). In the study, severe limitations were experienced for the Lanczos-type methods due to the orthogonality constraints (also see [43]), for the CG type methods to compute several eigenpairs simultaneously, and for incorporating preconditioning techniques (see also [25]).

In [2], we obtained a trace (or subspace) version of (1.6):

$$\sum_{i=1}^{k} \lambda_i = \frac{1}{2} \inf_{U^{\mathrm{T}}V = I_k} \mathrm{trace}(U^{\mathrm{T}}KU + V^{\mathrm{T}}MV), \tag{1.8}$$

as well as Cauchy-like interlacing inequalities for a structure-preserving projection H_{SR} of H. Based on this newly developed theory, here we have an opportunity to develop efficient numerical methods for the LR eigenvalue problem (1.1) in the much same way as the conjugate gradient and Lanczos methods for solving the large scale symmetric eigenvalue problem. In particular, we will show an important computational implication of the minimization principle (1.8) that is that it lends itself to seek approximations to a cluster of smallest eigenvalues with the positive sign λ_i $(1 \le i \le k)$ simultaneously through minimizing the objective function $\mathrm{trace}(U^{\mathrm{T}}KU + V^{\mathrm{T}}MV)$ subject to $U^{\mathrm{T}}V = I_k$ and that $\mathrm{span}(U)$ and $\mathrm{span}(V)$ are restricted inside two suitably built subspaces $\mathcal{U}$ and $\mathcal{V}$, respectively:

$$\sum_{i=1}^{k} \lambda_i \approx \frac{1}{2} \inf_{\substack{U^{\mathrm{T}}V = I_k \\ \mathrm{span}(U) \subseteq \mathcal{U},\, \mathrm{span}(V) \subseteq \mathcal{V}}} \mathrm{trace}(U^{\mathrm{T}}KU + V^{\mathrm{T}}MV), \tag{1.9}$$

where $\mathrm{span}(U)$ denotes the subspace spanned by the column vectors of U.

The minimization problem in the right-hand side of (1.9) doesn't look easy to solve at first sight. But we obtain a structure-preserving projection matrix H_{SR} and show that the sum of its first k smallest eigenvalues with the positive sign is the infimum. In this sense, H_{SR} is the best projection matrix from the given subspaces $\mathcal{U}$ and $\mathcal{V}$, and solving its eigenvalue problem yields the best approximations to λ_i $(1 \le i \le k)$ and their associated eigenvectors. Moreover, H_{SR} has the same block structure as H. With these new developments, we will be able to construct efficient numerical algorithms that can compute several smallest eigenvalues with the positive sign

of H simultaneously. Indeed we will present our versions of locally optimal conjugate gradient type algorithms, including blocked versions for computing these smallest eigenvalues simultaneously and preconditioned versions for speedy convergence. An extended presentation about the steepest descent-like methods and their applications in *ab initio* calculation of optical absorption spectra appeared [36]. We point out that these new algorithms are not straightforward applications of the standard steepest decent and nonlinear conjugate gradient algorithms, but improved ones to take advantage of the best projection matrix H_{SR} we have uncovered. All these are made possible by our new theory, parallel to some of the well-known and important results for the symmetric eigenvalue problem [22, 32, 37].

The rest of this paper is organized as follows. Section 2 presents an algorithm to construct approximate eigenpairs for H, given a pair of approximate deflating subspaces $\{\mathcal{U}, \mathcal{V}\}$. It is derived from the result in [2] for the case when the subspaces do consist of a pair of deflating subspaces. Section 3 and appendix A discuss how to construct the best approximations to some of the eigenpairs of H, given a pair of approximate deflating subspaces $\{\mathcal{U}, \mathcal{V}\}$. The results in section 3 justifies the algorithm in section 2 from a different perspective. In section 4, we apply newly established minimization principles in [2] to derive CG type algorithms for computing a set of the smallest eigenvalues with the positive sign. In section 5, we present numerical results to illustrate the convergence behaviors of CG methods. Concluding remarks are in section 6.

Notation. We will follow the notation as specified at the end of section 1 in [2]. In particular, K, $M \in \mathbb{R}^{n\times n}$ are assumed, by default, to be symmetric positive semi-definite and one of which is definite, unless explicitly stated differently.

2 Approximate deflating subspaces

Recall that $\{\mathcal{U}, \mathcal{V}\}$ is a *pair of deflating subspaces* of $\{K, M\}$ if [2, subsection 2.2]

$$K\mathcal{U} \subseteq \mathcal{V} \quad \text{and} \quad M\mathcal{V} \subseteq \mathcal{U}. \tag{2.1}$$

Each such a pair will yield a subset of H's eigenvalues and corresponding eigenvectors associated with the eigenvalues in the subset [2, subsection 2.2]. But in practical computations, rarely pairs of exact deflating subspaces are known, only approximate ones. The question then arises: how to compute approximate eigenpairs of H given a pair of *approximate* deflating subspaces.

Let $\{\mathcal{U}, \mathcal{V}\}$ be a pair of *approximate* deflating subspaces with $\dim\mathcal{U} = \dim\mathcal{V} = \ell$ such that $W \stackrel{\text{def}}{=} U^{\text{T}}V$ is nonsingular. In [2, section 2], we defined a structure-preserving projection

$$H_{\text{SR}} = \begin{pmatrix} 0 & W_1^{-\text{T}}U^{\text{T}}KUW_1^{-1} \\ W_2^{-\text{T}}V^{\text{T}}MVW_2^{-1} & 0 \end{pmatrix}. \tag{2.2}$$

of H onto the pair of the subspaces $\{\mathcal{U}, \mathcal{V}\}$, where $W_i \in \mathbb{R}^{\ell\times\ell}$ are from factorizing $W = W_1^{\text{T}}W_2$ and nonsingular. This H_{SR} in many ways, as will become clear later, play the same role for H as the Rayleigh quotient matrix for the symmetric eigenvalue problem.

Theorem 2.6 in [2] shows how to construct the eigenpairs of H from those of H_{SR} when $\{\mathcal{U}, \mathcal{V}\}$ *is* a pair of deflating subspaces of $\{K, M\}$. The way of construction there naturally leads us to propose the following algorithm.

> **Algorithm 2.1.** Given the basis matrices U and V of an approximate deflating subspaces $\{\mathcal{U}, \mathcal{V}\}$ of $\{K, M\}$, this algorithm returns approximate eigenvalues and eigenvectors for H as follows.
>
> 1. Construct H_{SR} as in (2.2) if $U^{\text{T}}V$ is nonsingular;

2. Compute the eigenpairs $\left\{\hat{\lambda}, \begin{pmatrix} \hat{y} \\ \hat{x} \end{pmatrix}\right\}$ of H_{SR};

3. The computed eigenvalues $\hat{\lambda}$ approximate some eigenvalues of H, and the associated approximate eigenvectors can be recovered as $\begin{pmatrix} VW_2^{-1}\hat{y} \\ UW_1^{-1}\hat{x} \end{pmatrix}$.

In view of [2, Theorem 2.7], as far as the eigenvalue problem of H is concerned, in theory any one of H_{SR} associated with a given pair of *approximate* deflating subspaces is just as good as another. Numerically, however, we should pick basis matrices that are sufficiently well-conditioned, like with orthonormal columns.

REMARK **2.1.** For this subsection, our default assumption on $K, M \in \mathbb{R}^{n\times n}$ is not required. $\diamond$

3 Best approximations by a pair of subspaces

Two most important aspects in solving a large scale eigenvalue problem are

1. building subspaces which the desired eigenvectors (or invariant subspaces) are close to, and

2. seeking "*best possible*" approximations from the suitably built subspaces.

In this section, we shall address the second aspect for our current problem at hand, i.e., seeking "*best possible*" approximations to a few smallest eigenvalues with the positive sign of H and their associated eigenvectors from given pair of subspaces. We will prove that H_{SR} provides best approximations. We leave the first aspect to the later sections when we present our computational algorithms.

The concept of "*best possible*" comes with a quantitative measure as to what constitutes "*best possible*". There may not be such a measure in general. But for the eigenvalue problem here, each of the minimization principles we established in [2] provides a quantitative measure.

Recall the default assumption that K, $M \in \mathbb{R}^{n\times n}$ are symmetric positive semi-definite and one of them is definite. Let $\{\mathcal{U}, \mathcal{V}\}$ be a pair of approximate deflating subspaces of $\{K, M\}$ and $\dim(\mathcal{U}) = \dim(\mathcal{V}) = \ell$. Motivated by the minimization principles in [2] we would seek

1. the best approximation to λ_1 in the sense of

$$\inf_{x\in\mathcal{U},\, y\in\mathcal{V}} \rho(x, y) \tag{3.1}$$

   and its associated approximate eigenvector;

2. the best approximations to λ_j $(1 \le j \le k)$ in the sense of

$$\frac{1}{2} \inf_{\substack{\text{span}(\hat{U})\subseteq\mathcal{U},\text{span}(\hat{V})\subseteq\mathcal{V} \\ \hat{U}^{\mathrm{T}}\hat{V}=I_k}} \text{trace}(\hat{U}^{\mathrm{T}}K\hat{U} + \hat{V}^{\mathrm{T}}M\hat{V}) \tag{3.2}$$

   and their associated approximate eigenvectors. Necessarily $k \le \ell$.

To this end, we divide our investigation into two cases. Let U, $V \in \mathbb{R}^{n\times\ell}$ be the basis matrices of $\mathcal{U}$ and $\mathcal{V}$, respectively, and set $W = U^{\mathrm{T}}V$. The two cases are

1. $W = U^{\mathrm{T}}V$ is nonsingular;

2. $W = U^{\mathrm{T}}V$ is singular.

For the first case, i.e., $W = U^{\mathrm{T}}V$ is nonsingular. Factorize $W = W_1^{\mathrm{T}}W_2$, where $W_i \in \mathbb{R}^{\ell\times\ell}$ are nonsingular. How this factorization is done is not essential mathematically. But it is included to accommodate cases when such a factorization may offer certain conveniences. In general, simply taking $W_1 = W^{\mathrm{T}}$ and $W_2 = I_\ell$ or $W_1 = I_\ell$ and $W_2 = W$ may just be good enough.

For the best approximation to λ_1 by (3.1), we note that any $x \in \mathcal{U}$ and $y \in \mathcal{V}$ can be written as $x = U\hat{u}$ and $y = V\hat{v}$ for some $\hat{u}$, $\hat{v} \in \mathbb{R}^\ell$ and vice versa. Therefore, we have

$$\begin{aligned}\rho(x,y) &= \frac{\hat{u}^{\mathrm{T}}U^{\mathrm{T}}KU\hat{u} + \hat{v}^{\mathrm{T}}V^{\mathrm{T}}MV\hat{v}}{2|\hat{u}^{\mathrm{T}}W\hat{v}|} \\ &= \frac{\hat{x}^{\mathrm{T}}W_1^{-\mathrm{T}}U^{\mathrm{T}}KUW_1^{-1}\hat{x} + \hat{y}^{\mathrm{T}}W_2^{-\mathrm{T}}V^{\mathrm{T}}MVW_2^{-1}\hat{y}}{2|\hat{x}^{\mathrm{T}}\hat{y}|},\end{aligned} \tag{3.3}$$

where $\hat{x} = W_1\hat{u}$ and $\hat{y} = W_2\hat{v}$. By [2, Theorem 3.1], the quantity in (3.1) is the smallest eigenvalue with the positive sign of H_{SR} defined earlier in section 2:

$$H_{\mathrm{SR}} = \begin{pmatrix} 0 & W_1^{-\mathrm{T}}U^{\mathrm{T}}KUW_1^{-1} \\ W_2^{-\mathrm{T}}V^{\mathrm{T}}MVW_2^{-1} & 0 \end{pmatrix} \in \mathbb{R}^{2\ell\times 2\ell}. \tag{2.2}$$

Now turn to the best approximations to λ_j $(1 \le j \le k)$ by (3.2). Note that any $\hat{U}$ and $\hat{V}$ such that $\mathrm{span}(\hat{U}) \subseteq \mathcal{U}$, $\mathrm{span}(\hat{V}) \subseteq \mathcal{V}$, and $\hat{U}^{\mathrm{T}}\hat{V} = I_k$ can be written as

$$\hat{U} = UW_1^{-1}\hat{X}, \quad \hat{V} = VW_2^{-1}\hat{Y},$$

where $\hat{X}$, $\hat{Y} \in \mathbb{R}^{\ell\times k}$ and $\hat{X}^{\mathrm{T}}\hat{Y} = I_k$, and vice versa. Hence we have

$$\hat{U}^{\mathrm{T}}K\hat{U} + \hat{V}^{\mathrm{T}}M\hat{V} = \hat{X}^{\mathrm{T}}W_1^{-\mathrm{T}}U^{\mathrm{T}}KUW_1^{-1}\hat{X} + \hat{Y}^{\mathrm{T}}W_2^{-\mathrm{T}}V^{\mathrm{T}}MVW_2^{-1}\hat{Y}$$

and thus

$$\begin{aligned}&\inf_{\substack{\mathrm{span}(\hat{U})\subseteq\mathcal{U},\mathrm{span}(\hat{V})\subseteq\mathcal{V}\\ \hat{U}^{\mathrm{T}}\hat{V}=I_k}} \mathrm{trace}(\hat{U}^{\mathrm{T}}K\hat{U} + \hat{V}^{\mathrm{T}}M\hat{V}) \\ &\qquad = \inf_{\hat{X}^{\mathrm{T}}\hat{Y}=I_k} \mathrm{trace}(\hat{X}^{\mathrm{T}}W_1^{-\mathrm{T}}U^{\mathrm{T}}KUW_1^{-1}\hat{X} + \hat{Y}^{\mathrm{T}}W_2^{-\mathrm{T}}V^{\mathrm{T}}MVW_2^{-1}\hat{Y}).\end{aligned} \tag{3.4}$$

By [2, Theorem 3.2], we know that the right-hand side of (3.4) is the sum of the k smallest eigenvalues with the positive sign of H_{SR}.

In summary, the *best approximations to the first k eigenvalues with the positive sign of H within the pair of approximate deflating subspaces are the eigenvalues of H_{SR}*. Algorithmically, denote by μ_j $(j = 1,\ldots,\ell)$ the eigenvalues with the positive sign of H_{SR} in ascending order and by $\hat{z}_j$ the associated eigenvectors, i.e., $0 \le \mu_1 \le \cdots \le \mu_\ell$, and

$$H_{\mathrm{SR}}\hat{z}_j = \mu_j\hat{z}_j, \quad \hat{z}_j = \begin{pmatrix}\hat{y}_j \\ \hat{x}_j\end{pmatrix}. \tag{3.5}$$

It can be verified that

$$\rho(UW_1^{-1}\hat{x}_j, VW_2^{-1}\hat{y}_j) = \mu_j \quad \text{for } j = 1,\ldots,\ell.$$

Naturally, according to Algorithm 2.1, we take $\lambda_j \approx \mu_j$ and the corresponding approximate eigenvectors of H as

$$\widetilde{z}_j \equiv \begin{pmatrix}\widetilde{y}_j \\ \widetilde{x}_j\end{pmatrix} = \begin{pmatrix}VW_2^{-1}\hat{y}_j \\ UW_1^{-1}\hat{x}_j\end{pmatrix} \quad \text{for } j = 1,\ldots,\ell. \tag{3.6}$$

In practice, not all of the approximate eigenpairs $(\mu_j, \widetilde{z}_j)$ are equally accurate to the same level. Usually the first few pairs are more accurate than the rest.

For the ease of reference, we summarize the findings for the first case of nonsingular $W = U^{\mathrm{T}}V$ into the following theorem.

Theorem 3.1. *Suppose that one of K, $M \in \mathbb{R}^{n\times n}$ is definite. Let $\{\mathcal{U}, \mathcal{V}\}$ be a pair of approximate deflating subspaces of $\{K, M\}$ with $\dim(\mathcal{U}) = \dim(\mathcal{V}) = \ell$, and let U, $V \in \mathbb{R}^{n\times\ell}$ be the basis matrices of $\mathcal{U}$ and $\mathcal{V}$, respectively. If $W \overset{\text{def}}{=} U^{\mathrm{T}}V$ is nonsingular, then*

$$\sum_{j=1}^{k} \mu_j = \frac{1}{2} \inf_{\substack{\mathrm{span}(\hat{U})\subseteq\mathcal{U},\mathrm{span}(\hat{V})\subseteq\mathcal{V} \\ \hat{U}^{\mathrm{T}}\hat{V}=I_k}} \mathrm{trace}(\hat{U}^{\mathrm{T}}K\hat{U} + \hat{V}^{\mathrm{T}}M\hat{V}),$$

and the best approximations to λ_1 in the sense of (3.1) *or to λ_j ($1 \le j \le k$) in the sense of* (3.2) *are the eigenvalues $\{\mu_j\}$ of H_{SR} defined in* (2.2) *with the corresponding approximate eigenvectors given by* (3.6).

We recall that even though H_{SR} is not uniquely determined by the given subspaces $\mathcal{U}$ and $\mathcal{V}$, the approximate eigenpairs $(\mu_j, \widetilde{z}_j)$ are uniquely determined, as guaranteed by [2, Theorem 2.7].

It turns out the second case (namely W is singular) is much more complicated, but the conclusion is similar in that both optimization problems in (3.1) and (3.2) can still be solved through solving a smaller eigenvalue problem for a projection matrix $\hat{H}_{\mathrm{SR}}$ to be defined in appendix A. Due to its complexity in treatment, we defer the consideration for the second case to appendix A, where Theorem A.1 similar to Theorem 3.1 is obtained.

Remark **3.1.** The best approximation technique so far is based on the minimization principles in [2, Theorems 3.1 and 3.2]. Naturally one may wonder if a similar technique could be devised using the minimization principles in [2, Theorem 3.3] for the original LR eigenvalue problem (1.3) of [2]:

$$\begin{pmatrix} A & B \\ -B & -A \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} = \lambda \begin{pmatrix} u \\ v \end{pmatrix}.$$

But that seems hard, if at all possible. The difficulty lies in that there appears no good way to define a proper projection matrix of $\begin{pmatrix} A & B \\ B & A \end{pmatrix}$ or of $\begin{pmatrix} A & B \\ -B & -A \end{pmatrix}$ onto the given subspaces. $\diamond$

4 4-D CG algorithms

4.1 Partial gradients

The partial gradients of the Thouless functional $\rho(x, y)$ with respect to x and y will be needed later for minimization. To find the gradients, we perturb x and y to $x+p$ and $y+q$, respectively, where p and q are assumed tiny in magnitude. Assuming $x^{\mathrm{T}}y \neq 0$, we have for sufficiently tiny p and q, up to the first order in p and q,

$$\begin{aligned}
\rho(x+p, y+q) &= \frac{(x+p)^{\mathrm{T}}K(x+p) + (y+q)^{\mathrm{T}}M(y+q)}{2|(x+p)^{\mathrm{T}}(y+q)|} \\
&= \frac{x^{\mathrm{T}}Kx + 2p^{\mathrm{T}}Kx + y^{\mathrm{T}}My + 2q^{\mathrm{T}}My}{2|x^{\mathrm{T}}y + p^{\mathrm{T}}y + q^{\mathrm{T}}x|} \\
&= \frac{x^{\mathrm{T}}Kx + 2p^{\mathrm{T}}Kx + y^{\mathrm{T}}My + 2q^{\mathrm{T}}My}{2\,|x^{\mathrm{T}}y|} \left[1 - \frac{p^{\mathrm{T}}y + q^{\mathrm{T}}x}{x^{\mathrm{T}}y}\right] \\
&= \rho(x, y) + \frac{1}{x^{\mathrm{T}}y}\, p^{\mathrm{T}} \left[Kx - \rho(x, y)\, y\right] + \frac{1}{x^{\mathrm{T}}y}\, q^{\mathrm{T}} \left[My - \rho(x, y)\, x\right].
\end{aligned}$$

Therefore the partial gradients of $\rho(x,y)$ with respect to x and y are given by

$$\nabla_x \rho = \frac{1}{x^{\mathrm{T}} y}\left[Kx - \rho(x,y)\, y\right], \quad \nabla_y \rho = \frac{1}{x^{\mathrm{T}} y}\left[My - \rho(x,y)\, x\right]. \tag{4.1}$$

There is a close relation between these two partial gradients and the residual:

$$Hz - \rho(x,y) z \equiv \begin{pmatrix} 0 & K \\ M & 0 \end{pmatrix} \begin{pmatrix} y \\ x \end{pmatrix} - \rho(x,y) \begin{pmatrix} y \\ x \end{pmatrix} = x^{\mathrm{T}} y \begin{pmatrix} \nabla_x \rho \\ \nabla_y \rho \end{pmatrix}. \tag{4.2}$$

Namely the block vector obtained by stacking $\nabla_x \rho$ over $\nabla_y \rho$ is parallel to the residual.

4.2 4-D search

The line search is a common approach in the process of optimizing a function value. For our case, we are interested in solving

$$\inf_{x,y} \rho(x,y) = \inf_{x,y} \frac{x^{\mathrm{T}} K x + y^{\mathrm{T}} M y}{2|x^{\mathrm{T}} y|} \tag{4.3}$$

in order to compute λ_1 and its associated eigenvector of H.

From the theoretical point of view, this task of minimizing $\rho(x,y)$ may end up with no optimal arguments because possibly no x and y attend the infimum, unless both K and M are definite. One may argue that in this case, λ_1 is already known, i.e., $+0$, when the infimum cannot be attained and it happens if one of K and M is singular. Naturally one can compute corresponding eigenvectors (by, e.g., the inverse iteration) and deflate out the eigenvalues 0. But in practice, that one of them is singular may not be known *a priori*, except that both are semi-definite is usually known from the problem setup. A likely scenario would be that one may still attempt to minimize $\rho(x,y)$ anyway. What would happen then? First numerically rarely a matrix is exactly singular. This means that the singular K or M is not actually singular (even might be slightly indefinite). With carefully written computer codes, one may safely regard the singular one barely definite. We find from our numerical tests that with a without approximately H^{-1}, any computation by minimizing $\rho(x,y)$ can still yield meaningful numerical results: the computed λ_1 is very tiny, as tiny as about down to $O(\|K\| + \|M\|)\boldsymbol{u}$, and one of x and y is negligible compared to the other, where $\boldsymbol{u}$ is the machine unit roundoff. Therefore, despite of the implied theoretical impasse by [2, Theorem 3.1] when one of K and M is singular, attempting to minimize $\rho(x,y)$, with a suitable without, is still a worthwhile thing to do in seeking λ_1 and its associated eigenvector of H.

Given a search direction $\begin{pmatrix} q \\ p \end{pmatrix}$ from the current position $\begin{pmatrix} y \\ x \end{pmatrix}$, the basic idea of the standard line search[2] is to look for the best possible scalar argument t on the line

$$\left\{ \begin{pmatrix} y \\ x \end{pmatrix} + t \begin{pmatrix} q \\ p \end{pmatrix} \,:\, t \in \mathbb{R} \right\} \tag{4.4}$$

to minimize ρ. Carrying out the line search, i.e., minimizing ρ along the line (4.4), is rather straightforward through differentiating $\rho(x+tp, y+tq)$ [3]. However we decided to give up this

[2]Since ρ is homogeneous of degree 0, i.e., $\rho(tx,ty) \equiv \rho(x,y)$ for any scalar t, minimizing ρ along the line (4.4) is in fact minimizing ρ in $\left\{ \alpha \begin{pmatrix} y \\ x \end{pmatrix} + \alpha t \begin{pmatrix} q \\ p \end{pmatrix} \,:\, \alpha,\, t \in \mathbb{R} \right\}$ which in general form a plan in $\mathbb{R}^{2n}$ spanned by $\begin{pmatrix} y \\ x \end{pmatrix}$ and $\begin{pmatrix} q \\ p \end{pmatrix}$, excluding the line $\left\{ t \begin{pmatrix} q \\ p \end{pmatrix} \,:\, t \in \mathbb{R} \right\}$. Therefore the standard line search becomes a defacto plane search for ρ. To be consistent with the standard terminology in optimization, we still call it *the line search*.

standard idea for reasons to be detailed in a moment. Instead, we shall look for four scalars α, β, s, and t to minimize

$$\rho(\alpha x + sp, \beta y + tq) = \frac{(\alpha x + sp)^{\mathrm{T}} K (\alpha x + sp) + (\beta y + tq)^{\mathrm{T}} M (\beta y + tq)}{2|(\alpha x + sp)^{\mathrm{T}} (\beta y + tq)|}.$$

This no longer performs a *line search*, but a *4-dimensional subspace search* (or, *4-D search* for short):

$$\inf_{\alpha,\beta,s,t} \rho(\alpha x + sp, \beta y + tq) = \min_{u \in \mathrm{span}(U),\ v \in \mathrm{span}(V)} \rho(u, v), \tag{4.5}$$

within the *4-dimensional subspace*

$$\left\{ \begin{pmatrix} \beta y + tq \\ \alpha x + sp \end{pmatrix} \quad \text{for all scalars } \alpha,\ \beta,\ s, \text{ and } t \right\}, \tag{4.6}$$

where $U = (x, p)$ and $V = (y, q)$. The right-hand side of (4.5) can be solved by the methods given in section 3 if $U^{\mathrm{T}}V$ is nonsingular (the common case) or in appendix A if $U^{\mathrm{T}}V$ is singular (the rare case).

We prefer our 4-D search to the standard line search along the line (4.4) for the following reasons:

1. While we have no formal proof, it seems that the standard line search cannot be recasted into a (much) smaller eigenvalue problem of a matrix having the same block structure as H: zero diagonal blocks and symmetric off-diagonal blocks.

2. The standard line search is not readily extensible to the subspace search, a crucial technique for our development for simultaneously computing few smallest eigenvalues with the positive sign and corresponding eigenvectors of H.

3. The standard line search yields the best possible approximation along the line (4.4) that is contained in the 4-dimensional subspace over which our 4-D search minimizes. Starting with the same p and q, the solution by our 4-D search is and can be much better at about the same cost.

4. Although we restricted our developments so far on real K and M, they are actually valid for Hermitian K and M after minor changes, i.e., replacing all transposes $(\cdot)^{\mathrm{T}}$ by complex conjugate transposes $(\cdot)^{\mathrm{H}}$. When K and M are Hermitian and some of their entries are complex, our 4-D search is truly a 4-D search over a 4-dimensional subspace in $\mathbb{C}^{2n}$ (the $2n$-dimensional Euclidean vector space over the complex field), whereas the standard line search does not minimize ρ over a straight line in $\mathbb{C}^{2n}$ because t is restricted to be real. Conceivably the standard line search solution could be even worse in the complex case.

4.3 4-D CG algorithms

The minimization principle in [2, (1.8)/(1.11)], and the newly established one in [2, Theorem 3.2] make it tempting to apply memory-efficient nonlinear CG algorithms [28] to solve LR (a.k.a. RPA) eigenvalue problems. Not surprisingly, such applications had been attempted in [25, 27] based on the Thouless functional ϱ in (1.5). Conceivably when only one eigenvalue and its associated eigenvector are requested, it matters little, if any, to apply CG to (1.4) for the Hamiltonian matrix in (1.2) or to (1.6) for H in (1.1). But it is a very different story if more than one eigenpairs are requested, in which case block algorithms become necessary. It seems hard, if at all possible, to create a block CG algorithm for the Hamiltonian matrix eigenvalue

problem (1.2) directly, even with our new minimization principle in [2, Theorem 3.3] for the same reason as we pointed out in Remark 3.1. On the other hand, the developments in section 3 and appendix A make it possible for designing efficient block CG algorithms to compute the first few smallest eigenvalues with the positive sign λ_j and their corresponding eigenvectors simultaneously, based on the minimization principle in [2, Theorem 3.2] and the Cauchy-like interlacing inequalities in [2, Theorem 3.4]. This is the precise reason we prefer to work with H.

The *locally optimal CG algorithm* [33, 39] was born as a result of

1. the observation that the next approximation from applying a (classical) nonloinear CG algorithm [28] to an optimization problem lies in the subspace spanned by the most recent approximation, the most recent searching direction, and the gradient at the most recent approximation, and

2. that subspace is the same as the one spanned by the two most recent approximations and the gradient, and thus

3. we should compute the next approximation as the optimal solution within that subspace.

It has been noted that the locally optimal CG algorithm is often better suited for solving large scale Hermitian eigenvalue problems, especially with a proper preconditioner [19, 20] than the (classical) nonlinear CG algorithms (see also [30]. It converges fast, has no parameters to worry about, and is still easy to implement. Inspired by this, we present in what follows our locally optimal CG algorithms (with or without preconditioners).

For many nonlinear optimization problems, even solving simple line searches poses challenges. But for the eigenvalue problem for H, thanks to Theorem 3.1 and Theorem A.1, the optimal approximate solution within a pair of subspaces of dimension higher than 1 is easily computed, very much like the case for the standard Hermitian eigenvalue problem for which Knyazev [20] proposed the *Locally Optimal Block Preconditioned Conjugate Gradient* method (LOBPCG). Theorem 3.1 and Theorem A.1 enable us to go for block CG algorithms as well.

Algorithm 4.1 below summarizes four *locally optimal 4-D CG algorithms* in one. We attach "4-D" to them because of their relation to the 4-D search idea in subsection 4.2. Their creation follows the idea of "*local optimality*" in the locally optimal CG algorithm in that each step the optimal solution is searched within the subspace spanned by the two most recent approximations and the partial gradients. Each of the four algorithms is realized through adjusting its integer parameter k and preconditioner Φ:

- *Locally Optimal 4-D CG algorithm* (LO4DCG): $k = 1$ and

$$\Phi = \begin{pmatrix} 0 & I_n \\ I_n & 0 \end{pmatrix}; \tag{4.7}$$

- *Locally Optimal Preconditioned 4-D CG algorithm* (LOP4DCG): $k = 1$ and $\Phi \approx (H - \mu I_{2n})^{-1}$;

- *Locally Optimal Block 4-D CG algorithm* (LOB4DCG): $k > 1$ and Φ as in (4.7);

- *Locally Optimal Block Preconditioned 4-D CG algorithm* (LOBP4DCG): $k > 1$ and[3] $\Phi \approx (H - \mu I_{2n})^{-1}$.

[3]Other possibilities include picking Φ to vary with CG steps and with the desired λ_j, i.e., Φ in (4.9) could be made dependent on i and j, for fast convergence. But doing so likely increase preconditioning cost.

The key iterative step in these locally optimal 4-D CG algorithms is to seek the best possible approximations in the subspace spanned by the two most recent approximations and the (preconditioned) gradients at the most recent approximations, except for the first iterative step for which the search subspace is simply spanned by the most recent approximations and the (preconditioned) gradients at the approximations. A straightforward application would be to search the next approximations within

$$\operatorname{span}\left\{\begin{pmatrix} y_j^{(i)} \\ x_j^{(i)} \end{pmatrix}, \begin{pmatrix} y_j^{(i-1)} \\ x_j^{(i-1)} \end{pmatrix}, \begin{pmatrix} q_j \\ p_j \end{pmatrix}, \quad 1 \le j \le k\right\}, \tag{4.8}$$

where the superscript $(i-1)$ and (i) indicate that they are for the $(i-1)$st and ith iterations, respectively, and

$$\begin{pmatrix} q_j \\ p_j \end{pmatrix} = \Phi \begin{pmatrix} \nabla_x \rho \\ \nabla_y \rho \end{pmatrix}\Bigg|_{(x,y)=(x_j^{(i)}, y_j^{(i)})}, \tag{4.9}$$

and Φ is a preconditioner. For the first iteration, the vectors in (4.8) with the superscript $(i-1)$ should be deleted from the list because they are not available yet. To utilize the best approximation methods in section 3 and appendix A, we modify this approach by using the search space

$$\operatorname{span}\left\{\begin{pmatrix} y_j^{(i)} \\ 0 \end{pmatrix}, \begin{pmatrix} y_j^{(i-1)} \\ 0 \end{pmatrix}, \begin{pmatrix} q_j \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ x_j^{(i)} \end{pmatrix}, \begin{pmatrix} 0 \\ x_j^{(i-1)} \end{pmatrix}, \begin{pmatrix} 0 \\ p_j \end{pmatrix} \quad 1 \le j \le k\right\}.$$

Breaking each vector into two in such a way is a common technique today in developing structure-preserving algorithms (see, e.g., [18, 23]). We are now ready to give our four locally optimal CG algorithms collectively in one.

Algorithm 4.1. The *locally optimal block preconditioned 4-D CG algorithms:*

0 Given initial approximations X_0 and Y_0 having k columns such that columns of $Z_0 = \begin{pmatrix} Y_0 \\ X_0 \end{pmatrix}$ are approximate eigenvectors of H associated with λ_j, $1 \le j \le k$.
1 for $i = 0, 1, \ldots$ until convergence:
2 $\quad \rho_j = \rho((X_i)_{(:,j)}, (Y_i)_{(:,j)})$, $1 \le j \le k$;
3 $\quad P_i = KX_i - Y_i \operatorname{diag}(\rho_1, \ldots, \rho_k)$, $Q_i = MY_i - X_i \operatorname{diag}(\rho_1, \ldots, \rho_k)$;
3.1 $\quad \begin{pmatrix} Q_i \\ P_i \end{pmatrix} \leftarrow \Phi \begin{pmatrix} P_i \\ Q_i \end{pmatrix}$ if the preconditioner Φ is given;
4.1 $\quad$ For $i = 0$: $U = (X_i, P_i)$, $V = (Y_i, Q_i)$;
4.2 $\quad$ For $i > 0$: $U = (X_i, X_{i-1}, P_i)$, $V = (Y_i, Y_{i-1}, Q_i)$;
4.3 $\quad$ Orthogonalize the columns of U and V;
4.4 $\quad W = U^{\mathrm{T}}V = W_1^{\mathrm{T}} W_2$;
5 $\quad$ Construct H_{SR} as in (2.2) (assume W is nonsingular);
6 $\quad$ Compute the k smallest eigenvalue with positive sign of H_{SR}, and the associated eigenvectors as in (3.5);
7 $\quad X_{i+1} = UW_1^{-1}(\hat{x}_1, \ldots, \hat{x}_k)$, $Y_{i+1} = VW_2^{-1}(\hat{y}_1, \ldots, \hat{y}_k)$;
8 $\quad$ Normalize each column of $Z_{i+1} = \begin{pmatrix} Y_{i+1} \\ X_{i+1} \end{pmatrix}$.
9 end

A few comments are in order for Algorithm 4.1:

1. At Line 2, evaluations of ρ_j are needed only for sweep $i = 0$; for $i \geq 1$, they are the k smallest eigenvalues with the positive sign of H_{SR} in the previous sweep.

2. For the convergence test, we can use the relative residual norm

$$\frac{\|Hz_j^{(i)} - \rho(x_j^{(i)}, y_j^{(i)})z_j^{(i)}\|}{\left[\|H\| + \rho(x_j^{(i)}, y_j^{(i)})\right] \|z_j^{(i)}\|}$$

   to determine if the approximate eigenpair $(\rho(x_j^{(i)}, y_j^{(i)}), z_j^{(i)})$ has converged to a desired accuracy, where $z_j^{(i)} = (Z_i)_{(:,j)}$, $x_j^{(i)} = (X_i)_{(:,j)}$, and $y_j^{(i)} = (Y_i)_{(:,j)}$, and $\|\cdot\|$ is some matrix/vector norm, e.g., the ℓ_1-vector norm and ℓ_1-operator norm.

3. U and V constructed at Line 4.1 or Line 4.2 may be ill-conditioned, especially when near convergence because then the gradients tend to the zero vector and X_i and Y_i are almost converged. To ensure that U and V are well-conditioned for better numerical stability, we may have to orthogonalize their columns via, e.g., the (classical/modified) Gram-Schmidt orthogonalization process. This is the reason we have Line 4.3 there.

4. From Line 5 to Line 8, we leave out the case when $U^{\text{T}}V$ is singular for simplicity. Actual implementation should include the case for which the optimal solution has been given in detail in appendix A. Specifically, instead of H_{SR} as in (2.2), we compute $\hat{H}_{\text{SR}}$ as in (A.5) and its $\min\{k, r\}$ smallest eigenvalues with the positive sign and the associated eigenvectors as in (A.7), and finally compute $Z_{i+1} = (\widetilde{z}_1, \ldots, \widetilde{z}_{\min\{k,r\}})$ by (A.8) – (A.10), where r is the (numerical) rank of W.

   There are two factors that affect the (non)singularity of $U^{\text{T}}V$: 1) the choices of U and V as the basis matrices of $\mathcal{U} = \text{span}(X_i, X_{i-1}, P_i)$ and $\mathcal{V} = \text{span}(Y_i, Y_{i-1}, Q_i)$, respectively, and 2) the angle $\angle(\mathcal{U}, \mathcal{V})$ between the two subspaces. The first factor can be alleviated by picking orthonormal bases as suggested at Line 4.3, albeit an expensive step. A similar issue came up in the standard LOBPCG [20], too, and essentially the same idea as in Line 4.3 was suggested in [16]. But the second factor is an intrinsic one [2, Lemma 2.2] and we must then resort the technique detailed in appendix A. Even with the technique, a complete and robust implementation must confront the important issue of deciding the numerical rank of $U^{\text{T}}V$ to balance convergence speed and numerical stability. This will be one of the issues that we will look into in our future study.

5. At Line 6, LAPACK's subroutine `xSYSVD` can be used to solve the eigenvalue problem of H_{SR} because of its small size. In theory half of the eigenvalues of H_{SR} have the positive sign and the other half are opposite. But when the off-diagonal blocks of H_{SR} is semi- or barely definite, some of the tiny eigenvalues with the positive sign may be computed by `xSYSVD` negative or even complex with tiny magnitude. This can be easily detected and corrected by a simple post-processing.

6. At Line 8, we can simply scale each column of Z_{i+1} to be a unit vector in some vector norm.

7. Sometimes it can be helpful to use a k that is somewhat bigger than the actual number of requested eigenpairs for the acceleration of convergence.

8. The algorithm without Line 4.2, using Line 4.1 for all i, and without the preconditioner Φ is reminiscent of the so-called *Simultaneous Rayleigh Quotient Minimization Method* (SIRQIT) due to Longsine and McCormick [24] for the standard Hermitian eigenvalue problem.

9. Φ as in (4.7) gives the plain 4-D CG algorithm (i.e., without preconditioning). An efficient preconditioner to compute the eigenvalues of H close to a prescribed point μ is

$$\Phi = (H - \mu I_{2n})^{-1}.$$

Then the vectors p_j and q_j defined by (4.9) can be computed through approximately solving a linear system with the coefficient matrix $(H - \mu I_{2n})^{-1}$ in practice. Note that the arrangement of the two n-entry blocks in the vector applied to by Φ is not mistaken. In fact the vector is parallel to the corresponding residual vector as given by (4.2). The modified directions are parallel to the ones obtained from one step of the inverse power iteration on the residual. When μ is closer to the desired eigenvalues than any others, the preconditioned directions should have "larger" components in the desired eigenvectors than the ones obtained without preconditioning. Since we are particularly interested in the smallest eigenvalues with the positive sign, $\mu = 0$ is often an obvious choice. Then

$$\Phi \begin{pmatrix} \nabla_x \rho \\ \nabla_y \rho \end{pmatrix} = \begin{pmatrix} 0 & M^{-1} \\ K^{-1} & 0 \end{pmatrix} \begin{pmatrix} \nabla_x \rho \\ \nabla_y \rho \end{pmatrix} = \begin{pmatrix} M^{-1} \nabla_y \rho \\ K^{-1} \nabla_x \rho \end{pmatrix} =: \begin{pmatrix} q \\ p \end{pmatrix}.$$

In this case, both p and q can be computed by using the conjugate gradient method [10, 13]. The search direction in the x-component depends only on $\nabla_x \rho$ while the search direction in the y-component depends only on $\nabla_y \rho$. This in part also justifies the correct block ordering in the vector applied to by Φ in (4.9).

10. The rough flop counts per step, excluding the pre-conditioning part as it may vary with implementation and ignoring those of $O(k^3)$ and $O(nk)$ or less, are as follows.

    (a) Residual computations at Line 3: $2k$ matrix-vector multiplications by K or M;

    (b) Orthogonalzations by, e.g., modified Gram-Schmidt process, at Line 4.3 [13, p.232]: $16nk^2$ (for $i = 0$) or $36nk^2$ (for $i > 0$) flops;

    (c) Forming H_{SR}: $4k$ (for $i = 0$) or $6k$ (for $i > 0$) matrix-vector multiplications by K or M, plus $8nk^2$ (for $i = 0$) or $18nk^2$ (for $i > 0$) flops;

    (d) Forming W at Line 4.4: $8nk^2$ (for $i = 0$) or $18nk^2$ (for $i > 0$) flops;

    (e) Forming X_{i+1} and Y_{i+1} at Line 7: $4nk^2$.

    In summary, the cost per step, excluding the pre-conditioning part, is $6k$ (for $i = 0$) or $8k$ (for $i > 0$) matrix-vector multiplications by K or M, plus $36nk^2$ (for $i = 0$) or $76nk^2$ (for $i > 0$) flops.

5 Numerical examples

In this section, we present some numerical experiment results obtained within the MATLAB environment to illustrate the essential convergence behaviors of locally optimal 4-D CG algorithms in section 4. More tests on much larger scale LR problems will be in our future work. Just recently, in collaboration with computational chemists, in [36] we successfully solve an LR problem with $2n = 5,650,410$ by the *4-D Block Steepest Descent* method (4DBSD) which is Algorithm 4.1 with Line 4.2 deleted and Line 4.1 used for all i with no preconditioner.

For the first example, we use the pair of matrices K and M of H (1.1) generated from the linear response analysis of the density matrix calculated by the Quantum ESPRESSO, an electronic structure calculation code that implements density functional theory (DFT) using plane-waves as a basis set and pseudopotentials [11]. For simplicity, we use a synthesized pair of

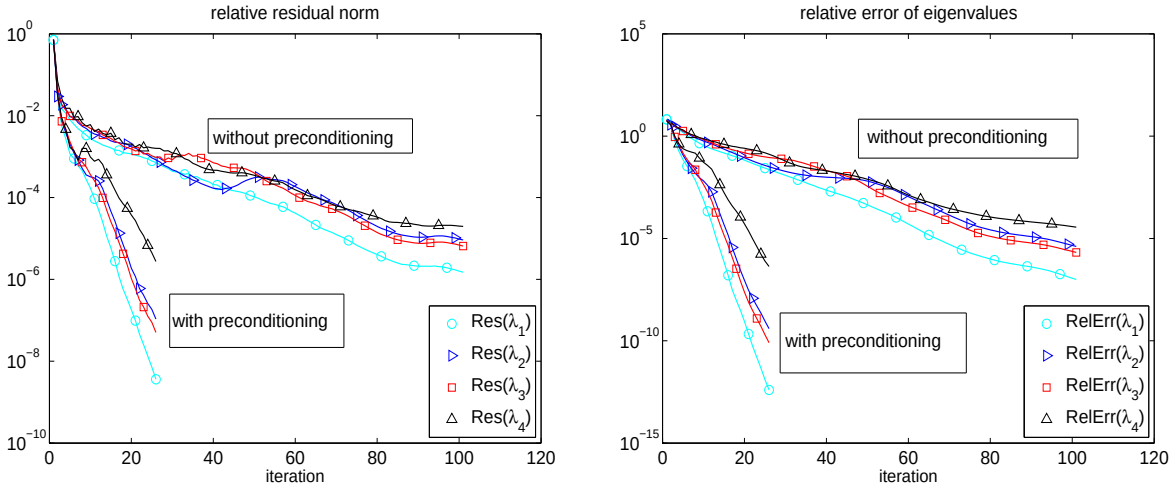

Figure 5.1: The convergence behaviors of the locally optimal block 4-D CG algorithms with/without preconditioning for computing the 4 smallest positive eigenvalues (excitation states) of a synthesized bi-atomic molecule: relative residual norms (left) and relative eigenvalue errors (right).

matrices K and M for the sodium dimer Na_2, namely a simple biatomic molecule. Such small molecules are often used as benchmark tests to assess various simulation models, functionals and methods (for example see [26]). Both K and M are symmetric positive definite and of order $n = 1862$.

Our goal is to compute 4 smallest positive eigenvalues $0 < \lambda_1 < \lambda_2 < \lambda_3 < \lambda_4$ and corresponding eigenvectors z_1, z_2, z_3, z_4 of H. The initial approximate eigenvectors of z_i are chosen as $(e_j^{\mathrm{T}}, e_j^{\mathrm{T}})^{\mathrm{T}}$ for $j = 1, 2, 3, 4$. The relative residual norms and relative eigenvalue errors for the jth approximate eigenpair $(\lambda_j^{(i)}, z_j^{(i)})$ at the ith iterative step to the exact jth eigenpairs (λ_j, z_j) are defined by

$$\frac{\|Hz_j^{(i)} - \lambda_j^{(i)} z_j^{(i)}\|_1}{(\|H\|_1 + \lambda_j^{(i)})\|z_j^{(i)}\|_1} \quad \text{and} \quad \frac{|\lambda_j^* - \lambda_j^{(i)}|}{|\lambda_j^*|},$$

respectively, where λ_j^* are computed by the QR algorithm (via MATLAB's function `eig`) and considered to be the "exact" eigenvalues. The preconditioner is chosen to be

$$\Phi = H^{-1} = \begin{pmatrix} 0 & M^{-1} \\ K^{-1} & 0 \end{pmatrix}.$$

The preconditioned vectors $q_i := M^{-1}p_i$ and $p_i := K^{-1}q_i$ are computed by the CG method [10, 13]. Often very crude approximations are good enough. In this example, we solve both equations with stopping tolerance 10^{-2} or maximum 20 iterations.

Figure 5.1 shows the relative residual norms and the relative eigenvalue errors of a MATLAB implementation of the locally optimal block 4-D CG algorithm with and without preconditioning (Algorithm 4.1 with $k = 4$). We observe that the significant difference in convergence rate with and without preconditioning.

In this example, for each CG step there are $2k = 8$ linear systems with coefficient matrix K or M. On average, they are approximately solved by a combined $32k = 128$ linear CG steps (or 16 linear CG steps per linear system). That is to say that each LOP4DCG uses $32k$ matrix-vector multiplications by K or M for the preconditioning part.

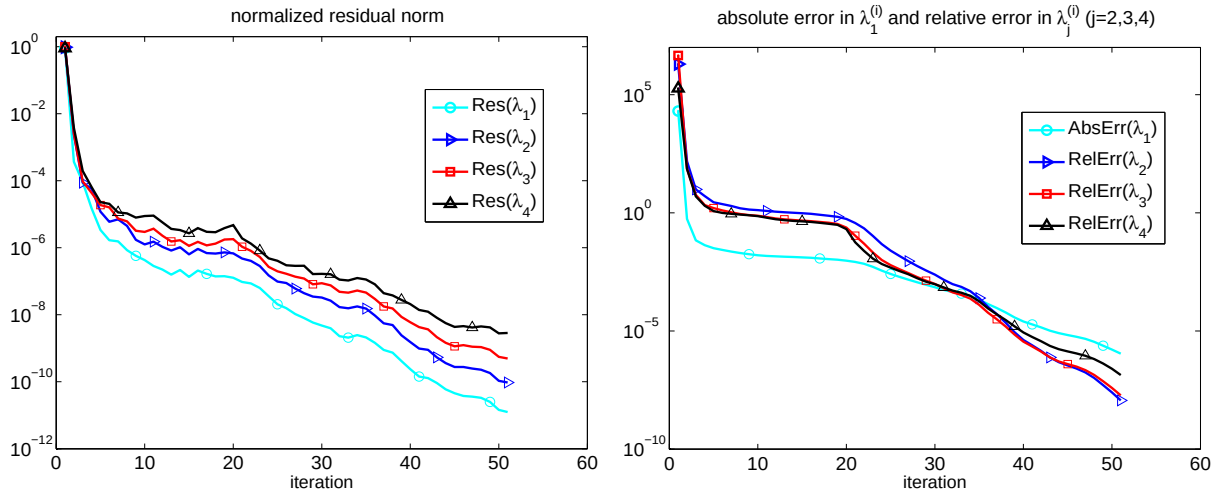

Figure 5.2: The convergence behaviors of the locally optimal block 4-D CG algorithms with preconditioning for computing the 4 smallest eigenvalues with the positive sign for the artificially constructed H: relative residual norms (left) and absolute or relative eigenvalue errors (right).

The next example is artificially constructed with a singular K: all $K_{(i,i+1)} = K_{(i+1,i)} = -1$ for $1 \le i \le n-1$, $K_{(j,j)} = 2$ for $2 \le j \le n-1$, $K_{(1,1)} = K_{(n,n)} = 1$, and $K_{(i,j)} = 0$ elsewhere, $M = \mathrm{diag}(1, 2, \ldots, n)$, and $n = 2000$. We use the locally optimal block 4-D CG algorithm to compute the first 4 smallest eigenvalues with the positive sign. We find that without a preconditioner, convergence is very slow. Figure 5.2 shows the relative residual norms and the absolute error in $\lambda_1^{(i)}$ (since $\lambda_1 = 0$) and relative errors in $\lambda_j^{(i)}$ $(j = 2, 3, 4)$ by Algorithm 4.1 with $k = 4$ using the preconditioner $\Phi = H^{-1}$ implemented again by the CG method with stopping tolerance 10^{-2} or maximum 50 iterations. We point out that our numerical history indicates that the stopping tolerance 10^{-2} was never reached in solving linear systems with coefficient matrix K, i.e., CG took 50 steps for each of such a system. But we still observe decent convergence rate towards the designed eigenvalues.

6 Concluding remarks

Basing on the theoretical foundation laid out in [2] for the LR (a.k.a. RPA) eigenvalue problem (1.1) in computational quantum chemistry and physics, we developed a 4-D search technique which enhances the standard line search method in optimization and then devise locally optimal CG methods that are capable of computing the first few smallest eigenvalues with the positive sign and corresponding eigenvectors simultaneously.

The numerical examples in section 5 demonstrate the effectiveness of the new algorithms, especially with suitable preconditioners. Extended numerical experiment results on a block 4-D steepest descent type method for the first-principle calculation of the excitation states of large molecules is presented in [36]. However, we do not have any precise estimate on rates of convergence yet.

In both [2] and this paper, we have focused on the case where the LR (RPA) eigenvalue problem has only real eigenvalues with eigenvalues 0 allowed. There are cases in which imaginary eigenvalues occur. For example, the positive-definiteness condition of $A + B$ and/or $A - B$ is not met in [31, 34]. The development of efficient numerical methods for treating such large scale problems is a subject of future study.

A Best approximations: the singular case

This appendix continues the investigation in section 3 to seek best approximate eigenpairs of H for given $\{\mathcal{U}, \mathcal{V}\}$, a pair of approximate deflating subspaces of $\{K, M\}$ with $\dim(\mathcal{U}) = \dim(\mathcal{V}) = \ell$. In section 3, we have treated the case in which $W \stackrel{\text{def}}{=} U^{\mathrm{T}}V$ is nonsingular, where U, $V \in \mathbb{R}^{n\times\ell}$ are the basis matrices of $\mathcal{U}$ and $\mathcal{V}$, respectively. In what follows, we will treat the case in which W is singular.

Suppose that W is singular, and factorize

$$W = W_1^{\mathrm{T}} W_2, \quad W_i \in \mathbb{R}^{r\times\ell}, \quad r = \mathrm{rank}(W) < \ell. \tag{A.1}$$

So both W_i have full row rank. Factorize[4]

$$W_i^{\mathrm{T}} = Q_i \begin{pmatrix} R_i \\ 0 \end{pmatrix} \quad \text{for } i = 1, 2, \tag{A.2}$$

where $R_i \in \mathbb{R}^{r\times r}$, $Q_i \in \mathbb{R}^{\ell\times\ell}$ $(i = 1, 2)$ are nonsingular.

Consider the best approximation to λ_1 by (3.1). We still have (3.3):

$$\rho(x, y) = \frac{\hat{u}^{\mathrm{T}}U^{\mathrm{T}}KU\hat{u} + \hat{v}^{\mathrm{T}}V^{\mathrm{T}}MV\hat{v}}{2|\hat{u}^{\mathrm{T}}W\hat{v}|}, \tag{3.3}$$

where $x = U\hat{u}$ and $y = V\hat{v}$ for some $\hat{u}$, $\hat{v} \in \mathbb{R}^{\ell}$. Note the correspondence between $x \in \mathcal{U}$ and $\hat{u} \in \mathbb{R}^{\ell}$ and that between $y \in \mathcal{V}$ and $\hat{v} \in \mathbb{R}^{\ell}$ are one-one. Let $\hat{x} = W_1\hat{u} \in \mathbb{R}^r$ and $\hat{y} = W_2\hat{v} \in \mathbb{R}^r$. Since $r < \ell$, $\hat{u}$ is not uniquely defined by $\hat{x}$; neither is $\hat{v}$ by $\hat{y}$. But use (A.2) to see that

$$\hat{u} = Q_1^{-\mathrm{T}} \begin{pmatrix} R_1^{-\mathrm{T}}\hat{x} \\ u \end{pmatrix}, \quad \hat{v} = Q_2^{-\mathrm{T}} \begin{pmatrix} R_2^{-\mathrm{T}}\hat{y} \\ v \end{pmatrix},$$

where u, $v \in \mathbb{R}^{\ell-r}$ are arbitrary. Partition

$$Q_1^{-1}U^{\mathrm{T}}KUQ_1^{-\mathrm{T}} = \begin{matrix} \\ r \\ \ell-r \end{matrix} \begin{matrix} \begin{matrix} r & \ell-r \end{matrix} \\ \begin{pmatrix} K_{11} & K_{12} \\ K_{12}^{\mathrm{T}} & K_{22} \end{pmatrix} \end{matrix}, \quad Q_2^{-1}V^{\mathrm{T}}MVQ_2^{-\mathrm{T}} = \begin{matrix} \\ r \\ \ell-r \end{matrix} \begin{matrix} \begin{matrix} r & \ell-r \end{matrix} \\ \begin{pmatrix} M_{11} & M_{12} \\ M_{12}^{\mathrm{T}} & M_{22} \end{pmatrix} \end{matrix}. \tag{A.3}$$

We have

$$\hat{u}^{\mathrm{T}}U^{\mathrm{T}}KU\hat{u} = \begin{pmatrix} R_1^{-\mathrm{T}}\hat{x} \\ u \end{pmatrix}^{\mathrm{T}} \begin{pmatrix} K_{11} & K_{12} \\ K_{12}^{\mathrm{T}} & K_{22} \end{pmatrix} \begin{pmatrix} R_1^{-\mathrm{T}}\hat{x} \\ u \end{pmatrix},$$
$$\hat{v}^{\mathrm{T}}V^{\mathrm{T}}MV\hat{v} = \begin{pmatrix} R_2^{-\mathrm{T}}\hat{y} \\ v \end{pmatrix}^{\mathrm{T}} \begin{pmatrix} M_{11} & M_{12} \\ M_{12}^{\mathrm{T}} & M_{22} \end{pmatrix} \begin{pmatrix} R_2^{-\mathrm{T}}\hat{y} \\ v \end{pmatrix}.$$

Given $\hat{x}$, $\hat{u}^{\mathrm{T}}U^{\mathrm{T}}KU\hat{u}$ is minimized at these u such that $K_{22}u = -K_{12}^{\mathrm{T}}R_1^{-\mathrm{T}}\hat{x}$. This equation always has a solution because that $Q_1^{-1}U^{\mathrm{T}}KUQ_1^{-\mathrm{T}}$ is positive semi-definite implies $\mathrm{span}(K_{12}^{\mathrm{T}}) \subseteq \mathrm{span}(K_{22})$, and its solution is not unique if K_{22} is singular. But the non-uniqueness does not matter as far as the minimal value of $\hat{u}^{\mathrm{T}}U^{\mathrm{T}}KU\hat{u}$ is concerned. The same can be said about $\hat{v}^{\mathrm{T}}V^{\mathrm{T}}MV\hat{v}$. In fact,

$$\min_u \hat{u}^{\mathrm{T}}U^{\mathrm{T}}KU\hat{u} = \hat{x}^{\mathrm{T}}R_1^{-1}\left(K_{11} - K_{12}K_{22}^{\dagger}K_{12}^{\mathrm{T}}\right)R_1^{-\mathrm{T}}\hat{x}, \tag{A.4a}$$
$$\min_v \hat{v}^{\mathrm{T}}V^{\mathrm{T}}MV\hat{v} = \hat{y}^{\mathrm{T}}R_2^{-1}\left(M_{11} - M_{12}M_{22}^{\dagger}M_{12}^{\mathrm{T}}\right)R_2^{-\mathrm{T}}\hat{y}, \tag{A.4b}$$

[4] Computationally, this can be realized by the QR decompositions of W_i^{T}. For more generality in presentation, we do not assume have to be QR decompositions.

where $K_{22}^\dagger$ and $M_{22}^\dagger$ are the Moore-Penrose inverses of K_{22} and M_{22}, respectively. The minimums in (A.4) are attained at those u and v satisfying

$$K_{22}u = -K_{12}^{\mathrm{T}} R_1^{-\mathrm{T}} \hat{x}, \quad M_{22}v = -M_{12}^{\mathrm{T}} R_2^{-\mathrm{T}} \hat{y}.$$

Finally, the quantity in (3.1) is

$$\inf_{\hat{x},\hat{y}} \frac{\hat{x}^{\mathrm{T}} R_1^{-1} \left(K_{11} - K_{12}K_{22}^\dagger K_{12}^{\mathrm{T}}\right) R_1^{-\mathrm{T}} \hat{x} + \hat{y}^{\mathrm{T}} R_2^{-1} \left(M_{11} - M_{12}M_{22}^\dagger M_{12}^{\mathrm{T}}\right) R_2^{-\mathrm{T}} \hat{y}}{2|\hat{x}^{\mathrm{T}}\hat{y}|}$$

which, by [2, Theorem 3.1], is the smallest eigenvalue with the positive sign of $\hat{H}_{\mathrm{SR}}$:

$$\hat{H}_{\mathrm{SR}} = \begin{pmatrix} 0 & R_1^{-1} \left(K_{11} - K_{12}K_{22}^\dagger K_{12}^{\mathrm{T}}\right) R_1^{-\mathrm{T}} \\ R_2^{-1} \left(M_{11} - M_{12}M_{22}^\dagger M_{12}^{\mathrm{T}}\right) R_2^{-\mathrm{T}} & 0 \end{pmatrix} \in \mathbb{R}^{2r\times 2r}. \quad \text{(A.5)}$$

Now we turn to the best approximations to λ_j $(1 \le j \le k)$ by (3.2). Assume (A.1) and (A.2). Any $\hat{U}$, $\hat{V} \in \mathbb{R}^{n\times k}$ such that $\mathrm{span}(\hat{U}) \in \mathcal{U}$, $\mathrm{span}(\hat{V}) \in \mathcal{V}$, and $\hat{U}^{\mathrm{T}}\hat{V} = I_k$ can be written as

$$\hat{U} = UQ_1^{-\mathrm{T}}\widetilde{U}, \quad \hat{V} = VQ_2^{-\mathrm{T}}\widetilde{V},$$

where $\widetilde{U}$, $\widetilde{V} \in \mathbb{R}^{\ell\times k}$ that make $\hat{U}^{\mathrm{T}}\hat{V} = I_k$, and vice versa. We note that necessarily

$$k = \mathrm{rank}(I_k) = \mathrm{rank}(\hat{U}^{\mathrm{T}}\hat{V}) \le \mathrm{rank}(W) = r.$$

We first look into what constraint is needed on $\widetilde{U}$ and $\widetilde{V}$ in order to enforce $\hat{U}^{\mathrm{T}}\hat{V} = I_k$. To this end, we partition

$$\widetilde{U} = \begin{matrix} r \\ \ell-r \end{matrix} \overset{k}{\begin{pmatrix} \widetilde{U}_1 \\ \widetilde{U}_2 \end{pmatrix}}, \quad \widetilde{V} = \begin{matrix} r \\ \ell-r \end{matrix} \overset{k}{\begin{pmatrix} \widetilde{V}_1 \\ \widetilde{V}_2 \end{pmatrix}}.$$

We have

$$\hat{U}^{\mathrm{T}}\hat{V} = \widetilde{U}^{\mathrm{T}}Q_1^{-1}W_1^{\mathrm{T}}W_2Q_2^{-\mathrm{T}}\widetilde{V} = \widetilde{U}^{\mathrm{T}} \begin{pmatrix} R_1 \\ 0 \end{pmatrix} (R_2^{\mathrm{T}}, 0)\widetilde{V} = \widetilde{U}_1^{\mathrm{T}} R_1 R_2^{\mathrm{T}} \widetilde{V}_1.$$

Let $\hat{X} = R_1^T\widetilde{U}_1$, $\hat{Y} = R_2^{\mathrm{T}}\widetilde{V}_1 \in \mathbb{R}^{r\times k}$. Then $\hat{U}^{\mathrm{T}}\hat{V} = I_k$ is equivalent to $\hat{X}^{\mathrm{T}}\hat{Y} = I_k$ which will be enforced henceforth, while $\widetilde{U}_2$ and $\widetilde{V}_2$ are arbitrary. Assume the partitioning in (A.3). We have

$$\hat{U}^{\mathrm{T}}K\hat{U} = \widetilde{U}^{\mathrm{T}}Q_1^{-1}U^{\mathrm{T}}KUQ_1^{-\mathrm{T}}\widetilde{U} = \begin{pmatrix} R_1^{-\mathrm{T}}\hat{X} \\ \widetilde{U}_2 \end{pmatrix}^{\mathrm{T}} \begin{pmatrix} K_{11} & K_{12} \\ K_{12}^{\mathrm{T}} & K_{22} \end{pmatrix} \begin{pmatrix} R_1^{-\mathrm{T}}\hat{X} \\ \widetilde{U}_2 \end{pmatrix},$$

$$\hat{V}^{\mathrm{T}}M\hat{V} = \widetilde{V}^{\mathrm{T}}Q_1^{-1}V^{\mathrm{T}}KVQ_1^{-\mathrm{T}}\widetilde{V} = \begin{pmatrix} R_2^{-\mathrm{T}}\hat{Y} \\ \widetilde{V}_2 \end{pmatrix}^{\mathrm{T}} \begin{pmatrix} M_{11} & M_{12} \\ M_{12}^{\mathrm{T}} & M_{22} \end{pmatrix} \begin{pmatrix} R_2^{-\mathrm{T}}\hat{Y} \\ \widetilde{V}_2 \end{pmatrix}.$$

Given $\hat{X}$ and $\hat{Y}$, it can be verified that

$$\min_{\widetilde{U}_2} \mathrm{trace}(\hat{U}^{\mathrm{T}}K\hat{U}) = \mathrm{trace}(\hat{X}^{\mathrm{T}}R_1^{-1}[K_{11} - K_{12}K_{22}^\dagger K_{12}^{\mathrm{T}}]R_1^{-\mathrm{T}}\hat{X}), \quad \text{(A.6a)}$$

$$\min_{\widetilde{V}_2} \mathrm{trace}(\hat{V}^{\mathrm{T}}M\hat{V}) = \mathrm{trace}(\hat{Y}^{\mathrm{T}}R_2^{-1}[M_{11} - M_{12}M_{22}^\dagger M_{12}^{\mathrm{T}}]R_2^{-\mathrm{T}}\hat{Y}) \quad \text{(A.6b)}$$

with the minimums attained at those $\widetilde{U}_2$ and $\widetilde{V}_2$ satisfying

$$K_{22}\widetilde{U}_2 = -K_{12}^{\mathrm{T}}R_1^{-\mathrm{T}}\hat{X}, \quad M_{22}\widetilde{V}_2 = -M_{12}^{\mathrm{T}}R_2^{-\mathrm{T}}\hat{Y}.$$

Therefore the quantity in (3.2) is

$$\inf_{\hat{X}^{\mathrm{T}}\hat{Y}=I_k} \operatorname{trace}\left(\hat{X}^{\mathrm{T}}R_1^{-1}[K_{11}-K_{12}K_{22}^{\dagger}K_{12}^{\mathrm{T}}]R_1^{-\mathrm{T}}\hat{X}+\hat{Y}^{\mathrm{T}}R_2^{-1}[M_{11}-M_{12}M_{22}^{\dagger}M_{12}^{\mathrm{T}}]R_2^{-\mathrm{T}}\hat{Y}\right)$$

which, by [2, Theorem 3.2], is the sum of the k smallest eigenvalues with the positive sign of $\hat{H}_{\mathrm{SR}}$ defined by (A.5).

In summary, the *best approximations to some of the eigenvalues of H within the pair of approximate deflating subspaces are the eigenvalues of $\hat{H}_{\mathrm{SR}}$*. Denote by μ_j $(j=1,\ldots,r)$ the eigenvalues with the positive sign of $\hat{H}_{\mathrm{SR}}$ in ascending order and by $\hat{z}_j$ the associated eigenvectors:

$$\hat{H}_{\mathrm{SR}}\hat{z}_j=\mu_j\hat{z}_j,\quad \hat{z}_j=\begin{pmatrix}\hat{y}_j\\ \hat{x}_j\end{pmatrix}. \tag{A.7}$$

Following the derivations above, we conclude

$$\rho(\widetilde{x}_j,\widetilde{y}_j)=\mu_j\quad \text{for } j=1,\ldots,r,$$

where

$$\widetilde{x}_j=UQ_1^{-\mathrm{T}}\begin{pmatrix}R_1^{-\mathrm{T}}\hat{x}_j\\ u_j\end{pmatrix},\quad \widetilde{y}_j=VQ_2^{-\mathrm{T}}\begin{pmatrix}R_2^{-\mathrm{T}}\hat{y}_j\\ v_j\end{pmatrix} \tag{A.8}$$

for u_j and v_j satisfying

$$K_{22}u_j=-K_{12}^{\mathrm{T}}R_1^{-\mathrm{T}}\hat{x}_j,\quad M_{22}v_j=-M_{12}^{\mathrm{T}}R_2^{-\mathrm{T}}\hat{y}_j. \tag{A.9}$$

Naturally the approximate eigenvectors of H should be taken as

$$\widetilde{z}_j=\begin{pmatrix}\widetilde{y}_j\\ \widetilde{x}_j\end{pmatrix}\quad \text{for } j=1,\ldots,r. \tag{A.10}$$

For easy of reference, we summarize our findings into the following theorem.

Theorem A.1. *Suppose that one of K, $M\in\mathbb{R}^{n\times n}$ is definite. Let $\{\mathcal{U},\mathcal{V}\}$ be a pair of approximate deflating subspaces of $\{K,M\}$ with $\dim(\mathcal{U})=\dim(\mathcal{V})=\ell$, and let U, $V\in\mathbb{R}^{n\times\ell}$ be the basis matrices of $\mathcal{U}$ and $\mathcal{V}$, respectively. Suppose that $W\overset{\mathrm{def}}{=}U^{\mathrm{T}}V$ is singular and let $\hat{H}_{\mathrm{SR}}$ be defined by* (A.5). *Then the best approximations to λ_1 in the sense of* (3.1) *or to λ_j $(1\le j\le k)$ in the sense of* (3.2) *are the corresponding eigenvalues μ_j of $\hat{H}_{\mathrm{SR}}$ defined in* (A.5)*, with the corresponding approximate eigenvectors given by* (A.8) – (A.10).

In [2, Theorem 2.7], we proved the approximate eigenpairs are unique for given $\{\mathcal{U},\mathcal{V}\}$ with nonsingular $U^{\mathrm{T}}V$, even though there are infinitely many different H_{SR} associated with the pair of subspaces. We are facing with the same question for $\hat{H}_{\mathrm{SR}}$ in whose construction there are three non-unique choices:

$$\begin{cases}1.\ \text{Factorizations in (A.2) are not unique.}\\ 2.\ \text{Factorization } W=W_1^{\mathrm{T}}W_2 \text{ in (A.1) is not unique.}\\ 3.\ \text{Basis matrices } U \text{ and } V \text{ are not unique.}\end{cases} \tag{A.11}$$

The question would arise if different $\hat{H}_{\mathrm{SR}}$ could produce different approximate eigenpairs. This is addressed by the following theorem.

Theorem A.2. *Suppose that one of K, $M \in \mathbb{R}^{n\times n}$ is definite. Let $\mathcal{U}$ and $\mathcal{V}$ be two subspaces of $\mathbb{R}^n$ of dimension ℓ with basis matrices U, $V \in \mathbb{R}^{n\times\ell}$, respectively. Suppose that $W = U^{\mathrm{T}}V$ is singular and define $\hat{H}_{\mathrm{SR}}$ by* (A.5)*. Then*

1. *the approximate eigenvalues, i.e., the eigenvalues of $\hat{H}_{\mathrm{SR}}$, are invariant with respect to any of the non-uniqueness listed in* (A.11) *for constructing $\hat{H}_{\mathrm{SR}}$;*

2. *the approximate eigenvectors by* (A.8) – (A.10) *are invariant with respect to any of the non-uniqueness listed in* (A.11) *if and only if both K_{22} and M_{22} are definite.*

Proof. To see the first conclusion, we notice that the infimum (3.2) only depends on $\{\mathcal{U}, \mathcal{V}\}$ and is invariant with respect to any of the non-uniqueness in (A.11) for $1 \le k \le r$. Since the infimum is the sum of the first k smallest eigenvalues with the positive sign of $\hat{H}_{\mathrm{SR}}$, let k go from 1 to r to conclude that the eigenvalues with the positive sign of $\hat{H}_{\mathrm{SR}}$ are invariant with respect to any of the non-uniqueness in (A.11); so are all eigenvalues of $\hat{H}_{\mathrm{SR}}$.

For the second conclusion, let us first select one choice for each of them in (A.11), namely basis matrices U and V, a factorization $W = W_1^{\mathrm{T}}W_2$ in (A.1), and two factorizations in (A.2). Let $H_0 \overset{\mathrm{def}}{=} \hat{H}_{\mathrm{SR}}$ with these selected choices, and suppose that both K_{22} and M_{22} are definite. We shall now prove that the approximate eigenvectors are invariant with respect to any variation to the selected ones. Along the way, we will also see the definiteness of K_{22} and M_{22} does not change with the variations, either.

1. *Invariance with respect to different choices of factorizations in* (A.2). Any factorizations other than the given ones in (A.2) can be written as

$$W_i^{\mathrm{T}} = Q_i \begin{pmatrix} S_{i1} & \\ & S_{i2} \end{pmatrix} \begin{pmatrix} S_{i1}^{-1}R_i \\ 0 \end{pmatrix}, \tag{A.12}$$

   for some nonsingular $S_{i1} \in \mathbb{R}^{r\times r}$, $S_{i2} \in \mathbb{R}^{(\ell-r)\times(\ell-r)}$. Denote by $H_1 \overset{\mathrm{def}}{=} \hat{H}_{\mathrm{SR}}$ with given U, V and (A.1), and (A.12). Perform substitutions

$$Q_i \begin{pmatrix} S_{i1} & \\ & S_{i2} \end{pmatrix} \leftarrow Q_i,\ S_{i1}^{-1}R_i \leftarrow R_i,\ S_{1i}^{-1}K_{ij}S_{1j}^{-\mathrm{T}} \leftarrow K_{ij},\ S_{2i}^{-1}M_{ij}S_{2j}^{-\mathrm{T}} \leftarrow M_{ij}$$

   to see $H_0 = H_1$ and that the approximate eigenvectors for H by (A.8) – (A.10) do not change. Also the definiteness of K_{22} and M_{22} does not change with the variation in (A.12).

2. *Invariance with respect to different choice of factorization $W = W_1^{\mathrm{T}}W_2$.* Any factorization other than the given one in (A.1), can be written as

$$W = W_1^{\mathrm{T}}SS^{-1}W_2 = (S^{\mathrm{T}}W_1)^{\mathrm{T}}(S^{-1}W_2) \tag{A.13}$$

   for some nonsingular $S \in \mathbb{R}^{r\times r}$. Define $H_1 \overset{\mathrm{def}}{=} \hat{H}_{\mathrm{SR}}$ with given U, V, and (A.13) and

$$(S^{\mathrm{T}}W_1)^{\mathrm{T}} = Q_1 \begin{pmatrix} R_1S \\ 0 \end{pmatrix}, \quad (S^{-1}W_2)^{\mathrm{T}} = Q_2 \begin{pmatrix} R_2S^{-\mathrm{T}} \\ 0 \end{pmatrix}. \tag{A.14}$$

   Since we just proved the invariance with respect to different choices of factorizations in (A.2), it suffices to prove that the approximate eigenvectors obtained through H_0 and H_1 are the same. Upon using substitutions $R_1S \leftarrow R_1$ and $R_2S^{-\mathrm{T}} \leftarrow R_2$, we find

$$H_1 = \left(S^{-1} \oplus S^{\mathrm{T}}\right) H_0 \left(S^{-1} \oplus S^{\mathrm{T}}\right)^{-1},$$

   and thus the relationships between the eigenvectors for H_0 and H_1. It can then be verified that the approximate eigenvectors obtained through H_0 and H_1 via (A.8) – (A.10) are the same. Also the definiteness of K_{22} and M_{22} does not change with the variation in (A.13).

3. *Invariance with respect to different choices of basis matrices.* Given basis matrices U and V of $\mathcal{U}$ and $\mathcal{V}$, respectively, any other basis matrices can be written as UR and VS for some nonsingular R, $S \in \mathbb{R}^{\ell\times\ell}$. Define $H_1 \overset{\text{def}}{=} \hat{H}_{\text{SR}}$ with UR and VS, and

$$W = (UR)^{\mathrm{T}}(VS) = R^{\mathrm{T}}U^{\mathrm{T}}VS = (W_1R)^{\mathrm{T}}(W_2S), \tag{A.15}$$

and

$$(W_1R)^{\mathrm{T}} = R^{\mathrm{T}}Q_1\begin{pmatrix}R_1\\0\end{pmatrix}, \quad (W_2S)^{\mathrm{T}} = S^{\mathrm{T}}Q_2\begin{pmatrix}R_2\\0\end{pmatrix}. \tag{A.16}$$

By the two invariance properties we just proved, it suffices to prove that the approximate eigenvectors obtained through H_0 and H_1 are the same. Perform substitutions

$$UR \leftarrow U, \quad VS \leftarrow V, \quad R^{\mathrm{T}}Q_1 \leftarrow Q_1, \quad S^{\mathrm{T}}Q_2 \leftarrow Q_2$$

to see $H_0 = H_1$ and that the approximate eigenvectors for H by (A.8) – (A.10) do not change. Again the definiteness of K_{22} and M_{22} does not change with the variation from U to UR and from V to VS.

Finally if K_{22} is singular, then u_j satisfying the first equation in (A.9) is not unique. In fact, if u_j is one, any $u_j + g$ is another, for any g in the kernel of K_{22}. So the defining equation in (A.8) for $\widetilde{x}_j$ gives

$$\widetilde{x}_j = UQ_1\begin{pmatrix}R_1^{-\mathrm{T}}\hat{x}_j\\-K_{22}^{\dagger}K_{12}^{\mathrm{T}}R_1^{-\mathrm{T}}\hat{x}_j + g\end{pmatrix} \tag{A.17}$$

leading to different approximate eigenvectors as g varies within the kernel of K_{22}. The same thing happens if M_{22} is singular. □

The proof of Theorem A.2 exposes the cause for the approximate eigenvectors by (A.8) – (A.10) not to be uniquely determined, namely, one of the equations in (A.9) may have infinitely many solutions[5]. When that's the case, we can either always take

$$u_j = -K_{22}^{\dagger}K_{12}^{\mathrm{T}}R_1^{-\mathrm{T}}\hat{x}_j, \quad v_j = -M_{22}^{\dagger}M_{12}^{\mathrm{T}}R_1^{-\mathrm{T}}\hat{y}_j$$

or settle the non-uniqueness by

$$\min_{g,h}\left\{\|K\widetilde{x}_j - \mu_j\widetilde{y}_j\|_2^2 + \|M\widetilde{y}_j - \mu_j\widetilde{x}_j\|_2^2\right\} \tag{A.18}$$

over all g in the kernel of K_{22} and h in the kernel of M_{22}, upon noticing (A.17) and

$$\widetilde{y}_j = VQ_2^{-\mathrm{T}}\begin{pmatrix}R_2^{-\mathrm{T}}\hat{y}_j\\-M_{22}^{\dagger}M_{12}^{\mathrm{T}}R_2^{-\mathrm{T}}\hat{y}_j + h\end{pmatrix}. \tag{A.19}$$

Finally it can be seen that (A.18) is a least squares problem in g and h. The next theorem says that there are Cauchy-like interlacing inequalities for $\widehat{H}_{\text{SR}}$, too.

Theorem A.3. *Assume the conditions of* Theorem A.1. *Then*

$$\lambda_i \le \mu_i \le \lambda_{i+2n-2\ell} \quad \textit{for } 1 \le i \le r, \tag{A.20}$$

where $\lambda_{i+2n-2\ell} = \infty$ *if* $i + 2n - 2\ell > n$.

[5]By default, one of K and M is definite. Thus at most one of K_{22} and M_{22} is singular.

Proof. Suppose for the moment that both K and M are definite. Recall the equivalence between the eigenvalue problem (1.1) and the one for [2]

$$\boldsymbol{A} - \lambda \boldsymbol{B} \equiv \begin{pmatrix} M & 0 \\ 0 & K \end{pmatrix} - \lambda \begin{pmatrix} 0 & I_n \\ I_n & 0 \end{pmatrix}. \tag{A.21}$$

$\boldsymbol{A}$ is symmetric positive definite if K and M are. Let

$$Z = \begin{pmatrix} VQ_2^{-\mathrm{T}}(R_2^{-1} \oplus I_{\ell-r}) & \\ & UQ_1^{-\mathrm{T}}(R_1^{-1} \oplus I_{\ell-r}) \end{pmatrix}$$

which has full column rank. It can be verified, upon using (A.3), that

$$Z^{\mathrm{T}}\boldsymbol{A}Z = \begin{pmatrix} \widehat{M} & 0 \\ 0 & \widehat{K} \end{pmatrix}, \quad Z^{\mathrm{T}}\boldsymbol{B}Z = \begin{pmatrix} 0 & \widehat{I} \\ \widehat{I} & 0 \end{pmatrix},$$

where

$$\begin{aligned} \widehat{M} &= \begin{pmatrix} R_2^{-\mathrm{T}} & \\ & I_{\ell-r} \end{pmatrix} \begin{pmatrix} M_{11} & M_{12} \\ M_{12}^{\mathrm{T}} & M_{22} \end{pmatrix} \begin{pmatrix} R_2^{-1} & \\ & I_{\ell-r} \end{pmatrix}, \\ \widehat{K} &= \begin{pmatrix} R_1^{-\mathrm{T}} & \\ & I_{\ell-r} \end{pmatrix} \begin{pmatrix} K_{11} & K_{12} \\ K_{12}^{\mathrm{T}} & K_{22} \end{pmatrix} \begin{pmatrix} R_1^{-1} & \\ & I_{\ell-r} \end{pmatrix}, \\ \widehat{I} &= \begin{pmatrix} I_r & \\ & 0 \end{pmatrix} \in \mathbb{R}^{\ell \times \ell}. \end{aligned}$$

$Z^{\mathrm{T}}\boldsymbol{A}Z$ is positive definite because $\boldsymbol{A}$ is. Note the eigenvalues of $\boldsymbol{B} - \lambda\boldsymbol{A}$ are $\pm\lambda_i^{-1}$ and

$$-\lambda_1^{-1} \le -\lambda_2^{-1} \le \cdots \le -\lambda_n^{-1} < \lambda_n^{-1} \le \cdots \le \lambda_2^{-1} \le \lambda_1^{-1}.$$

Denote the eigenvalues[6] of $Z^{\mathrm{T}}\boldsymbol{B}Z - \lambda Z^{\mathrm{T}}\boldsymbol{A}Z$ by $\pm\sigma_i$ ordered as

$$-\sigma_1 \le \cdots \le -\sigma_r < -\sigma_{r+1} = \cdots = -\sigma_\ell = 0 = \sigma_\ell = \cdots = \sigma_{r+1} < \sigma_r \le \cdots \le \sigma_1.$$

Now apply Cauchy's interlacing inequalities (extended for the generalized eigenvalue problem) to $\boldsymbol{B} - \lambda\boldsymbol{A}$ and $Z^{\mathrm{T}}\boldsymbol{B}Z - \lambda Z^{\mathrm{T}}\boldsymbol{A}Z$ to get for $1 \le i \le \ell$

$$\lambda_i^{-1} \ge \sigma_i \ge \begin{cases} \lambda_{i+2n-2\ell}^{-1}, & \text{if } i + 2n - 2\ell \le n, \\ 0, & \text{otherwise.} \end{cases}$$

Equivalently $\lambda_i \le \sigma_i^{-1} \le \lambda_{i+2n-2\ell}$ for $1 \le i \le \ell$. It remains to show that $\mu_i = \sigma_i^{-1}$ for $1 \le i \le r$. To this end, we let

$$Z_1 = \begin{pmatrix} I_r & 0 \\ -K_{22}^{-1}K_{12}^{\mathrm{T}}R_1^{-1} & I_{\ell-r} \end{pmatrix}, \quad Z_2 = \begin{pmatrix} I_r & 0 \\ -M_{22}^{-1}M_{12}^{\mathrm{T}}R_2^{-1} & I_{\ell-r} \end{pmatrix}.$$

It can be verified that $Z_1^{\mathrm{T}}\widehat{I}Z_2 = \widehat{I}$ and

$$\begin{aligned} Z_1^{\mathrm{T}}\widehat{K}Z_1 &= \begin{pmatrix} R_1^{-1}\widehat{K}_{11}R_1^{-\mathrm{T}} & 0 \\ 0 & K_{22} \end{pmatrix}, \quad \widehat{K}_{11} = K_{11} - K_{12}K_{22}^{-1}K_{12}^{\mathrm{T}}, \\ Z_2^{\mathrm{T}}\widehat{M}Z_2 &= \begin{pmatrix} R_2^{-1}\widehat{M}_{11}R_2^{-\mathrm{T}} & 0 \\ 0 & M_{22} \end{pmatrix}, \quad \widehat{M}_{11} = M_{11} - M_{12}M_{22}^{-1}M_{12}^{\mathrm{T}}. \end{aligned}$$

[6]These eigenvalues are the same as those of $(Z^{\mathrm{T}}\boldsymbol{A}Z)^{-1/2}(Z^{\mathrm{T}}\boldsymbol{B}Z)(Z^{\mathrm{T}}\boldsymbol{A}Z)^{-1/2} = \begin{pmatrix} 0 & C \\ C^{\mathrm{T}} & 0 \end{pmatrix}$, where $C = \widehat{M}^{-1/2}\widehat{I}\widehat{K}^{-1/2}$. Thus σ_i are the singular values of C.

The eigenvalues of $Z^{\mathrm{T}}\boldsymbol{B}Z - \lambda Z^{\mathrm{T}}\boldsymbol{A}Z$ are the same as these of

$$(Z_1 \oplus Z_2)^{\mathrm{T}} \left(Z^{\mathrm{T}}\boldsymbol{B}Z - \lambda Z^{\mathrm{T}}\boldsymbol{A}Z\right)(Z_1 \oplus Z_2)$$

which is a 4×4 block matrix pencil and becomes, after switching its second and third row and its second and third column,

$$\begin{pmatrix} 0 & I_r \\ I_r & 0 \end{pmatrix} \oplus 0_{2(\ell-r)\times 2(\ell-r)} - \lambda \begin{pmatrix} R_2^{-1}\widehat{M}_{11}R_2^{-\mathrm{T}} & 0 \\ 0 & R_1^{-1}\widehat{K}_{11}R_1^{-\mathrm{T}} \end{pmatrix} \oplus \begin{pmatrix} M_{22} & 0 \\ 0 & K_{22} \end{pmatrix}.$$

Thus the nonzero eigenvalues of $Z^{\mathrm{T}}\boldsymbol{B}Z - \lambda Z^{\mathrm{T}}\boldsymbol{A}Z$ are the same as these of

$$\begin{pmatrix} 0 & I_r \\ I_r & 0 \end{pmatrix} - \lambda \begin{pmatrix} R_2^{-1}\widehat{M}_{11}R_2^{-\mathrm{T}} & 0 \\ 0 & R_1^{-1}\widehat{K}_{11}R_1^{-\mathrm{T}} \end{pmatrix}$$

which in turn are the same as the reciprocals of the eigenvalues of $\widehat{H}_{\mathrm{SR}}$, i.e., $\sigma_i = \mu_i^{-1}$, as expected.

Consider now that K is singular. Let $K(\epsilon) = K + \epsilon I_n$ which is definite, where $\epsilon > 0$. Define accordingly $H(\epsilon)$ and its eigenvalues $\pm\lambda_i(\epsilon)$, $\widehat{H}_{\mathrm{SR}}(\epsilon)$ and its eigenvalues $\pm\mu_i(\epsilon)$. By what we just proved, we have

$$\lambda_i(\epsilon) \le \mu_i(\epsilon) \le \lambda_{i+2n-2\ell}(\epsilon) \quad \text{for } 1 \le i \le r. \tag{A.22}$$

From the construction of $\widehat{H}_{\mathrm{SR}}$ above, we see that $\lim_{\epsilon\to 0^+} \widehat{H}_{\mathrm{SR}}(\epsilon)$ exists and the limit is the $\widehat{H}_{\mathrm{SR}}$ given by (A.5). Since eigenvalues are continuous functions of matrix entries, letting $\epsilon \to 0^+$ in (A.22) to get the desired inequalities in (A.20). □

Remark **A.1.** Noticeably our treatment above is much more complicated than the nonsingular case in section 3. Certainly an argument can be made not to use $\{\mathcal{U}, \mathcal{V}\}$ with a singular W at all because [2, Lemma 2.1] says that W is nonsingular if $\{\mathcal{U}, \mathcal{V}\}$ is exact. But in practice, especially at the beginning of an iterative process, it is hard to guarantee this is so at all time. Our treatment, albeit complicated, shows that the optimums in (3.1) and (3.2) can still be realized. An alternative and much simpler treatment for the singular case at a tradeoff of achieving only suboptimal approximations to (3.1) and (3.2) is as follows. Suppose (A.1) and (A.2). We have

$$(R_1^{-1}, 0)Q_1^{-1}U^{\mathrm{T}}VQ_2^{-\mathrm{T}}\begin{pmatrix} R_2 \\ 0 \end{pmatrix} = I_r.$$

After substitutions

$$U \leftarrow UQ_1^{-\mathrm{T}}\begin{pmatrix} R_1 \\ 0 \end{pmatrix}, \quad V \leftarrow VQ_2^{-\mathrm{T}}\begin{pmatrix} R_2 \\ 0 \end{pmatrix},$$

two new subspaces $\mathcal{U}$ and $\mathcal{V}$ with dimension r are born with new basis matrices U and V satisfying $U^{\mathrm{T}}V = I_r$, returning to the nonsingular W case in section 3. ◇

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References

[1] E. Anderson, Z. Bai, C. Bischof, S. Blackford, J. Demmel, J. Dongarra, J. Du Croz, A. Greenbaum, S. Hammarling, A. McKenney, and D. Sorensen. *LAPACK Users' Guide (third ed.)*. SIAM, Philadelphia, 1999.

[2] Z. Bai and R.-C. Li. Minimization principle for linear response eigenvalue problem I: Theory. submitted, 2011.

[3] Z. Bai and R.-C. Li. Minimization principle for linear response eigenvalue problem with applications. Technical Report 2011-06, Department of Mathematics, University of Texas at Arlington, May 2011. Available at `http://www.uta.edu/math/preprint/`.

[4] P. Benner, H. Faßbender, and M. Stoll. A Hamiltonian Krylov-Schur-type method based on the symplectic Lanczos process. *Linear Algebra Appl.*, 435(3):578–600, 2011.

[5] P. Benner and H. Faßbender. An implicitly restarted symplectic Lanczos method for the Hamiltonian eigenvalue problem. *Linear Algebra Appl.*, 263:75–111, 1997.

[6] P. Benner, V. Mehrmann, and H. Xu. A numerically stable structure preserving method for computing the eigenvalues of real Hamiltonian or symplectic pencils. *Numer. Math.*, 78:329–357, 1998.

[7] D. Bohm and D. Pines. A collective description of electron interactions: III. Coulomb interactions in a degenerate electron gas. *Phys. Rev.*, 92:609–625, 1953.

[8] A. Bunse-Gerstner and V. Mehrmann. A symplectic QR like algorithm for the solution of the real algebraic Riccati equation. *IEEE Trans. Automat. Control*, 31(12):1104–1113, 1986.

[9] B. E. Chi. The eigenvalue problem for collective motion in the random phase approximation. *Nuclear Physics*, A146(2):449–456, 1970.

[10] J. Demmel. *Applied Numerical Linear Algebra*. SIAM, Philadelphia, PA, 1997.

[11] P. Giannozzi *et al*. QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials. *J. Phys. Condensed Matter*, 21:395502, 2009. Available at `http://www.quantum-espresso.org/`.

[12] U. Flaschka, W.-W. Lin, and J.-L. Wu. A KQZ algorithm for solving linear-response eigenvalue equations. *Linear Algebra Appl.*, 165:93 – 123, 1992.

[13] G. H. Golub and C. F. Van Loan. *Matrix Computations*. Johns Hopkins University Press, Baltimore, Maryland, 3rd edition, 1996.

[14] M. Gruning, A. Marini, and X. Gonze. Exciton-plasmon states in nanoscale materials: breakdown of the Tamm-Dancoff approximation. *Nano Letters*, 9:2820–2824, 2009.

[15] M. Gruning, A. Marini, and X. Gonze. Implementation and testing of Lanczos-based algorithms for random-phase approximation eigenproblems. Technical report, arXiv:1102.3909v1, February 2011.

[16] U. Hetmaniuk and R. Lehoucq. Basis selection in LOBPCG. *J. Comput. Phys.*, 218(1):324 – 332, 2006.

[17] M. E. Hochstenbach. A Jacobi-Davidson type method for the product eigenvalue problem. *J. Comput. Appl. Math.*, 212:46–62, 2008.

[18] Kevin J. Kerns and Andrew T. Yang. Preservation of passivity during RLC network reduction via split congruence transformations. *IEEE Trans. Computer-Aided Design of Integrated Circuits and Systems*, 17(7):582–591, July 1998.

[19] A. V. Knyazev. A preconditioned conjugate gradient method for eigenvalue problems and its implementation in a subspace. *Internat Series Numer. Math.*, 96:143–154, 1991.

[20] A. V. Knyazev. Toward the optimal preconditioned eigensolver: Locally optimal block preconditioned conjugate gradient method. *SIAM J. Sci. Comput.*, 23(2):517–541, 2001.

[21] D. Kressner. A periodic Krylov-Schur algorithm for large matrix products. *Numer. Math.*, 103:461–483, 2006.

[22] R.-C. Li. Accuracy of computed eigenvectors via optimizing a Rayleigh quotient. *BIT*, 44(3):585–593, 2004.

[23] R.-C. Li and Z. Bai. Structure-preserving model reductions using a Krylov subspace projection formulation. *Commun. Math. Sciences*, 3(2):179–199, 2005.

[24] D. E. Longsine and S. F. McCormick. Simultaneous Rayleigh-quotient minimization methods for $Ax = \lambda Bx$. *Linear Algebra Appl.*, 34:195–234, 1980.

[25] M. J. Lucero, A. M. N. Niklasson, S. Tretiak, and M. Challacombe. Molecular-orbital-free algorithm for excited states in time-dependent perturbation theory. *J. Chem. Phys.*, 129(6):064114, 2008.

[26] M. A. Marques, A. Castro, and A. Rubio. Assessment of exchange-correlation functionals for the calculation of dynamical properties of small clusters in time-dependent density functional theory. *J. Chem. Phys.*, 115(7):3006–3014, 2001.

[27] A. Muta, J.-I. Iwata, Y. Hashimoto, and K. Yabana. Solving the RPA eigenvalue equation in real-space. *Progress Theoretical Physics*, 108(6):1065–1076, 2002.

[28] J. Nocedal and S. Wright. *Numerical Optimization*. Springer, 2nd edition, 2006.

[29] J. Olsen, H. J. Aa. Jensen, and P. Jørgensen. Solution of the large matrix equations which occur in response theory. *J. Comput. Phys.*, 74(2):265 – 282, 1988.

[30] E. E. Ovtchinnikov. Jacobi correction equation, line search, and conjugate gradients in Hermitian eigenvalue computation I: Computing an extreme eigenvalue. *SIAM J. Numer. Anal.*, 46(5):2567–2592, 2008.

[31] P. Papakonstantinou. Reduction of the RPA eigenvalue problem and a generalized Cholesky decomposition for real-symmetric matrices. *EPL (Europhysics Letters)*, 78(1):12001, 2007.

[32] B. N. Parlett. *The Symmetric Eigenvalue Problem*. SIAM, Philadelphia, 1998.

[33] B. T. Polyak. *Introduction to optimization*. Optimization Software, New York, 1987.

[34] P. Ring, Z.-Y. Ma, N. Van Giai, D. Vretenar, A. Wandelt, and L.-G. Gao. The time-dependent relativistic mean-field theory and the random phase approximation. *Nuclear Physics A*, 249:249–268, 2001.

[35] P. Ring and P. Schuck. *The Nuclear Many-Body Problem*. Springer-Verlag, New York, 1980.

[36] D. Rocca, Z. Bai, R.-C. Li, and G. Galli. A block variational procedure for the iterative diagonalization of non-Hermitian random-phase approximation matrices. *J. Chem. Phys.*, 136:034111, 2012.

[37] G. W. Stewart and Ji-Guang Sun. *Matrix Perturbation Theory*. Academic Press, Boston, 1990.

[38] R. E. Stratmann, G. E. Scuseria, and M. J. Frisch. An efficient implementation of time-dependent density-functional theory for the calculation of excitation of large molecules. *J. Chem. Phys.*, 109:8218–8824, 1998.

[39] I. Takahashi. A note on the conjugate gradient method. *Inform. Process. Lett.*, 5:45–49, 1965.

[40] D. J. Thouless. Vibrational states of nuclei in the random phase approximation. *Nuclear Physics*, 22(1):78–95, 1961.

[41] D. J. Thouless. *The quantum mechanics of many-body systems*. Academic, 1972.

[42] S. Tretiak, C. M. Isborn, A. M. N. Niklasson, and M. Challacombe. Representation independent algorithms for molecular response calculations in time-dependent self-consistent field theories. *J. Chem. Phys.*, 130(5):054111, 2009.

[43] S. Tretiak and S. Mukamel. Density matrix analysis and simulation of electronic excitations in conjugated and aggregated molecules. *Chem. Rev.*, 102:3171–3212, 2002.

[44] E. V. Tsiper. Variational procedure and generalized Lanczos recursion for small-amplitude classical oscillations. *JETP Letters*, 70(11):751–755, 1999.

[45] E. V. Tsiper. A classical mechanics technique for quantum linear response. *J. Phys. B: At. Mol. Opt. Phys.*, 34(12):L401–L407, 2001.

[46] H. A. van der Vorst. A generalized Lanczos scheme. *Math. Comp.*, 39(160):559–561, 1982.

[47] D. S. Watkins. *The Matrix Eigenvalue Problems: GR and Krylov Subspace Methods*. SIAM, Philadelphia, 2007.

[48] J. H. Wilkinson. *The Algebraic Eigenvalue Problem*. Oxford University Press, Oxford, 1965.