Solving quantum impurities with entanglement-based observables

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Quantum many-body systems exhibit fascinating many-body phenomena when the small interacting impurity changes the physics of a large noninteracting environment. The characterisation of such strongly-correlated non-perturbative effects is particularly challenging due to the infinite size of the environment, and the inability of local correlators to capture the build-up of long-ranged entanglement in the system. Here, we devise an entanglement-based observable – the purity of the impurity – as a witness for the formation of strong correlations. We showcase the utility of our scheme when exactly solving the open Kondo box model in the small box limit, corresponding to all-electronic dot–cavity devices. Thus, we characterise the experimentally-observed metal-to-insulator phase transition in the system and identify how the (conducting) dot-lead Kondo singlet is quenched by an (insulating) intra-impurity singlet formation. Our work showcases that quantum information observables serve as excellent order parameters for the characterisation of strong correlation, and motivates experimental observation of the purity of mesoscopic devices as entanglement witnesses.

Quantum many-body systems are an important and challenging frontier of physics due to the exponential size of their Hilbert space. The system’s ground state can be dominated by many-body interactions and thus profoundly removed from its simpler single-particle limit, i.e., nonlocal entanglement manifests between the system’s constituents. Correspondingly, finding the strongly-correlated ground state is equivalent to solving complex optimisation problems in the context of quantum annealing [1]. Alongside theoretical and numerical studies of quantum many-body systems, analogue quantum simulators serve as an alternative method to emulate strongly-correlated effects using experimentally controllable devices [2]. Examples of such simulators include trapped atoms [3] or ions [4, 5], light-matter systems [6], as well as electronic [7] or superconducting [8] mesoscopic devices.

Among strongly-correlated systems, quantum impurity problems hold a prominent position [9]. They engender a simple case where many-body interactions manifest only at a specific spot in space, i.e., at the quantum impurity. Nevertheless, by coupling the impurity to a large reservoir of noninteracting particles, strong correlations manifest in the whole system. Understanding quantum impurity problems is key to understanding quantum many-body systems as a whole. For example, in Dynamical Mean Field Theory (DMFT), quantum many-body systems are mapped to (more easily tractable) quantum impurity systems [10,11]. Quantum impurities also model a large class of quantum simulators, which are composed of confined regions of space embedded within a larger experimental environment.

Let us consider the Kondo effect, where a localised magnetic impurity is screened by an electron cloud, forming a so-called many-body Kondo singlet [12]. The effect arises in both strongly-correlated materials and mesoscopic quantum devices: in a bulk metal, it induces a decrease in the resistivity due to the pinning of the screening electrons around the impurity [13,14], while in mesoscopic quantum dots, it leads to an open transport channel and an enhancement in conductivity [15–18]. The latter conductivity peak has been detected in single- and multi-channel quantum dots already since the late ’90s [19,21]. Yet, only very recently the length scale of the macroscopic Kondo screening cloud was measured [22]. Note, however, that these observations correspond to concrete physical indicators for the formation of a Kondo singlet, but they do not directly measure the entanglement of the Kondo impurity with its environment. Current efforts focus on the direct characterisation of the amount of entanglement in Kondo singlets in quantum dots, e.g., by measuring the dot’s entropy [22,25].

The more complex the internal structure of a quantum impurity is, the broader the variety of many-body phenomena that can arise. An example of such a structured quantum impurity problem involves an electronic dot-cavity system [26]. The model’s impurity consists of a quantum dot that is coupled to an electronic cavity, i.e., to a discrete set of electronic noninteracting levels. The dot-cavity impurity is weakly coupled to three electron leads [26]. Note that the dot-cavity resembles a two-dot system [27,28], where one of the two dots is large and noninteracting, i.e., it is equivalent to a so-called Kondo box [29] in the large level-spacing limit. The dot-cavity model was used to describe the transport signatures of a mesoscopic device, that showcased a metal-to-insulator phase transition. The transition was postulated to arise when a dot-cavity molecular singlet forms and quenches the conducting Kondo effect of the dot [30]. Thus far, only approximate methods were used to describe the dot-cavity phase transition [26], leaving several open questions concerning the accuracy of the interpretation, as well as the role of entanglement in this system.

In this work, we report on an exact study of the dot-
cavity system and its nonlocal entanglement characteristics. We devise the purity of subparts of the structured impurity as entanglement witnesses to the formation of strong correlations in the system. Specifically, we use the purity of the dot and that of the dot-cavity to resolve whether the dot forms a many-body Kondo singlet with its leads or isolates from the environment by forming a singlet with the cavity. Our study, furthermore, showcases a numerical non-perturbative method for the study of quantum impurity systems that combines the strengths of both Numerical Renormalisation Group (NRG) and Matrix Product States (MPS) formalism. The former is used for mapping the infinite reservoirs to 1D chains, thus systematically truncating irrelevant degrees of freedom of the system. The latter efficiently delivers entanglement information in the system. Our results motivate the detection of the purity of subparts of a quantum system, as well as to an electronic cavity (red) with tunnel-coupling $\Omega$.

**Purity.** We are interested in the entanglement of a quantum impurity with its environment, see Fig. 1(a). As a measure for the entanglement, we will use its purity, together with the purity of its subparts. In a bipartite system $A \otimes B$, the purity $P_A$ of a subsystem $A$ (e.g., of the impurity) reads

$$P_A = \text{Tr} \left[ \rho_A^2 \right],$$

where $\rho_A = \text{Tr}_B (\rho)$ is the reduced density matrix of subsystem $A$ obtained by tracing out subsystem $B$ (the environment). In the Schmidt basis $\{|\alpha\rangle\}$ of $\rho_A$, with Schmidt coefficients $\lambda_n^2$, we can rewrite $\rho_A = \sum_n \lambda_n^2 \langle \alpha | \alpha \rangle$. As the trace is a basis-independent operation, the purity reads $P_A = \sum_n \lambda_n^4$ and is therefore directly related to the entanglement entropy of subsystem $A$, $S_A = - \sum_n \lambda_n^4 \ln (\lambda_n^2)$. Correspondingly, we can use it as a measure for entanglement: when $A$ is decoupled from its environment, then $P_A = 1$; if $A$ is fully mixed with the environment, then $P_A = 1/n$, where $n$ is the size of the Hilbert space of $A$, e.g., 1/4 for a single spinful electron site, and 1/16 for a system with two spinful electron sites. Crucially, when a singlet forms between $A$ and $B$, then the purity is $P_A = 1/2$.

**Model.** The effective Hamiltonian of the dot-cavity system reads [26, 30] (cf. Fig. 1(b))

$$H = H_{\text{dot}} + H_{\text{cav}} + H_{\text{coup}} + H_{\text{leads}} + H_{\text{tun}}.$$

The dot Hamiltonian $H_{\text{dot}} = \sum_{\sigma} \epsilon_{\sigma} n_{\sigma \downarrow} + U n_{\uparrow} n_{\downarrow}$ describes an Anderson impurity [34], with a spin-degenerate electron level at energy $\epsilon_0$ and electron-electron charging energy $U$, where $n_{\sigma \downarrow}$ denotes the dot's occupation number with spin $\sigma \in \{\uparrow, \downarrow\}$. The cavity Hamiltonian is $H_{\text{cav}} = \sum_{j \sigma} c_{j \sigma}^\dagger f_{j \sigma}^\dagger f_{j \sigma}$, where the index $j$ labels spin-degenerate electron levels with equally-spaced energies $\epsilon_j = \epsilon_{\text{cav}} + j \delta$ with spacing $\delta$, and $f_{j \sigma}^\dagger$ ($f_{j \sigma}$) is the fermionic cavity creation (annihilation) operator of the $j$th level. Note that due to the screening within the large spatial extent of mesoscopic electronic cavi- ties [26, 30], no electron-electron repulsion term is introduced in $H_{\text{cav}}$. The dot and cavity are tunnel-coupled $H_{\text{coup}} = \sum_{j \sigma} \Omega_j d_j^\dagger f_{j \sigma}^\dagger + \text{H.c.}$, with energy-dependent coupling amplitudes $\Omega_j$. The environment is composed of three leads $H_{\text{leads}} = \sum_{\ell \sigma} \epsilon_{\ell \sigma} k_{\ell \sigma}^\dagger k_{\ell \sigma} + \sum_{\kappa \sigma} \epsilon_{\kappa \sigma} c_{\kappa \sigma}^\dagger c_{\kappa \sigma}$, where we denote $c_{k_{\ell \sigma}}$ ($c_{\kappa \sigma}^\dagger$) the fermionic annihilation (creation) of an electron with momentum $k$ and spin $\sigma$ in the leads left and right to the dot; $\ell \in \{L, R\}$. The $c_{k_{\ell \sigma}}$ ($c_{\kappa \sigma}^\dagger$) operator acts on the cavity lead and is defined analogously. The tunnelling Hamiltonian term $H_{\text{tun}} (H_{\text{tun}} = H_{\text{tun}} + H_{\text{tun}})$ couples the closed dot-cavity system to the leads. We assume that the dot is coupled to the left and right leads, with energy-independent tunnelling amplitudes $t_{\text{tun}}$; $H_{\text{tun}} = \sum_{\ell \sigma} t_{\ell \sigma} d_{\ell \sigma}^\dagger c_{\ell \sigma} + \text{H.c.}$.

Similarly, the cavity is tunnel-coupled to its own lead as $33$ $H_{\text{cav}} = \sum_{\ell \sigma} t_{\ell \sigma}^\dagger d_{\ell \sigma} c_{\ell \sigma} + \text{H.c.}$ with an energy-independent tunnelling amplitude $t_{\text{tun}}$.

As discussed in Ref. [26], the closed dot-cavity system, i.e., Eq. (2) with $t_{\ell \sigma}, t_{\text{tun}}, t_{\text{tun}}=0$, forms an “artificial molecule” impurity. Specifically, as a function of dot and cavity energies (set experimentally by tuning voltage gates), it exhibits a charge stability diagram with re-
regions dominated by Coulomb or exchange blockade, separated by resonance lines where the particle number on the molecule is ill-defined, see Fig. 1(c). The exchange blockade region involves the formation of a dot-cavity singlet, when sufficiently-large level spacing in the cavity δ ≫ Ω / U is taken.

NRG-MPS for quantum impurities. Coupling the dot-cavity molecule to its leads can change the occupation of the molecule and transport through it. Indeed, in experiments [31], Kondo transport is observed, separated by exchange-blockade regions whenever the cavity couples strongly to the dot’s electron spin. Until now, however, signatures of strong correlation with the leads were derived only via approximate methods, e.g., with a generalisation of the Meir-Wingreen formula for non-proportional coupling [29], via a perturbative treatment in $H_{\text{tun}}$, or by using a Green’s function equations of motion approach [26]. Here, we treat the system-environment couplings exactly. Instead, we approximate the environments by 1D Wilson chains [36, 37], and we explore the properties of the open dot-cavity system using an MPS formalism [38], denoted by open rounded squares. As a next step, we map the remaining P lead and its tunnel-coupling to a 1D semi-infinite Wilson chain [36] [see Fig. 2(a)]

$$H_{\text{chain}} = \sum_{\sigma} \tau_{\text{d}} d_{\text{d} \sigma} c_{n \sigma} + \sum_{n=1}^{\infty} \sum_{\sigma} t_{dn} c_{n-1 \sigma} c_{n \sigma} + H.c.,$$

where $c_{n \sigma}$ is the annihilation operator of a fermion with spin σ at site n of the P-chain. Similarly, we transform the terms including the cavity lead into a Wilson chain $H_{\text{cav}} = \sum_{\sigma} \tau_{\text{c}} f_{\text{b} \sigma} + \sum_{n} \sum_{\sigma} \tau_{\text{c} \sigma} b_{n \sigma} b_{n+1 \sigma} + H.c.,$ where $b_{n \sigma}$ is the annihilation operator in the cavity lead defined analogously to $c_{n \sigma}$. From the Wilson transformation [36], the nearest-neighbour hopping coefficients along the chain are exponentially decaying in the chain site number $t_{dn} \propto \Lambda^{-n/2}$ for $n \gg 1$.

An impurity problem with environments that are transformed into Wilson chains is commonly studied with the NRG technique [36, 40, 41]. Note that in the standard NRG procedure, the transformed 1D environment is slowly added to the system by iteratively attaching chain sites to the finite chain. Additionally, as the Wilson transformation involves a different energy scale at each added site, the length of the studied chain (iteration depth) encodes the effective temperature (or resolution) of the resulting ground state. Correspondingly, the order in which sites are added to the chain, in a multi-terminal case like ours, may strongly impact the NRG results. In our work, we instead calculate equilibrium properties of a finite chain with a Variational algorithm for MPS (VMPS) [31, 32]. The VMPS algorithm treats each site identically, thus leading to more stable and reliable results. As sites beyond some point in the Wilson chain become marginal [37], we truncate both the dot and cavity leads at a finite length N (here $N=40$), obtaining a 1D finite chain that approximates the open dot-cavity system. Nevertheless, recall that each chain site has a $d=4$ dimensional Hilbert space with empty $|0\rangle$, full $|\uparrow\rangle$, or singly occupied $|\uparrow\downarrow\rangle$ basis states. Therefore, the number of coefficients required to characterise a pure state of the chain grows as $d^N$, hitting the current computational limits at relatively small N. On this account, we work with MPS to characterise states and operators [Fig. 2(b)], which allows for a controlled truncation of the number of required coefficients based on the entanglement information between different chain sites. This truncation harnesses a Singular Value Decomposition (SVD) compression that caps the maximal number of Schmidt values describing each link in the chain, dubbed bond dimension D. Only thus, are we able to reach chains of length $N_{\text{tot}} \geq 80$ sites [32].

Note that even though our method does not treat the impurity-environment coupling perturbatively, its performance depends on the system parameters. Specifically, the ability of the VMPS to find the correct strongly-correlated ground state depends both on the finite chains’ lengths (as in NRG due to the Wilson transformation) and the MPS truncation. As the impurity-environment coupling becomes smaller, the required resolution to find
The Kondo ground state increases, and thus also the required chain length and bond dimension. In the following, we establish that the purity is a good measure for observing the formation of the strongly-correlated Kondo singlet. Hence, per system parametrisation, we systematically explore convergence of our variational ground-state search by following the dot’s purity $P_d$ as a function of the two truncation knobs $\delta$. Furthermore, we observe that it is sufficient to consider a cavity with a single spinful energy level $\epsilon_c=\epsilon_{cav}$, in the experimentally-relevant regime where $\delta \gg 1292/U$.

**Results.** Having obtained the converged ground state of the whole system (impurity plus environment) per parametrisation, we start by revisiting the charge stability diagram, see Fig. 3(a). The occupation $n_{tot}=(n_d+n_c)$ of the dot-cavity subsystem (evaluated on the obtained ground state) qualitatively matches that obtained by the exact diagonalisation treatment of the closed dot-cavity system. Crucially, we observe a region competing with the dot’s Coulomb blockade regimes, where due to the dot-cavity tunnel-coupling, the cavity has an odd occupation. This hints towards the fact that in this region a dot-cavity spin-singlet forms (exchange-blockade) also in the open system. Quantitatively, we observe that the coupling to the environment shifts the charge instability lines to slightly higher energies, as expected from (self-energy) renormalisation of the closed systems’ levels by high-order coupling to the environment.

To verify the formation of a dot-cavity spin-singlet and the existence of an exchange blockade region, we analyse the spin configuration of the dot-cavity molecule. To this end, we define and calculate their spin-spin correlation

$$C(d, c) = \frac{1}{2} \sum_{\sigma} \langle n_{d\sigma} \rangle \langle n_{c\bar{\sigma}} \rangle - \langle n_{d\sigma} n_{c\bar{\sigma}} \rangle,$$

which returns 1 (-1) when the spins on the dot and cavity are aligned (anti-aligned). We plot the resulting $C(d, c)$ in Fig. 3(b): (i) when the dot has an even occupation, no dot-cavity spin-spin correlations appear; (ii) when the dot has an odd occupation and the cavity has an even occupation, we observe weak spin-spin alignment. In this region, we expect that the dot anti-aligns with the lead electrons (Kondo singlet). The ensuing strong dot-lead correlation leaves space for the cavity electrons (holes) to partially redistribute into the leads in the full (empty) cavity level configuration. The spin of these residual particles tends to align with the spin of the electron occupying the dot [33]. (iii) In the (1,1) occupation region, we observe strong spin anti-alignment behaviour, in agreement with the closed system exact diagonalisation results. The latter clearly confirms the hypothesised formation of a molecular dot-cavity singlet [26, 30].

The results presented so far do not identify signatures of strong coupling to the environment. To observe these, propose to use quantum information inspired observables, namely the dot and dot-cavity purities $P_d$, $P_{dc}$, see Figs. 3(c) and (d), and cf. Eq. 1. In MPS formalism, the purity of a subsystem composed of a few sites is readily calculated with a few matrix multiplications [33]. Combining the information from both purities, we can deduce much about the correlation in the full system. For example, in region (i) both $P_d \rightarrow 1$ and $P_{dc} \rightarrow 1$, i.e., the electrons on the dot is decoupled from all lead and cavity electrons, and the combined dot-cavity electrons are also decoupled from the lead electrons. This decoupling highlights that in region (i), the ground state does not show strong correlations between the impurity and the environment. Similarly, in region (iii), the dot-cavity “molecule” decouples from the leads ($P_{dc} \rightarrow 1$). Yet a $P_d \approx 1/2$ attests that the dot electron is in a spin-singlet configuration with its cavity counterpart. Crucially, in region (ii), $P_d \approx P_{dc} \approx 1/2$ marks that the dot forms a singlet, but that this singlet is not affected by the inclusion of the cavity’s Hilbert space, i.e., we have a clear evidence for the formation of a Kondo singlet between the
dot and lead electrons. Note that we modelled the quantum dot with an Anderson Impurity Hamiltonian instead of the Kondo Hamiltonian considered in Ref. 32, i.e. we did not a priori assume the formation of Kondo effects. Thereby, our method does not merely verify the Kondo singlet formation for the Anderson model; it additionally fully characterises the phase transitions to other many-body entanglement regimes in the structured impurity of the open Kondo box.

Our work showcases three important milestones: (a) the power of NRG-MPS approaches for solving multi-channels quantum impurity problems as insinuated in Refs. 31, 32, (b) the applicability of quantum information based observables for studying strongly-correlated systems, and (c) the conclusive demonstration of the physics behind the dot-cavity metal-to-insulator transition. Moreover, our approach highlights how the purity of different subparts can be harnessed to study complex quantum impurity systems. Employing minimal and sufficient entanglement witnesses (such as the purities proposed here) in experiments is an important next step for the quantum simulation field, where tomography of small subsystems is readily accessible. Future work will explore the generalization of our approach to out-of-equilibrium and mixed state systems.

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1. Convergence behaviour of the NRG–MPS technique

The quality of a Matrix Product State (MPS) or operator (MPO) representation is affected by the extent of the singular value decomposition (SVD) truncation, namely by choosing the bond dimension \(D\). There is no general rule for determining the minimal \(D\) that ensures a converged physical result. Hence, trials with different bond dimensions are performed. Convergence properties of physical observables might indicate a minimal bond dimension that is still able to capture the physics of the system. Similarly to the MPS bond dimension, also the length of the Wilson chain \(N\) must be sufficiently large in order to observe the formation of a Kondo singlet, see discussion in the main text. We checked the results of the dot-cavity model presented in the main text for convergence. For example, in Fig. S1(a), we observe a strong dependence of the dot purity \(P_d\) on the bond dimension. At high dot-lead coupling \(t_d/U \gg 0\), the dot fully mixes with the environment, and \(P_d \approx 1/4\) is minimal for any bond dimension. For lower coupling strengths, the purity gradually increases to approximately 1/2, marking the formation of a Kondo singlet. The jumps in \(P_d\) observed in the 0.2\(U < t_d < 0.3U\) range showcase the aforementioned limitation of the MPS method: a finite MPS bond dimension is not sufficient to capture the physics of the system. Indeed, a purity value indicating the presence of a dot-lead Kondo singlet is expected for any \(t_d\) in the low coupling regime.

In Fig. S1(b), we repeat the convergence analysis of the \(P_d\) for varying Wilson chain length \(N\) for both the dot and cavity leads. We see that the weaker the dot-lead tunnel-coupling is, the longer \(N\) has to be to capture the Kondo-associated entanglement features established in the system. This is a known requirement in NRG since the Wilson chain relates to the energy resolution of the lead. A finite chain truncates the information corresponding to coupling with lead electrons near the Fermi level. These low-energy electrons are the ones dominating the hybridisation with the dot in the low tunnel-coupling regime.

Following our calibration, in the results presented in the main text, we set the dot-lead tunnelling amplitude at \(t_d=0.25U\), as well as use MPS bond dimension \(D=100\) and an NRG chain length \(N=40\) for both the dot and cavity leads, i.e., where the purity value is close to 1/2, and the expected dot-lead Kondo singlet is detected.

2. Truncation to a single cavity level

In the main text, we define and discuss the cavity Hamiltonian \(H_{cav}\), whose spin-degenerate electron levels \(\epsilon_{c}^{(j)}=\epsilon_{cav}+j\delta\) have constant spacing \(\delta\). For the presented results, however, we suffice to include only a single cavity level. In this section, we check the validity of this approximation. In Fig. S2, we compare the outcome of our VMPS analysis between a dot-cavity system with a single-level cavity and a two-level one. We perform the comparison for a dot located at \(\epsilon_{d}=-U/2\), where the competition between Kondo and dot-cavity singlet formation occurs.

In Figs. S2(a) and (b), we show that for \(\epsilon_{c}>-U/2\) the single-level cavity exhibits the same physics as the two-level cavity. As discussed in the main text, at \(\epsilon_{c}=0\) [cf. region (i)], the cavity is approximately occupied by a single electron and the dot-cavity molecule decouples from the environment \([P_{d}c_{\downarrow}\rightarrow 1]\). For a cavity approximately empty or full, the purities are \(P_{d} \approx P_{d\downarrow} \approx 1/2\). Hence, we conclude that the cavity does not impact the many-body strong hybridisation between the dot and its environment, namely the dot-lead Kondo singlet.

A different picture emerges for \(\epsilon_{c}<-U/2\). In region (ii), for the single-level cavity case, the occupation \(\langle n_{c}\rangle=2\) saturates and the system remains in the dot-lead...
singlet configuration. For the two-level case, a third electron occupies the cavity \( \langle n_c \rangle >2 \). At \( \epsilon_c \approx -U \), \( P_d \) and \( P_{dc} \) of the two-level cavity [cf. region (iii)] are equal to the \( P_d \) and \( P_{dc} \) at \( \epsilon_c \approx 0 \) [cf. region (i)]. Thus, in the \( \langle n_c \rangle \approx 3 \) configuration the unpaired cavity electron couples to the dot, exactly as in the \( \langle n_c \rangle \approx 1 \) configuration, and establishes a molecular dot-cavity singlet quenching the Kondo effect.

With these results, we conclude that it is sufficient to consider a cavity with single spinful energy level \( \epsilon_c = \epsilon_{cav} \) in the \( \epsilon_c \in [-U/2, U/2] \) interval for characterising the many-body physics of the electronic dot-cavity impurity. Indeed, in this interval, the physics of the single- and multi-level cavity is the same. Additionally, the two-level cavity for \( \epsilon_c < -U/2 \) manifests the same phenomena as in the \( \epsilon_c \in [-U/2, U/2] \) interval. Thus, the physics in the multi-level cavity case only depends on the presence of an unpaired electron in the cavity, regardless of the number of occupied cavity levels.

3. Dot-cavity alignment in the Kondo regime

As discussed in the main text [cf. Fig. 3 of the main text, region (ii)], whenever the dot and lead form a Kondo singlet, we observe a limited dot-cavity spin alignment, see also Fig. S3(a), region (ii). This effect coexists with the strong dot-lead hybridisation and implies that the cavity occupation is not perfectly quantised. A necessary condition for the dot and cavity spin to align is that the cavity is not perfectly full (empty) for positive (negative) \( \epsilon_c \). Such behaviour is corroborated by our calculation of \( \langle n_c \rangle \), see Fig. S3(b). The cavity electrons (holes) could redistribute into the leads, the dot, or both.

In Fig. S3(b), we observe that the dot occupation is constant in the (ii) region. Thus, in the Kondo dot-lead configuration, the cavity electrons (holes) partially redistribute into the leads in the full (empty) cavity configuration. In the Kondo singlet, a continuous second-order scattering of lead electrons near the Fermi level occurs. Thus, for a dot electron in the \( \sigma \) spin configuration, a lead electron in \( \bar{\sigma} \) scatters in and out of its energy level \( |\bar{\psi}\rangle \), leaving the latter free for a fraction of time. When the cavity is tuned near the Fermi level, the \( \bar{\sigma} \) cavity electron (hole) partially redistributes in the free lead level. The \( \bar{\sigma} \) cavity electron (hole) level is therefore partially empty. Hence, dot and cavity showcase a marginal spin-spin alignment behaviour. Our results show that the NRG-MPS method is capable of identifying such higher-order processes.

4. Purity calculation in MPS formalism

In this section, we show how to readily calculate the purity of a subsystem using a MPS formalism. The density matrix \( \rho_A \) of a subsystem \( A \) is defined as

\[
\rho_A = \text{Tr}_B(\rho_{AB}) = \sum_k (I_A \otimes \langle \sigma_k |_B ) \rho_{AB} (I_A \otimes | \sigma_k \rangle_B ) ,
\]

(S1)

where \( \rho_{AB} \) is the density matrix of the full system \( A \otimes B \), \( I_A \) is the identity operator on \( A \), and \( \{ | \sigma_k \rangle_B \} \) is an orthonormal basis of \( B \). In our work, we deal with density matrices of a 1D discrete system (chain), whose pure state can be written in Fano form

\[
|\psi\rangle = \sum_\sigma c_{\sigma_1,\sigma_2,...,\sigma_N} |\sigma_1 \rangle \otimes |\sigma_2 \rangle \otimes ... \otimes |\sigma_N \rangle ,
\]

(S2)

where \( \{ | \sigma_i \rangle \} \) is a basis of the Hilbert space of the \( i^{th} \) chain site, and \( c_{\sigma_1,\sigma_2,...,\sigma_N} \) are the amplitudes of the specific tensorial basis state. The reduced density matrix of, e.g., the subsystem composed of the first chain reads

\[
\rho_1 = \text{Tr}_{\text{env}}(\rho) = \sum_{\sigma_2,...,\sigma_N} (I_1 \otimes (|\sigma_2 ... \sigma_N \rangle \langle \psi | \langle I_1 | \sigma_2 ... \sigma_N \rangle)) ,
\]

(S3)
where with $env$ we indicate the environment $|\sigma_2\rangle \otimes \ldots |\sigma_N\rangle \equiv |\sigma_2, \ldots, \sigma_N\rangle$ and $I_1$ the identity operator of the 1st site. In MPS formalism, the pure state as in Eq. (S2) is expressed as

$$|\psi\rangle = \sum_{\sigma} M^{\sigma_1} \ldots M^{\sigma_N} |\sigma_1\rangle \otimes |\sigma_2\rangle \otimes \ldots \otimes |\sigma_N\rangle,$$  (S4)

where $\{M^{\sigma_i}\}$ is a set of matrices (the MPS matrices) characterising the $i$th site, see Ref. [38] for a detailed discussion. The calculation of $\rho_1$ in MPS formalism is particularly efficient if the chain is of the so-called “orthonormal” form [38]. Under this condition, the MPS sets of matrices $i = 2, \ldots, N$ satisfy the orthogonality properties $\sum_{\sigma_i} (M^{\sigma_i})^\dagger M^{\sigma_i} = I_i$, and the calculation of $\rho_1$ reduces to

$$\rho_1 = \sum_{\sigma_1, \sigma_1'} M^{\sigma_1} \left( M^{\sigma_1'} \right)^\dagger |\sigma_1\rangle \langle \sigma_1'|.$$  (S5)

Analogously, the density matrix of the $i$th site of the chain reduces to

$$\rho_i = \sum_{\sigma_i, \sigma_i'} M^{\sigma_i} \left( M^{\sigma_i'} \right)^\dagger |\sigma_i\rangle \langle \sigma_i'|,$$  (S6)

from which the square of the density matrix reads

$$\rho_i^2 = \sum_{\sigma_i, \sigma_i', \sigma_i''} M^{\sigma_i} \left( M^{\sigma_i'} \right)^\dagger M^{\sigma_i''} \left( M^{\sigma_i''} \right)^\dagger |\sigma_i\rangle \langle \sigma_i'|.$$  (S7)

The purity $P_i$ of the $i$th chain site in MPS formalism therefore reads

$$P_i = \text{Tr}(\rho_i^2) = \sum_{\sigma_i, \sigma_i'} M^{\sigma_i} \left( M^{\sigma_i'} \right)^\dagger \left[ M^{\sigma_i} \left( M^{\sigma_i'} \right)^\dagger \right]^\dagger.$$  (S8)