Compressed representation of Kohn-Sham orbitals via selected columns of the density matrix

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Given a set of Kohn-Sham orbitals from an insulating system, we present a simple, robust, efficient and highly parallelizable method to construct a set of, optionally orthogonal, localized basis functions for the associated subspace. Our method explicitly uses the fact that density matrices associated with insulating systems decay exponentially along the off-diagonal direction in the real space representation. Our method avoids the usage of an optimization procedure, and the localized basis functions are constructed directly from a set of selected columns of the density matrix (SCDM). Consequently, the only adjustable parameter in our method is the truncation threshold of the localized basis functions. Our method can be used in any electronic structure software package with an arbitrary basis set. We demonstrate the numerical accuracy and parallel scalability of the SCDM procedure using orbitals generated by the Quantum ESPRESSO software package. We also demonstrate a procedure for combining SCDM with Hockney’s algorithm to efficiently perform Hartree-Fock exchange energy calculations with near linear scaling.

INTRODUCTION

Kohn-Sham density functional theory (KSDFT) is the most widely used electronic structure theory for molecules and systems in condensed phase. In KSDFT the many body electronic structure properties are, in principle, exactly mapped into a fictitious single particle system. The Kohn-Sham orbitals, which are orthonormal eigenfunctions of the single particle Kohn-Sham Hamiltonian operator, can describe various physical quantities such as density, energy, and atomic forces. However, it is expensive to compute and store the Kohn-Sham orbitals of large systems, which are spatially delocalized. Let \( N \) be the number of degrees of freedom and \( N_e \) be the number of electrons in the system. The cost for storing the Kohn-Sham orbitals is \( \mathcal{O}(N N_e) \), and the cost for computing them is generally \( \mathcal{O}(N N_e^2) \), or “cubic scaling” assuming \( N \sim \mathcal{O}(N_e) \). In modern KSDFT calculations the Hartree-Fock exact exchange term is also often taken into account in the form of hybrid functional. The computational cost for this step not only scales cubically but also has a large pre-constant, which limits the application of hybrid functional calculations to hundreds of atoms.

In order to reduce both the storage requirements and the cost associated with subsequent computations, it is important to realize that the Kohn-Sham orbitals are not unique. Any non-degenerate linear transformation of the set of Kohn-Sham orbitals yields exactly the same physical properties of a system. In other words, the physically relevant quantity is the subspace spanned by the Kohn-Sham orbitals. Various efforts have been made to utilize this fact and to find a set of localized orbitals to form compressed representation of a Kohn-Sham subspace. For example, the Marzari-Vanderbilt construction of maximally localized Wannier functions (MLWFs) uses a nonlinear optimization approach to find a unitary transformation of the Kohn-Sham subspace into a set of orthogonal functions localized in real space. The locality, or the “nearsightedness” principle, is guaranteed for insulating systems with a finite HOMO-LUMO gap. The localized orbitals have wide applications in chemistry and physics. For instance, localized orbitals can be be used to construct linear scaling methods for solving KSDFT using local and semi-local functionals, and for calculations using hybrid functionals.

In this manuscript, we present an alternative method to the widely used MLWFs to compute a set of localized orbitals associated with the Kohn-Sham subspace for insulating systems. Our method is simple, robust, efficient and highly parallelizable. Our method explicitly uses the fact that for insulating systems the single particle density matrix is exponentially localized along the off-diagonal direction in the real space representation. Our algorithm finds a set of localized orbitals by directly using selected columns of the density matrix (SCDM) associated with the Kohn-Sham orbitals. Consequently, by construction the SCDM are localized in the real space representation. As opposed to the MWLFs which are orthonormal, the SCDM are not orthonormal in general. However, a set of orthonormal and localized functions spanning the Kohn-Sham subspace can be obtained via a simple linear transformation of the SCDM.

The only adjustable parameter in our method is the truncation threshold for the localized functions, and our method can be used in any electronic structure software package with any basis set, ranging from planewaves to Gaussian basis sets, provided that the Kohn-Sham orbitals can be represented on a real space grid. One striking feature of our method is its simplicity: a prototype
sequential implementation takes just a few lines of code. The construction of the SCDM only involves simple linear algebra routines such as a rank-revealing QR factorization and matrix-matrix multiplication. Therefore, the parallel implementation for computing the SCDM can straightforwardly scale, for the problem size we tested, to more than one thousand processors. This enables the computation of localized basis functions for the self-consistent treatment of the Hartree-Fock terms in KS-DFT calculations with hybrid exchange-correlation functionals. As an application, we also demonstrate a procedure for combining SCDM with Hockney’s algorithm to efficiently compute the Hartree-Fock exchange energy.

THEORY

For insulating systems, the locality of the single particle density matrix along the off-diagonal direction can be in general observed in the real space representation. For the sake of clarity, in this manuscript we explicitly require the Kohn-Sham orbitals to be represented on a real space grid defined as below. In some cases the real space representation may not be necessary, and we postpone such a discussion to the conclusion.

Let \( \{ \psi_j(x) \} \) be a set of Kohn-Sham orbitals which satisfy the orthonormality condition

\[
\int \psi_j^*(x) \psi_{j'}(x) \, dx = \delta_{jj'},
\]

and we have access to \( \psi_j(x) \) evaluated at a set of discrete grid points \( \{ x_i \} \). Let \( \{ \omega_i \} \) be a set of positive integration weights associated with the grid points \( \{ x_i \} \), then the discrete orthonormality condition corresponding to Eq. (1) is given by

\[
\sum_{i=1}^{N} \psi_j(x_i) \psi_{j'}(x_i) \omega_i = \delta_{jj'}.
\]

Let \( \psi_j = [\psi_j(x_1), \psi_j(x_2), \ldots, \psi_j(x_N)]^T \) be a column vector, and \( \tilde{\Psi} = [\psi_1, \ldots, \psi_N] \) be a matrix of size \( N \times N_c \). In this paper we call \( \tilde{\Psi} \) the real space representation of the Kohn-Sham orbitals. We also define a diagonal matrix \( W = \text{diag}[\omega_1, \ldots, \omega_N] \).

It should be noted that the real space representation of Kohn-Sham orbitals can be obtained with any type of basis set, and therefore our method is applicable for any electronic structure software package. For instance, if the Kohn-Sham orbitals are represented using the plane wave basis functions, their real space representation can be obtained on a uniform grid efficiently with the fast Fourier transform (FFT) technique. In such case, \( \omega_i \)'s take the same constant value for all \( i \). For general basis sets such as Gaussian basis functions or localized atomic orbitals, let \( \{ \chi_k(x) \} \) be the collection of basis functions (usually \( M \ll N \)). We first evaluate each basis function on the real space grid as \( \chi_k = [\chi_k(x_1), \chi_k(x_2), \ldots, \chi_k(x_N)]^T \), and denote by \( X = [\chi_1, \ldots, \chi_M] \) the collection of all basis functions. \( X \) is a matrix of size \( N \times M \). Then the Kohn-Sham orbitals can be obtained as the linear combination of basis functions as \( \tilde{\Psi} = XZ \). Here the matrix \( Z \) of size \( M \times N_c \), and is usually obtained by solving a generalized eigenvalue problem. For these general basis functions and in particular in all-electron calculations, the grid points are usually chosen to be non-uniform to improve the accuracy of numerical quadrature, and correspondingly the weights \( \omega_i \)'s are non-uniform as well.

We define \( \Psi = W^{1/2} \tilde{\Psi} \) to be the set of weighted Kohn-Sham orbitals represented in the real space, in which case the discrete orthonormality condition in Eq. (2) becomes \( \Psi^* \Psi = I \), where \( I \) is an identity matrix of size \( N_c \). We now seek a compressed basis for the span of \( \Psi \), denoted by the set of vectors \( \Phi = [\phi_1, \ldots, \phi_{N_c}] \), such that each \( \phi_i \) is localized. The single particle density matrix is defined as \( P = \Psi \Psi^* \). The nearsightedness principle states that for insulating systems, each column of the matrix \( P \) is localized. As a result, selecting any linearly independent subset of \( N_c \) of them will yield a localized basis for the span of \( \Psi \). However, picking \( N_c \) random columns of \( P \) may result in a poorly conditioned basis if, for example, there is too much overlap between the selected columns. Therefore, we would like a means for choosing a well conditioned set of columns, denoted \( C \), to use as the localized basis. Intuitively we expect such a basis to select columns to minimize overlaps with each other when possible.

To achieve this we utilize the so called interpolative decomposition. Given an \( N \times N \) matrix \( A \) with rank \( k \), such a factorization seeks to find a permutation matrix \( \Pi \), a subset of \( k \) columns of \( A \) whose indices form the set \( C \) and a matrix \( T \) such that

\[
A \Pi = A_{C,C} \begin{bmatrix} I & T \end{bmatrix},
\]

and \( \|T\| \) should be small. The interpolative decomposition can be computed via the (strong) rank revealing QR factorization. Such a factorization computes \( A \Pi = Q \begin{bmatrix} R_1 & R_2 \end{bmatrix} \) where \( Q \) is an \( N \times k \) orthonormal matrix, \( R_1 \) is an upper triangular matrix and \( \Pi \) is a permutation matrix. The permutation \( \Pi \) is chosen to keep \( R_1 \) well conditioned. The interpolative decomposition is a powerful technique, and can be used to build low rank approximations for a matrix \( A \) with many small singular values (i.e. \( A \) has many columns that are nearly linearly dependent). In such cases, algorithms for constructing these factorizations often choose \( k \) to ensure that a certain relative accuracy in the approximation of \( A \) is achieved. However, in our situation we know \( P \) is exactly of rank \( N_c \) and are thus not concerned with approximation error.

It may not be feasible to compute an interpolative decomposition of \( P \) directly because we would have to con-
structure $P$ explicitly. The storage cost of $P$ is $N^2$, and computing a partial rank revealing QR factorization of $P$ scales as $O(N_e N^2)$, which is prohibitively expensive. Randomized algorithms exist that would reduce this cost, for example\cite{Halko2011, Halko2012}, but they do not achieve the desired computational scaling. To achieve the desired scaling, observe that for any rank revealing QR factorization of $\Psi$,

$$\Psi^* \Pi = QR$$

where $Q$ is an $N_c \times N_e$ matrix with orthonormal columns, then

$$P \Pi = (\Psi Q) R.$$  

It can be readily verified that $\Psi Q$ is an $N \times N_e$ matrix with orthonormal columns. Therefore, rather than computing a rank revealing QR factorization of $P$ we may equivalently compute a rank revealing QR factorization of $\Psi^*$. This computation scales as $O(N_e^2 N)$. The permutation matrix $\Pi$ reveals the selection of columns $C$, and the SCDM can be computed as

$$\tilde{\Phi} = \Phi_{C,.} = \Psi \Psi_{C,.}.$$ 

Using the SCDM we may recover the density matrix in a simple manner. Since $P$ is a rank $N_e$ Hermitian and positive semi-definite matrix and $P_{C,C}$ has linearly independent columns we may write $P$ in the form $P = P_{C,C} D P_{C,C}^*$, where $D$ is an $N_e \times N_e$ matrix. By restricting $P$ to the $C$ row and column indices, $P_{C,C} = P_{C,C} D P_{C,C}^*$ we observe that $D = (P_{C,C})^{-1}$ is uniquely determined. Therefore,

$$P = P_{C,C} (P_{C,C})^{-1} P_{C,C}. \tag{4}$$

Eq. (4) also suggests a method for the construction of orthonormal and localized basis functions. If we let

$$P_{C,C} = LL^*$$

be a Cholesky factorization of $P_{C,C}$, then

$$\Phi = \tilde{\Phi} L^{-*}$$

has orthogonal columns and they may be used as a basis for the span of $\Psi$. In this case we may also write $P = \Phi \Phi^*$. The orthogonality of $\Phi$ follows from $P$ being an orthogonal projector. Based on the locality of the columns of $P$ and because the permutation $\Pi$ picks columns of $P$ that form a well conditioned basis, $P_{C,C}$ is sparse and only lower triangular entries of $L$ near the diagonal have significant magnitude. Therefore, this orthogonalization step does not significantly impact the localization of the basis functions.

Concisely, we construct the SCDM $\tilde{\Phi}$ or the orthogonalized SCDM $\Phi$ via the algorithm below.

1. Compute the index set $C$ associated with an interpolative decomposition of $\Psi^*$ via a rank revealing QR decomposition.

2. Compute the SCDM $\tilde{\Phi} = P_{C,.} = \Psi (\Psi_{C,.})^*$ as the new localized basis

3. Compute the orthogonal basis $\Phi$ by solving $\Phi L^* = \tilde{\Phi}$

This algorithm is completely deterministic and can be applied to any local or nonlocal basis set. In general, when computing the rank revealing QR factorization for an interpolative decomposition, a so called strong rank revealing QR factorization is technically required. However, for the types of systems we are interested in a more traditional rank revealing QR factorization, such as the one implemented in LAPACK\cite{Lapack} as DGEQP3 suffices. The use of a Cholesky factorization allows us to avoid explicit inversion of $P_{C,C}$ and instead use triangular solves to either apply the spectral projector or orthogonalize the basis.

The overall cost of the algorithm is $O(N_e^2 N + N_e^3)$ to build $\Phi$ or $\tilde{\Phi}$. The cost of the necessary rank revealing QR is $O(N_e^2 N)$ and the dominant cost in $N_e$ that has no dependence on $N$ is the Cholesky factorization, which costs $O(N_e^3)$. The cost of forming the columns of $\Phi$ is $O(N_e^2 N)$ as is the cost of constructing $\Phi$ via triangular solves. Because the algorithm is constructed from simple and standard linear algebra routines it is easy to parallelize. Specifically, steps one, two and four in the preceding algorithm may each use a parallel version, e.g. from ScaLAPACK\cite{Scalapack}, of the respective linear algebra routine. Because the required Cholesky factorization is of a $N_e \times N_e$ matrix it is often not necessary to parallelize that portion of the algorithm, though one certainly could. By utilizing common factorizations and operations the algorithm immediately benefits from improvements to the underlying, serial or parallel, linear algebra routines and does not require any specialized code. One example of this would be use of recent variants on computing a parallel rank revealing QR factorizations such as the communication avoiding version discussed in\cite{Dong2019}. The efficiency of a parallel version will be briefly discussed in the section on numerical results.

To illustrate the simplicity of the algorithm we present a serial implementation of the algorithm in MATLAB\cite{Matlab}. The following two lines of code implement the above algorithm and compute the SCDM. The discrete input $\Psi$ is represented as $\Psi_{ai}$ and $N_e$ is the column dimension of $\Psi$, and therefore the number of localized basis functions. When complete, the matrix $P_{C,e}$ contains the localized SCDM.
If a set of orthonormal and localized orbitals are to be computed, the following three lines can be added, and the matrix Phi contains the orthogonalized and localized SCDM.

\[
\begin{align*}
\Phi &= \Phi_C(\text{Chol}(\Sigma)); \\
\Sigma &= \Phi_C(\text{Chol}(S)); \\
\text{Chol} &= \text{Chol}(S); \\
\Phi &= \Phi_C/\text{Chol}.
\end{align*}
\]

**NUMERICAL RESULTS**

*Localized basis functions*

We now demonstrate the effectiveness of our algorithm from both qualitative and quantitative points of view. For all of our numerical experiments we used QUANTUM ESPRESSO to compute the eigenfunctions. The SCDM are then computed from the Kohn-Sham orbitals for the occupied states. Fig. 1 (a) and (b) show one of the orthogonalized SCDM obtained from a silicon crystal on a cubic domain with 512 atoms, and a snapshot of a water system with 64 molecules. The kinetic energy cut-off for the silicon crystal is 10 Ry, and for water is 75 Ry. For all calculations we use the Troullier-Martins pseudopotential with the Perdew-Burke-Ernzerhof (PBE) functional. The orbitals are very localized in the real space and resemble the shape of MLWFs. Our method automatically finds the centers of all localized orbitals, which for the silicon crystal are in the middle of Si-Si bond, and for water is closer to the oxygen atoms than to the hydrogen atoms.

The locality of the SCDM is demonstrated by computing the locality ratio. We define this as the ratio between the volume of a cuboid domain that contains the support of the basis function after truncating the functions below a certain relative threshold and the volume of the whole domain. Fig. 2 shows the average locality ratio of the SCDM basis functions, both before and after orthogonalization, for each test system as the relative truncation threshold is varied. We clearly see the exponential decay of the SCDM for silicon and water. When the relative truncation value is \(10^{-2}\), the support of each SCDM only occupies \(1 \sim 2\%\) of the computational domain, therefore significantly reducing the storage cost for the orbitals. Furthermore, as anticipated the orthogonalization procedure does not impact the locality of the functions, and in some cases even serves to further localize them. The good localization properties also implies that \(P_{C,C}\) should be a well conditioned matrix. In fact, we observe that the condition number is only 3.18 for the silicon crystal and 2.83 for the water molecules.

\[
\begin{align*}
\text{R} & = \text{R}C(\text{Chol}(S)); \\
\text{Chol} & = \text{Chol}(S); \\
\Phi & = \Phi_C/\text{Chol}.
\end{align*}
\]

**Computation of Hartree-Fock exchange energy**

We now use the orthogonalized SCDM to compute the Hartree-Fock exchange energy with, under some mild assumptions, linear scaling cost. The Hartree-Fock exchange energy is invariant under unitary transformation of the Kohn-Sham orbitals, and can be computed as

\[
E_x = -\frac{1}{2} \sum_{i,j=1}^{N_e} \iint \phi_i(x) \phi_j(x) \phi_j(y) \phi_i(y) \frac{d x \, d y}{x - y}.
\]

Here \(\{\phi_i\}_{i=1}^{N_e}\) can be the Kohn-Sham orbitals, or any unitary transformation of them. When Eq. 5 is computed using the Kohn-Sham orbitals \(\{\psi_k\}_{k=1}^{N_e}\) directly the standard method is to use the plane wave basis set and solve Poisson’s equation for each pair of Kohn-Sham orbitals. This results in having to solve \(O(N_s^2)\) dis-
crete versions of Poisson’s equation with periodic boundary conditions and enforcing zero mean in the solution, which may be done via use of the Fast Fourier Transform (FFT). The overall cost for computing the energy is then $O(N^2eN \log N)$, where the $\log N$ factor arises from the FFT. In a hybrid functional calculation such as PBE0 functionals\[33\] the cost of this step can be dominating compared to a calculation using semi-local exchange-correlation functionals.

By using the orthogonalized and truncated SCDM instead of the delocalized Kohn-Sham orbitals the computational cost can be significantly reduced, at the expense of introducing some controllable error. Let $\hat{\Phi} = [\hat{\phi}_1, \ldots, \hat{\phi}_{N_e}]$ denote the truncated version of $\Phi$. This truncation introduces some error, but the relative truncation value may be chosen to achieve any desired accuracy. After truncation we may simply neglect pairs of $\hat{\phi}_i$ and $\hat{\phi}_j$ with disjoint support, or if their product is sufficiently small on the global domain. When each $\hat{\phi}_i$ is localized the $O(N^e_0)$ terms in Eq. (5) is reduced to $O(N_e)$ significant terms. The computational cost can be further reduced by noting that the solution of Poisson’s equation is only needed on the support of $\hat{\phi}_i \hat{\phi}_j$. This fact may be used to reduce the size of each FFT by using Hockney’s algorithm\[32\]. Hockney’s algorithm is a fast and direct method that does not introduce any additional approximation error. A brief presentation of the main idea of Hockney’s method is provided in the supporting information.

Given the above techniques we still have to determine the support of $\hat{\phi}_i \hat{\phi}_j$ for all $i, j$ pairs, which scales as $O(N^2_e)$. However, if the vectors are appropriately stored this operation amounts to a few logical operations per pair of $\hat{\phi}$ and will not be a dominant portion of the computational cost. Ignoring this cost and assuming that as the number of atoms and size of a molecule grows the support of the basis functions remains constant yields an overall computational scaling of $O(N_e)$. Here the constant will dominantly depend on the size of FFTs required by Hockney’s algorithm.

To demonstrate the efficiency and accuracy of computing the Hartree-Fock exchange energy using the orthogonalized SCDM we construct a quasi one-dimensional silicon crystal extended along the $z$-direction. The total number of atoms varies from 32 to 512 and for each problem size $N_e$ is twice the number of atoms. The kinetic energy cutoff and other parameters are the same as for the aforementioned silicon crystal. All calculations are performed on a single computational node. Fig. 3 shows the time to compute the Hartree-Fock exchange energy when using the localized and truncated basis functions. Our error criteria is the relative error in the computed exchange energy. We truncate the localized basis functions at two different values, and achieve relative error of $4\% \sim 5\%$ in Fig. 3 (a) and $0.3\% \sim 0.4\%$ in Fig. 3 (b). We observe that using the localized and truncated functions greatly reduces the computational time even when factoring in the cost of performing the localization to obtain the orthogonalized SCDM. The cost for the localization scales as $O(N N^2_e)$ but is over an order of magnitude faster (even more so if a parallel version is used) than computing the exchange energy directly using Kohn-Sham orbitals. It is also clear that the use of localized basis functions without using Hockney’s algorithm (“full FFTs”) reduces the observed computational cost to $O(N N_e \log N)$, while using Hockney’s algorithm (“Hockney”) further reduces the computational cost to $O(N_e)$. For the largest problem size the speedup in computing the exchange energy using the localized basis functions and Hockney’s algorithm relative to the computational time using Kohn-Sham orbitals is about a factor of 300 when the relative error is $4\% \sim 5\%$, and a factor of 90 when the relative error is $0.3\% \sim 0.4\%$.

FIG. 3: Time to compute the Hartree-Fock exchange energy and the localized basis functions. The localized basis functions were truncated to achieve (a) $4\% \sim 5\%$ and (b) $0.3\% \sim 0.4\%$ relative error in the energy computation.
FIG. 4: Parallel scaling for computing the orthogonalized SCDM. The times given are the average over 10 computations, and the error bars are one standard deviation away from the mean.

Parallel computation

The above discussion featured results for the localization procedure and the subsequent computation of the Hartree-Fock exchange energy when using a single machine. For the largest quasi one-dimensional problem considered $\Psi$ is a $777600 \times 1024$ matrix. To demonstrate how the algorithm may be parallelized we implemented a simple parallel version in FORTRAN using ScaLAPACK for the parallel rank-revealing QR. The pivoted QR, matrix multiplication to form $\hat{\Phi}$, and the triangular solve to form $\Phi$ were all done using parallel versions of the respective algorithms. We make note of the fact that the parallel timings given here are not on machines of the same type as those used previously for the examples, so the absolute times are not directly comparable. Computing the orthogonalized SCDM for the aforementioned problem using this code and 1024 processors took under 1.5 seconds. This is not including the time to distribute $\Psi$ amongst the processors, which took 1.3 seconds, nor the time to read $\Psi$ from disk. In Figure 4 we show the scaling of this parallel implementation for computing the orthogonalized SCDM. Once again we omit the time taken to distribute the matrix. Here we see that for the given problem size the method scales close to ideally on up to 1024 processors.

CONCLUSION

We have presented a simple procedure to obtain compressed Kohn-Sham orbitals by directly using SCDM. The nearsightedness principle guarantees the locality of the SCDM for insulating systems. The computation of the orthogonalized SCDM is a simple and fast procedure that may be done immediately following the computation of $\Psi$ to build a well localized orthogonal basis for the Kohn-Sham orbital subspace. Because the algorithm is built out of a few very simple linear algebra routines, it is simple to implement, parallelize, and include in electronic structure software packages.

Our work can be extended in several directions. Besides the computation of the Hartree-Fock exchange energy, the high parallel scalability of the SCDM procedure can enable the computation of localized basis functions for the self-consistent treatment of the Hartree-Fock exchange terms in KSDFT calculations with hybrid functionals.

In this work we explicitly require the Kohn-Sham orbitals to be represented on a real space grid. This is very natural for electronic structure software packages based on planewave and finite difference methods, but may or may not be natural for other basis sets such as localized atomic orbitals. In fact, the locality of the single particle density matrix along the off-diagonal direction holds in general when the density matrix is represented using localized basis functions. Therefore it would be possible to directly find the localized representation of the single particle density matrix represented using localized basis functions and skip the real space representation. The numerical consequence of this procedure should be carefully tested.

The cost of computing the SCDM may be reduced via a randomized version of the algorithm. For the examples presented here this method slightly reduced the quality of the localized basis functions and consequently was not used. However, it may still be beneficial for very large problems. The SCDM may also be a useful tool for achieving linear scaling electronic structure calculations for insulating systems, as well as higher level quantum chemical treatment at the post-DFT level. Our idea is not limited to the compression of Kohn-Sham orbitals, but may also be generalized for the compression of pair products of Kohn-Sham orbitals in excited state calculations.

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