ELSEVIER

Computational Materials Science 2 (1994) 326–340

COMPUTATIONAL MATERIALS SCIENCE

Large scale electronic structure calculations using the Lanczos method

Lin-Wang Wang, Alex Zunger

National Renewable Energy Laboratory, Golden, CO 80401, USA

Received 17 February 1994; accepted 24 February 1994

Abstract

The orthogonality requirement in either iterative diagonalization or conjugate gradient approaches to the single particle Schrödinger equation $\hat{H}\psi = E\psi$ leads to an overall N^3 scaling of the effort with the number N of atoms. We show that the Lanczos method circumvents this problem even when applied to all occupied states. Our implementation shows that the method is stable, exact, scales as N^2 for N around a few hundreds, and is thus optimally suited for such mid-size (100 – 1000 atoms) quantum systems. The analogy between the basic Lanczos equations and Anderson's localization in a disordered one-dimensional tight-binding chain is pointed out and used to gain some insights into improved convergence and stability of the method. For a 900-atom Si cluster tested here using pseudopotentials and a plane wave basis, the Lanczos method is about an order of magnitude faster than the state-of-the-art preconditioned conjugate gradient method using the same pseudopotentials and basis set.

1. Introduction

While recent developments in computational strategies [1] enable first principles electronic structure calculations for systems with up to 100 atoms, rapid experimental advances are constantly shifting interest to quantum systems calculations of large quantum systems [5], but not for *total* energy calculations that require *all* occupied eigensolutions. Although there are several promising proposals for total-energy electronic structure method with a linear-in-size (N) scaling of the effort [6–10], these are still in their formative stages and the cross-over size

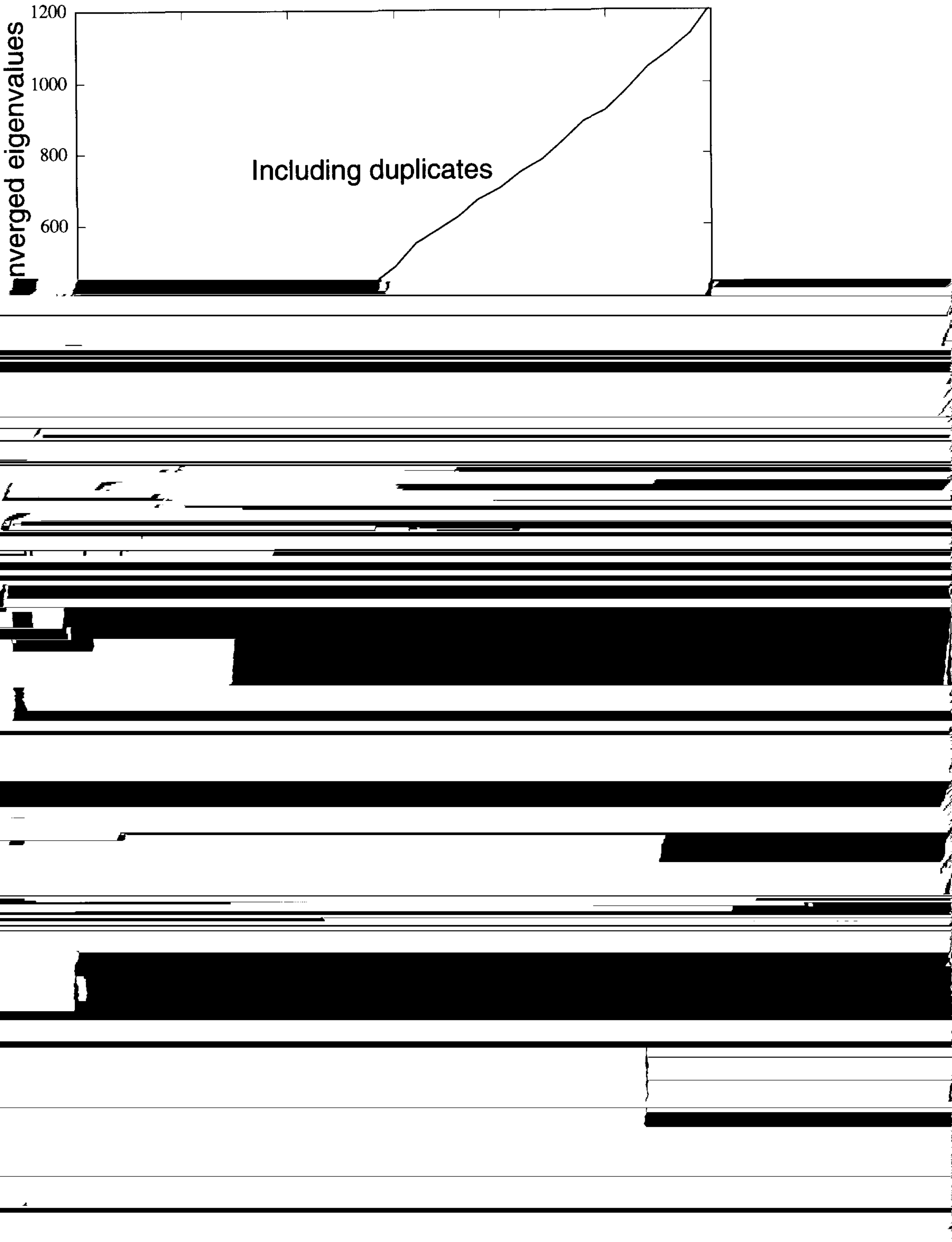

Fig. 5. The Number of converged eigenvalues as functions of the Lanczos iteration index i. Since this system has 120 occupied

from Fig. 4, most of the eigenstates have been converged far before N_t is reached, so for each E_r^c it is worth doing a few (5 to 10) inverse it-

erations for different matrix dimensions M (<

converged eigenstates generated at each sweep decay as a geometrical series.

(2) The Lanczos procedure described here is

stable. It guarantees that each converged eigen-

360Mbyte.

To test whether this procedure is stable, we

have repeated the calculation of the largest system $Si_{617}H_{316}$ using different starting random wavefunctions $u_1(r)$ in the Lanczos iteration (Eq. (6)). The two sets of the eigenvalues

within 0.0001% of the Lanczos results. For systems larger than Si…, the computational

times denoted by "E" in Table 2 have been estimated from the known scaling of the orthogonalization part (N^3 scaling) and the $H\psi(r)$ part (N^2 scaling). It can be seen from Table 2

References

[1] M.C. Payne, M.P. Teter, D.C. Allan, T.A. Arias and

[11] C. Lanczos, J. Res. Nat. Bur. Stand. 45 (1950) 255.

A1133.