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# Determination of the Mott insulating transition by the multi-reference density functional theory

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## Abstract

It is shown that a momentum boost technique applied to the extended Kohn–Sham scheme enables the computational determination of the Mott insulating transition. Self-consistent solutions are given for correlated electron systems by first-principles calculations defined by the multi-reference density functional theory, in which the effective short-range interaction can be determined by the fluctuation reference method. An extension of the Harriman construction is made for the twisted boundary condition in order to define the momentum-boost technique in the first-principles manner. For an effectively half-filled-band system, the momentum-boost method tells that the period of a metallic ground state by the local density approximation (LDA) calculation is shortened to the least period of the insulating phase, indicating the occurrence of the Mott insulating transition.

## 1. Introduction

Detection of the Mott insulating transition is a desirable function for first-principles calculations, which has been demanded for years. A recently developed multi-reference density functional theory with the fluctuation reference method defines a self-consistent first-principles calculation, in which a short-range correlation effect is explicitly included [1–3]. This technique is a generalization of the Kohn–Sham scheme of the electronic structure calculation [4, 5]. Incorporation of the effective many-body system to determine the total energy and the single-particle charge density of the electronic state became possible through (1) the introduction of a fluctuation-counting term and (2) the reformulation of the exchange correlation energy functional as a residual exchange correlation energy functional. The effective Hamiltonian appearing in the theory is a kind of Hubbard model [6] or Anderson model [7]. The LDA +  $U$  Hamiltonian [8] is derived as an approximation for the residual exchange correlation functional [9].

In this short paper, I show a technique to determine the Mott insulating phase in this first-principles calculation. The method is an application of the momentum boost technique known in the literature [10, 11]. To introduce the momentum boost method in the density functional

theory, one needs to show the  $N$  representability. This is done by extending the Harriman construction [12, 13] for the twisted boundary condition. A test calculation will be shown using a simple artificial system, which may be represented by a one-dimensional Hubbard model. In the last part, I will summarize the present work.

## 2. Momentum boost technique

I consider a Born–von–Karman boundary condition with a twist. Consider an array of atoms in one dimension and let  $L$  be the number of atoms in the direction, which is called the  $x$  direction below. Formally using a phase  $2\pi\Phi/\Phi_0$  with  $0 \leq \Phi < \Phi_0$  and a unit flux  $\Phi_0$ , I introduce a twisted boundary condition in this  $x$  direction:

$$\Psi(x_1, \dots, (\mathbf{r}_j + La\mathbf{e}_x, \sigma_j), \dots, x_N) = \exp(2\pi i\Phi/\Phi_0)\Psi(x_1, \dots, (\mathbf{r}_j, \sigma_j), \dots, x_N), \quad (1)$$

$$c_{N+1,\sigma}^\dagger = \exp(2\pi i\Phi/\Phi_0) c_{1,\sigma}^\dagger. \quad (2)$$

Here, I used a combined coordinate  $x_j = (\mathbf{r}_j, \sigma_j)$  with the space and spin coordinates.  $\mathbf{e}_x$  is a unit vector and  $a$  is the lattice constant in the  $x$  direction.  $\Psi$  is a many-body wavefunction and  $c_{i,\sigma}^\dagger$  is a creation operator defined with a properly determined Wannier basis  $\phi_i(\mathbf{r})$ . Indeed, determining a Wannier transformation by fixing a gauge in the unitary transformation [14], the set of  $\phi_i(\mathbf{r})$  is uniquely determined in each self-consistent step of the extended Kohn–Sham scheme [2].

Using another gauge transformation, the twisted boundary condition is represented by shifted  $k$  vectors,  $\mathbf{k} = (\frac{2\pi m}{La} + \frac{2\pi\Phi}{La\Phi_0}, k_y, k_z)$ , for the single-particle Bloch orbitals. Here  $m$  is an integer within a range of  $0 \leq m < L$ . From the single-particle energy  $\varepsilon_n(\mathbf{k})$ , one may construct a tight-binding model written in  $c_{i,\sigma}^\dagger$  and  $c_{i,\sigma}$ .

Determining the eigen energy of the effective fermion system, one has an energy-flow diagram as a function of  $\Phi$ . If the system is a metal, the flow given by adiabatic connection of the ground state should show a long extended AB period, while the period has to be  $\Phi_0$ , when there is a charge gap due to formation of a Mott gap. Although the density functional theory utilized in the present work is the ground-state formulation, one can have a signal of the change in the period as the disappearance of a cusp in the lowest-energy flow. In the case of the Mott insulator, the flux line becomes a smooth curve without any cusp. The appearance of the isolated lowest flow line implies uniqueness of the ground state separated by a gap from charge excitations in the whole range of twist and thus indicates the formation of the Mott gap. If one has difficulty in finding the change in the period with a three-dimensional sample, one may consider a one-dimensional setup using a needle-like sample.

Before discussing the result, I show that the  $N$  representability of the present problem is guaranteed. In the present setup, only the boundary condition in the  $x$  direction is a twisted one. Thus I consider a slice of the charge density  $\tilde{\rho}(x) = \rho(\mathbf{r})$  fixing  $y$  and  $z$  coordinates for simplicity. We are seeking  $\varphi_k(x)$  satisfying

$$\tilde{\rho}(x) = \sum_k \lambda_k |\varphi_k(x)|^2, \quad (3)$$

where  $k$  is an integer,  $0 \leq \lambda_k \leq 1$ , and  $\sum_k \lambda_k = N$ . In the twisted boundary condition, however,  $\tilde{\rho}(x + La)$  obeys the periodic boundary condition as

$$\begin{aligned} \tilde{\rho}(x + La) &= \int d\sigma_1 dx_2 \cdots dx_N \Psi((\mathbf{r}_1 + La\mathbf{e}_x, \sigma_1), x_2, \dots, x_N)^* \\ &\quad \times \Psi((\mathbf{r}_1 + La\mathbf{e}_x, \sigma_1), x_2, \dots, x_N) \end{aligned}$$

$$\begin{aligned}
&= \int d\sigma_1 dx_2 \cdots dx_N \Psi((\mathbf{r}_1, \sigma_1), x_2, \dots, x_N)^* \Psi((\mathbf{r}_1, \sigma_1), x_2, \dots, x_N) \\
&= \tilde{\rho}(x).
\end{aligned} \tag{4}$$

Here, integration with respect to  $\sigma_j$  should be interpreted as a summation. Thus, we can readily prepare the orbital  $\varphi_k(x)$  obeying the twisted boundary condition as,

$$\varphi_k(x) = \frac{\tilde{\rho}(x)^{1/2}}{N} \exp(ikf(x) + i2\pi\Phi/\Phi_0), \tag{5}$$

with

$$f(x) = \frac{2\pi}{N} \int_0^{La} \tilde{\rho}(x') dx'. \tag{6}$$

We can immediately show that a set of  $\varphi_k(x)$  forms a complete orthonormal and that

$$\sum_k \lambda_k |\varphi(x)|^2 = \frac{1}{N} \tilde{\rho}(x) \sum_k \lambda_k = \tilde{\rho}(x). \tag{7}$$

Using the orbital wavefunction  $\varphi_k(x)$  obeying the twisted boundary condition, we can show the existence of a many-body state  $|\Psi\rangle$  whose coordinate expression is the single Slater determinant made of  $\varphi_k(x)$ .

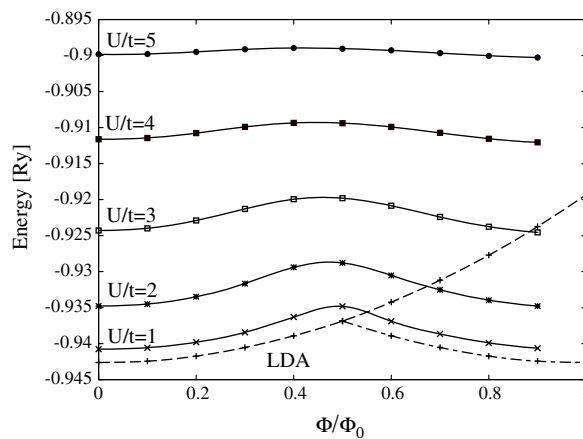
### 3. One-dimensional hydrogen array

As a test calculation, I consider a one-dimensional hydrogen array. The system is denoted by an outer unit cell with ten atoms for the many-body calculation and an inner unit cell with a single atom for the single-particle problem. The lattice parameters for the inner cell are  $a = 2 \text{ \AA}$  and  $b = c = 10 \text{ \AA}$ . The determination of  $U$  may be achieved by setting a reference calculation [2]. Here, to show the change in the period explicitly, I consider  $U/t$  as a parameter and perform only the self-consistent calculation for the extended Kohn–Sham system. Here,  $t$  is the value of the nearest-neighbour transfer parameter. Note that the present system is represented by a tight-binding model with long-range hopping terms. For the exchange–correlation energy functional, I utilized the Perdew–Zunger parameterization of the Ceperley–Alder diffusion Monte Carlo data [15]. The Troullier–Martins soft pseudopotential is used with a cutoff energy of 20 (Ryd) [16]. This setup is confirmed to be accurate enough for the discussion below by increasing the parameters. The numerical diagonalization with the Lanczos algorithm is used to obtain the many-body state of the first-principles Hubbard model.

The result of the momentum boost is depicted in figure 1. For  $U = 0$ , the LDA calculation shows a crossing in energy flow lines. In this case, the lowest energy flow can be traced, since the constrained LDA calculation fixing the filling of each  $k$  point is available. Once a finite  $U$  is introduced, the many-body calculation automatically concludes the lowest branch of the energy flows. In this system, we see a continuous change in the energy flow which has an energy gap structure at  $\Phi = \Phi_0/2$ . This result is qualitatively the same as the single-band Hubbard model with only the nearest-neighbour hopping [11]. If the fluctuation reference method is applied precisely, a finite value of  $U$  is expected, since the inter-atomic distance of  $2 \text{ \AA}$  is in a strong-correlation regime for the hydrogen molecule [2]. Thus the present result gives a concrete test for the method of determination of the Mott insulator from first principles.

### 4. Summary

I have shown that the momentum boost method is formulated rigorously in density functional theory. The  $N$  representability is shown for the twisted boundary condition. Using the multi-reference density functional theory (MR-DFT), we are able to construct an effective interacting



**Figure 1.** The ground-state energy of a hydrogen array system as a function of the phase  $\Phi$  in the boundary condition. The system has ten atoms. The value of  $U/t$  is set to be 0, 1, 2, ..., 5 with  $t$  being the nearest-neighbour transfer parameter. For the case of  $U = 0$  (LDA calculation), crossing of the flow lines occurs at  $\Phi = \Phi_0/2$ , while the crossing becomes anti-crossing due to the charge-gap formation for finite  $U$ .

fermion system, which may undergo the Mott insulating transition. The momentum boost technique is applicable for this problem to detect the transition. In a realistic system, the formation of the Mott gap by applied pressure or by effective carrier doping could be seen as a change in the period of the lowest energy flow.

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