

Operator approach in the linearized augmented-plane-wave method: Efficient electronic-structure calculations including forces

S. Goedecker and K. Maschke

Ecole Polytechnique Fédérale de Lausanne, Institut de Physique Appliquée, CH-1015 Lausanne, Switzerland

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The most efficient algorithms for the calculation of the electronic and structural properties of solids and molecules are based on iterative diagonalization methods or the Car-Parrinello molecular-dynamics approach. In these methods, the Hamiltonian is used as an operator and it is therefore not necessary to know the explicit form of the Hamiltonian matrix. We will show that the full-potential linearized augmented-plane-wave (LAPW) method can be formulated in operator form, and that this formulation allows very efficient electronic-structure calculations. We furthermore derive an approach for the calculation of forces in density-functional calculations, which eliminates complications arising from the self-consistency requirement. We then apply this approach to the LAPW method.

INTRODUCTION

The linearized augmented-plane-wave (LAPW) method^{1,2} in its full-potential versions³ is a reliable first-principles method of calculating electronic properties of periodic solids. It can be applied to materials containing all the elements of the Periodic Table. The pseudopotential method,⁴ which is the main alternative, has the disadvantage that the pseudopotentials require an unrealistically large number of plane waves for certain elements because their pseudopotential has a hard core. For elements with soft core pseudopotentials, it turns out that pseudopotential methods in operator^{5,6} form are usually faster than the LAPW method, especially if the Kleinman-Bylander⁷ separable form is chosen. We will show that the LAPW method can be reformulated as an operator method and that the numerical effort is then comparable to the numerical effort for a separable pseudopotential. One thus can handle large numbers of plane waves at low computational cost.

In the second part of this paper we will discuss the efficient calculation of forces in the LAPW operator method. The quantum-mechanical forces acting on the nuclei in solids are an important quantity in structure calculations. If forces can be calculated directly, the ground-state geometry of the nuclei can be found much more efficiently and accurately than by simply varying the positions of the nuclei and perhaps calculating forces by numerical differentiation of the total energy. Furthermore, they are required in quantum-mechanical molecular-dynamics simulations.⁵ We show that in the case of the LAPW method a simple analytical formula for the forces can be found and discuss how it can efficiently be implemented. The obstacles which prevented the use of the LAPW method in large-scale electronic structure calculations such as quantum-mechanical molecular-dynamics calculations should hereby be removed.

REFORMULATION OF THE LAPW METHOD

Let us, for simplicity, first consider a single atom at the origin in the unit cell and a Bloch wave function $\Phi_{\mathbf{k}}(r)$. For simplicity we will, in the following, drop the index \mathbf{k} and we will use L to denote the tuple l, m . The wave function in the muffin-tin sphere is given by

$$\begin{aligned}\Phi_{\text{MFT}}(\mathbf{r}) &= \sum_L \alpha_L \phi_L^1 + \beta_L \phi_L^2 \\ &= \sum_L \alpha_L R_L^1(r) Y_L(\hat{\mathbf{r}}) + \beta_L R_L^2(r) Y_L(\hat{\mathbf{r}}).\end{aligned}\quad (1)$$

In the usual LAPW method R^1 is the solution of the radial Schrödinger equation at a reference energy and R^2 is its derivative with respect to energy. We choose R^1 and R^2 to be linear combinations of their LAPW counterparts such that

$$R_L^1(r_c) = 1, \quad \frac{\partial R_L^1(r_c)}{\partial r} = 0, \quad (2)$$

$$R_L^2(r_c) = 0, \quad \frac{\partial R_L^2(r_c)}{\partial r} = 1, \quad (3)$$

where r_c is the radius of the muffin-tin sphere. The space spanned by these transformed radial functions is, of course, exactly the same as the space spanned by the original ones.

In the interstitial region the wave function Φ_{IST} is written as a superposition of plane waves

$$\Phi_{\text{IST}} = \sum_{\mathbf{K}} c_{\mathbf{K}} e^{i\mathbf{K}\cdot\mathbf{r}},$$

with $\mathbf{K} = \mathbf{k} + \mathbf{G}$ where \mathbf{G} is a reciprocal-lattice vector. We now require that the wave function and its gradient be continuous across the boundary of the sphere for all angular momentum up to l_{max} . This gives

$$\alpha_L = \int Y_L^* \Phi_{\text{IST}}|_{|\mathbf{r}|=r_c} d\Omega, \\ \beta_L = \int Y_L^* \hat{\mathbf{n}} \cdot \nabla \Phi_{\text{IST}}|_{|\mathbf{r}|=r_c} d\Omega.$$

We define projectors which return the value and radial derivative of a plane wave on the surface of the muffin-tin sphere as

$$p_L^{1*}(\mathbf{K}) = \int Y_L^* e^{i\mathbf{K}\cdot\mathbf{r}}|_{|\mathbf{r}|=r_c} d\Omega = 4\pi i^l j_l(Kr_c) Y_L^*(\hat{\mathbf{K}}), \\ p_L^{2*}(\mathbf{K}) = \int Y_L^* \hat{\mathbf{n}} \cdot \nabla e^{i\mathbf{K}\cdot\mathbf{r}}|_{|\mathbf{r}|=r_c} d\Omega \\ = \frac{4\pi i^l}{2l+1} [l j_{l-1}(Kr_c) - (l-1) j_{l+1}(Kr_c)] Y_L^*(\hat{\mathbf{K}}),$$

where $\hat{\mathbf{n}}$ is the unit outwardly directed radial vector. The expansion coefficients of the muffin-tin region are then given by scalar products

$$\alpha_L = \sum_{\mathbf{K}} p_L^{1*}(\mathbf{K}) c_{\mathbf{K}}, \quad (4)$$

$$\beta_L = \sum_{\mathbf{K}} p_L^{2*}(\mathbf{K}) c_{\mathbf{K}}. \quad (5)$$

We can now eliminate the coefficients α_L and β_L by the continuity requirements (4) and (5) for the wave function and the derivative. The plane-wave expansion coefficients $c_{\mathbf{K}}$ are then the only variational degrees of freedom and the Hamiltonian and overlap matrix are only labeled by \mathbf{K} . After some algebraic manipulations sketched in Appendix A we obtain the following Hamiltonian and overlap matrix $H_{\mathbf{K},\mathbf{K}'}$ and $S_{\mathbf{K},\mathbf{K}'}$:

$$H_{\mathbf{K},\mathbf{K}'} = \Omega \frac{1}{2} \delta_{\mathbf{K},\mathbf{K}'} \mathbf{K}^2 - \frac{1}{2} \mathbf{K} F_{\mathbf{K}-\mathbf{K}'} \\ + \int_{\text{IST}} V(\mathbf{r}) e^{-i(\mathbf{K}-\mathbf{K}')\cdot\mathbf{r}} d\mathbf{r} \\ + \sum_{L,L'} \sum_{\kappa,\kappa'} p_L^{\kappa}(\mathbf{K}) H_{L,L'}^{\kappa,\kappa'} p_{L'}^{\kappa'*}(\mathbf{K}'), \quad (6)$$

$$S_{\mathbf{K},\mathbf{K}'} = \Omega \delta_{\mathbf{K},\mathbf{K}'} - F_{\mathbf{K}-\mathbf{K}'} + \sum_L \sum_{\kappa,\kappa'} p_L^{\kappa}(\mathbf{K}) S_{L,L'}^{\kappa,\kappa'} p_{L'}^{\kappa'*}(\mathbf{K}'), \quad (7)$$

where

$$F_{\mathbf{q}}(r_c) = \int_{\text{MFT}} e^{-i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} \\ = 4\pi \frac{\sin(qr_c) - qr_c \cos(qr_c)}{q^3}, \quad (8)$$

$$S_{L,L'}^{\kappa,\kappa'} = \int_0^{r_c} (R_L^{\kappa} R_{L'}^{\kappa'}) r^2 dr, \quad (9)$$

$$H_{L,L'}^{\kappa,\kappa'} = \delta_{L,L'} \frac{1}{2} \left[\int_0^{r_c} \left[r \frac{\partial R_L^{\kappa}}{\partial r} \right] \left[r \frac{\partial R_{L'}^{\kappa'}}{\partial r} \right] dr \right. \\ \left. + l(l+1) \int_0^{r_c} R_L^{\kappa} R_{L'}^{\kappa'} dr \right] \\ + \int_0^{r_c} R_L^{\kappa}(r) Y_L^*(\hat{\mathbf{r}}) V(\mathbf{r}) Y_{L'}(\hat{\mathbf{r}}) R_{L'}^{\kappa'}(r) dr. \quad (10)$$

Similarly, in the case of several atoms in the cell at positions $R_1, \dots, R_{N_{\text{at}}}$, and for nonoverlapping muffin-tin spheres, the Hamiltonian and overlap matrix can be written as

$$H_{\mathbf{K},\mathbf{K}'} = \Omega \frac{1}{2} \delta_{\mathbf{K},\mathbf{K}'} \mathbf{K}^2 - \frac{1}{2} \mathbf{K} F_{\mathbf{K}-\mathbf{K}'}^{\text{MFT}} \mathbf{K}' \\ + \int_{\text{IST}} V(\mathbf{r}) e^{-i(\mathbf{K}-\mathbf{K}')\cdot\mathbf{r}} \\ + \sum_{\mu} \sum_{L,L'} \sum_{\kappa,\kappa'} p_{\mu,L}^{\kappa}(\mathbf{K}) H_{\mu,L,L'}^{\kappa,\kappa'} p_{\mu,L'}^{\kappa'*}(\mathbf{K}'), \quad (11)$$

$$S_{\mathbf{K},\mathbf{K}'} = \Omega \delta_{\mathbf{K},\mathbf{K}'} - F_{\mathbf{K}-\mathbf{K}'}^{\text{MFT}} + \sum_{\mu} \sum_L \sum_{\kappa,\kappa'} p_{\mu,L}^{\kappa}(\mathbf{K}) S_{\mu,L,L'}^{\kappa,\kappa'} p_{\mu,L'}^{\kappa'*}(\mathbf{K}'), \quad (12)$$

where

$$F_{\mathbf{K}-\mathbf{K}'}^{\text{MFT}} = \sum_{R_{\mu}} F_{\mathbf{K}-\mathbf{K}'}(\mathbf{R}_{\mu}, r_{c\mu}).$$

$F_{\mathbf{K}}(\mathbf{R}_{\mu}, r_{c\mu})$ is the Fourier transform of a step function which is unity in the muffin-tin sphere centered at \mathbf{R}_{μ} with radius $r_{c\mu}$ and zero elsewhere. The projectors $p_{\mu,L}^{\kappa}$ project out the value and radial derivative of a wave function on the muffin-tin sphere centered at \mathbf{R}_{μ} . The parameters $H_{L,L'}^{\kappa,\kappa'}(\mu)$ and $S_{L,L'}^{\kappa,\kappa'}(\mu)$ describe the properties of the atom at \mathbf{R}_{μ} .

This formulation differs in two points from the conventional LAPW formulation. First, as was mentioned above, the radial functions R_L^{κ} and $R_L^{\kappa'}$ are chosen in a different way. Second, the kinetic energy is written as $\nabla\Phi^* \cdot \nabla\Phi$ instead of $\Phi\nabla^2\Phi^*$. This form of the kinetic energy will simplify the calculation of the forces because it does not require boundary terms arising from the discontinuity of the second derivative on the surface of the muffin-tin sphere.⁸ Furthermore the Hamiltonian matrix is automatically Hermitian. This form of the kinetic energy was also chosen by Slater in his original formulation of the APW method.⁹ As a consequence of these two differences the parameters $H_{\mu,L,L'}^{\kappa,\kappa'}$ and $S_{\mu,L,L'}^{\kappa,\kappa'}$ are not identical to the usual potential parameters.¹

BASIC VARIABLES FOR THE OPERATION COUNT

We first distinguish between the numerical effort associated with the muffin-tin and the interstitial regions. The part associated with the muffin-tin region is small since it is linear with respect to the size of the system. It becomes even less important if the frozen-core approximation¹⁰ is used, which is known to be very well satisfied. Since the calculation of forces in the following paragraph is considerably simplified if the frozen-core approximation is used, we will assume from now on that all calculations are done in the frozen-core approximation.

The time-consuming part comes, then, from the interstitial region. The basic variables defining the numerical effort in this region are N_{at} , the number of atoms in the cell; N_{PW} , the number of plane waves; N_{el} , the number of valence electron orbitals.

Those three quantities are extensive quantities. One can define the corresponding intensive quantities as $n_{\text{PW}} = N_{\text{PW}}/N_{\text{at}}$ and $n_{\text{el}} = N_{\text{el}}/N_{\text{at}}$. Another important term is the number of separable matrix terms in (6) and (7), which can be written as $O_{\mathbf{K},\mathbf{K}'} = f(\mathbf{K})g(\mathbf{K}')$. If l is running from 0 to l_{max} and m from $-l$ to l , there are $(l_{\text{max}}+1)^2$ possible combinations. A typical value for

l_{\max} in a LAPW calculation is eight. If the muffin-tin sphere is small, it is clear that less multipoles must be included in the expansion of the potential in the muffin-tin sphere. The number of plane waves which is required increases, however. As a rule of thumb, the smallest wavelength of the plane-wave expansion should not be larger than r_c . In the following operation count we will only include terms with a dependence which is higher than quadratic with respect to the size of the system N_{at} . Furthermore, we will not discuss cubic terms $N_{\text{el}}^2 N_{\text{PW}}$ which arise from orthogonalization or similar procedures in some iterative diagonalization methods.

OPERATION COUNT

Setting up the Hamiltonian and overlap matrix and solving the generalized eigenvalue problem, as well as calculating the coefficients $\alpha_L(\mathbf{R}_\mu)$ and $\beta_L(\mathbf{R}_\mu)$, which are needed to calculate the charge distribution in the muffin-tin sphere, takes most of the CPU time in a typical LAPW program. Iterative diagonalization methods turned out to be the most efficient in electronic-structure calculations,¹¹ where only the lowest eigenstates are calculated. In most iterative methods the explicit matrices are never needed but the matrices must only be known as operators, i.e., one must be able to calculate matrix-times-vector products. The calculation of these products usually takes most of the time within the diagonalization routine. In the operator method the dependence with respect to the number of plane waves is reduced from n_{PW}^2 to n_{PW} for large systems as shown in Appendix B. This gives considerably more flexibility in choosing the muffin-tin radii. The ability to use a large number of plane waves n_{PW} is useful whenever small muffin-tin radii are needed, which may happen in the following cases.

(1) Systems containing simultaneously very large and very small atoms. This causes difficulties because the muffin-tin radii must all be roughly of the same size to avoid a nearly singular overlap matrix which gives rise to numerical problems.

(2) Systems containing semicore states. For these systems, the two variational degrees of freedom for each l, m component given by R_l^1 and R_l^2 are not sufficient to describe semicore states. One must either use modified linear theories¹² with more degrees of freedom or include the semicore states as valence states. If the muffin-tin radius is chosen sufficiently small both valence and semicore states can be treated with the same panel.¹³

Even in standard systems, the method should usually be faster. In the conventional implementation the dominating part is, for typical values, the term $N_{\text{at}}^3 n_{\text{PW}}^2 (l_{\max} + 1)^2$ which arises from setting up the matrix as shown in Appendix B. This is to be compared with the term

$$N_{\text{at}}^3 n_{\text{PW}} n_{\text{it}} n_{\text{el}} (l_{\max} + 1)^2$$

(Appendix B) in the operator approach. Since $n_{\text{it}} n_{\text{el}}$ is usually less than n_{PW} the speed up is given by the ratio of these two numbers.

FORCES

If a complete basis set is used, which is, however, never the case in numerical calculations, the force acting on a nucleus at \mathbf{R}_ν is given by the Hellman-Feynman theorem:¹⁴

$$-\mathbf{f}_{\text{HF}}(\mathbf{R}_\nu) = Q_\nu \nabla_{\mathbf{R}_\nu} \left[\int \frac{\rho(r')}{|\mathbf{R}_\nu - \mathbf{r}'|} + \sum_{\mu \neq \nu} \frac{Q_\mu}{|\mathbf{R}_\nu - \mathbf{R}_\mu|} \right],$$

where ρ is the electronic charge density, \mathbf{R}_μ the position of the nuclei μ , and Q_μ their charge. In all current computational methods using Gaussians or Slater orbitals as basis functions, as well as in the LAPW, linearized-muffin-tin orbital (LMTO) and pseudopotential method additional terms, called Pulay corrections,¹⁵ \mathbf{f}_p must be added. The actual force \mathbf{f} acting on the nucleus which is given by the derivative of the total energy with respect to the position of the ν th nucleus is then

$$\mathbf{f}(\mathbf{R}_\nu) = \mathbf{f}_{\text{HF}}(\mathbf{R}_\nu) + \mathbf{f}_p(\mathbf{R}_\nu).$$

Even though \mathbf{f}_p is a correction term, it is desirable to include it because in this way forces are given with an accuracy corresponding to the accuracy of energies.¹⁶ \mathbf{f}_{HF} , on the other hand, has only the accuracy of a charge density which is much lower. Unfortunately the analytical form for the Pulay corrections are sometimes fairly complicated and numerically expensive. In the next section we derive a formula for the forces, which is based on the eigenvalues only, and which allows us to calculate the forces in the LAPW method easily.

A GENERAL FORMULA FOR FORCES IN A SELF-CONSISTENT CALCULATION

Let us consider a system of N_{at} nuclei whose positions are given by $\mathbf{R}_1, \dots, \mathbf{R}_{N_{\text{at}}}$ together with its N_{el} occupied electronic wave functions $\Phi_1, \dots, \Phi_{N_{\text{el}}}$. Omitting the trivial nuclei-nuclei interaction term, the total electronic energy E_{tot} of the system is given in the local-density approximation (LDA) (Ref. 17) by

$$E_{\text{tot}} = \sum_{j=1}^{N_{\text{el}}} \langle \Phi_j | (T + V^{\text{ion}} + V^{\text{sc}}) | \Phi_j \rangle - \frac{1}{2} \int \int \frac{\rho(r)\rho(r')}{|\mathbf{r} - \mathbf{r}'|} + \int \varepsilon(\rho(r)) - \mu(\rho(r))\rho(r), \quad (13)$$

where V^{ion} is the ion potential

$$V^{\text{ion}}(r) = \sum_{\mu=1}^{N_{\text{at}}} \frac{Q_\mu}{|\mathbf{r} - \mathbf{R}_\mu|}$$

and V^{sc} the self-consistent potential

$$V^{\text{sc}} = \int \frac{\rho(r')}{|\mathbf{r} - \mathbf{r}'|} + \mu(\rho(r)).$$

Remember that we are using the frozen-core approximation. This means that ρ is the valence-electron density and Q_μ is the ionic charge, i.e., the charge of the nucleus minus the charge of the core electrons. Let us now consider an infinitesimally different ionic configuration $\mathbf{R}_1 + \Delta\mathbf{R}_1, \dots, \mathbf{R}_N + \Delta\mathbf{R}_N$. This implies a new ionic po-

tential $V^{\text{ion}} + \Delta V^{\text{ion}}$, new wave functions $\Phi_j + \Delta\Phi_j$, and a new charge density which up to first order is given by

$$\rho + \Delta\rho = \sum_j |\Phi_j|^2 + \sum_j \Phi_j^* \Delta\Phi_j + \Delta\Phi_j^* \Phi_j.$$

Under the assumption that occupied and unoccupied levels do not cross, and using $\partial(\epsilon\rho)/\partial\rho = \mu$, it is now easy to show that the first-order change ΔE_{tot} is given by

$$\begin{aligned} \Delta E_{\text{tot}} = & \sum_{j=1}^{N_{\text{el}}} \langle \Phi_j | (T + V^{\text{ion}} + V^{\text{sc}}) | \Delta\Phi_j \rangle \\ & + \langle \Delta\Phi_j | (T + V^{\text{ion}} + V^{\text{sc}}) | \Phi_j \rangle \\ & + \langle \Phi_j | \Delta V^{\text{ion}} | \Phi_j \rangle. \end{aligned}$$

This is an interesting result. Since there is no term $\langle \Phi_j | \Delta V^{\text{sc}} | \Phi_j \rangle$, it tells us that V^{sc} does not change to first order with the position of the nuclei and that the change in the total energy is given by the sum of the changes in the Kohn-Sham eigenvalues E_i for a fixed potential V^{sc} (i.e., independent of the position of the nuclei),

$$\Delta E_{\text{tot}} = \sum_{j=1}^{N_{\text{el}}} \Delta E_j = \sum_{j=1}^{N_{\text{el}}} \Delta \langle \Phi_j | (T + V^{\text{ion}} + V^{\text{sc}}) | \Phi_j \rangle. \quad (14)$$

The complicated self-consistent problem is thus reduced to a much simpler eigenvalue problem. The eigenvalues are given by

$$E_j(\mathbf{R}_1, \dots, \mathbf{R}_{N_{\text{at}}}) = \frac{\sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}, \mathbf{K}'}^j H_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}, \mathbf{K}'}^j}{\sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}, \mathbf{K}'}^j S_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}, \mathbf{K}'}^j}. \quad (15)$$

The condition that V^{sc} be fixed is implied in Eq. (15) since the Hamiltonian matrix is defined for a fixed potential. To calculate the force in the x direction acting on the atom at position $\mathbf{R}_v = (x_v, y_v, z_v)$ we must calculate $\sum_j dE_j/dx_v$. When calculating this derivative we have to take into consideration two dependencies: First, parts of H and S depend explicitly on the nuclei positions; second, the expansion coefficients $c_{\mathbf{K}}^j$ of the wave functions Φ_j depend on the ion positions, i.e.,

$$\frac{dE_j}{dx_v} = \frac{\partial E_j}{\partial x_v} + \sum_{\mathbf{K}} \frac{\partial E_j}{\partial c_{\mathbf{K}}^j} \frac{dc_{\mathbf{K}}^j}{dx_v}. \quad (16)$$

The coefficients $c_{\mathbf{K}}^j(\mathbf{R}_1, \dots, \mathbf{R}_{N_{\text{at}}})$ are the coefficients of the solution for the configuration $\mathbf{R}_1, \dots, \mathbf{R}_{N_{\text{at}}}$. This guarantees that the orbitals are orthogonal for any configuration. The explicit form of the $c_{\mathbf{K}}^j$'s is, of course, not known. However, because of the eigenvalue condition $\partial E_j / \partial c_{\mathbf{K}}^j = 0$, the second term in (16) is zero and the explicit form of the $c_{\mathbf{K}}^j$'s is not needed. We therefore have (putting $\langle \Phi_j | S | \Phi_j \rangle = 1$)

$$\frac{dE_{\text{tot}}}{dx_v} = \sum_j \frac{dE_j}{dx_v} = \sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}, \mathbf{K}'}^j \left[\frac{\partial H_{\mathbf{K}, \mathbf{K}'}}{\partial x_v} - E_j \frac{\partial S_{\mathbf{K}, \mathbf{K}'}}{\partial x_v} \right] c_{\mathbf{K}, \mathbf{K}'}^j. \quad (17)$$

Equation (17) is the central formula for the calculation of

forces. It can be considered as a generalization of the Hellmann-Feynman theorem, since it is also valid in the case where the basis functions depend on the positions of the nuclei. Let us note that such a simple formula can only be found for the sum of the Hellmann-Feynman and Pulay forces but not for the Pulay force alone. What remains to be done is to calculate (17) in the case of the LAPW method just by using elementary calculus.

TREATMENT OF FORCES IN MOLECULAR-DYNAMICS SIMULATION

In Car-Parrinello molecular-dynamics simulations the wave functions Φ_j are usually not exact eigenfunctions for the self-consistent potential. Even in this case (14) and (15) still hold if one considers the quantities

$$E_j = \langle \Phi_j | (T + V^{\text{ion}} + V^{\text{sc}}) | \Phi_j \rangle$$

not as eigenvalues, but as expectation values. The expansion coefficients $c_{\mathbf{K}}^j$ and the positions of the nuclei are considered as independent variables in Car-Parrinello molecular-dynamics simulations. Therefore one does not calculate the total derivative of the total energy with respect to \mathbf{R}_μ but only the partial derivative, and the second term in (16) must not be included. As a consequence of this, the orthogonality requirement for the wave functions is not automatically satisfied but must explicitly be included. The force acting on the nuclei is thus given by (17) under the additional requirement of orthogonality which can be included by Lagrange multiplier techniques.

If the wave functions are exact eigenfunctions of the self-consistent potential, the total energy is only a function of the positions of the nuclei. Since the total energy is invariant under translations the sum of all forces vanishes:

$$\begin{aligned} 0 &= \nabla_{\mathbf{r}} E_{\text{tot}}(\mathbf{R}_1 + \mathbf{r}, \dots, \mathbf{R}_{N_{\text{at}}} + \mathbf{r})|_{\mathbf{r}=0} \\ &= \sum_{\mu} \nabla_{\mathbf{R}_\mu} E_{\text{tot}} = - \sum_{\mu} \mathbf{f}(\mathbf{R}_\mu). \end{aligned}$$

In the case of Car-Parrinello molecular-dynamics simulations the sum of all forces does not vanish and the center of mass of the nuclei may be accelerated. This acceleration is, however, small since the ratio of the electron mass (which is not the physical mass) to the ion mass is small.

FORCES IN THE LAPW METHOD

The formulation of the Hamiltonian and overlap matrix (11) and (12) allows us now to calculate (17) easily. The matrix element $H_{\mathbf{K}, \mathbf{K}'}$ can obviously be rewritten as

$$\begin{aligned} H_{\mathbf{K}, \mathbf{K}'} &= \int \Theta_{\text{IST}}(\mathbf{r}) g_{\mathbf{K}, \mathbf{K}'}(\mathbf{r}) d\mathbf{r} \\ &+ \sum_{\mu} \sum_{L, L'} \sum_{\kappa, \kappa'} p_{\mu, L}^{\kappa}(\mathbf{K}) \left[\int \Theta_{\mu}(\mathbf{r}) g_{\mu, L, L'}^{\kappa, \kappa'}(\mathbf{r}) d\mathbf{r} \right] \\ &\quad \times p_{\mu, L'}^{\kappa'}(\mathbf{K}'), \end{aligned} \quad (18)$$

where

$$g_{\mathbf{K}, \mathbf{K}'}(\mathbf{r}) = \frac{1}{2} \nabla e^{i\mathbf{K} \cdot \mathbf{r}} \nabla e^{-i\mathbf{K}' \cdot \mathbf{r}} + e^{i\mathbf{K} \cdot \mathbf{r}} \nabla e^{-i\mathbf{K}' \cdot \mathbf{r}}, \quad (19)$$

$$g_{\mu,L,L'}^{\kappa,\kappa'}(\mathbf{r}) = \delta_{L,L'} \frac{1}{2} \nabla \phi_{\mu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\mu) \cdot \nabla \phi_{\mu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\mu) \\ + \phi_{\mu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\mu) V \phi_{\mu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\mu) . \quad (20)$$

The basis functions in the muffin-tin sphere $\phi_{\mu,L}^{\kappa}$ are defined by Eq. (1), Θ_{IST} is a step function which is unity in the interstitial region and zero elsewhere, and Θ_μ a step function which is unity in the μ th muffin-tin sphere and zero elsewhere. To evaluate the first term in (17),

$$\sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \nabla_{\mathbf{R}_\nu} H_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}'}^j , \quad (21)$$

we will assume that the wave function and its derivative as well as the potential be continuous across the muffin-

tin spheres. This is a good approximation because according to (4) and (5) these conditions are exactly fulfilled for the low- l components, which make up most of the wave function on the outside of the muffin-tin sphere. Under these circumstances the contributions to (21) from taking the derivative of (18) with respect to the Θ functions cancel since the gradient of Θ_{IST} is the negative of the gradient of Θ_μ . Note that we would get an additional surface contribution if we had written the kinetic energy as $\Psi^* \nabla^2 \Psi$ because $\nabla^2 \Psi$ is not continuous across the muffin-tin sphere. Taking the derivative of (18) with respect to the potential, we obtain the electron-ion part of the Hellmann-Feynman force,

$$-f_{\text{HF}}(R_\nu) = \sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \left[\int_{\text{IST}} e^{i\mathbf{K}\cdot\mathbf{r}} (\nabla_{\mathbf{R}_\nu} V) e^{-i\mathbf{K}'\cdot\mathbf{r}} d\mathbf{r} \right. \\ \left. + \sum_{L,L'} \sum_{\kappa,\kappa'} p_{\nu,L}^{\kappa}(\mathbf{K}) \left[\int_{\nu} \phi_{\nu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\nu) (\nabla_{\mathbf{R}_\nu} V) \phi_{\nu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\nu) d\mathbf{r} \right] p_{\nu,L'}^{\kappa'}(\mathbf{K}') \right] c_{\mathbf{K}'}^j \\ = Q_\nu \int \nabla_{\mathbf{R}_\nu} \frac{\rho(\mathbf{r}')}{|\mathbf{R}_\nu - \mathbf{r}'|} d\mathbf{r}' . \quad (22)$$

The subscript ν on the integral in (22) means that it is only taken over the ν th muffin-tin sphere. The remaining dependencies of (18) on the ion positions give Pulay forces. Differentiation of (18) with respect to $p_{\mu L}^{\kappa}(\mathbf{K}) p_{\mu L'}^{\kappa'}(\mathbf{K}')$ just brings down a term $i(\mathbf{K}-\mathbf{K}')$ and we obtain

$$\sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \left[-i(\mathbf{K}-\mathbf{K}') \sum_{L,L'} \sum_{\kappa,\kappa'} p_{\nu,L}^{\kappa}(\mathbf{K}) H_{\nu L, L'}^{\kappa,\kappa'} p_{\nu,L'}^{\kappa'}(\mathbf{K}') \right] c_{\mathbf{K}'}^j .$$

Finally, there is still a dependence because the basis functions in the muffin-tin sphere are centered on the nuclei. Using the fact that

$$\nabla_{\mathbf{R}_\nu} \phi_{\nu,L}^{\kappa}(\mathbf{r}-\mathbf{R}_\nu) = -\nabla \phi_{\nu,L}^{\kappa}(\mathbf{r}-\mathbf{R}_\nu) ,$$

we obtain a term

$$-\sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \sum_{L,L'} \sum_{\kappa,\kappa'} p_{\nu,L}^{\kappa}(\mathbf{K}) \left[\int_{\nu} \left\{ \delta_{L,L'} \nabla \left[\frac{1}{2} \nabla \phi_{\nu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\nu) \cdot \nabla \phi_{\nu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\nu) \right] \right. \right. \\ \left. \left. + \nabla \phi_{\nu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\nu) V \phi_{\nu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\nu) + \phi_{\nu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\nu) V \nabla \phi_{\nu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\nu) \right\} d\mathbf{r} \right] p_{\nu,L'}^{\kappa'}(\mathbf{K}') c_{\mathbf{K}'}^j .$$

The part arising from the kinetic energy vanishes because the gradient formula¹⁸ tells us that the derivative of a spherical harmonic contains only spherical harmonics whose l is lowered or raised by one. The part arising from the potential vanishes if the potential has spherical symmetry, but not in the general case. Let us denote this integral by $\mathbf{G}_{\nu,L,L'}^{\kappa,\kappa'}$ where

$$\mathbf{G}_{\nu,L,L'}^{\kappa,\kappa'} = \int \phi_{\nu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\nu) V \nabla \phi_{\nu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\nu) + \nabla \phi_{\nu,L}^{\kappa*}(\mathbf{r}-\mathbf{R}_\nu) V \phi_{\nu,L'}^{\kappa'}(\mathbf{r}-\mathbf{R}_\nu) d\mathbf{r} . \quad (23)$$

The second term in (17) can be handled in the same way and the total Pulay force is given by

$$-f_p(R_\nu) = \sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \left[i(\mathbf{K}-\mathbf{K}') \sum_{L,L'} \sum_{\kappa,\kappa'} p_{\nu,L}^{\kappa}(\mathbf{K}) (H_{\mu,L,L'}^{\kappa,\kappa'} - E_j \delta_{L,L'} S_{\mu,L}^{\kappa,\kappa'}) p_{\nu,L'}^{\kappa'}(\mathbf{K}') \right] c_{\mathbf{K}'}^j \\ - \sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \left[\sum_{L,L'} \sum_{\kappa,\kappa'} p_{\nu,L}^{\kappa}(\mathbf{K}) \mathbf{G}_{\nu,L,L'}^{\kappa,\kappa'} p_{\nu,L'}^{\kappa'}(\mathbf{K}') \right] c_{\mathbf{K}'}^j . \quad (24)$$

In the above expression (24) for the force we have neglected the implicit dependence of the potential parameters $H_{\mu,L,L'}^{\kappa,\kappa'}$ and $S_{\mu,L}^{\kappa,\kappa'}$ on the positions of the ions. This dependence comes from the fact that the form of atomiclike basis functions $\phi_{\mu,L}^1$ and $\phi_{\mu,L}^2$ in the muffin-tin region depends on the ion configuration. The error arising from neglecting this must, however, be very small as can be seen by the following argument. Let $\gamma_\mu(\mathbf{R}_1, \dots, \mathbf{R}_{N_{\text{at}}})$ be a parameter which describes the wave function in the μ th muffin-tin sphere. In principle γ_μ could be chosen such that the total energy is minimized for all possible configurations

$$\frac{\partial E_{\text{tot}}}{\partial \gamma_{\mu}} = 0. \quad (25)$$

In this case this dependence gives no contribution $\Delta f(R_{\nu})$ to the force because

$$\Delta f(R_{\nu}) = \sum_{\mu} \frac{\partial E_{\text{tot}}}{\partial \gamma_{\mu}} \nabla_{\mathbf{R}_{\nu}} \gamma_{\mu}(\mathbf{R}_1, \dots, \mathbf{R}_{N_{\text{at}}}) = 0.$$

Since the minimization of the total energy with respect to the wave functions R_l^1 and R_l^2 is intractable it is usually not done. There are, however, empirical rules which give potential parameters close to the optimal values. Experience shows that with this choice the total energy cannot significantly be lowered and we will therefore assume that (25) is fulfilled. If the wave functions are held fixed as in the case of our pseudopotential version of the LAPW method,¹³ Δf vanishes, of course, too.

To facilitate the numerical evaluation of (24) we pull the gradient over on the wave function

$$\begin{aligned} \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^* [i(\mathbf{K} - \mathbf{K}') p(\mathbf{K}) p^*(\mathbf{K}')] c_{\mathbf{K}'} &= \sum_{\mathbf{K}, \mathbf{K}'} (i\mathbf{K} c_{\mathbf{K}}^*) [p(\mathbf{K}) p^*(\mathbf{K}')] c_{\mathbf{K}'} + \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^* [p(\mathbf{K}) p^*(\mathbf{K}')] (-i\mathbf{K}' c_{\mathbf{K}'}) \\ &= 2 \operatorname{Re} \sum_{\mathbf{K}, \mathbf{K}'} (i\mathbf{K} c_{\mathbf{K}}^*) [p(\mathbf{K}) p^*(\mathbf{K}')] c_{\mathbf{K}'} . \end{aligned}$$

We then obtain the final result for the Pulay force

$$\begin{aligned} \mathbf{f}_p(R_{\nu}) &= -2 \operatorname{Re} \left[\sum_j \sum_{\mathbf{K}, \mathbf{K}'} i\mathbf{K} c_{\mathbf{K}}^{j*} \left[\sum_{L, L'} \sum_{\kappa, \kappa'} p_{\nu, L}^{\kappa}(\mathbf{K}) (H_{\mu, L, L'}^{\kappa, \kappa'} - E_j \delta_{L, L'} S_{\mu, L}^{\kappa, \kappa'}) p_{\nu, L'}^{\kappa'}(\mathbf{K}') \right] c_{\mathbf{K}'}^j \right] \\ &+ \sum_j \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^{j*} \left[\sum_{L, L'} \sum_{\kappa, \kappa'} p_{\nu, L}^{\kappa}(\mathbf{K}) \mathbf{G}_{\nu, L, L'}^{\kappa, \kappa'} p_{\nu, L'}^{\kappa'}(\mathbf{K}') \right] c_{\mathbf{K}'}^j . \end{aligned} \quad (26)$$

OPERATION COUNT FOR THE CALCULATION OF FORCES

The Hellmann-Feynman force can easily be calculated. One first builds up the charge density in real space and then solves Poisson's equation in Fourier space. To sum up the charge density in real space, all orbitals must be transformed into real space which takes order $N_{\text{el}} N_{\text{PW}} \ln(N_{\text{PW}})$ operations. All other operations are negligible. The Pulay force term (26) has only separable terms and the discussion given in Appendix B applies. It can be calculated with order $N_{\text{el}} N_{\text{PW}} (l_{\text{max}} + 1)^2$ operations. The total number of operations to calculate the forces acting on all the N_{at} atoms is, therefore, of the order of

$$N_{\text{el}} N_{\text{PW}} \ln(N_{\text{PW}}) + N_{\text{at}} N_{\text{el}} N_{\text{PW}} (l_{\text{max}} + 1)^2$$

which is similar to the operation count for a single iteration in the diagonalization.

CONCLUSIONS

We have shown that the full potential LAPW method can be reformulated in terms of convolution and separable matrices. In this form the numerical efficiency of the LAPW method is comparable to that of one of the most well-known full-potential electronic structure methods, which is the pseudopotential method by Kleinman and Bylander in the operator form. Since there are more convolution and separable terms than in the Kleinman-Bylander scheme, the prefactor for the operation count is higher. This should, however, be balanced by the fact that less plane waves are required in the LAPW method than in the pseudopotential method. Furthermore, we have shown that forces can be calculated in the operator

approach with an effort which is small compared to that of an electronic structure calculation.

The results for the forces are, with minor modifications, also valid for our pseudopotential version of the LAPW method.¹³ The main difference in the pseudopotential version is that in this case the muffin-tin radii must be chosen smaller to obtain good transferability. The field of large-scale electronic structure calculations such as Car-Parrinello molecular-dynamics simulations was up to now dominated by pseudopotential methods with the exception of the modified APW method¹⁹ of Soler and Williams. It is to be expected that this reformulation of the LAPW method will introduce the LAPW method in this field. Our approach to the calculation of forces in density-functional theory should also be useful in other than LAPW methods.

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APPENDIX A: MUFFIN-TIN INTEGRALS

The integral $\int \Phi^*(\mathbf{r}) \Phi(\mathbf{r})$ and $\int \nabla \Phi^*(\mathbf{r}) \cdot \nabla \Phi(\mathbf{r})$ must be calculated over the muffin-tin sphere for both the plane-wave expansion

$$\Phi(\mathbf{r}) = \sum_{\mathbf{K}} c_{\mathbf{K}} e^{i\mathbf{K} \cdot \mathbf{r}}$$

and the sum of atomiclike functions

$$\Phi(\mathbf{r}) = \sum_L R_L(r) Y_L(\hat{\mathbf{r}}),$$

where

$$R_L(r) = \alpha_L R_L^1(r) + \beta_L R_L^2(r).$$

(a) Plane waves,

$$\begin{aligned} \int \Phi^* \Phi &= \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^* c_{\mathbf{K}'} \int_{\text{sphere}} e^{i(\mathbf{K}-\mathbf{K}') \cdot \mathbf{r}} d\mathbf{r} \\ &= \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^* F_{\mathbf{K}-\mathbf{K}'} c_{\mathbf{K}'} , \end{aligned}$$

$$\int \nabla \Phi^* \cdot \nabla \Phi = \sum_{\mathbf{K}, \mathbf{K}'} c_{\mathbf{K}}^* \mathbf{K} F_{\mathbf{K}-\mathbf{K}'} \mathbf{K}' c_{\mathbf{K}'} ,$$

where $F_{\mathbf{K}-\mathbf{K}'}$ is given by (8).

(b) Atomiclike functions,

$$\int \Phi^* \Phi = \sum_{L, L'} \int_{\text{sphere}} R_L R_{L'} Y_L^* Y_{L'} d\mathbf{r} = \sum_L \int (R_L)^2 r^2 dr ,$$

$$\begin{aligned} \int \nabla \Phi^* \cdot \nabla \Phi &= \int \Phi^* \nabla \Phi \cdot d\mathbf{a} - \int \Phi^* \nabla^2 \Phi dr \\ &= \sum_L r_c^2 R_L(r_c) \frac{\partial R_L(r_c)}{\partial r} + l(l+1) \int (R_L)^2 dr - \int R_L \frac{\partial}{\partial r} \left[r^2 \frac{\partial}{\partial r} \right] R_L dr \\ &= \sum_L l(l+1) \int (R_L)^2 dr + \int \left[\frac{\partial R_L}{\partial r} \right]^2 r^2 dr^2 , \end{aligned}$$

where the identity

$$-\nabla^2 = \frac{\Lambda^2}{r^2} - \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \frac{\partial}{\partial r} \right]$$

was used. Λ is the angular momentum operator.

APPENDIX B: COMPARISON OF THE NUMBER OF OPERATIONS IN THE CONVENTIONAL IMPLEMENTATION AND THE OPERATOR IMPLEMENTATION OF THE LAPW METHOD

1. Conventional LAPW

The Hamiltonian matrix is first explicitly set up according to (11). This takes order

$$N_{\text{PW}}^2 N_{\text{at}} (l_{\text{max}} + 1)^2 = N_{\text{at}}^3 n_{\text{PW}}^2 (l_{\text{max}} + 1)^2$$

operations. The operation count for the overlap matrix is similar. If we assume that iterative diagonalization is used, the calculation of the N_{el} eigenstates will take

$$N_{\text{el}} N_{\text{PW}}^2 n_{\text{it}} = N_{\text{at}}^3 n_{\text{el}} n_{\text{PW}}^2 n_{\text{it}}$$

operations (a single matrix multiplication takes n_{PW}^2 operations), where n_{it} is typically about five. The calculation of the coefficients $\alpha_L(\mathbf{R}_\mu)$ and $\beta_L(\mathbf{R}_\mu)$ according to (4) and (5) for all electron orbitals takes order

$$N_{\text{el}} N_{\text{at}} N_{\text{PW}} (l_{\text{max}} + 1)^2 = N_{\text{at}}^3 n_{\text{el}} n_{\text{PW}} (l_{\text{max}} + 1)^2$$

operations.

2. Operator method

The matrix-times-vector multiplications are performed without calculating the matrices explicitly. Hamiltonian and overlap matrices consist of sums of the following three types of matrices.

(1) Diagonal matrices: $H_{\mathbf{K}, \mathbf{K}'} = \delta_{\mathbf{K}, \mathbf{K}'} f(\mathbf{K})$. Matrix-times-vector products can be calculated with $O(N_{\text{PW}})$ operations.

(2) Convolution matrices: $H_{\mathbf{K}, \mathbf{K}'} = f(\mathbf{K} - \mathbf{K}')$. Using two fast Fourier transformations,^{5,6} matrix-times-vector products can be calculated with $O(N_{\text{PW}} \ln(N_{\text{PW}}))$ operations. If N_{PW} is large (Appendix C) this is considerably faster than conventional matrix multiplication.

(3) Separable matrices: $H_{\mathbf{K}, \mathbf{K}'} = f(\mathbf{k})g(\mathbf{K}')$. Matrix-times-vector products can be calculated with $O(N_{\text{PW}})$ operations.

(4) Mixed type matrices:

$$H_{\mathbf{K}, \mathbf{K}'} = f(\mathbf{K})g(\mathbf{K} - \mathbf{K}')h(\mathbf{K}') .$$

Matrix-times vector products can be calculated with

$$O(N_{\text{PW}} + N_{\text{PW}} \ln(N_{\text{PW}}))$$

operations.

For one of the separable terms in (11) we will now discuss in more detail how the separable terms are handled. Let us consider the matrix multiplication

$$\sum_{\mathbf{K}'} \left[\sum_{L, L'} p_L^1(\mathbf{K}) H_{L, L'}^{11} p_{L'}^{1*}(\mathbf{K}') \right] c_{\mathbf{K}'} .$$

Changing the order of summation we calculate first

$$\alpha_{L'} = \sum_{\mathbf{K}'} p_{L'}^{1*}(\mathbf{K}') c_{\mathbf{K}'} .$$

for all L which takes $(l_{\text{max}} + 1)^2 N_{\text{PW}}$ operations. Then we calculate for all L

$$A_L = \sum_{L'} H_{L, L'}^{11} \alpha_{L'} .$$

The operation count of $(l_{\text{max}} + 1)^4$ for this is negligible since it does not involve any extensive variables. Finally

we calculate for all \mathbf{K}

$$\sum_L p_L^1(\mathbf{K}) A_L,$$

which again takes $(l_{\max} + 1)^2 N_{\text{PW}}$ operations. Taking into account the sum over all atoms in (11) the separable part takes $N_{\text{at}}(l_{\max} + 1)^2 N_{\text{PW}}$ operations. Neglecting the contribution from the diagonal part, the diagonalization takes, therefore, order

$$N_{\text{el}} n_{\text{it}} [N_{\text{PW}} \ln(N_{\text{PW}}) + N_{\text{at}} (l_{\max} + 1)^2 N_{\text{PW}}] \\ = N_{\text{at}}^2 n_{\text{el}} n_{\text{it}} [n_{\text{PW}} \ln(N_{\text{PW}}) + N_{\text{at}} (l_{\max} + 1)^2 n_{\text{PW}}]$$

operations. Note that the coefficients $\alpha_L(\mathbf{R}_\mu)$ (4) and $\beta_L(\mathbf{R}_\mu)$ (5) are obtained as a by-product of the matrix multiplication of the separable terms.

APPENDIX C: MATRIX-TIMES-VECTOR MULTIPLICATIONS FOR CONVOLUTION MATRICES

Conventional matrix-times-vector multiplication costs N_{PW}^2 operations for any matrix. If the matrix is a convolution matrix the multiplication can be done with fast Fourier techniques (FFT). One must perform two three-dimensional FFT's in a dealiased²⁰ form. Neglecting linear operations, the operation count for the method using FFT's goes as follows: Each three-dimensional FFT

applied to a cube containing $n^3 = N_{\text{PW}}$ \mathbf{K} points requires first n^2 one-dimensional FFT's along the x direction, then $2n^2$ one-dimensional FFT's along the y direction, and finally $4n^2$ one-dimensional FFT's along the z direction. Each FFT is of length $2n$ and requires $2(2n)\log_2(2n)$ operations if n is a multiple of 2. If this is not the case the first factor of 2 must be replaced by another factor of similar magnitude. Altogether we arrive, therefore, at

$$2[14n^3(2n)\log_2(2n)] = 56/3 N_{\text{PW}} \log_2(8N_{\text{PW}})$$

operations. Furthermore, one must take into account that with the Fourier technique one must take all the \mathbf{K} points in a cube, whereas with conventional multiplication one can take only the \mathbf{K} points in a sphere whose diameter is equal to the length of the edge of the cube. Since the ratio of the volumes of the cube and the sphere is roughly two, one must add another factor of 2 in the operation count for the Fourier technique. The fast Fourier method is, therefore, faster if

$$36N_{\text{PW}} \log_2(8N_{\text{PW}}) < N_{\text{PW}}^2.$$

The crossover point for the method is, therefore, at around $N_{\text{PW}} = 420$. The actual timing depends, of course, not only on the number of operations, but also on the execution speed. Highly vectorized programs for multiple FFT's are available on most vector machines.

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