Fast iterative diagonalization of nonlocal pseudopotential Hamiltonians using the fast Fourier transformation

S. Goedecker and K. Maschke
Institut de Physique Appliquée, Département de Physique, Ecole Polytechnique Fédérale de Lausanne, PHB-Ecublens, CH-1015 Lausanne, Switzerland
(Received 18 September 1989)

Many electronic-structure calculations are using nonlocal pseudopotentials in connection with iterative diagonalization methods. The most time-consuming steps in those calculations are the calculations of the n^2 matrix elements and the subsequent diagonalization of this matrix, which requires the repeated calculation of matrix vector products. Using conventional matrix multiplication this takes an order of n^2 operations. We present a new technique which significantly reduces the computational effort for **k** points of high symmetry. Since the Hamiltonian is defined as an operator, no individual matrix elements are required. Furthermore, it allows the use of the fast Fourier transform to do the matrix times vector multiplication.

INTRODUCTION

It is well known that the matrix multiplication $\underline{H}\mathbf{x}$, where $\underline{H}$ is the $n\times n$ Hamiltonian matrix expressed in plane waves, can be done with the help of a fast Fourier transformation with $n\log_2 n$ operations if the potential is local.[1] This method takes advantage of the fact that the kinetic energy is diagonal in Fourier space whereas the potential energy is diagonal in real space. To calculate the potential energy, one therefore first transforms into real space, multiplies with the potential, and then transforms back. For large n, this is significantly faster than conventional matrix multiplication. Unfortunately, no exact method has hitherto been known which allows the use of the fast Fourier transformation in connection with nonlocal potentials. This has prevented the use of nonseparable nonlocal potentials in large-scale computations such as molecular-dynamics simulations.[2]

THE ALGORITHM FOR NONLOCAL POTENTIALS

The nonlocal part of the pseudopotential Hamiltonian such as proposed by Bachelet, Hamann, and Schlüter[3] is of the form

$$H=\sum_{l=0}^{l_{\max}} |l\rangle V_l(r)\langle l| ,$$

where $l_{\max}$ is typically 2. The basic idea is now the following. To calculate $\underline{H}\Psi$, where $\Psi=\sum_{\mathbf{k}} c^{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{x}}$, one first splits up Ψ into components corresponding to the different angular moments and a remainder: $\Psi=\Phi_{l=0}+\Phi_{l=1}+\cdots+\Phi_{l=l_{\max}}+\Phi_r$. Then one calculates $\Phi_{l=0},\Phi_{l=1},\ldots,\Phi_{l=l_{\max}},\Phi_r$ in real space and multiplies each part with the corresponding local potential $V_l(r)$. Finally one transforms back into Fourier space. Since the transformation from real space into Fourier space can be done with $n\log_2 n$ operations, this part of the calculation will take an order of $n\log_2 n$ operations. We now show how the decomposition of the wave function Ψ into the angular momentum components can be efficiently implemented.

THE DECOMPOSITION OF THE WAVE FUNCTIONS INTO ANGULAR MOMENTUM COMPONENTS

We start with the well-known expansion

$$e^{i\mathbf{k}\cdot\mathbf{r}}=\sum_{l=0}^{\infty} i^l j_l(kr)\sum_{m=-l}^{l} Y_{lm}^*(\theta_{\mathbf{r}},\phi_{\mathbf{r}})Y_{lm}(\theta^{\mathbf{k}},\phi^{\mathbf{k}}) , \quad (1)$$

where **k** is the sum of the Bloch wave vector and a reciprocal-lattice vector. Let us furthermore expand the spherical Bessel functions into an arbitrary set of functions $f_{\lambda,\kappa}(r)$:

$$i^l j_l(kr)=\sum_{\lambda,\kappa}\alpha_{\lambda,\kappa,l}^{k} f_{\lambda,\kappa}(r) . \quad (2)$$

We have chosen to index f by two indices for reasons which will become apparent later on. Inserting (2) into (1) we obtain

$$e^{i\mathbf{k}\cdot\mathbf{r}}=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}\sum_{\lambda,\kappa}\alpha_{\lambda,\kappa,l}^{\mathbf{k}} Y_{lm}(\theta^{\mathbf{k}},\phi^{\mathbf{k}})f_{\lambda,\kappa}(r) \times Y_{lm}^*(\theta_{\mathbf{r}},\phi_{\mathbf{r}}) . \quad (3)$$

We can now interpret the coefficients $\alpha_{\lambda,\kappa,l}^{\mathbf{k}} Y_{lm}^*(\theta^{\mathbf{k}},\phi^{\mathbf{k}})$ as the coefficients of a matrix whose columns are labeled by **k** and whose rows are labeled by λ,κ,l,m. This matrix does the transformation from the basis set $e^{i\mathbf{k}\cdot\mathbf{r}}$ to the basis set $f_{\lambda,\kappa}(r)Y_{lm}(\theta_{\mathbf{r}},\phi_{\mathbf{r}})$. For arbitrary Ψ we can therefore write

$$\Phi_l(\mathbf{r})=\sum_{\lambda,\kappa,m}\beta_{\lambda,\kappa,l,m} f_{\lambda,\kappa}(r)Y_{lm}^*(\theta_{\mathbf{r}},\phi_{\mathbf{r}}) , \quad (4)$$

where

$$\beta_{\lambda,\kappa,l,m}=\sum_{\mathbf{k}}\alpha_{\lambda,\kappa,l}^{k} Y_{lm}(\theta^{\mathbf{k}},\phi^{\mathbf{k}})c^{\mathbf{k}} . \quad (5)$$

Equations (4) and (5) are the basic equations. We have now to find a basis set $f_{\lambda,\kappa}(r)$ which allows a fast evaluation of (4) and (5). Gonze and Vigneron[4] have proposed in a similar context to use polynomials as the expansion set for the spherical Bessel functions. We will choose a different basis set which allows an exact expansion of the spherical Bessel functions. If the Bloch wave vector is a high-symmetry point in the Brillouin zone, the **k**'s can be grouped into a relatively small number, n_c, of classes with the same length. We therefore use all the spherical Bessel functions corresponding to a length of **k** represented by one of the classes as the basis set:

$$f_{\lambda,\kappa}(r)=i^l j_\lambda(\kappa r) \ .$$

It follows from (2) that

$$\alpha^{\mathbf{k}}_{\lambda,\kappa,l}=\delta_{\lambda,l}\delta_{\kappa,k} \ .$$

Equation (5) then becomes

$$\beta_{\lambda,\kappa,l,m}=\delta_{\lambda,l}\sum_{\substack{\mathbf{k}\\(|\mathbf{k}|=\kappa)}} Y_{l,m}(\theta^{\mathbf{k}},\phi^{\mathbf{k}})c^{\mathbf{k}}$$
$$=\delta_{\lambda,l}\tilde{\beta}_{\kappa,l,m} \qquad (6)$$

and (4) becomes

$$\Phi_l(\mathbf{r})=\sum_\kappa\sum_m \tilde{\beta}_{\kappa,l,m} i^l j_l(\kappa r) Y^*_{lm}(\theta_{\mathbf{r}},\phi_{\mathbf{r}}) \ . \qquad (7)$$

Let us now discuss the number of operations which are necessary to evaluate (6) and (7). If there are n_c different classes, we have to calculate n_c different $\tilde{\beta}_{\kappa,l,m}$. Exact numbers for n_c are given in Table I for the case of the simple-cubic lattice. To calculate all $\tilde{\beta}_{\kappa,l,m}$, we need an order of n operations, where n is the number of plane waves. This is negligible. In (7) we have to perform a summation over an order of n_c terms. If we would calculate the wave function on all the n grid points in real space, we would arrive at an order of $n_c n$ operations. In practice, however, we have to calculate the wave function only in the region where the pseudopotential is not negligible, which can result in a significant reduction for cells with many atoms.

TABLE I. The number of classes of **k**'s with different length at the Γ point for a simple-cubic lattice.

Number of **k**'s	123	1419	14147	124487
Number of **k**'s with different length, n_c	8	42	189	802

CONCLUSIONS

We have presented an exact method to evaluate $\underline{H}\mathbf{x}$ which allows a considerable increase in speed in the calculation of the energy eigenvalues at high-symmetry points. The latter condition is ideally fulfilled in cluster calculations[3] where one usually is interested in the Γ point of a cubic structure. If the eigenstates are required for arbitrary Bloch wave vectors or if the structure has only low symmetry, Eqs. (4) and (5) can be the starting point for approximations. One can, for instance, group **k** vectors of similar lengths into one class. This would require only an order of $n^{4/3}$ operations for the decomposition.

[1]J. L. Martins and M. L. Cohen, Phys. Rev. B **37**, 6143 (1988).
[2]D. Hohl, R. O. Jones, R. Car, and M. Parrinello, J. Chem. Phys. **89**, 6823 (1988).
[3]G. B. Bachelet, D. R. Hamann, and M. Schlüter, Phys. Rev. B **26**, 4199 (1982).
[4]X. Gonze, J. P. Vigneron, and J. P. Michenaud, J. Phys. Condens. Matter **1**, 525 (1989).