Maximally Localized Dynamical Quantum Embedding for Solving Many-Body Correlated Systems

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ABSTRACT

Quantum computing opens new avenues for modelling correlated materials, notoriously challenging to solve due to the presence of large electronic correlations. Quantum embedding approaches, such as the dynamical mean-field theory, provide corrections to first-principles calculations for strongly correlated materials, which are poorly described at lower levels of theory. Such embedding approaches are computationally demanding on classical computing architectures, and hence remain restricted to small systems, which limits the scope of applicability. Hitherto, implementations on quantum computers are limited by hardware constraints. Here, we derive a compact representation, where the number of quantum states is reduced for a given system, while retaining a high level of accuracy. We benchmark our method for archetypal quantum states of matter that emerge due to electronic correlations, such as Kondo and Mott physics, both at equilibrium and for quenched systems. We implement this approach on a quantum emulator demonstrating a reduction of the required number of qubits.

Introduction

Correlated materials have attracted a widespread interest due to their broad range of complex properties and possible avenues for technological applications. Modelling correlated transition metal systems remains a challenge for standard approaches, such as first-principle based methods. With the discovery of high-temperature superconductivity, a large effort has been devoted to the study of archetypal theories for describing Mott insulators, leading to developments of theoretical tools for solving correlated materials accurately. An exact solution of the Hubbard model, which describes such correlated materials, is lacking in two or three dimensions. However, embedding approaches based on controllable approximations, such as the dynamical mean-field theory (DMFT)[1,2], have provided accurate predictions[3–5]. DMFT is a non-perturbative approach that allowed to obtain the phase diagram of the Mott-Hubbard transition. The marriage of density functional theory (DFT) and DMFT (DFT+DMFT) provides the work-horse for studying correlated materials.

DMFT is based on a self-consistent mapping between local properties of a given material onto a so-called Anderson impurity model (AIM), a correlated impurity embedded in an infinite non-interacting bath. The AIM can be solved by high-level many-body methods, such as the continuous-time Monte Carlo approach (CTQMC). The latter provides an exact solution within statistical error bars, but is limited to the imaginary time representation. The evaluation of real-frequency spectral quantities requires the ill-defined analytical continuation[6,7]. Cluster extensions of DMFT suffer from the *fermionic sign-problem* when inter-orbital hybridizations are present. Other solvers are based on the numerical renormalization group (NRG)[8], which allow real axis calculations and access to Kondo physics, but remain challenging to extend for multi-orbital systems. Another method that provides solutions for real frequencies is the exact diagonalisation (ED) approach, where a finite size discretization of the AIM is used, through representation of the infinite bath in terms of a small number of effective *bath-sites*. In typical implementations, the bath size (N_b) is restricted because of the exponential growth of the Hilbert space with the total number of sites N_s (bath sites and impurity orbitals). Nonetheless, Lanczos-based algorithms allows to deal with large Hilbert spaces, where the discretization at low temperature[9,10] is fine enough to compute observables accurately. Some success has been achieved by ED for multi-orbital systems with three or five orbitals[11–13], but in general the limitation in the bath size limits the scope of applicability for realistically describing transition metal oxides. Towards the achievement of the largest number of bath sites, ED calculations have been extended to handle the single impurity embedding problem, allowing up to O(100)[14–17] and O(300)[18] uncorrelated bath sites. Although the latter approaches allow the obtaining of approximations of the zero temperature Green's function, the building of systematically high energy excited states remains challenging.

Moreover, scaling the precision of the latter approach on classical computers is not feasible due to the exponential increase of the Hilbert space with the number of bath sites. In view of the recent progress in the implementation of ED on quantum computers[19], the latter approach has gained interest due to the possibility of scaling linearly on quantum computers. However, due to current hardware limitations (noise and decoherence issues), such algorithms are limited to a small number of qubits and short quantum circuits on the currently available noisy intermediate-scale quantum (NISQ) computers. Hence it is important to develop an ED based solver that can obtain precise results with a reduced number of bath sites, since the required number of qubits is proportional to this.

In this light, here we present the maximally localized dynamical embedding embedding (MLDE) methodology to solve the AIM for DMFT, which extends the ED method by adding electron-electron interactions also in the bath. Our method provides a maximally localized quantum impurity model, where the non-local self-energy component of the correlation due to interactions in the bath remains minimal, and hence the AIM minimally breaks locality (DMFT is a purely local theory). MLDE provides the benefit that the environment used in the quantum embedding approach is described by propagating correlated electrons (instead of free electrons in DMFT). This firstly improves the hybridization fitting procedure compared to ED due to the polynomial increase of the number of fitting parameters with number of bath sites. Secondly, the number of poles in the MLDE hybridization function increases exponentially with the number of bath sites, compared to a linear increase for ED. This allows to obtain converged system properties with a much smaller number of bath sites than the one required by ED. This is reminiscent of the representation of correlated electrons by a Green's function embedding approach, where correlations are described by hidden fictitious additional fermionic degrees of freedom[20,21].

This representation has hence the potential to improve the scope of applicability of the quantum embedding approach, whilst limiting the small number of bath sites. We report that quantum impurity models with as few as 3 bath sites can reproduce both the Kondo regime and the Mott transition and obtain good agreement for dynamical magnetic susceptibilities, poising this approach as a candidate to describe 2-particle excitations such as excitons in correlated systems, with applications for high-temperature superconductors[22,23]. We also present a quantum computing algorithm for MLDE, and show that with a number of qubits, available on current hardware, one can achieve a fine description of correlations in materials.

Results

From the Anderson impurity model to maximally localized dynamical embedding

Within DMFT, the lattice model is mapped to an effective Anderson impurity model (AIM) where a correlated atom is connected to a non-interacting bath with the hybridization function $\Delta(\omega)$ as shown in Fig 1.a.

In the ED approximation, the continuum bath is represented with a finite number of effective sites (Fig.1.b). Typically, for a fixed set of the Hamiltonian parameters (see Eq.1 Methods section), the AIM is solved (1) by using a Lanczos algorithm to converge the ground state and excited states[9,12] which contribute to the thermal average. Once the eigenstates are obtained, the dynamical and static observables are computed. As previously stated, the number of bath sites is severely limited because of the exponential scaling of the Hilbert space with the number of sites. To improve the performance of the algorithm, without increasing the number of bath sites, we introduced a two-body interaction between the bath electrons (Fig.1.c). The resulting approximation is represented by an extended ED solver where the non-local component of the correlation potential remain minimal. Thus the ED method has been extended to a maximally localized dynamical embedding (MLDE) model where the same accuracy is obtained with a reduced number of bath sites. (see Methods section for the formal mathematical definition of the model).

Comparison between MLDE and CTQMC solver

We now turn to a simple test of the MLDE equations for a typical AIM. The bath hybridization used as a test case is obtained from a typical correlated material (The hybridization function is available in Extended Data Figure 1), and the impurity energy is set to $\varepsilon_f = -U/2$ to stay at half-filling. We perform a benchmark of the MLDE approach with respect to both a continuous-time Monte Carlo and Lanczos solver, which both provide in this case the same answer used as a reference. Both the imaginary part (Fig.2.a) and real part (Fig.2.b) obtained by MLDE with as few as $N_b = 3$ bath sites provide a very good agreement with the exact solution. We considered both the nearly free electron (NFE) limit ($U < 10$) and the atomic limit ($U > 10$), and in both cases the MLDE solution is consistently in agreement with the exact answer. Noteworthy, the strength of correlation and overall physical properties are also well captured by MLDE with $N_b = 2$, whereas the ED solver with the same number of bath sites largely overestimates the strength of correlation (see Extended Data Figure 2). MLDE captures well the delocalization-localization transition (see Fig.2.b-c), where the MLDE with $N_b = 2$ only differs near the transition while MLDE with $N_b = 3$ is exact (Fig. 2.c).

Kondo physics

We extend further the application of MLDE to a realistic study case of the deposition of a correlated Kondo molecule on a gold surface (see Fig.3.a). Stable organic radical molecules exhibit a Kondo peak in the low-temperature experimental conductance, which is due to the presence of a single unpaired electron in the highest occupied molecular orbital[24–26]. In the gas phase these molecules are paramagnetic. Due to their low spin-orbit interaction and small hyperfine splitting, they are expected to exhibit long spin-coherence times, and therefore have potential as building blocks of molecular spintronics applications[26]. When brought in contact with a metal surface, the system corresponds to a single impurity Anderson model (SIAM) and has been modeled in the past using CTQMC or NRG as impurity solvers[27,28]. To demonstrate the capability of MLDE to describe Kondo physics, we choose the 1,3,5-triphenyl-6-oxoverdazyl (TOV) organic radical molecule, which, when deposited on a Au substrate has been shown experimentally to exhibit a Kondo temperature of about ≈ 37K[24]. Compared to other radical molecules on surfaces, this molecule has the advantage to have a well-defined contact geometry, where the molecule lies flat on the surface. We use the same simulation setup and parameters as in a previous study[27] so that also the hybridization function of the SIAM is the same (see Extended Data Figure 5). Describing Kondo physics at low temperature is a notoriously difficult problem for quantum impurity solvers. In particular, the collapse of energy scales in the Kondo limit prevents typical Lanczos or ED solvers from capturing the Kondo resonance, as the finite discretization tends to introduce fictitious gaped states at very low temperature. We performed calculation at $T = 5$K. In the Kondo regime (see Fig.3.b) the hybridization remains constant up to the lowest frequency. We note that $\Delta_1(i\omega_n) = \Delta_2(i\omega_n)$, which confirms that the embedding source field $\delta\Delta(i\omega_n)$ remains negligible. Furthermore, MLDE fares better with $N_b = 4$ than the best possible fit obtained by the usual discretized Lanczos approach ($N_b = 12$, triangles in Fig.3.b), as the hybridization vanishes at small frequencies (in the molecule, it remains constant). The imaginary part of the self-energy obtained by MLDE shows a Fermi liquid type behavior at small frequencies (see Fig.3.c), whereas the self-energy obtained by ED shows an artificial Mott singularity, due to the bath discretization. We note that this low temperature was beyond the reach of our CTQMC solver ($\beta = 35000$ Ryd^{-1}). Below the Kondo temperature, we recover the Fermi liquid behavior of the self-energy (see Fig.3.d). As the MLDE representation is compact, it opens a large degree of possible manipulation once the AIM is established. In particular, we extended the calculation to the time dynamics of the Kondo molecule with the Keldysh formalism[29] after a magnetic quench, where at time $t_0 = 0$ the molecule is magnetically polarized along the e_z axis, and the external magnetic field released for $t > t_0$. The magnetic moment enters in a precession dynamics with the frequency equal to the Kondo temperature (see Fig.3.e). The estimated Kondo temperature(T_K) which we achieved with different methods, e.g. from the time dynamics ($T_K \approx 33.64$) and from perturbation theory[30,31] ($T_K \approx 33$, see subsection MLDE for Kondo molecule in the Methods section), which provides a further check of the validity of the MLDE approach. The dynamics can also be resolved below T_K, where we observe additional harmonics, reminiscent from the Kondo Zeeman splitting at $t = t_0$[32].

Mott transition

We have so far focused on simple AIM systems in the absence of mean-field corrections to the hybridization. We now turn to the dynamical mean-field MLDE approach, applied to the Mott transition where the local Green's function is defined with the density of state of the two-dimensional square-lattice Hubbard model (see Fig.4.a). Using the MLDE as a solver for DMFT, we recover the well-known metal-insulator transition (MIT) associated with the charge localization induced by the local Hubbard repulsion U. Here, the DMFT self-consistency is enforced in Matsubara frequency. We recover with a simple MLDE and $N_b = 3$ bath sites the spectral function of the Hubbard model, with the usual features (lower and upper Hubbard bands, quasi-particle peak below the transition $U_c \approx 9$, Mott gap for $U > U_c$). We note that interestingly the spectral function obtained by MLDE also shows the satellite peaks at the gap edge for $U > U_c$. This feature is typically difficult to obtain with analytically continued spectral function from Matsubara quantity. Indeed the inner peak near the Mott gap edge are associated with the real part of the self-energy, and in particular, is derived from an actual quasi-particle solution[33].
The MLDE charge gap Δ reproduces the known trend $\Delta = U - W$, which provides a further test of the theory. The benchmark with the converged solution obtained by CTQMC is accurate (see Fig.4.b), and the MLDE solution essentially within the error bars of the CTQMC for $U/t = 12$. Across the Mott transition, the agreement between the Green's functions remains good (see Fig.4.c).

Bethe-Salpeter Equation

We extended the calculations to the Bethe-Salpeter Equation (BSE) formalism, applied to the MLDE solution. In particular we calculate the local irreducible vertex Γ (see Section Vertex calculation in MLDE in the Methods section), which enables the calculation of the non-local dynamical magnetic susceptibility $\chi^{mag}(\omega)$ within MLDE. We performed calculations for $U/t = 12$ at various temperatures, to explore the behavior of magnetic excitations across the Mott gap melting. Within the Mott gap phase at temperature $T/t = 0.025$ (Fig.5.a), the Neel fluctuations are local in momentum at $Q_{\text{Neel}} = (\pi, \pi)$. As expected, the spin fluctuations are largely located at Q_{Neel} in the Mott phase ($T = 0.025t$). We observe that the antiferromagnetic magnetic fluctuations become gradually more incoherent as the temperature is increased throughout (Fig.5.b). Interestingly, we observe a large degree of incoherence at all considered temperature, which is reminiscent of the large scattering rate of the archetypal Mott system La2CuO4. At very large temperature (Fig.5.c), the fluctuations are fully incoherent, and the spectrum uniform across the Brillouin zone.

We note that a known challenge for the BSE approach is the extraction of the local irreducible vertex Γ, which is obtained by calculating the two-electron response function $G^{(2)}$. Such a quantity requires traditionally computationally demanding collection of statistics by CTQMC and few other alternatives exist. The vertex can be calculated in the Lehman representation (see Section Vertex calculation in MLDE in the Methods section), but requires sampling over the whole Hilbert space, which is not possible for AIM with more than few bath sites. In MLDE this task is largely simplified as the Hilbert space remains compact. We provide a benchmark of the local vertex Γ with CTQMC for both $U/t = 6$ and $U/t = 12$ (see respectively Figs.5.e and 5.g), in both cases the agreement is quantitative. This agreement is also obtained in the fermionic representation of the local magnetic susceptibility $\chi_{i\nu,i\nu}$ (see Figs.5.d and 5.f). In conclusion, our MLDE solver can be integrated in ab-initio DFT+DMFT, where the irreducible vertex and the self-energy are the key quantities which are ultimately used to compute any observable. We ensure correctness of the approximation up to the two particle response function, while for higher order response functions the technique is limited as compared as ED.

MLDE for multiorbital system

So far, we have considered a single orbital case as a proof of concept of the formalism. To highlight the advantage of the MLDE solver, we now turn to the application to a multi-orbital case. The ED solver has been notoriously limited for the case of multi-orbital systems[34] as it would require a large number of bath sites, whereas CTQMC solver would be affected by the sign problem triggered by the non-zero off-diagonal terms of the hybridization between the orbitals. Therefore, in this section, we demonstrate the ability to treat multi-orbital systems within MLDE framework. As a test system, we applied DFT+DMFT on $LaNiO_3$, whose nontrivial contributions stemming from electronic correlations have been studied using CTQMC solver[35]. In particular, The crystal field splits the Ni d orbital into two maniforlds t_{2g} (composed of d_{xy}, d_{xz}, d_{yz} orbital) and e_g ($d_{z^2}, d_{x^2-y^2}$); here, the e_g are active orbitals and pose this compound as our ideal candidate of two orbitals system.

Our main focus is the comparison between a standard DFT+DMFT implementation using CTQMC and ED impurity solver with our MLDE environment with 3 bath sites. As shown in Fig 6a, a good agreement is achieved between CTQMC and MLDE when considering the local self energy for the first iteration of the DMFT loop.

We also show the local self energy obtained with ED using 3 bath sites. Notice that, differently from the results obtained by MLDE, the ED solver does not well capture the correct self energy behavior. A better agreement between the ED solver and the CTQMC would be achieved at the cost of adding more bath sites to increase the number of parameters and density of poles in the hybridization function required for the fit of the hybridization, as it is done in MLDE.

Lastly, we consider the local Green's function for the final iteration of the self-consistent DMFT loop. Notice that while we only include 3 sites in the bath, the MLDE Green's function is in a good agreement compared to CTQMC results.

MLDE on a quantum computer

Since MLDE reduces the number of bath sites compare to ED, it opens the oportunity to run Hamiltonian simulation on Noisy Intermediate-Scale Quantum computer(NISQ) with a reduced number of qubits. Here we develop a quantum algorithm to run MLDE on NISQ devices and demonstrate it using a quantum simulator on the benchmark system considered in Fig. 2. To calculate the Green's function on a quantum computer, we use the variational quantum eigensolver (VQE) based method presented by Rungger et al[36], which was demonstrated to run on currently available hardware as it is rather resilient to noise. Details of the method are presented in the Sec. 5 of the method. Here we show results obtained with our implementation within the Quest quantum simulator[37]. To demonstrate that the quantum MLDE algorithm gives the same accuracy as the classical MLDE algorithm, we apply it on the same Anderson impurity model shown in Fig. 2. The system has one impurity site and two bath sites, which we map to a 6 qubit system using a Jordan-Wigner transform[38]. In Fig. 2.d we show that the self-energy for $U = 6$ computed with the quantum computing algorithm agrees very very well with the result obtained using the classical computing algorithm. We have verified that the same is true for all values of U. This, therefore, demonstrates the functionality of our quantum algorithm for MLDE.

Discussion

We now turn to the discussion of MLDE and its advantages and limitations with respect to either other methods or known limits. Indeed, the MLDE non-local self energy vanishes, as expected, when: the local impurity correlation is naught ($U_{imp} = 0$), the bath correlations are naught, or when the impurity-bath coupling is naught ($V = 0$). In between the latter limits, we note that

MLDE is an approximation of the exact AIM self-energy, on the same ground as the standard ED technique. Although MLDE mitigates to a large extent the bath discretisation error present in the standard ED approach, it introduces another error due to non-local components in the self-energy between the impurity and the bath orbitals. MLDE and ED converge to the same exact solution in the limit where the bath discretisation involves a large number of bath orbitals.

In this regard, we showed in this work that MLDE also performs optimally in the intermediate regime, where typically emergent quantum state of matters occur, i.e. Kondo physics, Mott physics, and correlated Fermi liquids. MLDE always performs better than ED (for the same number of bath sites) when the non local components in the self-energy are minimised. The reason for the better performance are twofold: MLDE improves the hybridization fitting procedure compared to ED due to the polynomial increase of the number of fitting parameters with number of bath sites, and the number of poles in the MLDE hybridization function increases exponentially with the number of bath sites, compared to a linear increase for ED. This allows to obtain converged system properties with a much smaller number of bath sites than the one required by ED. In MLDE, the non-local self-energy provides a figure of merit of the obtained solution. A quantitative way to check that the MLDE is converged to a good set of parameters is to systematically evaluate the self-energy for increasing number of bath sites, in the same way as it is done for ED.

A further advantage is that MLDE can be applied in the realm of quantum computing on NISQ devices, where, with a reduced number of required qubits, MLDE can allow the simulation of transition metal systems on a quantum computer, with minor errors induced by the bath discretization. Note that on classical computers the computational cost increases exponentially with system size, both for ED and MLDE, while quantum computers can potentially reduce this to a polynomial scaling[39,40]. On near term quantum computers the noise limits the size of calculations that can be done in practice, so that only very small proof of concept DMFT systems have been demonstrated on hardware[36]. Since MLDE significantly reduces the number of sites needed to describe a given physical system, it allows to reduce the required number of qubits as well as the depth of the quantum circuits. Therefore, in the near term, it is a potential framework to run larger physical systems on NISQ devices, while in the long term, the reduction of the required number of sites will be beneficial also in the fault-tolerant quantum computing regime.

Methods

Overview of the MLDE method

Within DMFT, the effective AIM is subject to a self-consistency condition which relates the Green's function of the impurity model $G(i\omega_n)$ to the so-called Weiss-field $\mathscr{G}_0^{-1}(i\omega_n)$, which completely characterizes the AIM. For the single-band case, the Hamiltonian of the AIM reads:

$$H = \sum_{ij\sigma} \varepsilon_{ij\sigma} \hat{d}^{\dagger}_{i\sigma} \hat{d}_{j\sigma} + \sum_{i\sigma} V_i (\hat{d}^{\dagger}_{i\sigma} \hat{f}_{\sigma} + hc) + U \hat{n}_{f\uparrow} \hat{n}_{f\downarrow} + \sum_{\sigma} \varepsilon_f \hat{f}^{\dagger}_{\sigma} \hat{f}_{\sigma} \tag{1}$$

where $d^{\dagger}_{p\sigma}$ ($d_{p\sigma}$) creates (destroys) a particle with spin σ in the d-orbitals of the uncorrelated bath ($p \in [1, N_b]$) and $f^{\dagger}_{\sigma}$ (f_{σ}) creates (destroys) a spin σ particle on the impurity, U is the static Coulomb repulsion on the impurity and V is the tunneling amplitude between the impurity and the bath.

In the MLDE approach we add an additional general two-electron interaction to the bath sites, and the interaction vertex reads:

$$H_{\text{int}} = \sum_{\sigma_1,\sigma_2} \sum_{i,j,k,l} U^{(2)}_{i,j,k,l} \hat{d}^{\dagger}_{i\sigma_1} \hat{d}^{\dagger}_{j\sigma_2} \hat{d}_{k\sigma_2} \hat{d}_{l\sigma_1} \tag{2}$$

The $U^{(2)}$ tensor is a fictitious two-body interaction between bath electrons introduced to enlarge the number of fitting parameters for a given number of bath sites, and also to enlarge the number of poles in the hybridization function. Although the free electron propagator takes a simple polynomial form, e.g. $G_0^{-1} = i\omega_n - \varepsilon_f - \sum_i V_i^2 / (i\omega_n - \varepsilon_i)$, a correlated Green's function is instead described by a mapping to an exponentially long one-dimensional chain within the Krylov space[41]. In particular, when the bath is correlated, the Weiss field incorporates the dressed propagator of the bath electrons $\tilde{G}^{(1)}_{\text{d}}$:

$$G_0^{-1} = i\omega_n - \varepsilon_f - \underbrace{\boldsymbol{V}^{\dagger} \tilde{\boldsymbol{G}}^{(1)}_{\text{d}} \boldsymbol{V}}_{\boldsymbol{\Delta}^{(1)}}, \tag{3}$$

where the hybridization to the bath is denoted as $\Delta^{(1)}$. We emphasize that at this level of the theory, the self energy of the impurity Σ_f remains naught, and hence we have a fully local theory from the perspective of the impurity. However, as we introduce a local correlation U on the impurity, two effects occur: i) the propagator of the bath $\tilde{G}_d^{(1)}$ is dependent on the correlation on the impurity, and hence changes to $\tilde{G}_d^{(2)}$ and ii) a non-local part of the self-energy Σ_{fd} between the impurity and the bath emerges:

$$G_{imp}^{-1} = i\omega_n - \varepsilon_f - \underbrace{(\mathbf{V}+\mathbf{\Sigma}_{fd})^{\dagger}\tilde{\mathbf{G}}_{\mathrm{d}}^{(2)}(\boldsymbol{V}+\mathbf{\Sigma}_{fd})}_{\mathbf{\Delta}^{(2)}} - \mathbf{\Sigma}_f \tag{4}$$

We have now a set of embedding equations, which leads to a generalized Dyson equation

$$\boldsymbol{G}^{-1} - \boldsymbol{G}_0^{-1} = \delta\mathbf{\Delta} - \mathbf{\Sigma}_f, \tag{5}$$

where the $\delta\Delta$(z) is a source field that stems from the non-local correlations. Indeed, the latter term can be rationalized following a simple argument via the Migdal energy functional, which in MLDE reads

$$U\langle \hat{n}_{\uparrow}\hat{n}_{\downarrow}\rangle = \frac{1}{2}\left(Tr\left(\Sigma_f G_f\right) + \underbrace{Tr\left(\mathbf{\Sigma}_{fd}\boldsymbol{G}_{fd}\right)}_{\bar{Z}} \right) \tag{6}$$

Part of the correlation energy spills effectively on the bath[42], leading to a correlation leakage term $\bar{Z}$.

The DMFT equations can be recovered when the leakage potential and leakage correlation energy are small. Note that the obtained DMFT equations are for a self-energy with interactions in the bath, which differs from the one obtained without interactions in the bath, and which only becomes equal to the exact self-energy for an infinitely large number of bath sites. As the tuning of the bath propagator allows for a very large set of parameters, these constraints can be successfully enforced via Lagrange parameters (see the cost function defined in Eq. 19) in the fit of the DMFT hybridization, to maximize the locality of the embedding, with a concomitant exponential improvement of the bath discretization errors. It is known that in high dimension, or when a system is strongly correlated, the electron self-energy is well separable into a local dynamical part and a static non-local contribution[43]. In this respect, we reabsorb the embedding potential into a shift of the static part of the MLDE self energy, which ensures that the Migdal energy remains exact.

Details of the MLDE derivation

In this section, we provide an extended derivation of the MLDE formalism briefly introduced in the previous section. To this end, we start with reviewing the case of a non-correlated bath. We then proceed to introduce the quantities of interest in the MLDE formalism, where bath interactions are included. We also provide the pseudo-code where the algorithm is described in more detail.

Case of non correlated bath

In the case of the non correlated bath, shown in Fig.1.a, the AIM is described by the Hamiltonian of Eq. 1. Then the free-electron propagator is

$$G_0^{-1}(i\omega_n) = i\omega_n - \varepsilon_f - \Delta(i\omega_n). \tag{7}$$

In the ED approximation, the continuum bath is represented with a finite number of effective sites. Hereby, we consider and AIM in a start geometry, where the hybridization $\Delta(i\omega_n)$ is discretized as follows:

$$\Delta^{ED}(i\omega_n) = \sum_d \frac{V_d^2}{i\omega_n - \varepsilon_d} \tag{8}$$

In the case of correlated impurity, $U \neq 0$, the problems acquire a many-body nature, and the electron propagation is affected by the scattering events, therefore correlations between the electrons in the system. We then introduce the interacting propagator of the impurity of the AIM in the function of the hopping terms V_d between impurity and bath sites and onsite energies ε_d of the bath sites:

$$(G^{ED})_{imp}^{-1}(i\omega_n) = i\omega_n - \varepsilon_f - \Delta^{ED}(i\omega_n) - \Sigma^{ED}(i\omega_n) \tag{9}$$

The Dyson equation for the impurity reads:

$$(G^{ED})_{imp}^{-1}(z) = G_0^{-1}(z) - \Sigma_f^{ED}(z) \tag{10}$$

For our convenience, we report the Green's function and the self-energy Σ of the problem, in a matrix formalism Hence:

$$\mathbf{\Sigma}^{ED} = \left(\begin{array}{c|c} \Sigma_{\mathrm{f}}^{ED} & 0 \\ \hline 0 & 0 \end{array}\right) \quad \text{and} \quad (\boldsymbol{G}^{ED})^{-1} = \left(\begin{array}{c|c} (i\omega_n - \varepsilon_f - \Sigma_f^{ED}) & -\mathbf{V} \\ \hline -\mathbf{V} & (i\omega_n - \varepsilon_d) \end{array}\right) \tag{11}$$

Notice that their dimension is $N_b + 1$, with N_b being the number of bath sites.

Case of correlated bath

In the maximally localized dynamical embedding approach (MLDE), we extend the formalism of the ED approximation to include correlations in the bath sites, adding a two-body interaction term of Eq. 2 in the Hamiltonian. Hence, the MLDE Hamiltonian reads as:

$$H=\sum_{ij\sigma}\varepsilon_{ij\sigma}\hat{d}^{\dagger}_{i\sigma}\hat{d}_{j\sigma}+\sum_{i\sigma}V_i(\hat{d}^{\dagger}_{i\sigma}\hat{f}_{\sigma}+hc)+U\hat{n}_{f\uparrow}\hat{n}_{f\downarrow}+\sum_{\sigma}\varepsilon_f\hat{f}^{\dagger}_{\sigma}\hat{f}_{\sigma}+\sum_{\sigma_1,\sigma_2}\sum_{i,j,k,l}U^{(2)}_{i,j,k,l}\hat{d}^{\dagger}_{i\sigma_1}\hat{d}^{\dagger}_{j\sigma_2}\hat{d}_{k\sigma_2}\hat{d}_{l\sigma_1} \tag{12}$$

We emphasize that this formalism can be extended further by the addition of higher order terms (e.g. if a three-rank tensor is introduced, the number of parameters scales at most as N_b^6) being limited only by the dimension of the Hilbert space. Therefore our method introduces a great advantage over the ED solver, where the number of fitting parameters scales only as N_b^2, and therefore a larger number of bath sites is needed for the increase of the accuracy.

Consistent with the previous section, we start from the definition of the free-electron propagator in Eq. 3, where the hybridisation to the bath is denoted as $\Delta^{(1)}$ and includes a Green's function for the bath $\tilde{G}^{(1)}_{\mathrm{d}}$ which is interacting rather then being a free-electron propagator as in Eq. 8 (more details in the pseudocode **Algorithm 1**). We emphasise that in case of non correlated bath the number of poles in the hybridization scales linearly with N_b, while the inclusion of a correlated bath introduces an exponential scaling of the number of poles in the hybridization function with N_b. Notice that at this level of the theory, the self-energy of the impurity Σ_f remains nought, and hence we have a fully local theory from the perspective of the impurity.

We now turn to the case of non-zero correlation on the impurity where the interacting propagator on the bath $G^{(2)}_{\mathrm{d}}$ is affected by the correlation in the impurity and a non-local part of the self-energy Σ_{fd} between the impurity and the bath emerges.

To better clarify the formalism introduced in Eq.(4) so far, we recall the Green's function and the self-energy in their matrix form. Hence:

$$\boldsymbol{G}(i\omega_n)=\left(\begin{array}{c|c} G_{\mathrm{f}}(i\omega_n) & \boldsymbol{G}_{\mathrm{fd}}(i\omega_n)\\ \hline \boldsymbol{G}_{\mathrm{df}}(i\omega_n) & \boldsymbol{G}_{\mathrm{d}}(i\omega_n)\end{array}\right)\quad\text{and}\quad \boldsymbol{\Sigma}(i\omega_n)=\left(\begin{array}{c|c} \Sigma_{\mathrm{f}}(i\omega_n) & \boldsymbol{\Sigma}_{\mathrm{fd}}(i\omega_n)\\ \hline \boldsymbol{\Sigma}_{\mathrm{df}}(i\omega_n) & \boldsymbol{\Sigma}_{\mathrm{d}}(i\omega_n)\end{array}\right) \tag{13}$$

Notice that to simplify the notation, the repeated indices are dropped. Therefore $\boldsymbol{G}_{dd}=\boldsymbol{G}_d$. A comparison with the matrices defined in the case of non-correlated bath, provides further clarification on the consequences of introducing interactions in the bath sites. The block of the self-energy matrix related to the bath has now an non-zero contribution by definition of our model. A less trivial observation concern the off-diagonal terms which are non zero, due to the interaction leaked in the bonds between impurity and bath sites. Therefore we can distinguish different contribution to the correlation energy, and in terms of Migdal energy functional, it reads:

$$E_U^{tot}=\frac{2}{\beta}\sum_{i\omega_n}\mathrm{Tr}\left[\boldsymbol{G}^{\uparrow}(i\omega_n)\left(\boldsymbol{\Sigma}^{\uparrow}(i\omega_n)-\boldsymbol{\Sigma}^{\uparrow}(\infty)\right)\right]+\frac{2}{\beta}\mathrm{Tr}\left(\boldsymbol{\rho}^{\uparrow}\boldsymbol{\Sigma}^{\uparrow}(\infty)\right) \tag{14}$$

$$E_U^{bath}=\frac{2}{\beta}\sum_{i\omega_n}\mathrm{Tr}\left[\boldsymbol{G}_d^{\uparrow}(i\omega_n)\left(\boldsymbol{\Sigma}^{\uparrow}(i\omega_n)-\boldsymbol{\Sigma}^{\uparrow}(\infty)\right)_d\right]+\frac{2}{\beta}\mathrm{Tr}\left(\boldsymbol{\rho}_d^{\uparrow}\boldsymbol{\Sigma}_d^{\uparrow}(\infty)\right) \tag{15}$$

$$E_U^{imp}=\frac{2}{\beta}\sum_{i\omega_n}G_f^{\uparrow}(i\omega_n)\left(\Sigma_f^{\uparrow}(i\omega_n)-\Sigma_f^{\uparrow}(\infty)\right)+\frac{2}{\beta}\rho_f^{\uparrow}\Sigma_f^{\uparrow}(\infty) \tag{16}$$

where ρ is the density matrix. Part of the correlation energy spills effectively on the bath[44], leading to a correlation leakage term, which is defined dynamically

$$z_U^{int}(i\omega_n)=\frac{\left(\boldsymbol{\Sigma}_{fd}(i\omega_n)\cdot\boldsymbol{G}_{fd}(i\omega_n)\right)^2}{|i\omega_n|} \tag{17}$$

and statically as a result of the average over matsubara frequencies:

$$E_U^{leakage}=\frac{2}{\rho_f}\left(E_U^{tot}-E_U^{bath}-E_U^{imp}\right) \tag{18}$$

The locality principle behind the DMFT equations can be recovered when the leakage potential $\delta\Delta(z)$ and leakage correlation energy are small. We clarify that the leakage field $\bar{Z}$ defined in Eq. 6 is $\bar{Z}=E_U^{leakage}R_f/2$. As the tuning of the bath propagator allows for a very large set of parameters, these constraints can be successfully enforced via Lagrange parameters in the fit of the

DMFT hybridization, to maximize the locality of the embedding, with a concomitant exponential improvement of the bath discretization errors. To that end, the cost function of the minimization procedure is defined as:

$$d = \sum_{i\omega_n} \frac{|\Delta^{(1)} - \Delta_{target}|^2 + |\Delta^{(2)} - \Delta_{target}|^2 + \alpha_3 |\Delta^{(1)} - \Delta^{(2)}|^2}{|i\omega_n|^{\alpha_1}} + \lambda_s |E_U^{leakage}| + \lambda_d \sum_{i\omega_n} z_U^{int}(i\omega_n) \tag{19}$$

where the parameters $\alpha_1, \alpha_3, \lambda_d, \lambda_s$ are properly tuned according to the case of study to improve the minimisation procedure and Δ_{target} is the hybridisation of the continuous bath. We noticed that in most of the cases, the leakage field $E_U^{leakage}$ turns out to be naturally small. Indeed this follows from the constraint that $|\Delta^{(1)} - \Delta^{(2)}|$ has to be minimal. Therefore the fitting process tries to reduce the leakage by construction. It is worth it to notice, however, that all the terms in the cost function are important to give more flexibility to the minimisation process and avoid it to get stuck into local minima. Moreover, the first and the last term allow to have a better control on the low frequencies contributions and increase the precision of the description of phenomena occurring at a low energy scale, e.g. Kondo physics.
To ensures that the Migdal energy remains exact, we reabsorb the leakage field into a shift of the static part of the MLDE self-energy. This is an allowed procedure as it is known that in high dimension, or when a system is strongly correlated, the electron self-energy is well separable into a local dynamical part and a static non-local contribution[43].
To summarise, in the MLDE formalism, non-local correlations are introduced to enlarge the parameters for the fitting of the parameters. This represents an improvement over existent ED formalism , as the increased number of fitting parameters allow the use of less bath sites. The non-locality is kept small through a Lagrange parameter in the cost function. To recover consistency with MLDE, the leaked (non-local) correlated energy is reabsorbed in the impurity self-energy. More details on the different steps of the algorithm are shown in the pseudo-code (**Algorithm 1**) reported below.

Algorithm 1 MLDE solver

inputs: $\Delta_{target}, \varepsilon_f, \alpha, \alpha_3, \lambda_s, \lambda_d, U, N_b$

Require: $U_{imp} = 0$

call solver: input: Δ_{target}, output $\boldsymbol{\varepsilon}^{(1)}, \boldsymbol{V}^{(1)}, \boldsymbol{G}^{(1)}$

$\boldsymbol{\Sigma}^{(1)} \leftarrow i\omega_n \boldsymbol{I} - \boldsymbol{\varepsilon}^{(1)} - \left(\boldsymbol{G}^{(1)}\right)^{-1}$ (Dyson Equation)

$\tilde{\boldsymbol{G}}_d^{(1)} \leftarrow (i\omega_n \boldsymbol{I})^{-1} - \boldsymbol{\varepsilon}_d^{(1)} - \boldsymbol{\Sigma}_d^{(1)}$

$\boldsymbol{\Delta}^{(1)} \leftarrow \boldsymbol{V}^{(1)\dagger} \tilde{\boldsymbol{G}}_d^{(1)} \boldsymbol{V}^{(1)}$

$d_1 \leftarrow \sum_n \frac{|\Delta^{(1)} - \Delta_{target}|^2}{|i\omega_n|^{\alpha}}$

Require: $U_{imp} \neq 0$

call solver: input: Δ_{target}, output $\boldsymbol{\varepsilon}^{(2)}, \boldsymbol{V}^{(2)}, \boldsymbol{G}^{(2)}$

$\boldsymbol{\Sigma}^{(2)} \leftarrow i\omega_n \boldsymbol{I} - \boldsymbol{\varepsilon}^{(2)} - \left(\boldsymbol{G}^{(2)}\right)^{-1}$ (Dyson Equation)

$\boldsymbol{\Sigma}_{int}^{(2)} \leftarrow -\boldsymbol{V}^{(2)} - \boldsymbol{\Sigma}_{fd}^{(2)}$

$\tilde{\boldsymbol{G}}_d^{(2)} \leftarrow \left(-\boldsymbol{\varepsilon}_d^{(2)} - \boldsymbol{\Sigma}_d^{(2)} + i\omega_n \boldsymbol{I}\right)^{-1}$

$\boldsymbol{\Delta}^{(2)} \leftarrow \boldsymbol{\Sigma}_{int}^{(2)\dagger} \tilde{\boldsymbol{G}}_d^{(2)} \boldsymbol{\Sigma}_{int}^{(2)}$

compute $E_U^{tot}, E_U^{bath}, E_U^{imp}$ and $E_U^{leakage}$

$d_2 \leftarrow \sum_n \frac{|\Delta^{(2)} - \Delta_{target}|^2}{|i\omega_n|^{\alpha}}$

$d_3 \leftarrow \sum_n \frac{|\Delta^{(2)} - \Delta^{(1)}|^2}{|i\omega_n|^{\alpha}}$

$d_4 \leftarrow |E_U^{leakage}|$

$d_5 \leftarrow \sum_{i\omega_n} |z_U^{int}(i\omega_n)|$

$d \leftarrow d_1 + d_2 + \alpha_3 d_3 + d_4 \lambda_s + d_5 \lambda_d$

$\Sigma_f \leftarrow \Sigma_f + E_U^{leakage}$

MLDE error scaling

As we stated previously, we find that in our calculations the non-local self energy contribution remains small. Additionally, we note that the non-local self energy vanishes in known limits of the theory: when the local impurity correlation is naught

($U_{imp} = 0$), when the bath correlations are naught ($U_b = 0$)(Here for simplicity we denote as U_b the bath fictitious interactions), or when the impurity-bath coupling is naught ($V = 0$). In any of these limits, the non-local self energy vanishes and the MLDE distance measure is optimal. The first two cases are identical in nature, as they are mirror of each others, and both corresponds to the usual DMFT implementation where a correlated sub-system is embedded within a non-correlated one. We now turn the discussion to the case of small V coupling within MLDE. To address this scenario, we consider the smallest possible example system, where the local density of states of the impurity is identical in a free electron bath and in a correlated bath:

- **First system (with bath interactions):** the impurity is connected to a single interacting bath orbital with local interaction U_b. The green's function of the interacting bath has poles at $-U_b/2$ and $U_b/2$ (we stay at half-filling).
- **Second system (no bath interactions):** the impurity is coupled to two non-interacting bath orbitals with energies $-U_b/2, U_b/2$

In both cases, the local density of states of the bath is set to half-filling (see Extended Data Figure 3.a). In Fig. S1b, we report the MLDE scaling error as a function of leakage for different impurity to bath coupling V. As mentioned above and expected, for $V = 0$ MLDE is exact and the self energies of both approaches identical. Upon increasing V, we compare the self energies obtained by both methods, as MLDE is an approximation in this limit, and compare the error obtained in the self energy with the leakage spilling. As shown in Extended Data Figure 3.b, in this simple limit the MLDE error obtained in the self energy correlates with the leakage. This is an illustration of the scaling of the MLDE error in a simple known limit, near the limit where MLDE is exact (weak bath correlations or weak impurity-bath coupling).

Benchmark in real frequency

Previously, we preferred to benchmark the MLDE with ED and CTQMC, in the Matsubara formalism, rather than in real frequency. Indeed, the spectral functions are notoriously known for being a poor benchmark when using the continuous fraction method, used for the benchmark ED solver (as the number of bath sites is large, Lehmann representation of the GF is not achievable). In particular, reliable benchmarks can be obtained via the Keldysh formalism and Fourier transforming the time evolution to real frequency[45], but require extensive computing resources for the large systems used as the benchmark. In this regard, we compared the spectral functions obtained by the MLDE for the Mott transitions with the one obtained by ED (see Extended Data Figure 4). However, a comparison in the real axis is less reliable due to the continuous fraction. Moreover, the ill-conditioned maxent approach prevents us from detailed comparison on the real axis between CTQMC and MLDE. Therefore, we have benchmarked the different solvers in the Matsubara formalism.

Vertex calculations in MLDE

In this section, we provide further insights on the application of MLDE to the calculation of two particle Green's function quantities. In particular, the dynamical susceptibility fermionic matrix is computed by inverting the Bethe-Salpeter Equation (BSE)

$$\chi(q,i\Omega_n)_{(i\nu\sigma)(i\nu'\sigma')} = [(\chi^0(q,i\Omega_n)^{-1} + \Gamma^{imp}(i\Omega_n)]^{-1}_{(i\nu\sigma)(i\nu'\sigma')}, \tag{20}$$

where :

$$\chi(q,i\Omega)^0_{(i\nu\sigma)(i\nu'\sigma')} = -\delta_{i\nu i\nu'}\delta_{\sigma\sigma'}\sum_k g_{k\sigma}(i\nu)g_{k+q,\sigma'}(i\nu+i\Omega), \tag{21}$$

and Γ^{imp} is the impurity vertex obtained by inverting the BSE for the MLDE impurity problem. The impurity susceptibility reads:

$$G^{(2)imp}_{(i\nu\sigma)(i\nu'\sigma')}(i\Omega) = \int_0^\beta d\tau_1 d\tau_2 d\tau_3 d\tau_4 e^{i\nu(\tau_1-\tau_2)+i\nu'(\tau_3-\tau_4)+i\Omega(\tau_1-\tau_4)} \left\langle \mathscr{T}_\tau c^\dagger_\sigma(\tau_1)c_\sigma(\tau_2)c^\dagger_{\sigma'}(\tau_3)c_{\sigma'}(\tau_4)\right\rangle \tag{22}$$

$$\chi^{imp}_{(i\nu\sigma)(i\nu'\sigma')}(i\Omega) = G^{(2)imp}_{(i\nu\sigma)(i\nu'\sigma')}(i\Omega) - \beta\delta_{0\Omega}G(i\nu)G(i\nu'). \tag{23}$$

In our work, we compute the two-particle response function G^2 via the Lehman representation[46,47]:

$$G^{(2)imp}_{i\nu i\nu'}(i\Omega) = \frac{1}{Z}\sum_{ijkl}\sum_{\Pi}\Phi(E_i,E_j,E_k,E_l;\omega_{\Pi_1},\omega_{\Pi_2},\omega_{\Pi_3})\mathrm{sign}(\Pi)\langle i|\mathscr{O}_{\Pi_1}|j\rangle\langle j|\mathscr{O}_{\Pi_2}|k\rangle\langle k|\mathscr{O}_{\Pi_3}|l\rangle\langle l|c_\sigma|i\rangle \tag{24}$$

$$\Phi(E_i,E_j,E_k,E_l;\omega_1,\omega_2,\omega_3) = \int_0^\beta d\tau_1\int_0^{\tau_1} d\tau_2\int_0^{\tau_2} d\tau_3 e^{-\beta E_i+\tau_1(E_j-E_i)+\tau_2(E_k-E_j)+\tau_3(E_l-E_k)}e^{i(\omega_1\tau_1+\omega_2\tau_2+\omega_3\tau_3)}, \tag{25}$$

where we have introduced the closure relation, and define $O_1 = c_\sigma$, $O_2 = c^\dagger_\sigma$ and $O_3 = c_{\sigma'}$. E_i are the eigenvalues of the MLDE hamiltonian, Π are the triplet permutations, and the summation holds over all states of the Hilbert space.

The magnetic susceptibility is obtained as

$$\chi^{mag}(q, i\Omega_n) = \frac{1}{2} \sum_\sigma \chi(q, i\Omega_n)_{(i\nu,\sigma)(i\nu',\sigma)} - \chi(q, i\Omega_n)_{(i\nu,\sigma)(i\nu',-\sigma)}. \tag{26}$$

In principle, the minimization of the single particle leakage does not guarantee the minimization of the leakage for the two particle irreducible vertex. For the case we have studied, even without imposing a constraint on the two particle leakage, the agreement with CTQMC is good. We propose here a diagrammatic argument to support our findings, in particular, that MLDE also provides a good estimate of higher order response functions.

Firstly, the lowest order diagram contributing to the irreducible vertex is given by (see Ref.[48]):

$$\Gamma^{(1)}_{abcd} = U_{abcd}. \tag{27}$$

Since in MLDE, there are no direct many-body interaction terms in the Hamiltonian between the impurity and the bath, this diagram is equal to zero by construction for the cross term contribution. Furthermore, the MLDE non-local self-energy connecting the bath and the impurity can be expressed in term of the reducible vertex[49]:

$$\Sigma(14) = -U(12'3'1')G(1'4')G(23')G(2'3)F(4'234) \tag{28}$$

where F is the reducible vertex, which is related to the irreducible vertex by the Bethe-Salpeter equation

$$F(1234) = \Gamma(1234) + \Gamma(122'3')\chi^0(3'2'4'3')F(3'4'34). \tag{29}$$

Here we use the usual short-handed notations $1 = (\tau_1, l_1, \sigma_1)$ and assume sum over repeated indices. We now consider the case of $\Sigma(14)$ corresponding to the non-local self-energy that connects the impurity to the bath degrees of freedom. When the left-hand side of Eq. 28 is minimized in MLDE (reduction of the one-particle leakage), it naturally imposes that F remains small (for non-local contributions). Moreover, we see from Eq. 29 that minimizing F also imposes a minimization of the non-local terms in Γ, that connects the impurity to the bath. In summary, when the non-local self-energy is minimal, MLDE provides a good approximation of the irreducible vertex used throughout to calculate the dynamical magnetic susceptibility.

Finally, we emphasize that it is usually difficult to provide energy cutoffs on the low energy converged states of the Hamiltonian (typically in Lanczos or other iterative approaches), as the Boltzmann statistics only truncates one of the sums (on index i), whereas all eigenstates are required for the other summation. MLDE provides an ideal candidate for such calculations, as the number of bath sites can be kept small with very little cost in accuracy. We note indeed that the previous equation can be arranged to show a computational complexity growing with the cube of the number of eigenstates n, where n itself grows exponentially with the number of bath sites.

MLDE for Kondo molecule

The density of states (DOS) for different temperatures is shown in Extended Data Figure 6a. The fingerprint Kondo peak around zero energy can be seen at low temperatures. We emphasize that this feature is notoriously hard to obtain when using an ED solver as the formalism is affected by the small broadening factor needed to correctly capture this Kondo physics fingerprint. The Kondo temperature can be obtained from the relation[30,31] obtained from perturbation theory, which reads

$$T_K = -\frac{\pi Z}{4} \Im \Delta(i\omega_n)|_{\omega_n \to 0} \tag{30}$$

With the computed quasi-particle weight ($Z = 0.026$) and value of the hybridization in the limit of zero frequency (see Extended Data Figure 6.b), we obtain a Kondo temperature of $T = 33.8$ K, which is comparable with the experimental one at ≈ 37K[24]. We notice that temperature effect corrections to the Fermi liquid regime can be more easily studied in the Matsubara self-energy (see Extended Data Figure 6.b). In this representation, the quadratic behavior of the self-energy is apparent at low energy scales.

DFT+DMFT for $LaNiO_2$: Extension of MLDE to multiorbital

DFT+DMFT calculation is performed within Questaal package[50]. We used the local density approximation (LDA) functional with a k-mesh 10x10x10 points. The DMFT subspace is defined by the projection on Ni-d orbitals. The filled t_{2g} manifold is treated at the Hartree Fock level, and the e_g is treated using DMFT. In order to solve the Anderson impurity model composed of e_g orbital, we use MLDE with 2 impurity sites and 3 sites in the bath. The comparison with an exact solver is done using

CTQMC solver in TRIQS Package[51]. The interaction between the 2 e_g orbitals is given by the rotationally invariant Slater Hamiltonian

$$H_{int} = \frac{1}{2} \sum_{ijlk,\sigma\sigma'} U_{ijkl} c^{\dagger}_{i\sigma} c^{\dagger}_{j\sigma'} c_{k\sigma'} c_{l\sigma} \tag{31}$$

The tensor U_{ijkl} is given in term of Slater integral

$$U_{ijkl} = \sum_{k=0}^{2} \alpha_k^{ijlk} F^{2k} \tag{32}$$

Where α_k^{ijlk} are the Racah-Wigner numbers and F_k are the Slater integrals.

In the literature, we often use the parameter corresponding to the Hubbard interaction U and Hund's coupling J to express F^k with the relations

$$U = F^0 \tag{33}$$

$$J = \frac{F^2 + F^4}{14} \tag{34}$$

$$\frac{F^4}{F^2} = 10/16 \tag{35}$$

In this letter, we use the parameters $U = 7$ and $J = 1$ as suggested in[35].

MLDE quantum computing algorithm

In this section, we present a quantum algorithm to run MLDE on noisy intermediate-scale quantum (NISQ) computers. We perform a Jordan-Wigner (JW) transformation[38] to map the MLDE Hamiltonian (Eq. 12) to a qubit representation. After the transformation, the Hamiltonian is in the form of a linear combination of Pauli tensors. With the JW transformation, the required number of qubits is two times the number of sites in the system, the factor two being due to spin.

To compute the Green's function, we use a variational quantum eigensolver (VQE) based approach as presented in[36] since it is rather resilient to noise. The quality of the quantum computing solution corresponds largely to the ability of the quantum circuit to represent the eigenstates of H, and hence obtain accurate energies and amplitudes of the peaks in the Green's function. One, therefore, needs a state preparation ansatz able to accurately represent the ground state as well as the excited states with one electron added or removed[36]. For the results presented in the Results section, we use a so-called Hamiltonian variational ansatz (HVA)[52] to represent the eigenstates of H. Note that we obtain a similar accuracy also with different types of circuit ansatz, such as a so-called hardware efficient ansatz[53]. A general quantum state $|\psi(\boldsymbol{\theta})\rangle$ is prepared with a circuit ansatz as $|\psi\boldsymbol{\theta}\rangle = \hat{U}(\boldsymbol{\theta})\,|0\rangle$, where $|0\rangle$ is the initial state corresponding to all qubits set to the zero state, and $\hat{U}(\boldsymbol{\theta})$ is the unitary generated by the state preparation quantum circuit. Here $\boldsymbol{\theta}$ is a vector of parameters, which determine the specific state generated by the unitary. For the HVA $U(\boldsymbol{\theta})$ has the form

$$\hat{U}(\boldsymbol{\theta}) = \prod_{k}^{n_{\text{layers}}} \prod_{j} e^{-i\theta_j^k \hat{P}_j} \tag{36}$$

where P_j is a Pauli tensor in H after JW transformation and θ_j^k a real-valued parameter. The multiplication over j goes over all Pauli terms in the Hamiltonian. The ansatz corresponds to n_{layers} repetitions of individual blocks. In a given block, each Pauli tensor term in H, P_j, is included in the ansatz as imaginary exponentiation with the parameter θ_j^k, which can be readily implemented on a quantum computer[52]. The accuracy of the ansatz generally improves with increasing n_{layers}. We verified that for the system considered in the section MLDE on a quantum computer, $n_{\text{layesr}} = 2$ gives good accuracy.

To obtain the state preparation circuit parameters for the ground state, we minimize the cost function corresponding to the ground state energy

$$E_{\text{GS}} = \min_{\boldsymbol{\theta}} E(\boldsymbol{\theta}) = \min_{\boldsymbol{\theta}} \langle \psi(\boldsymbol{\theta}) | H | \psi(\boldsymbol{\theta}) \rangle \,. \tag{37}$$

To obtain the state preparation circuit parameters for the excited states there are several possibilities[54–56]. Here we use the method proposed in[55], where a penalty term is added to the Hamiltonian to impose orthogonality of each excited state to the lower energy states. To calculate the l^{th} excited state, $|\psi_l\rangle = |\psi(\boldsymbol{\theta}_l)\rangle$, with energy E_l, one therefore minimises

$$E_l = \min_{\boldsymbol{\theta}} \left[\langle \psi(\boldsymbol{\theta})|H|\psi(\boldsymbol{\theta})\rangle + \sum_{j=0}^{l-1} \alpha_j \left\langle \psi_j|\psi(\boldsymbol{\theta})\right\rangle^2 \right]. \tag{38}$$

Here the positive-valued α_j are arbitrary parameters, set in a way to optimize the convergence of the algorithm. Once all required state preparation parameters $\boldsymbol{\theta}_l$ are computed, the Green's function is obtained using the Lehman representation, and with it, the self-energy[36].

Data availability

Source data and code to generate the data for all the figures are available via Code Ocean[57].

Code availability

The code is available via Code Ocean[57].

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Author contributions statement

CW designed the research. CL and FJ equally contributed to the work. IR and FJ developed the quantum computing algorithm. All authors contributed to the code design, results production and analysis. All authors reviewed the manuscript.

Competing Interests Statement

The authors declare no competing interests.

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Figure 1. From AIM to MLDE. (a) Mapping of the lattice model (with inter-site hopping parameter t and on-site correlation U) onto a local impurity model, with a correlated atom (pink sphere) embedded in a non-interacting bath (blue shaded area) with energy ε as for the Anderson impurity model (AIM). The four possible configurations describe the quantum evolution of the atom. Electrons may hop from the atom to the bath via the frequency dependent hybridization function $\Delta(\omega)$, which plays the role of a dynamical mean field. b) Discretization of the continuum bath in non-interacting bath sites coupled to the interacting impurity through the hopping function V_i. This is the picture related to the ED impurity solver. c) Cartoon related to the Maximally Localised Dynamical Embedding (MLDE) solver where an interacting impurity is coupled to interacting bath sites.

Figure 2. MLDE benchmark and application on quantum computer. (a) Imaginary and (b) real part of the MLDE with $N_b = 3$ self-energy (dashed lines) obtained from on-site correlation U in range $2-15$ for a test hybridisation function for a half-filled impurity (units are arbitrary) and compared with continuous time Monte Carlo (continuous line). The scales are in arbitrary units. c) Quasi-particle weight obtained by MLDE with $N_b = 2$ and $N_b = 3$ compared to the continuous time Monte Carlo. The scales are in arbitrary units. d) Imaginary part of the self-energy as function of the Matsubara frequencies for the MLDE system presented in panel (b) and $U = 6$ ($N_b = 2$), computed using the quantum computing algorithm ("MLDE-VQE", blue solid curve), and compared to the results obtained with the conventional computing algorithm ("MLDE-exact", red dashed curves). $\mathfrak{Im}$ and $\mathfrak{Re}$ stand respectively for the imaginary and real part of the complex functions. The latter notations are used throughout the manuscript.

Figure 3. Kondo physics. (a) TOV organic molecule deposited on a gold substrate at $T = 5$K. (b) Imaginary parts of the molecule's hybridisation function (AIM), represented by MLDE $\Delta_{1,2}(i\omega_n)$, and as obtained by the ED solver with $N_b = 12$. (c) Imaginary part of the self-energy obtained by MLDE (squares) and ED (triangles). (d) Imaginary part of the self-energy in real axis frequencies obtained with MLDE and ED. (e) The inset shows the time dynamics of the magnetically quenched system, at a temperature $T = 100$K ($T = 20$K) above (below) the Kondo temperature ($T_K \approx 37$K). The main panel outlines the Fourier transform of the dynamics at $T = 100$K showing a peak at $\omega = 0.0029$eV ($= 33.64$ K).

Figure 4. Mott transition. (a) Spectral function obtained for the square lattice Hubbard model solved by MLDE ($N_b = 3$ for all calculations) with increasing values of the on-site dimensionless correlation ratio U/t. (b) Imaginary part of the self-energy obtained for $U/t = 12$ at different MLDE iteration of the self-consistent cycle compared with CQMC solutions. (c) Converged imaginary part of the MLDE Green's function for different values of U/t (symbols) compared with the DMFT CTQMC solution (dashed lines).

Figure 5. Dynamical susceptibility. Momentum resolved spin susceptibility obtained by Bethe Salpeter with the vertex calculated with MLDE in the Hubbard model with $U/t = 12$, at increasing temperature $T/t = 0.025$ (a), $T/t = 1$ (b), $T/t = 10$ (c). d) Magnetic susceptibility χ and e) irreducible vertex Γ resolved in fermionic frequency $i\nu$ obtained by MLDE (continuous line) and compared with the exact vertex (dashed line) at temperature $T/t = 0.025$ and $U/t = 6$.

Figure 6. MLDE multiorbital correlated $LaNiO_3$: Self energy of the first iteration of the self-consistent DMFT loop (panel a) and the Green's function for the final iteration (panel b) for the e_g orbitals of the Ni ion in the $LaNiO_3$ (d_{z^2} in red and $d_{x^2-y^2}$ in blue). In both case, we compare the quantities obtained with MLDE with 3 sites in the bath (solid line) with the quantities obtained with CTQMC (dashed line).

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