Accelerating the convergence of self-consistent linearized augmented-plane-wave calculations

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The applicability of Broyden's second method for accelerating the convergence of self-consistent electronic-structure calculations based on the linearized augmented-plane-wave method is discussed in terms of a W(001) surface calculation. It is found that its use results in a significant improvement in the convergence of the calculation, and based on this it is concluded that its use should increase the size of the systems for which such calculations are feasible.

INTRODUCTION

In electronic-structure calculations one seeks a selfconsistent solution of the Kohn-Sham equations. This is generally done iteratively as follows. Given an input charge density ρ_{in} , the corresponding potential is constructed and the resulting Kohn-Sham equations for the density-functional wave functions are solved. These yield an output density ρ_{out} , which in general differs from the input. Using the input and output densities of this and the previous iterations one then constructs an input for the next iteration ρ_{in} , the goal being to find a density which when input to the self-consistent loop yields an output equal to it within a specified tolerance.

One of the most efficient (in terms of the number of iterations required) methods for constructing the new input density is Broyden's modified Newton-Raphson method.¹ This technique and modified versions of it have been used in electronic-structure calculations on a variety of systems almost invariably with a significant reduction in the number of iterations needed to achieve the desired self-consistency.²⁻⁴ As far as we know, however, no attempt has been made until now to use Broyden's method to accelerate the convergence of self-consistent linearized augmented-plane-wave (LAPW) calculations. This is perhaps a result of the representation of the charge density in the general-potential LAPW method.⁵⁻⁷ This representation divides space into an interstitial region I, and regions inside atom-centered spheres S_n . In the interstitial region the charge density is expanded in symmetrized plane waves while in the spheres it is expanded on a discrete radial mesh in terms of symmetrized spherical harmonics, i.e., lattice harmonics. Thus

$$\rho(\mathbf{r}) = \begin{cases} \sum_{s} \rho_{s} \phi_{s}(\mathbf{r}), & \mathbf{r} \in I \\ \\ \sum_{j} \rho_{nj}(r_{n}) K_{nj}(\hat{\mathbf{r}}_{n}), & \mathbf{r} \in S_{n} \end{cases}$$
(1)

where the ϕ_s are symmetrized plane waves, the K_{nj} are lattice harmonics on site *n* and \mathbf{r}_n is with respect to the position of atom *n*. This representation introduces two

complications. The first is that it gives the charge density in terms of a large number N of parameters, ρ_i , $\rho = \{\rho_s, \rho_{nj}(r_n)\}$, often tens of thousands. The second is that the nature of the dependence of the charge density on some of the parameters (those describing the interstitial volume) is radically different from its dependence on the others (the numerical meshes). The first complication makes any scheme requiring the storage, inversion, or multiplication of $N \times N$ matrices undesirable, while the second makes it questionable whether a scheme such as Broyden's method which treats all the parameters on the same footing will be effective. Thus self-consistent LAPW calculations have generally been converged by taking linear combinations of the last iterations input and output densities, i.e.,

$$\boldsymbol{\rho}_{\rm in}^{(m+1)} = (1-\alpha)\boldsymbol{\rho}_{\rm in}^{(m)} + \alpha \boldsymbol{\rho}_{\rm out}^{(m)} , \qquad (2)$$

where α is a mixing parameter. This scheme will be referred to as straight mixing. For surfaces and other systems with large unit cells it is often necessary to use small values of α for the procedure to be stable, $\alpha \leq 0.1$ being common. Schemes using linear combinations of the input and output densities of the last two iterations are sometimes used to improve on this.⁸ However, often unacceptably slow rates of convergence are still obtained, especially for large unit cells with low symmetry, as in surface calculations.

Recently, Srivastava⁹ developed a scheme for implementing Broyden's second method which avoids the storage of $N \times N$ matrices as well as requiring no $N \times N$ matrix inversions or multiplications. In view of the fact that in many cases the size of systems on which electronic-structure calculations can be performed is limited by the computing time required, it was of interest to study the applicability of this approach in LAPW calculations. Here we report its first use in this context and discuss its efficiency in terms of a test calculation on a moderately large (N = 4744) system described below. It is concluded that the method is very effective in accelerating the convergence of these calculations and that its use may result in substantial reductions in the number of iterations required to treat other systems.

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BROYDEN'S SECOND METHOD

Since the implementation of Broyden's second method is discussed in detail in Ref. 9, we only summarize it here. As discussed above the goal is to construct an input charge density $\rho^{(m+1)}$ such that $\mathbf{F}^{(m+1)}$ is zero, where

$$\mathbf{F}^{(m)} = \boldsymbol{\rho}_{\text{out}}^{(m)} - \boldsymbol{\rho}_{\text{in}}^{(m)} . \tag{3}$$

The strategy used is to set

$$\boldsymbol{\rho}_{\rm in}^{(m+1)} = \boldsymbol{\rho}_{\rm in}^{(m)} - \underline{\boldsymbol{G}}^{(m)} \mathbf{F}^{(m)} , \qquad (4)$$

where $\underline{G}^{(m)}$ is an approximation to the inverse Jacobian matrix which the method attempts to improve after each iteration using the information contained in ρ_{in} and F. According to Broyden's second method the updating procedure for \underline{G} is

$$\underline{G}^{(m)} = \underline{G}^{(m-1)} + \frac{[\rho_{in}^{(m)} - \rho_{in}^{(m-1)} - \underline{G}^{(m-1)}(\mathbf{F}^{(m)} - \mathbf{F}^{(m-1)})](\mathbf{F}^{(m)} - \mathbf{F}^{(m-1)})^{T}}{(\mathbf{F}^{(m)} - \mathbf{F}^{(m-1)})^{T}(\mathbf{F}^{(m)} - \mathbf{F}^{(m-1)})}$$
(5)

Thus

$$\boldsymbol{\rho}_{in}^{(m+1)} = \boldsymbol{\rho}_{in}^{(m)} - \underline{\boldsymbol{G}}^{(1)} \mathbf{F}^{(m)} - \sum_{j=2}^{m} \mathbf{U}^{(j)} \mathbf{V}^{T(j)} \mathbf{F}^{(m)}$$
(6)

with

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$$\mathbf{U}^{(i)} = -\underline{G}^{(1)}(\mathbf{F}^{(i)} - \mathbf{F}^{(i-1)}) + \boldsymbol{\rho}_{in}^{(i)} - \boldsymbol{\rho}_{in}^{(i-1)} - \sum_{j=2}^{i-1} \mathbf{V}^{T(j)}(\mathbf{F}^{(i)} - \mathbf{F}^{(i-1)}) \mathbf{U}^{(j)} , \qquad (7)$$

$$\mathbf{V}^{T(i)} = \frac{(\mathbf{F}^{(i)} - \mathbf{F}^{(i-1)})^T}{(\mathbf{F}^{(i)} - \mathbf{F}^{(i-1)})^T (\mathbf{F}^{(i)} - \mathbf{F}^{(i-1)})} .$$
(8)

Thus the scheme as implemented in Ref. 9 only requires the storage of a relatively small number of vectors containing the information in the input and output charge densities of the previous iterations.

In the calculations presented here the initial guess $\underline{G}^{(1)}$ for the inverse Jacobian was taken to be a constant diagonal matrix so that the input to the second iteration was effectively constructed using straight mixing. Other initial guesses could be envisioned, but constructing them for the LAPW representation is not straightforward since one must ensure that the resulting scheme conserves the total electronic charge. This is guaranteed only if the initial guess is charge conserving.

THE TEST SYSTEM

Here the efficiency of Broyden's method is examined using a tungsten (001) surface calculation. The model used for this surface consists of a periodic array of fivelayer W(001) slabs at the bulk lattice spacing separated by five empty layers. The resulting unit cell is 10 times as large as a bulk unit cell. The calculations were performed using about 410 basis functions, which is somewhat underconverged for this system. This underconvergence is not expected to have any material effect on the relative performance of the mixing schemes. The k-space sampling was done using a set of three special points¹⁰ in the irreducible wedge of the two-dimensional Brillouin zone. Exchange and correlation were treated using the Wigner formula.11

In order to study the convergence of the calculation three quantities are monitored. These are the total energy, the average potential in the center of the vacuum layer (with respect to the Fermi energy), which is sensitive to the surface dipole, and the distance between the input and output densities, d, defined by

$$d = \left[\int_{\Omega} d^3 r (\rho_{\rm in} - \rho_{\rm out})^2 / \Omega\right]^{1/2}, \qquad (9)$$

where Ω is the volume of the unit cell.

In Figs. 1 and 2 the distance in the charge density, d, and the error in the total energy (with respect to the converged energy), respectively, are shown for each iteration as obtained using Broyden's method started with 10% straight mixing and using straight mixing. The initial charge density was constructed from overlapping atomic charge densities. In spite of the fact that the mixing parameter, $\alpha = 0.1$, is close to optimum for this system, it is apparent that Broyden's method is significantly more effective than straight mixing in converging the calculation.

The performance of the straight-mixing scheme is limited by instabilities which appear as the mixing parameter is increased causing the iterative process to diverge. In



FIG. 1. The distance $[in 10^{-3} \text{ electrons/a.u.}^3, \text{ cf. Eq. (9)}]$ between the input and output charge densities as a function of iteration number.



FIG. 2. The error in the total energy with respect to the final converged energy.

surface calculations one such instability involves an oscillation of charge between the surface layers and the interiors of the slabs. In Fig. 3 the vacuum potential, which is sensitive to such charge displacements, is shown at each iteration. By inspecting Fig. 3 it may be noted that the oscillations present in the vacuum potential using straight mixing with $\alpha = 0.1$ are suppressed when Broyden's method is used. This is a manifestation of the adaptiveness of the technique which allows it to rapidly converge the calculation while avoiding instabilities.

Since Broyden's method is a linear method one might expect its performance to suffer if it is started when the charge density is very far from the converged charge density. In some surface calculations faster convergence is obtained at the end of the calculation if the Broyden's matrix is restarted after the first few iterations, thereby discarding the information contained in the input and output charge densities of these iterations. We have also observed this in some calculations on bulk f-band materials.



FIG. 3. The planar-averaged potential at the center of the vacuum relative to the Fermi energy.

CONCLUSION

Based on the rapid convergence obtained using Broyden's method in general-potential LAPW calculations, we believe the method to be very effective in accelerating the convergence of such calculations in general. It is expected that the use of the method will lead to significant reductions in the computing requirements for selfconsistent electronic-structure calculations and should increase the size of the systems which can be treated.

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