Localisation Lengths in Density Functional Theory and Quantum Monte Carlo

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Introduction

The question of whether a wavefunction represents a metal or an insulator is easily answered within a one-electron, independent particles context, but in a many-body system it is less clear. The development of the Berry-Phase theory of polarisation [1] and localisation [2] has, however, allowed calculation of these quantities central to metallic and insulating behaviour, and recent work [3] has extended these to many-body systems. We investigate here the behaviour of the localisation length, a quantity representing the quadratic spread of an extended wavefunction, expected to be finite in an insulator and infinite in a metal. We use Density Functional Theory and Quantum Monte Carlo to study its behaviour in model systems comprising periodic arrays of quantum dots. We describe the circumstances under which a metal insulator transition is accompanied by a divergence in the localisation length, and we present a derivation of the many-body expressions for polarisation and localisation which stresses the connection to many-body Wannier functions.

Many Body Wannier Functions

In one-electron theory, polarisation and localisation are associated with the position and spread of the maximally localised Wannier functions. The same approach can be applied to many-body Bloch functions $\Psi_{\mathbf{k}}$ which result from translational symmetry of the whole Hamiltonian by single electron coordinates, to form many-body Wannier functions $W_{\mathbf{M}}$. In this localised many-body basis, the position operator is well-defined and its first and second moments can be calculated in terms of the variation of the many-body wavefunction with the twist vector $\mathbf{k}$ over the cell:

$$|W_{\mathbf{M}}\rangle = \frac{1}{\sqrt{V_k}} \int \mathrm{d}\mathbf{k}\, e^{-i\mathbf{k}.\mathbf{R}_{\mathrm{M}}} |\Psi_{\mathbf{k}}\rangle \tag{1}$$

so the expectation value of the centre of mass operator $\hat{\mathbf{X}} = \sum_j \mathbf{r}_j$ is

$$\langle W_{\mathbf{0}}|\hat{\mathbf{X}}|W_{\mathbf{0}}\rangle = \frac{i}{V_k} \int \mathrm{d}\mathbf{k} \left\langle \Phi_{\mathbf{k}} \middle| \frac{\partial \Phi_{\mathbf{k}}}{\partial \mathbf{k}} \right\rangle \tag{2}$$

where $\Phi_{\mathbf{k}}$ is the periodic part of $\Psi_{\mathbf{k}}$. We define the spread functional Ω as the variance of this operator:

$$\Omega = \langle W_{\mathbf{0}}|\, \hat{\mathbf{X}}^2\, |W_{\mathbf{0}}\rangle - \langle W_{\mathbf{0}}|\, \hat{\mathbf{X}}\, |W_{\mathbf{0}}\rangle^2 \tag{3}$$

which after some manipulations is split into gauge-invariant and gauge dependent parts Ω_I and $\widetilde{\Omega}$, where

$$\begin{aligned} \Omega_I &= -\frac{1}{V_k} \int \mathrm{d}\mathbf{k} \left(\left\langle \Phi_{\mathbf{k}} \middle| \frac{\partial^2 \Phi_{\mathbf{k}}}{\partial \mathbf{k}^2} \right\rangle - \left| \left\langle \Phi_{\mathbf{k}} \middle| \frac{\partial \Phi_{\mathbf{k}}}{\partial \mathbf{k}} \right\rangle \right|^2 \right) \\ \text{and} \qquad \widetilde{\Omega} &= \sum_{\mathbf{M}\neq\mathbf{0}} \left| \left\langle W_{\mathbf{M}} | \hat{\mathbf{X}} | W_{\mathbf{0}} \right\rangle \right|^2 \end{aligned} \tag{4}$$

To put this in a form in which the quantities can be evaluated within existing methods, an ansatz wavefunction is constructed for a larger cell, assuming short range correlations, to produce a 'single-point' formula for the spread per electron in the N electron system of side L_i in direction i, relying only on the many-body wavefunction Ψ at $\mathbf{k} = 0$:

$$\langle r_i^2 \rangle_c = -\frac{1}{N} \left(\frac{L_i}{2\pi} \right)^2 \ln \left| \left\langle \Psi \middle| e^{-i\mathbf{G}_i.\hat{\mathbf{X}}} \middle| \Psi \right\rangle \right|^2 \tag{5}$$

Evaluation in DFT and QMC

Density Functional Theory does not provide a true many-body ground state wavefunction, but localisation lengths can be calculated by forming an ansatz Slater determinant Ψ from the occupied orbitals at $\mathbf{k}$ points on a grid with spacings $\delta\mathbf{k}_i$, and evaluating the above expression for this wavefunction from the modulus squared of the complex number z_N . The determinant formed from the overlaps is very sparse and can be evaluated in terms of overlaps of individual Bloch functions:

$$z_N(\delta\mathbf{k}_i) = \left\langle \Psi \middle| e^{-i\delta\mathbf{k}_i.\hat{\mathbf{X}}} \middle| \Psi \right\rangle = \prod_{\mathbf{k}} \det S(\mathbf{k}) \tag{6}$$

where $S(\mathbf{k})$ is a matrix of overlaps between the periodic parts $u_{n\mathbf{k}}$ of the Bloch functions of the occupied bands n, whose terms are given by

$$S_{nn'}(\mathbf{k}) = \begin{cases} \langle u_{n\mathbf{k}}|\, u_{n'\mathbf{k}+\delta\mathbf{k}}\rangle \\ \langle u_{n\mathbf{k}}|\, e^{-i\mathbf{G}'.\mathbf{r}}\, |\, u_{n'\mathbf{k}+\delta\mathbf{k}-\mathbf{G}'}\rangle \end{cases} \tag{7}$$

where the latter expression is used if $\mathbf{k}+\delta\mathbf{k}$ is outside the Brillouin zone for $\mathbf{G}'$ such that $\mathbf{k}+\delta\mathbf{k}-\mathbf{G}'$ is inside. In a plane wave basis with coefficients $c_{n\mathbf{k}}$, z_N can be evaluated easily from the plane wave coefficients:

$$S_{nn'} = \sum_{\mathbf{G}} c^*_{n\mathbf{k}}(\mathbf{G})\, c_{n'(\mathbf{k}+\delta\mathbf{k}-\mathbf{G}')}(\mathbf{G}-\mathbf{G}') \tag{8}$$

where $\mathbf{G}' = 0$ if $\mathbf{k}+\delta\mathbf{k}$ is in the first B.Z.

In Quantum Monte Carlo [4], the complex number $z_N(\mathbf{G}_i)$ is evaluated for each of the electron configurations $\{\mathbf{r}_j\}$ generated during a VMC run, by evaluating the local operators $\exp -i\mathbf{G}_i.\hat{\mathbf{X}}$ at each timestep and averaging. This method runs into difficulties in large systems when the magnitude of the mean of $|z_N|$ becomes very small, as it is being averaged from numbers of unit magnitude and hence fixed variance. In this situation, using a run length M such that $\sqrt{M} \gg 2/|z_N|$ ensures $\Delta\langle x^2\rangle_c \ll \langle x^2\rangle_c$.

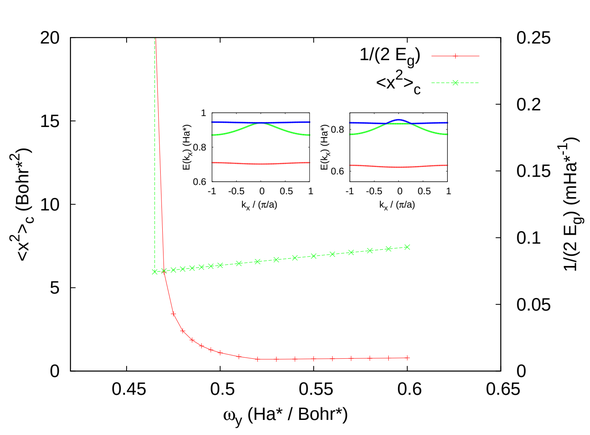
Localisation Lengths over Metal-Insulator Transitions

Previous work [2] has shown that the localisation length is a good indicator of metallic or insulating behaviour over a metal-insulator transition: a divergence to infinity was observed where the system becomes metallic. Additionally, localisation of Wannier functions is known to be closely related to the system's energy gap, so one might expect that, approaching a metal-insulator transition, as the gap tends to zero, the localisation length will diverge to infinity. We show by a number of examples with different bandstructures that this is only the case in particular circumstances and in general depends on the nature of the states which meet at the transition. We illustrate how different properties of the bandstructure in DFT affect localisation, and in QMC we show the effect of including correlation via a Jastrow factor.

Quasi-1D Chains

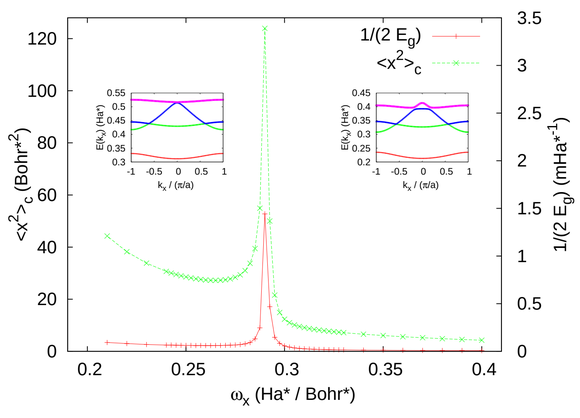
We simulate a quasi-1D chain of 2D dots along x using a strong barrier potential and a large unit cell in the y-direction. Including only k_y=0 but a range of k_x's in the first Brillouin zone produces a simplified bandstructure in which the effect of the symmetry properties of the Bloch functions can be distinguished. The two situations, shown below as (a) and (b), differ by the spatial symmetries of the states above and below the direct gap. In both cases the strength of the confining potential ω is tuned down such that a band-insulator to metal transition occurs at a particular strength ω_M. In (a) the bands which meet have different symmetries under reflection in the x-axis so there is no Hamiltonian matrix element between them for any k_x, whereas in (b) they are of the same symmetry so there is a matrix element for all k_x other than $k_x = 0$. In both cases the direct gap closes to zero, but only in (b) do we see a divergence in $\langle x^2\rangle_c$ around ω_M. This emphasises the importance of a nonzero matrix element between the states either side of the gap if a divergence is to be observed.

(a)



1D Chain of 4-electron dots: Localisation spread $\langle x^2\rangle_c$ and inverse direct energy gap $1/(2E_g)$ as ω is reduced. Inset: Energy bands $E(k_x)$ just before (left) and shortly after ω_M (right). System remains metallic after transition as the bands pass through each other.

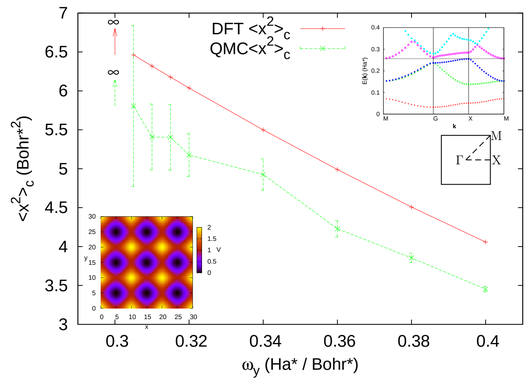
(b)



1D Chain of 6-electron dots: Localisation spread $\langle x^2\rangle_c$ and inverse direct energy gap $1/(2E_g)$ as ω is reduced. Insulating either side of ω_M but metallic at ω_M. Inset: as above but bands repel after crossing and system becomes insulating again.

2D Arrays

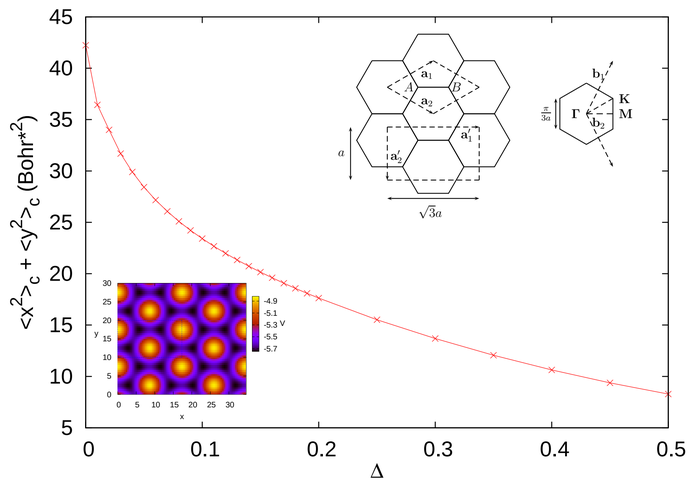
In this arrangement we restore 2D symmetry in the potentials, with a square lattice with each unit cell containing a single dot. Now, with six electrons per dot, the s band and the two p bands are filled and there is an indirect gap to the next band up. By reducing the confining potential ω we again drive the system over a metal-insulator transition but this time the smallest direct gap remains finite as the indirect gap falls to zero and the bands overlap. Consequently, no divergence is seen in $\langle x^2\rangle_c$ which merely grows as the confinement is reduced. The addition of correlation via a Jastrow factor within QMC reduces the localisation length significantly but does not otherwise affect its behaviour in this situation.



2D Array: Localisation spread in DFT and direct and indirect energy gaps, and localisation length in QMC with Jastrow factor. Inset: Dispersion curve for 2D array, showing Fermi surface and energy gap between Γ and **X** (above right). Reciprocal cell of square lattice showing high symmetry points (middle right). External potential in a sample of the supercell (below left)

Graphene Model

The hexagonal lattice corresponding to a model of the graphene system provides a situation with a direct gap which can be tuned down to zero in a realistic system. The system contains two types of sites, A and B, with the potential on the A sites scaled by $(1+\Delta)$ relative to B.
At nonzero Δ there is an energy gap and a finite localisation length, which can be interpreted as the electrons being localised to the lower energy sites. As $\Delta \to 0$, the energy gap closes to be strictly zero, and the localisation length diverges even though the lowest bands are fully occupied.



Graphene model: Localisation spread $\langle x^2\rangle_c + \langle y^2\rangle_c$ as potential asymmetry is increased. Inset: Potential for $\Delta = 0$ (bottom). Real and reciprocal lattices of the 2D system (top). The gap closes at **K** when $\Delta = 0$.

Conclusions

In conclusion, we have presented a variation on previous presentations of localisation lengths in periodic systems, which highlights the connection to many-body Wannier functions, and examined the behaviour of the localisation length in a variety of 2D model systems.
Calculations of localisation lengths in model 2D periodic systems have shown that the effect of symmetry properties on the possibility of hybridisation between bands is crucial to whether a band crossing shows up as a diverging localisation length. This will extend to 3D, although in real materials metal-insulator transitions rarely result from closure of a direct gap, so the chance of being able to observe such behaviour is small. Many-body Wannier functions also have a range of other possible applications, due to their properties as a localised yet still correlated description of the wavefunctions of extended insulators, such as in orbital magnetisation. They are also related to the polarisation fluctuations underlying finite size errors within Quantum Monte Carlo.

References

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[3] I. Souza, T. Wilkens, and R. Martin, Phys. Rev. B 62, 1666 (2000).

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