

Nonequilibrium quantum transport framework for spintronic devices with dynamical correlations

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Two-terminal spintronic devices remain challenging to model under realistic operating conditions, where the interplay of complex electronic structures, correlation effects, and bias-driven nonequilibrium dynamics may significantly impact charge and spin transport. Existing *ab initio* methods either capture bias-dependent transport but neglect dynamical correlations or include correlations but are restricted to equilibrium or linear-response regimes. To overcome these limitations, we present a framework for steady-state quantum transport, combining density functional theory (DFT), the nonequilibrium Greens' function (NEGF) method, and dynamical mean-field theory (DMFT). The framework is then applied to Cu/Co/vacuum/Cu and an Fe/MgO/Fe tunnel junction. In Co, correlations drive a transition from Fermi-liquid to non-Fermi-liquid behavior under finite bias, due to scattering of electrons with electron-hole pairs. This leads to incoherent contributions to the conductance that are observable in scanning tunneling spectroscopy experiments. In contrast, in the Fe/MgO/Fe junction, correlation effects are weaker: Fe remains close to equilibrium even at large biases. Nevertheless, inelastic scattering can still induce partly incoherent transport that modifies the device's response to the external bias. Overall, our framework provides a route to model spintronic devices beyond single-particle descriptions, while also suggesting new interpretations of experiments.

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I. INTRODUCTION

Spintronics exploits spin-dependent transport phenomena in materials, with applications in logic and data storage [1]. Typical devices are heterostructures formed by stacking thin magnetic and nonmagnetic layers. Among these, magnetic tunnel junctions (MTJs) [2–5] exhibiting tunneling magnetoresistance (TMR) [6–8] are already standard components in commercial read heads for hard-disk drives. However, the computational description of spintronic devices remains generally challenging due to their complex electronic structure, their open nature—allowing carriers to flow in and out of the active region—and the inherently nonequilibrium character of transport.

Ab initio studies typically rely on Kohn-Sham (KS) density functional theory (DFT) [9–11], within either the local spin density Approximation (LSDA) [12,13] or the generalized gradient approximation (GGA) [14–16] for the exchange-correlation functional, combined with the Landauer-Büttiker

(LB) approach [17–19] to quantum transport. In this framework, a device is treated as a central open region connected to electrodes that act as charge reservoirs, as shown in Fig. 1(a). The spin-dependent conductance is determined by the transmission of spin-polarized KS single-particle states from one electrode, through the central device region, to the opposite electrode [20]. Practical implementations include transfer-matrix methods [21,22], layer Korringa-Kohn-Rostoker approaches [23], and mode-matching techniques [24,25].

The most versatile and widely adopted implementation, however, is based on the nonequilibrium Green's function (NEGF) formalism [26–28] in its steady-state formulation, and is implemented using a variety of numerical approaches, including localized atomic-orbital bases [29–32], real-space finite-difference schemes [33], and exact or linear muffin-tin orbitals [34,35]. Commonly referred to as DFT + NEGF, this method can be naturally extended to the calculation of transport properties at finite bias, thereby going beyond the linear-response regime assumed in the original LB approach. As a result, it provides a device's current-voltage (*I*-*V*) characteristics [36–40] as well as other voltage-dependent observables, such as the spin-transfer torque [41–44].

DFT + NEGF relies on the assumption that the KS states represent a reasonable approximation of the excitation spectrum of a system. However, beyond issues of formal consistency [45], this assumption often fails even in practice, particularly for the 3d ferromagnets (Fe, Ni, Co, and related

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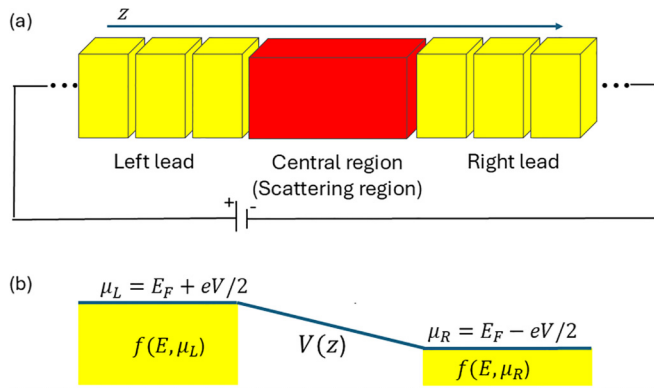


FIG. 1. Schematic illustration of a typical two-terminal transport setup. (a) A two-terminal spintronic device is modeled as infinite along the transport direction and partitioned into a central (scattering) region contacted by semi-infinite metallic leads. (b) Each lead acts as an electronic reservoir. Under a finite bias voltage V , the leads individually remain in equilibrium, with their electronic states occupied according to the Fermi-Dirac distribution functions, $f_{L(R)}(E, \mu_{L(R)})$, determined by the relative lead chemical potential $\mu_{L(R)}$. The electrostatic potential $V(z)$ drops inside the central region.

compounds) widely used in spintronic devices. Comparison with photoemission experiments shows that KS-DFT overestimates the spin splitting of the $3d$ states and produces majority-spin spectra that are too broad [46–49]. Moreover, it neglects finite-lifetime effects arising from electron-electron interaction, which broaden the electronic bands in both energy and momentum [46,50]. Finally, the phase-coherent picture of transport in terms of electron transmission may break down due to electron correlation effects. For instance, features observed in MTJs’ I - V characteristics have been interpreted as signatures of inelastic electron-electron or electron-magnon scattering [51–54]. While such effects can be included phenomenologically in DFT + NEGF, for example, via Büttiker probes [55,56], a truly *ab initio* treatment requires going beyond KS-DFT.

An improved description of the electronic structure of $3d$ metals can be achieved by combining DFT with dynamical mean-field theory (DMFT), in the so-called DFT + DMFT method [57–62]. Through the incorporation of dynamic, yet local-in-space, correlations, DMFT corrects the overestimation of spin-splitting and yields spectral properties in closer agreement with experiment [47,48,63–65]. Moreover, DMFT provides a true *ab initio* treatment of quasiparticle lifetimes and naturally captures incoherent effects arising from electron-electron scattering.

DMFT can be applied to study transport in devices using a layered formulation [66–73] in the linear-response regime. It can then be combined with DFT for *ab initio* calculations [74–78], building on embedding frameworks originally developed for point contacts [79–81] and molecular junctions [82–88]. The idea is to apply DMFT to a “correlated region,” defined within the central device region and hosting significant electron-electron interaction. Recently, these DFT + DMFT implementations for transport have also been generalized to “quasiequilibrium” conditions [65], valid when the voltage drop, albeit large in the device, remains small within the correlated region.

Genuine nonequilibrium conditions can be tackled by formulating DMFT within the NEGF framework using the Keldysh formalism [89–91]. Existing implementations, however, are typically designed for model bulk systems and focus on transient electron dynamics in the time domain. Studies of steady-state transport at finite bias in a device setup remain scarce and are generally limited to single-orbital models, employing ansätze for the Keldysh component of the many-body self-energy [92,93], workflows that compute only the retarded self-energy [94], or master-equation solvers [95]. Implementations for nonequilibrium bias-driven transport through realistic materials, described by multiple orbitals per atomic site, are still lacking, leaving electron correlation effects in current-carrying spintronic devices largely unexplored.

In this paper, we present a computational framework combining DFT, DMFT and NEGF in the steady-state regime to study transport properties of devices beyond the linear-response limit or quasiequilibrium ansätze adopted in previous DFT + DMFT transport implementations [65]. Our approach, that we called DFT + NEGF + DMFT, enables us to capture correlation effects induced by a finite bias, even in systems where a significant portion of the voltage drop occurs within the correlated region, leading to inherently nonequilibrium many-electron behavior.

Our framework is applied to two prototypical systems. The first one consists of a single Co monolayer sandwiched between Cu electrodes, with a gap introduced between one electrode and the Co layer to allow for a potential drop under bias. Our results demonstrate that nonequilibrium correlations in Co drive a qualitative transition from Fermi-liquid to non-Fermi-liquid behavior, arising from the inelastic scattering of electrons with collective electron-hole excitations. This reshapes the DFT-KS electronic structure and gives rise to incoherent contributions to the conductance, that can be measured through scanning tunneling spectroscopy (STS).

The second system represents the active part of an Fe/MgO/Fe MTJ. Here, the applied bias is found to have a much weaker effect on Fe, validating the quasiequilibrium approximations employed in previous studies [65]. Nevertheless, even in this limit, bias-induced inelastic scattering generates incoherent contributions to the current, which impact the device transport properties and performance.

The paper is organized as follows. In Sec. II, we introduce the method and describe its implementation. Computational details are provided in Sec. III. Results are presented in Sec. IV, focusing first on the Co/Cu system in Sec. IV A, and then on the Fe/MgO/Fe MTJ in Sec. IV B. Finally, we draw our conclusions in Sec. V.

II. METHOD AND IMPLEMENTATION

Our framework applies to the standard setup commonly used in quantum transport simulations of two-terminal devices under an applied bias voltage, as described in Sec. II A. It is formulated within the NEGF formalism in the steady-state limit, briefly reviewed in Sec. II B, where the relevant notations are defined. The implementation then proceeds through several key steps, as follows.

At the first level, the method relies on DFT + NEGF calculations to provide an effective single-particle description

of the device electronic structure, namely a static mean-field treatment under bias. This standard methodology is summarized in Sec. II C. From this starting point, we employ an embedding algorithm to extract a correlated subsystem via a projection scheme, detailed in Secs. II D 1 and II D 2. Within this subsystem, electron-electron interactions are treated beyond the static mean-field level. Conceptually, our embedding procedure can be viewed as a generalization of the band down-folding approaches commonly employed in bulk DFT + DMFT (e.g., Ref. [96]), adapted here to treat devices.

The central development of this work is the introduction of a practical finite-bias DMFT scheme that extends the equilibrium approach of Refs. [64,65,74] to nonequilibrium conditions, as explained in Sec. II D 3. This is achieved by employing a suitable nonequilibrium impurity solver (Sec. II D 6), which provides the many-body self-energies of the correlated subspace under bias. The self-energies are then embedded back into the full system (Sec. II D 5), enabling the calculations of observables such as the charge density, spectral functions, and in particular the steady-state current through the device (Sec. II E).

The framework is implemented in the SMEAGOL quantum transport package [32], which is interfaced with the SIESTA DFT code [97,98].

A. System setup

A two-terminal spintronic device, such as an MTJ, is modeled as infinite along the transport (z by convention) direction and divided into a central scattering region connected to semi-infinite metallic leads on the left- (L) and right- (R) hand side [28], as shown in Fig. 1(a). Periodic boundary conditions are applied in the transverse x - y plane, with transverse wave vector $\mathbf{k} = (k_x, k_y)$ spanning the two-dimensional Brillouin zone (BZ).

The partition into leads and the central region is implemented by using a linear combination of atomic orbitals (LCAO) basis set, in which each orbital is assigned to a specific part of the device. These basis orbitals are generally nonorthogonal. In principle, changes in the electronic structure of the central region feed back into the leads and vice versa, giving rise to interface effects. To capture these effects, a sufficient portion of each lead is included within the central region itself. As illustrated in Fig. 2(a), the formal lead/central region boundary is therefore not placed at the physical interface but rather a few layers deeper inside the leads, where the electronic structure has already relaxed to its bulk configuration.

The central region constitutes the active part of the device. In quantum transport calculations, it is typically assumed that electronic correlation effects are confined to this region, while the leads are treated within an effective single-particle description [26,99,100]. Thus each lead acts as an electronic reservoir, characterized by its chemical potential, $\mu_{L/R}$, and inverse temperature, $\beta_{L/R}$. When both leads have identical chemical potentials and temperatures—that is, $\mu_L = \mu_R = E_F$ and $\beta_L = \beta_R$ —the system is in thermodynamic equilibrium, described by the grand canonical ensemble, with E_F denoting the common Fermi energy.

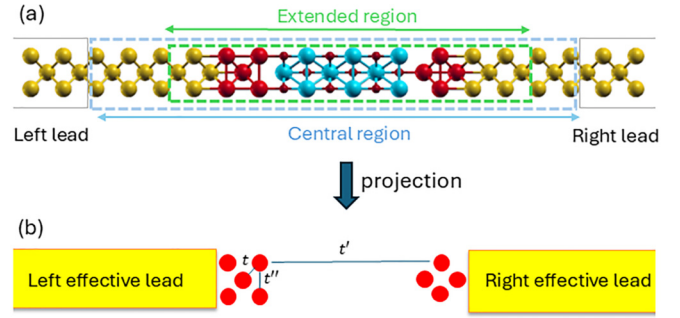


FIG. 2. The device setup used in this work. (a) The Fe/MgO/Fe junction sandwiched between simple metallic electrodes, introduced in Sec. IV B, shown as a prototypical example of a two-terminal device for DFT + NEGF calculations. Fe, Mg, and O atoms are represented by large red, cyan, and small red spheres, respectively, while the lead atoms are shown as yellow spheres. The Fe atoms define the correlated subspace. The formal lead/central region boundary is placed a few layers deeper inside the leads to ensure that interface effects are considered and that the correlated subspace does not interact directly with the leads, as required for Eq. (19) to hold. The “extended region” is indicated by the green dashed box. Atoms outside this region are not coupled to $\mathcal{C}$ due to the finite spatial extent of the basis orbitals. (b) The projection maps the original device onto an effective one, with renormalized hopping between correlated atoms and effective coupling to the leads.

The effect of a finite bias voltage, V , is modeled by shifting the chemical potentials of the leads such that $\mu_L = E_F + \frac{eV}{2}$ and $\mu_R = E_F - \frac{eV}{2}$, where e is the electron charge. Because of the metallic nature of the leads and the requirement of local charge neutrality, this shift corresponds to a rigid displacement of their entire band structures. Each lead individually remains in equilibrium, described by the Fermi-Dirac distribution functions $f_{L(R)}(E, \mu_{L(R)}) = [1 + e^{\beta(E - \mu_{L(R)})}]^{-1}$, while the central region experiences an electrostatic potential drop [see Fig. 1(b)]. This drives the system out of equilibrium and produces a steady-state charge current flowing through the central region from one lead to the other.

B. NEGF formalism for the steady state transport

The quantum transport properties of a two-terminal device in the steady state are computed from the spin (σ)-, energy (E)- and $\mathbf{k}$ -dependent NEGF of the central region. Its retarded, advanced, and lesser/greater components are given by [101]

$$G^{\sigma r}(E, \mathbf{k}) = [(E + i0^+)S(\mathbf{k}) - H^{\sigma}(\mathbf{k}) - \Sigma_L^{\sigma r}(E, \mathbf{k}) - \Sigma_R^{\sigma r}(E, \mathbf{k}) - \Sigma_{MB}^{\sigma r}(E, \mathbf{k})]^{-1}, \quad (1)$$

$$G^{\sigma a}(E, \mathbf{k}) = [G^{\sigma r}(E, \mathbf{k})]^{\dagger}, \quad (2)$$

and

$$G^{\sigma \lessgtr}(E, \mathbf{k}) = G^{\sigma r}(E, \mathbf{k})[\Sigma_L^{\sigma \lessgtr}(E, \mathbf{k}) + \Sigma_R^{\sigma \lessgtr}(E, \mathbf{k}) + \Sigma_{MB}^{\sigma \lessgtr}(E, \mathbf{k})]G^{\sigma a}(E, \mathbf{k}), \quad (3)$$

respectively. These satisfy the relation

$$G^{\sigma r}(E, \mathbf{k}) - G^{\sigma a}(E, \mathbf{k}) = G^{\sigma >}(E, \mathbf{k}) - G^{\sigma <}(E, \mathbf{k}), \quad (4)$$

so that only two of them are independent.

In Eq. (1), $H^\sigma(\mathbf{k})$ is the single-particle Hamiltonian of the central region, which also includes the electrostatic potential due to the voltage bias applied between the leads, and $S(\mathbf{k})$ is the orbital overlap. $\Sigma_{\text{MB}}^{\sigma r/a}(E, \mathbf{k})$ and $\Sigma_{\text{MB}}^{\sigma \lessgtr}(E, \mathbf{k})$ in Eqs. (1) and (3) are respectively the retarded/advanced and lesser/greater many-body self-energies, which account for electron-electron interaction. Instead, $\Sigma_{\text{L}}^{\sigma r/a}(E, \mathbf{k})$ ($\Sigma_{\text{R}}^{\sigma r/a}(E, \mathbf{k})$) and $\Sigma_{\text{L}}^{\sigma \lessgtr}(E, \mathbf{k})$ ($\Sigma_{\text{R}}^{\sigma \lessgtr}(E, \mathbf{k})$) are the retarded/advanced and lesser/greater embedding self-energies of the left-hand side (right-hand side) lead, which describe the effect of the leads on the central region. The self-energies satisfy a relation analogous to that of Eq. (4). The Hamiltonian, the orbital overlap, the Green's functions, and the self-energies are represented as square $N \times N$ matrices, where N is the number of basis orbitals within the central region. In principle, all of these matrices, except for $S(\mathbf{k})$, depend on the bias voltage, V . However, the explicit V -dependence is omitted for the sake of simplicity and will be indicated only when necessary.

Equation (1) is the Dyson equation. The retarded and advanced Green's functions, $G^{\sigma r}(E, \mathbf{k})$ and $G^{\sigma a}(E, \mathbf{k})$, describe the energy levels in the central region, given by the spin-resolved spectral function,

$$A^\sigma(E, \mathbf{k}) = i[G^{\sigma r}(E, \mathbf{k}) - G^{\sigma a}(E, \mathbf{k})]. \quad (5)$$

The integral of the diagonal matrix elements of $A^\sigma(E, \mathbf{k})$ over the transverse wave-number, $\mathbf{k}$, gives the density of states (DOS).

Equation (3) is sometimes called the Keldysh equation [102]. It describes the electron occupation of the central region. In particular, the integral of $G^{\sigma <}(E, \mathbf{k})$ over the energy and the wave-number gives the density matrix,

$$\rho^\sigma = \frac{1}{2\pi i} \frac{1}{\Omega_{\mathbf{k}}} \int_{\text{BZ}} d\mathbf{k} \int_{-\infty}^{\infty} dE G^{\sigma <}(E, \mathbf{k}), \quad (6)$$

where $\Omega_{\mathbf{k}}$ is the volume of the transverse BZ.

The retarded embedding self-energy, $\Sigma_{\text{L(R)}}^{\sigma r}(E, \mathbf{k})$, can be computed exactly using semianalytical [103] or iterative [104] schemes. Its real part gives the energy shifts of the central-region states, while the imaginary part

$$\Gamma_{\text{L(R)}}^\sigma(E, \mathbf{k}) = i[\Sigma_{\text{L(R)}}^{\sigma r}(E, \mathbf{k}) - \Sigma_{\text{L(R)}}^{\sigma r}(E, \mathbf{k})^\dagger], \quad (7)$$

describes their broadening due to coupling to the leads. The lesser/greater embedding self-energies $\Sigma_{\text{L(R)}}^{\sigma \lessgtr}(E, \mathbf{k})$ account for the inflow/outflow of charge from the L (R) lead to the central region. They can be obtained via the fluctuation-dissipation theorem [26],

$$\Sigma_{\text{L(R)}}^{\sigma <}(E, \mathbf{k}) = i f_{\text{L(R)}}(E, \mu_{\text{L(R)}}) \Gamma_{\text{L(R)}}^\sigma(E, \mathbf{k}), \quad (8)$$

$$\Sigma_{\text{L(R)}}^{\sigma >}(E, \mathbf{k}) = i[f_{\text{L(R)}}(E, \mu_{\text{L(R)}}) - 1] \Gamma_{\text{L(R)}}^\sigma(E, \mathbf{k}), \quad (9)$$

since the leads are charge reservoirs and remain in a local equilibrium. Upon the application of a bias voltage V , the embedding self-energies are rigidly shifted relative to their zero-bias values

$$\begin{aligned} \Sigma_{\text{L}}^{\sigma r/a(\lessgtr)}(E - eV/2, \mathbf{k}, V) &= \Sigma_{\text{L}}^{\sigma r/a(\lessgtr)}(E, \mathbf{k}, V = 0), \\ \Sigma_{\text{R}}^{\sigma r/a(\lessgtr)}(E + eV/2, \mathbf{k}, V) &= \Sigma_{\text{R}}^{\sigma r/a(\lessgtr)}(E, \mathbf{k}, V = 0), \end{aligned} \quad (10)$$

where the explicit V dependence has been reintroduced for clarity.

The many-body self-energy satisfies relations analogous to those in Eqs. (8) and (9) [28,83,105]:

$$\Sigma_{\text{MB}}^{\sigma <}(E, \mathbf{k}) = i F_{\text{MB}}^\sigma(E, \mathbf{k}) \Gamma_{\text{MB}}^\sigma(E, \mathbf{k}), \quad (11)$$

$$\Sigma_{\text{MB}}^{\sigma >}(E, \mathbf{k}) = i[F_{\text{MB}}^\sigma(E, \mathbf{k}) - 1] \Gamma_{\text{MB}}^\sigma(E, \mathbf{k}), \quad (12)$$

where $\Gamma_{\text{MB}}^\sigma(E, \mathbf{k}) = i[\Sigma_{\text{MB}}^{\sigma r}(E, \mathbf{k}) - \Sigma_{\text{MB}}^{\sigma r}(E, \mathbf{k})^\dagger]$ describes the broadening of the spectral function due to electron-electron interaction, and $F_{\text{MB}}^\sigma(E)$ denotes the occupation matrix of the central device region. However, unlike Eqs. (8) and (9), $F_{\text{MB}}^\sigma(E, \mathbf{k})$ generally differs from the Fermi function and reduces to it only at zero bias, namely in equilibrium.

Thus, in contrast to the leads, where the fluctuation-dissipation theorem provides a closed form for the retarded and lesser/greater self-energies, the many-body self-energies out of equilibrium must be calculated directly, accounting for the nonequilibrium occupation of the central region. This represents a significant challenge. In first-principles calculations, it has been addressed in a few works within the GW or three-body approximations of many-body perturbation theory, though these approaches have mostly been applied to molecular junctions [101,106] or quasione-dimensional systems [107–109]. In contrast, DFT + NEGF + DMFT studies of spintronic devices have largely been limited to zero-bias or special conditions, where the fluctuation-dissipation theorem is effectively recovered [64,74]. This motivates the need for a general implementation valid under arbitrary nonequilibrium conditions, as presented here.

C. DFT + NEGF

DFT + NEGF neglects the retarded and lesser many-body self-energies and employs the KS Hamiltonian, $H_{\text{KS}}^\sigma(\mathbf{k})$, as the single-particle Hamiltonian of the central region in Eq. (1). The embedding self-energies are likewise constructed from the corresponding KS Hamiltonians of the lead materials [110]. The exchange-correlation potential is usually treated within LSDA [12,13] or GGA [14–16] and acts as a static mean-field interaction. For ferromagnetic metals, relevant to the spintronic devices studied in this work, this yields an effective Stoner-like description of magnetism [111]. The Green's functions in DFT + NEGF are referred to as KS Green's functions and, throughout this paper, are denoted by lowercase symbols $g^{\sigma r/a(\lessgtr)}$ to distinguish them from the many-body Green's functions introduced in Eqs. (1)–(3).

The KS Hamiltonian of the central region depends functionally on the charge density ρ^σ , defined in Eq. (6) via the lesser Green's function. Consequently, Eqs. (1), (3), and (6) must be solved in a self-consistent manner. At finite bias, the electrostatic potential across the central region is obtained by solving the Poisson equation for the charge density at each iteration step and with the boundary conditions of Eq. (10) [32]. In certain systems, such as MTJs, where the voltage drop occurs almost entirely across the insulating barrier, the bias dependence can alternatively be incorporated into a non-self-consistent way by adding a linear ramp potential to the Hamiltonian. This will be the approach followed in this work, as discussed in Sec. III.

D. DMFT combined with DFT + NEGF

1. Projection onto the correlated subspace

The calculation of the many-body self-energy for all the N basis orbitals of a device's central region is practically infeasible. Often, this is also unnecessary, since in many cases electron-electron interaction beyond the static-mean field KS description is significant only for a few atoms or orbitals within the device. Therefore, we carry out a projection of the full central region onto a much smaller subspace, called the correlated subspace $\mathcal{C}$, which is spanned by a limited number of orbitals. The orthogonal complement of $\mathcal{C}$ within the central region is referred to as the “bath,” $\mathcal{B}$, and remains described at the KS level. Then, $\mathcal{B}$ can be further separated into two new effective leads, coupled to the correlated subspace $\mathcal{C}$, thereby providing a mapping from the original two-terminal device to a new, effective one, as shown in Fig. 2. DMFT will then be applied to treat this new system, and the obtained self-energies will finally be back-projected to the entire one (see Sec. IID 5).

Specifically, in the ferromagnetic transition metals used in spintronic devices, the valence states consist of the $4s$, $4p$, and $3d$ orbitals. The $4s$ and $4p$ are delocalized and form broad energy bands, so that their electronic behavior is well captured by the effective single-particle KS picture. In contrast, the $3d$ orbitals are more localized, energetically centered around the Fermi level, and therefore, moderately correlated. As such, they are selected to span $\mathcal{C}$. The size of $\mathcal{C}$ is $N_{\mathcal{C}} = 2 \times 5 \times N_{\text{TM}}$, where N_{TM} is the number of correlated transition metal atoms within our central region. The factor of 2 accounts for spin, and 5 corresponds to the number of $3d$ orbitals per atom. Conversely, the size of the bath is $N_{\mathcal{B}} = N - N_{\mathcal{C}}$.

The separation between $\mathcal{C}$ and $\mathcal{B}$ is achieved through a basis transformation that returns zero orbital overlap between $\mathcal{C}$ and $\mathcal{B}$ and also orthonormalizes the correlated orbitals inside $\mathcal{C}$, as detailed in Ref. [83]. This orthonormalization is convenient, as the impurity solvers employed in DMFT are typically formulated and implemented over an orthogonal basis set. In contrast, no orthonormalization is required for the bath orbitals, as the bath Green's function does not explicitly enter the DMFT solver.

Denoting the transformation matrix by $W(\mathbf{k})$, the Hamiltonian and overlap matrices transform as [83]

$$\begin{aligned} \bar{H}_{\text{KS}}^{\sigma}(\mathbf{k}) &= \begin{pmatrix} \bar{H}_{\mathcal{C}}^{\sigma}(\mathbf{k}) & \bar{H}_{\mathcal{C},\mathcal{B}}^{\sigma}(\mathbf{k}) \\ \bar{H}_{\mathcal{B},\mathcal{C}}^{\sigma}(\mathbf{k}) & \bar{H}_{\mathcal{B}}^{\sigma}(\mathbf{k}) \end{pmatrix} \\ &= W(\mathbf{k})^{\dagger} H_{\text{KS}}^{\sigma}(\mathbf{k}) W(\mathbf{k}), \end{aligned} \quad (13)$$

$$\begin{aligned} \bar{S}(\mathbf{k}) &= \begin{pmatrix} 1 & 0 \\ 0 & \bar{S}_{\mathcal{B}}(\mathbf{k}) \end{pmatrix} \\ &= W(\mathbf{k})^{\dagger} S(\mathbf{k}) W(\mathbf{k}). \end{aligned} \quad (14)$$

The explicit form of $W(\mathbf{k})$ used in our implementation requires the introduction of several auxiliary matrices, whose construction is algebraically involved. Since their explicit expressions are not required for the discussion that follows, we refer the reader to Eq. (10) of Ref. [83] and the Appendix therein for the full definition of $W(\mathbf{k})$ and the associated mathematical details.

In Eq. (13), $\bar{H}_{\mathcal{C}}^{\sigma}(\mathbf{k})$ ($\bar{H}_{\mathcal{B}}^{\sigma}(\mathbf{k})$) refers to the KS Hamiltonian of $\mathcal{C}$ ($\mathcal{B}$), while $\bar{H}_{\mathcal{C},\mathcal{B}}^{\sigma}(\mathbf{k})$ and $\bar{H}_{\mathcal{B},\mathcal{C}}^{\sigma}(\mathbf{k})$ are those corresponding to the couplings between $\mathcal{C}$ and $\mathcal{B}$. In contrast, the transformed overlap matrix, by construction, has the $\mathcal{C}$ -block equal to the identity matrix, reflecting the orthonormalization of the transformed basis within $\mathcal{C}$, and vanishing off-diagonal blocks, because of the zero-overlap condition between $\mathcal{C}$ and $\mathcal{B}$.

In second quantization, the KS Hamiltonian of $\mathcal{C}$ reads

$$\hat{H}_{\mathcal{C}}^{\sigma}(\mathbf{k}) = \sum_{i\lambda_i, j\lambda_j} [\bar{H}_{\mathcal{C}}^{\sigma}(\mathbf{k})]_{i\lambda_i, j\lambda_j} \hat{d}_{i\lambda_i\sigma}^{\dagger} \hat{d}_{j\lambda_j\sigma}, \quad (15)$$

where $\hat{d}_{i\lambda\sigma}^{\dagger}$ ($\hat{d}_{i\lambda\sigma}$) is the fermionic creation (annihilation) operator for an electron with spin σ in the λ th orbital of the i th atomic site. The indices i and j run over all atomic sites with correlated orbitals, while λ_i and λ_j label the correlated orbitals on the i th and j th atoms. For example, in a ferromagnetic transition metal with N_{TM} atoms having correlated $3d$ orbitals, we have $i, j = 1, \dots, N_{\text{TM}}$ and $\lambda_i, \lambda_j = 1, \dots, 5$. The diagonal terms in the Hamiltonian matrix, $[\bar{H}_{\mathcal{C}}^{\sigma}(\mathbf{k})]_{i\lambda_i, i\lambda_i}$, represent the orbital energies, while the off-diagonal ones, $[\bar{H}_{\mathcal{C}}^{\sigma}(\mathbf{k})]_{i\lambda_i, j\lambda_j}$ with $i \neq j$ ($i = j$ and $\lambda_i \neq \lambda_j$), describe inter- (intra) atomic hopping integrals for atoms within $\mathcal{C}$.

The Hamiltonian in Eq. (15) captures the electronic structure of $\mathcal{C}$ within the effective single-particle KS picture of DFT + NEGF. To incorporate electron-electron interactions, we include a multiorbital Hubbard-like interaction term, a choice justified by the predominantly local nature of the screened Coulomb interaction in transition metals. The resulting interacting Hamiltonian for $\mathcal{C}$ is

$$\begin{aligned} \hat{H}_{\mathcal{C},\mathcal{U}}(\mathbf{k}) &= \sum_{\sigma} \left[\hat{H}_{\mathcal{C}}^{\sigma}(\mathbf{k}) - \hat{H}_{\mathcal{C},dc}^{\sigma} + \right. \\ &\quad \left. + \frac{1}{2} \sum_{\substack{i,\lambda_1,\lambda_2,\lambda_3,\lambda_4 \\ \lambda_4,\sigma'}} U_{\lambda_1,\lambda_2,\lambda_3,\lambda_4} \hat{d}_{i\lambda_1\sigma}^{\dagger} \hat{d}_{i\lambda_2\sigma'}^{\dagger} \hat{d}_{i\lambda_4\sigma'} \hat{d}_{i\lambda_3\sigma} \right], \end{aligned} \quad (16)$$

where $U_{\lambda_1,\lambda_2,\lambda_3,\lambda_4}$ is the four-index interaction tensor. This can be parameterized in terms of the average effective Coulomb repulsion U and exchange J [112]

$$U = \frac{1}{(2l+1)^2} \sum_{\lambda_1,\lambda_2} U_{\lambda_1,\lambda_2,\lambda_1,\lambda_2}, \quad (17)$$

$$J = \frac{1}{2l(2l+1)} \sum_{\lambda_1 \neq \lambda_2} U_{\lambda_1,\lambda_2,\lambda_2,\lambda_1}. \quad (18)$$

The interaction parameters can, in principle, be computed from first principles using various methods, such as the constrained random phase approximation [113], constrained DFT [114], or constrained DMFT [115]. However, in the present work, their accurate determination is not of central importance. We therefore treat U and J as empirical parameters, taken from some of our previous works [48], as detailed in Sec. IV. The interaction matrix is then expressed in terms of the Slater integrals F^0 , F^2 , and F^4 (see Ref. [116]), with $U = F^0$, $J = (F^2 + F^4)/14$, and the ratio F^4/F^2 fixed to the atomic value, ~ 0.625 (Ref. [117]).

$\hat{H}_{\mathcal{C},dc}^{\sigma}$ is the double-counting correction, which subtracts the static correlation already included at the KS mean-field

level in $\hat{H}_C^\sigma(\mathbf{k})$. While its exact form is unknown, several approximate schemes have been proposed for bulk systems in equilibrium [60,62,118,119]. However, for open systems out of equilibrium, the proper treatment of double-counting remains an open problem, as further discussed in Sec. IID 6.

The same transformation applied to the Hamiltonian is also applied to the lead self-energy. In particular, if the system is constructed such that the leads are not directly coupled to $\mathcal{C}$ —a condition that can be ensured by including a sufficient number of lead layers within the central region, as in Fig. 2(b)—the lead self-energy acquires the block form

$$\begin{aligned}\bar{\Sigma}_{L(R)}^{\sigma r(<)}(E, \mathbf{k}) &= \begin{pmatrix} 0 & 0 \\ 0 & \bar{\Sigma}_{L(R),B}^{\sigma r(<)}(\mathbf{k}) \end{pmatrix} \\ &= W(\mathbf{k})^\dagger \Sigma_{L(R)}^{\sigma r(<)}(E, \mathbf{k}) W(\mathbf{k}).\end{aligned}\quad (19)$$

Finally, the sum, $\bar{\Sigma}_B(\mathbf{k}) = \bar{\Sigma}_{L,B}(\mathbf{k}) + \bar{\Sigma}_{R,B}(\mathbf{k})$ describes the total coupling of the bath orbitals to the device's leads.

2. Embedding self-energies for the correlated subspace

The KS Green's function in the transformed basis acquires the same block form as the KS Hamiltonian in Eq. (13), and it is defined as

$$\begin{aligned}\begin{pmatrix} \bar{g}_C^{\sigma r}(E, \mathbf{k}) & \bar{g}_{C,B}^{\sigma r}(E, \mathbf{k}) \\ \bar{g}_{B,C}^{\sigma r}(E, \mathbf{k}) & \bar{g}_B^{\sigma r}(E, \mathbf{k}) \end{pmatrix} \\ = \left[E \begin{pmatrix} 1 & 0 \\ 0 & \bar{S}_B(\mathbf{k}) \end{pmatrix} - \begin{pmatrix} \bar{H}_C^\sigma(\mathbf{k}) & \bar{H}_{C,B}^\sigma(\mathbf{k}) \\ \bar{H}_{B,C}^\sigma(\mathbf{k}) & \bar{H}_B^\sigma(\mathbf{k}) \end{pmatrix} \right. \\ \left. - \begin{pmatrix} 0 & 0 \\ 0 & \bar{\Sigma}_B^{\sigma r}(E, \mathbf{k}) \end{pmatrix} \right]^{-1}.\end{aligned}\quad (20)$$

Performing the matrix inversion yields the KS Green's function of $\mathcal{C}$

$$\bar{g}_C^{\sigma r}(E, \mathbf{k}) = [E - \bar{H}_C^\sigma(\mathbf{k}) - \bar{H}_{C,B}^\sigma(\mathbf{k}) \bar{g}_B^{\sigma r}(\mathbf{k}) \bar{H}_{B,C}^\sigma(\mathbf{k})]^{-1}, \quad (21)$$

where

$$\bar{g}_B^{\sigma r}(E, \mathbf{k}) = [E \bar{S}_B(\mathbf{k}) - \bar{H}_B^\sigma(\mathbf{k}) - \bar{\Sigma}_B^{\sigma r}(E, \mathbf{k})]^{-1} \quad (22)$$

is the retarded Green's function of the bath decoupled from $\mathcal{C}$.

Following Ref. [83], the bath can be further subdivided into N_{NI} basis orbitals that are coupled to $\mathcal{C}$, forming the so-called “extended region,” and the N_α (N_β) orbitals within the central region on the left (right) of $\mathcal{C}$, which are not coupled to $\mathcal{C}$ due to the finite spatial extent of the basis orbitals, as shown schematically in Fig. 2(a). Thus the bath and $\mathcal{C}$ -bath coupling Hamiltonians can be written in block forms as

$$\bar{H}_B^\sigma(\mathbf{k}) = \begin{pmatrix} H_{\alpha\alpha}^\sigma(\mathbf{k}) & \bar{H}_{\alpha,\text{NI}}^\sigma(\mathbf{k}) & H_{\alpha\beta}^\sigma(\mathbf{k}) \\ \bar{H}_{\alpha,\text{NI}}^{\sigma\dagger}(\mathbf{k}) & \bar{H}_{\text{NI}}^\sigma(\mathbf{k}) & \bar{H}_{\beta,\text{NI}}^{\sigma\dagger}(\mathbf{k}) \\ H_{\alpha\beta}^{\sigma\dagger}(\mathbf{k}) & \bar{H}_{\beta,\text{NI}}^\sigma(\mathbf{k}) & H_{\beta\beta}^\sigma(\mathbf{k}) \end{pmatrix}, \quad (23)$$

$$\bar{H}_{C,B}^\sigma(\mathbf{k}) = \begin{pmatrix} 0 & \bar{H}_{C,\text{NI}}^\sigma(\mathbf{k}) & 0 \end{pmatrix}, \quad (24)$$

while the bath overlap matrix, $\bar{S}_B(\mathbf{k})$, and the bath Green's function, $\bar{g}_B^{\sigma r}(E, \mathbf{k})$, can be written in the same form as Eq. (23). Note that the $H_{\alpha\beta}^\sigma$, $H_{\alpha\alpha}^\sigma$, and $H_{\beta\beta}^\sigma$ components are written without bars (cf. Eq. (16) in Ref. [83]), as they remain unchanged under the transformation in Eq. (13).

Next, we assume that $H_{\alpha\beta}^\sigma(\mathbf{k}) = 0$, which implies that the N_α orbitals to the left of $\mathcal{C}$ are not coupled to the N_β orbitals to the right, and

$$\bar{\Sigma}_B^{\sigma r}(E, \mathbf{k}) = \begin{pmatrix} \Sigma_{\alpha\alpha}^{\sigma r}(E, \mathbf{k}) & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \Sigma_{\beta\beta}^{\sigma r}(E, \mathbf{k}) \end{pmatrix}. \quad (25)$$

This condition is always enforced by the user in our calculations. Consequently, Eq. (21) simplifies to

$$\bar{g}_C^{\sigma r}(E, \mathbf{k}) = [E - \bar{H}_C^\sigma(\mathbf{k}) - \bar{\Sigma}_{C,\text{NI}}^{\sigma r}(E, \mathbf{k})]^{-1}, \quad (26)$$

where

$$\bar{\Sigma}_{C,\text{NI}}^{\sigma r}(E, \mathbf{k}) = \bar{H}_{C,\text{NI}}^\sigma(\mathbf{k}) \bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k}) \bar{H}_{\text{NI},C}^\sigma(\mathbf{k})^\dagger \quad (27)$$

is the effective retarded embedding self-energy for $\mathcal{C}$, with the Green's function of the NI part given by

$$\begin{aligned}\bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k}) \\ = [E \bar{S}_{\text{NI}}(\mathbf{k}) - \bar{H}_{\text{NI}}^\sigma(\mathbf{k}) - \bar{\Sigma}_{L,\text{NI}}^{\sigma r}(E, \mathbf{k}) - \bar{\Sigma}_{R,\text{NI}}^{\sigma r}(E, \mathbf{k})]^{-1}.\end{aligned}\quad (28)$$

Here, $\bar{\Sigma}_{L,\text{NI}}^{\sigma r}(E, \mathbf{k})$ and $\bar{\Sigma}_{R,\text{NI}}^{\sigma r}(E, \mathbf{k})$ are the embedding self-energies for NI from the left (α) and right (β) parts of the bath, respectively. These are defined analogously to Eqs. (62) and (63) of Ref. [83] as

$$\begin{aligned}\bar{\Sigma}_{L,\text{NI}}^{\sigma r}(E, \mathbf{k}) &= \bar{K}_{\text{NI},\alpha}^\sigma(E, \mathbf{k}) \bar{g}_{\alpha\alpha}^{\sigma r}(E, \mathbf{k}) \bar{K}_{\text{NI},\alpha}^\sigma(E, \mathbf{k})^\dagger, \\ \bar{\Sigma}_{R,\text{NI}}^{\sigma r}(E, \mathbf{k}) &= \bar{K}_{\text{NI},\beta}^\sigma(E, \mathbf{k}) \bar{g}_{\beta\beta}^{\sigma r}(E, \mathbf{k}) \bar{K}_{\text{NI},\beta}^\sigma(E, \mathbf{k})^\dagger,\end{aligned}\quad (29)$$

where

$$\bar{g}_{\alpha\alpha(\beta\beta)}^{\sigma r}(E, \mathbf{k}) = [K_{\alpha\alpha(\beta\beta)}^\sigma(E, \mathbf{k}) - \Sigma_{\alpha\alpha(\beta\beta)}^{\sigma r}(E, \mathbf{k})]^{-1} \quad (30)$$

is the Green's function of the α (β) block, $\bar{K}_{\text{NI},\alpha(\beta)}^\sigma(E, \mathbf{k}) = [E \bar{S}_{\text{NI},\alpha(\beta)}(\mathbf{k}) - \bar{H}_{\text{NI},\alpha(\beta)}^\sigma(\mathbf{k})]$ are the corresponding coupling matrices between NI and the left (right) subsystems and $K_{\alpha\alpha(\beta\beta)}^\sigma(E, \mathbf{k}) = [E \bar{S}_{\alpha\alpha(\beta\beta)}(\mathbf{k}) - \bar{H}_{\alpha\alpha(\beta\beta)}^\sigma(\mathbf{k})]$ corresponds to the left (right) block.

By substituting $\bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k})$ from Eq. (28) into the expression for $\bar{\Sigma}_{C,\text{NI}}^{\sigma r}(E, \mathbf{k})$ in Eq. (27) and by taking the imaginary part, we obtain the broadening matrices for $\mathcal{C}$ coupled to the new effective left and right leads

$$\begin{aligned}\bar{\gamma}_{L,C}^\sigma(E, \mathbf{k}) &= \bar{H}_{C,\text{NI}}^\sigma(\mathbf{k}) \bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k}) \bar{\Gamma}_{L,\text{NI}}^\sigma(E, \mathbf{k}) \\ &\quad \times \bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k})^\dagger \bar{H}_{C,\text{NI}}^\sigma(\mathbf{k})^\dagger,\end{aligned}\quad (31)$$

$$\begin{aligned}\bar{\gamma}_{R,C}^\sigma(E, \mathbf{k}) &= \bar{H}_{C,\text{NI}}^\sigma(\mathbf{k}) \bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k})^\dagger \bar{\Gamma}_{R,\text{NI}}^\sigma(E, \mathbf{k}) \\ &\quad \times \bar{g}_{\text{NI}}^{\sigma r}(E, \mathbf{k}) \bar{H}_{C,\text{NI}}^\sigma(\mathbf{k})^\dagger,\end{aligned}\quad (32)$$

as defined in Eqs. (71) and (72) of Ref. [83], with

$$\bar{\Gamma}_{L,\text{NI}}^\sigma(E, \mathbf{k}) = i[\bar{\Sigma}_{L,\text{NI}}^{\sigma r}(E, \mathbf{k}) - \bar{\Sigma}_{L,\text{NI}}^{\sigma r}(E, \mathbf{k})^\dagger], \quad (33)$$

$$\bar{\Gamma}_{R,\text{NI}}^\sigma(E, \mathbf{k}) = i[\bar{\Sigma}_{R,\text{NI}}^{\sigma r}(E, \mathbf{k}) - \bar{\Sigma}_{R,\text{NI}}^{\sigma r}(E, \mathbf{k})^\dagger]. \quad (34)$$

Finally, the lesser KS Green's function of $\mathcal{C}$ is given by

$$\begin{aligned}\bar{g}_C^{\sigma <}(E, \mathbf{k}) \\ = \bar{g}_C^{\sigma r}(E, \mathbf{k}) [\bar{\Sigma}_{L,C}^{\sigma <}(E, \mathbf{k}) + \bar{\Sigma}_{R,C}^{\sigma <}(E, \mathbf{k})] \bar{g}_C^{\sigma r}(E, \mathbf{k})^\dagger,\end{aligned}\quad (35)$$

where the lesser embedding self-energy for the left-hand side (right-hand side) effective lead is defined, analogously to Eq. (8), as

$$\bar{\Sigma}_{L(R),C}^{\sigma <}(E, \mathbf{k}) = i f_{L(R)}(E, \mu_{L(R)}) \bar{\gamma}_{L(R),C}^{\sigma}(E, \mathbf{k}). \quad (36)$$

Here, we use the chemical potential of the original electrodes. As such, while Eq. (36) is always valid in equilibrium, under nonequilibrium conditions it will hold only if the applied bias does not drop inside the portion of the device that has been incorporated into the new effective leads. This is an approximation inherent to our current implementation and should be taken into account, as it may limit its applicability. In principle, it can be easily relaxed, but doing so is involved within the present structure of the SMEAGOL code and is left for future work. For the systems studied here, the approximation, however, is well justified.

3. Nonequilibrium DMFT loop

The effective device is finally treated within DMFT. The retarded and the lesser Green's functions of C are defined analogously to Eq. (1) and (3) as,

$$\bar{G}_C^{\sigma r}(E, \mathbf{k}) = \bar{g}_C^{\sigma r}(E, \mathbf{k}) + \bar{g}_C^{\sigma r}(E, \mathbf{k}) \bar{\Sigma}_C^{\sigma r}(E) \bar{G}_C^{\sigma r}(E, \mathbf{k}), \quad (37)$$

$$\bar{G}_C^{\sigma <}(E, \mathbf{k}) = \bar{G}_C^{\sigma r}(E, \mathbf{k}) [\bar{\Sigma}_{L,C}^{\sigma <}(E, \mathbf{k}) + \bar{\Sigma}_{R,C}^{\sigma <}(E, \mathbf{k}) + \bar{\Sigma}_C^{\sigma <}(E)] \bar{G}_C^{\sigma r}(E, \mathbf{k})^\dagger. \quad (38)$$

Here, $\bar{g}_C^{\sigma r}(E, \mathbf{k})$ is the KS retarded Green's function defined in Eq. (26) and $\bar{\Sigma}_{L(R),C}^{\sigma <}(E, \mathbf{k})$ is the lesser embedding self-energy for the left (right) effective lead in Eq. (36). $\bar{\Sigma}_C^{\sigma r}(E)$ and $\bar{\Sigma}_C^{\sigma <}(E)$ are the many-body retarded and the lesser self-energies, which are $\mathbf{k}$ -independent and site diagonal due to the locality approximation of DMFT,

$$\bar{\Sigma}_C^{\sigma r(<)}(E) = \begin{pmatrix} \bar{\Sigma}_1^{\sigma r(<)}(E) & 0 & \dots & 0 \\ 0 & \bar{\Sigma}_2^{\sigma r(<)}(E) & \dots & 0 \\ 0 & 0 & \dots & \bar{\Sigma}_{N_{TM}}^{\sigma r(<)}(E) \end{pmatrix}. \quad (39)$$

Each $\bar{\Sigma}_i^{\sigma r(<)}(E)$ block represents the retarded (lesser) DMFT self-energy for atomic site i within the correlated subspace C . The index i runs over the N_{TM} transition-metal atoms forming the ferromagnetic layers in our spintronic devices. Each block is a 5×5 matrix, corresponding to the five $3d$ orbitals of these atoms.

The self-energies, $\bar{\Sigma}_C^{\sigma r}(E)$ and $\bar{\Sigma}_C^{\sigma <}(E)$, are obtained by mapping the correlated subspace onto a set of auxiliary impurity problems, one for each atom. In analogy with DMFT for zero-bias transport [73,74], this is achieved by enforcing the self-consistency condition that the so-called local Green's function of atom i coincides with the corresponding impurity's Green's function,

$$G_{imp,i}^{\sigma r(<)}(E) = \bar{G}_{loc,i}^{\sigma r(<)}(E) \equiv \frac{1}{\Omega_k} \int d\mathbf{k} \bar{G}_{C,i}^{\sigma r(<)}(E, \mathbf{k}). \quad (40)$$

Unlike the zero-bias case, where only the retarded component must be matched, under nonequilibrium conditions, the self-consistency condition applies to both the retarded and

lesser components due to the breakdown of the fluctuation-dissipation theorem (Sec. II B), except in certain special cases (Sec. II D 4).

In practice, the DMFT self-consistent cycle (schematically illustrated in Fig. 3) proceeds through the following main steps.

(i) Compute the many-body retarded and the lesser Green's functions of the correlated subsystem in Eqs. (37) and (38). For the first iteration, the many-body self-energies $\bar{\Sigma}_C^{\sigma r}(E)$ and $\bar{\Sigma}_C^{\sigma <}(E)$ are generally assumed to vanish.

(ii) For each atom i , extract the corresponding diagonal block $\bar{G}_{C,i}^{\sigma r(<)}(E, \mathbf{k})$ from $\bar{G}_C^{\sigma r(<)}(E, \mathbf{k})$, and compute the local retarded (lesser) Green's function.

(iii) Construct the retarded and lesser dynamical mean fields for each atom

$$\bar{G}_{DF,i}^{\sigma r}(E) = [\bar{G}_{loc,i}^{\sigma r}(E)^{-1} + \bar{\Sigma}_i^{\sigma r}(E)]^{-1}, \quad (41)$$

$$\bar{G}_{DF,i}^{\sigma <}(E) = \bar{G}_{DF,i}^{\sigma r}(E) \Delta_i^{\sigma <}(E) \bar{G}_{DF,i}^{\sigma r}(E)^\dagger, \quad (42)$$

where

$$\Delta_i^{\sigma <}(E) = [\bar{G}_{loc,i}^{\sigma r}(E)]^{-1} \bar{G}_{loc,i}^{\sigma <}(E) [\bar{G}_{loc,i}^{\sigma r}(E)^\dagger]^{-1} - \bar{\Sigma}_i^{\sigma <}(E). \quad (43)$$

is the lesser hybridization function.

(iv) Define the noninteracting impurity's retarded and lesser Green's functions as

$$g_{imp,i}^{\sigma r}(E) \equiv \bar{G}_{DF,i}^{\sigma r}(E), \quad g_{imp,i}^{\sigma <}(E) \equiv \bar{G}_{DF,i}^{\sigma <}(E). \quad (44)$$

(v) Solve each impurity problem using an appropriate nonequilibrium impurity solver. The specific solver employed in our work is detailed in Sec. II D 6. For each impurity, the solver yields the impurity's retarded and lesser many-body self-energies, denoted by $\Sigma_{imp,i}^{\sigma r}(E)$ and $\Sigma_{imp,i}^{\sigma <}(E)$, respectively, and the corresponding impurity's Green's functions,

$$G_{imp,i}^{\sigma r}(E) = [g_{imp,i}^{\sigma r}(E)^{-1} - \Sigma_{imp,i}^{\sigma r}(E)]^{-1}, \quad (45)$$

$$G_{imp,i}^{\sigma <}(E) = G_{imp,i}^{\sigma r}(E) [\Delta_i^{\sigma <}(E) + \Sigma_{imp,i}^{\sigma <}(E)] G_{imp,i}^{\sigma r}(E)^\dagger. \quad (46)$$

(vi) Update the DMFT retarded and lesser self-energies by setting $\bar{\Sigma}_i^{\sigma r}(E) = \Sigma_{imp,i}^{\sigma r}(E)$ and $\bar{\Sigma}_i^{\sigma <}(E) = \Sigma_{imp,i}^{\sigma <}(E)$ in Eq. (39). Return to step (i) and iterate until the self-consistency condition in Eq. (40) is satisfied.

4. Rigid shift approximation

In certain situations, the full nonequilibrium DMFT procedure described above can be simplified. In particular, when the correlated atoms are strongly coupled to one lead and are separated from the opposite lead by a sufficiently thick barrier—so that their interaction with that well-separated lead is negligible—they can be considered to be in equilibrium with the lead they are coupled to. In this limit, the fluctuation-dissipation theorem is effectively recovered.

This allows us to apply the "rigid shift approximation," introduced in Ref. [65]. Each block of the retarded and lesser correlated subspace's self-energies in Eq. (39) can be obtained

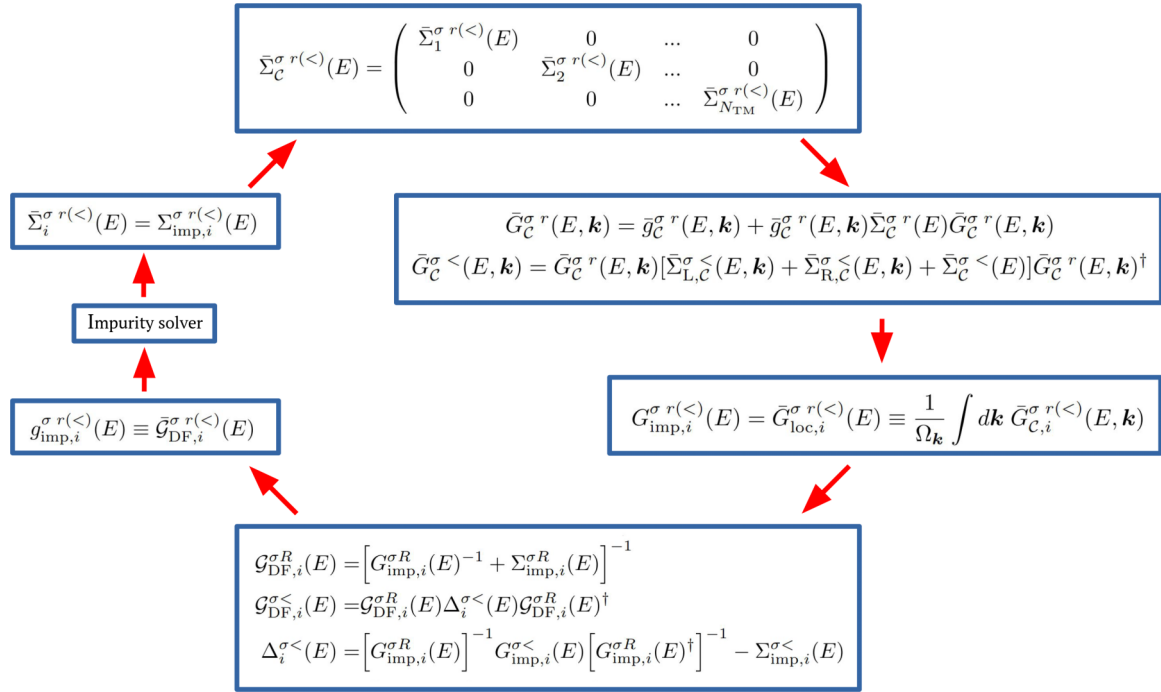


FIG. 3. Schematic representation of the DMFT loop.

through relations similar to Eqs. (10) and (8), namely,

$$\bar{\Sigma}_i^{\sigma r}(E \pm eV/2, V) = \bar{\Sigma}_i^{\sigma r}(E, V = 0), \quad (47)$$

$$\bar{\Sigma}_i^{\sigma <}(E, V) = if_{L(R)}(E, \mu_{L(R)}) \bar{\Gamma}_i^{\sigma}(E, V), \quad (48)$$

where $\bar{\Gamma}_i^{\sigma}(E, V) = i[\bar{\Sigma}_i^{\sigma r}(E, V) - \bar{\Sigma}_i^{\sigma r}(E, V)^\dagger]$. The minus (plus) sign in Eq. (47) and the Fermi function $f_{L(R)}$ in Eq. (48) are used when the atom i is coupled to the left-hand side (right-hand side) lead. By using this approach, only the retarded self-energy $\bar{\Sigma}_i^{\sigma r}(E, V = 0)$ at zero bias needs to be computed; the lesser and retarded components at finite bias can then be obtained directly, thus greatly reducing the computational effort.

In practice, however, we find that a straightforward application of the rigid-shift approximation often leads to poor current conservation, as it neglects the change in hybridization between the leads and the correlated orbitals induced by the electrostatic potential drop. To address this shortfall, we compute $\bar{\Sigma}_i^{\sigma r}(E, V)$ using an equilibrium calculation with this electrostatic potential added to the zero-bias Kohn-Sham Hamiltonian of the central region, and then obtain the corresponding lesser DMFT self-energy via Eq. (48).

5. Back-projection to the CR

Once the DMFT self-energy of $\mathcal{C}$ has been calculated, we can straightforwardly construct the self-energy of the full central region as

$$\bar{\Sigma}_{MB}^{\sigma r(<)}(E) = \begin{pmatrix} \bar{\Sigma}_{MB,C}^{\sigma r(<)}(E) & 0 \\ 0 & 0 \end{pmatrix}, \quad (49)$$

and apply the inverse transformation of Eq. (13) to express the many-body self-energy in the original basis,

$$\Sigma_{MB}^{\sigma r(<)}(E, \mathbf{k}) = W(\mathbf{k})^{-1\dagger} \bar{\Sigma}_{MB}^{\sigma r(<)}(E) W(\mathbf{k})^{-1}. \quad (50)$$

Then, $\Sigma_{MB}^{\sigma r(<)}(E, \mathbf{k})$ can then be used to compute the Green's functions defined in Eqs. (1) and (3), and thereby the spectral function [Eq. (5)], the density matrix [Eq. (6)], and the charge current defined in Sec. II E.

6. Impurity solver

At equilibrium, the impurity problem is often solved with high accuracy—up to stochastic noise—using continuous-time quantum Monte Carlo (CTQMC) methods [120], formulated on the imaginary frequency axis. To obtain real-frequency observables such as spectral functions, these results must be analytically continued to the real axis, a numerically ill-posed procedure [121]. Extending CTQMC methods to nonequilibrium systems is significantly more challenging, as it requires real-time simulations along the Keldysh contour [122–125], a task that suffers from severe sign problems and an exponential growth in computational cost with simulation time. Although recent advances, such as the steady-state inchworm CTQMC algorithm [94], have reduced the computational cost by several orders of magnitude and enabled the calculation of frequency-dependent steady-state quantities, applications have so far been limited to the single-orbital Anderson impurity model. Unfortunately, they remain computationally prohibitive for realistic multiorbital impurity problems.

Given our focus on transition metals, which present moderate correlations, we adopt a more computationally efficient impurity solver based on second-order perturbation theory in the electron-electron interaction [26,64,126]. This approach can be formulated directly on the real energy axis and has been shown in previous studies to produce results in good agreement with experimental photoemission spectra of transition metals thin films [48,64].

In our calculations, we assume that both the retarded and lesser components of the impurity Green's function, as well as the self-energy, are diagonal in orbital space. This significantly reduces the computational cost. For the systems considered here, this assumption is well justified due to their cubic symmetry and is routinely utilized in the literature [60,127]. In less symmetric systems, however, the diagonal approximation may not hold, and neglecting orbital off-diagonal elements introduces an approximation whose significance must be assessed on a case-by-case basis.

The retarded component of the impurity self-energy contains both first- and second-order terms in the Coulomb interaction,

$$\Sigma_{\text{imp},i\lambda}^{\sigma r}(E) \approx \Sigma_{\text{imp},i\lambda}^{\sigma r(1)} + \Sigma_{\text{imp},i\lambda}^{\sigma r(2)}(E), \quad (51)$$

while the lesser component only contains second-order terms,

$$\Sigma_{\text{imp},i\lambda}^{\sigma <}(E) \approx \Sigma_{\text{imp},i\lambda}^{\sigma <(2)}(E). \quad (52)$$

The subscript λ denotes the orbital of the impurity i , as in Eq. (16).

The first order contribution to the retarded self-energy takes the well-known Hartree-Fock form:

$$\Sigma_{\text{imp},i\lambda}^{\sigma r(1)}(E) = \sum_{i\lambda_1\sigma_1} U_{i\lambda i\lambda_1 i\lambda_1 i\lambda}^{\sigma\sigma_1\sigma_1} n_{i\lambda_1}^{\sigma_1} - \sum_{i\lambda_1} U_{i\lambda i\lambda_1 i\lambda_1 i\lambda}^{\sigma\sigma\sigma\sigma} n_{i\lambda_1}^{\sigma}, \quad (53)$$

which is local in time and therefore energy-independent. Here, $n_{i\lambda}^{\sigma} = -i \int \frac{dE}{2\pi} g_{\text{imp},i\lambda}^{\sigma <}(E)$ denotes the occupation of orbital λ with spin σ on impurity i , where $g_{\text{imp},i\lambda}^{\sigma <}(E)$ is the corresponding impurity Green's function defined in Eq. (44).

The second-order contributions to the retarded and lesser self-energies are given by

$$\begin{aligned} \text{Im}[\Sigma_{\text{imp},i\lambda}^{\sigma r(2)}(E)] = & -\pi \sum_{i\lambda_1 i\lambda_2 i\lambda_3 \sigma_1} U_{i\lambda i\lambda_1 i\lambda_2 i\lambda_3}^{\sigma\sigma_1\sigma\sigma_1} U_{i\lambda_3 i\lambda_2 i\lambda_1 i\lambda}^{\sigma_1\sigma_1\sigma\sigma} \int d\epsilon_1 d\epsilon_2 \rho_{i\lambda_1\sigma_1}(\epsilon_1) \rho_{i\lambda_2\sigma}(\epsilon_2) \rho_{i\lambda_3\sigma_1}(\epsilon_1 + \epsilon_2 - E) \\ & \times \{ \tilde{f}_{i\lambda_1\sigma_1}(\epsilon_1) \tilde{f}_{i\lambda_2\sigma}(\epsilon_2) + [1 - \tilde{f}_{i\lambda_1\sigma_1}(\epsilon_1) - \tilde{f}_{i\lambda_2\sigma}(\epsilon_2)] \tilde{f}_{i\lambda_3\sigma_1}(\epsilon_1 + \epsilon_2 - E) \} \\ & + \pi \sum_{i\lambda_1 i\lambda_2 i\lambda_3} U_{i\lambda i\lambda_1 i\lambda_2 i\lambda_3}^{\sigma\sigma\sigma\sigma} U_{i\lambda_2 i\lambda_3 i\lambda_1 i\lambda}^{\sigma\sigma\sigma\sigma} \int d\epsilon_1 d\epsilon_2 \rho_{i\lambda_1\sigma}(\epsilon_1 + \epsilon_2 - E) \rho_{i\lambda_2\sigma}(\epsilon_2) \rho_{i\lambda_3\sigma}(\epsilon_1) \\ & \times \{ \tilde{f}_{i\lambda_2\sigma}(\epsilon_2) \tilde{f}_{i\lambda_3\sigma}(\epsilon_1) + [1 - \tilde{f}_{i\lambda_2\sigma}(\epsilon_2) - \tilde{f}_{i\lambda_3\sigma}(\epsilon_1)] \tilde{f}_{i\lambda_1\sigma}(\epsilon_1 + \epsilon_2 - E) \} \end{aligned} \quad (54)$$

$$\begin{aligned} \Sigma_{\text{imp},i\lambda}^{\sigma <(2)}(E) = & 2\pi i \sum_{i\lambda_1 i\lambda_2 i\lambda_3 \sigma_1} U_{i\lambda i\lambda_1 i\lambda_2 i\lambda_3}^{\sigma\sigma_1\sigma\sigma_1} U_{i\lambda_3 i\lambda_2 i\lambda_1 i\lambda}^{\sigma_1\sigma_1\sigma\sigma} \int d\epsilon_1 d\epsilon_2 \rho_{i\lambda_1\sigma_1}(\epsilon_1) \rho_{i\lambda_2\sigma}(\epsilon_2) \rho_{i\lambda_3\sigma_1}(\epsilon_1 + \epsilon_2 - E) \\ & \times \tilde{f}_{i\lambda_1\sigma_1}(\epsilon_1) \tilde{f}_{i\lambda_2\sigma}(\epsilon_2) [1 - \tilde{f}_{i\lambda_3\sigma_1}(\epsilon_1 + \epsilon_2 - E)] \\ & - 2\pi i \sum_{i\lambda_1 i\lambda_2 i\lambda_3} U_{i\lambda i\lambda_1 i\lambda_2 i\lambda_3}^{\sigma\sigma\sigma\sigma} U_{i\lambda_2 i\lambda_3 i\lambda_1 i\lambda}^{\sigma\sigma\sigma\sigma} \int d\epsilon_1 d\epsilon_2 \rho_{i\lambda_1\sigma}(\epsilon_1 + \epsilon_2 - E) \rho_{i\lambda_2\sigma}(\epsilon_2) \rho_{i\lambda_3\sigma}(\epsilon_1) \\ & \times \tilde{f}_{i\lambda_2\sigma}(\epsilon_2) \tilde{f}_{i\lambda_3\sigma}(\epsilon_1) [1 - \tilde{f}_{i\lambda_1\sigma}(\epsilon_1 + \epsilon_2 - E)], \end{aligned} \quad (55)$$

while the real part of the retarded self-energy is obtained from the Kramers-Kronig relations. Here, $\rho_{i\lambda\sigma}(E) = -\frac{1}{\pi} \text{Im} g_{\text{imp},i\lambda}^{\sigma r}(E)$ denotes the spectral function of orbital λ with spin σ on impurity i , and $\tilde{f}_{i\lambda\sigma}(E)$ is the corresponding generalized Fermi function defined as

$$\tilde{f}_{i\lambda\sigma}(E) = \frac{g_{\text{imp},i\lambda}^{\sigma <}(E)}{2\pi \rho_{i\lambda\sigma}(E)}. \quad (56)$$

The numerical integrations (convolutions) are performed using the uniform mesh and the trapezoidal rule integration.

We note that the “bare” impurity Green's function g_{imp} enters the impurity self-energy, rather than the “dressed” Green's function, which would correspond to the Born approximation. A comparison, limited to the zero-bias case, between impurity solvers based on the dressed and bare impurity Green's functions can be found in Ref. [64]. That study shows that, although differences exist, they are generally small and negligible for the relatively small values of the interaction parameters employed in the present work. Furthermore, the dressing of the Green's function is effectively achieved through the self-consistent DMFT loop.

In Eq. (51), the first-order term corresponds to an on-site energy shift that adds to the double-counting correction in Eq. (16). In practice, these contributions can be absorbed into the definition of the bare Green's function, allowing one to evaluate explicitly only the second-order self-energy.

In previous zero-bias implementations [64] we considered two practical treatments. In the first, the sum of the first-order and double-counting contributions was approximated by the effective U potential of the DFT + U formulation by Dudarev *et al.* [128]. In this case, DFT + U calculations were used to obtain the corresponding KS Green's functions, and only the second-order self-energy was subsequently evaluated within the impurity solver. In the second approach, we instead used the standard KS DFT Green's function; neglecting the first-order term in the impurity solver effectively corresponds to approximating it by the LSDA/GGA exchange-correlation potential.

For ferromagnetic transition metals, both approaches give comparable results, albeit for different values of the interaction parameters. Larger values of U and J are required when using DFT + U than when using DFT Green's functions in the

evaluation of the self-energy. This reflects the opposite effects of the static U potential and the second-order self-energy: the former enhances the exchange splitting already overestimated by DFT, while the latter tends to reduce it. When the static U potential is included, a stronger interaction is therefore needed in the self-energy to counterbalance its effect on the exchange splitting.

Since a proper treatment of double-counting corrections in transport calculations at finite bias remains an open problem and the DFT + U calculations are numerically slow to converge for our systems, in the present work, we adopt the second approach and retain only the second-order self-energy evaluated using KS DFT Green's functions. At zero bias, this approximation yields results consistent with Refs. [65,74] for

$$I_{L(R)} = \frac{e}{h} \frac{1}{\Omega_k} \sum_{\sigma} \int dk \int_{-\infty}^{\infty} dE \text{Tr}[f_{L(R)}(E, \mu_{L(R)}) \Gamma_{L(R)}^{\sigma}(E, \mathbf{k}) A^{\sigma}(E, \mathbf{k}) + i \Gamma_{L(R)}^{\sigma}(E, \mathbf{k}) G^{\sigma <}(E, \mathbf{k})], \quad (57)$$

where $G^{\sigma <}(E, \mathbf{k})$, $A^{\sigma}(E, \mathbf{k})$, and $\Gamma_{L(R)}^{\sigma}(E, \mathbf{k})$ are defined in Eqs. (3), (5), and (7), respectively. Although Eq. (57) was originally derived assuming an orthonormal basis, it remains valid for the nonorthogonal case as well [129,130].

Since charge conservation implies that $I_L = -I_R$, the current through the CR is usually expressed in the symmetrized form, $I = (I_L - I_R)/2$, a convention adopted here. In practice, however, some care is required in the calculations. Although DMFT is a conserving many-body theory in the Baym-Kadanoff sense [62], ensuring charge conservation in practice can be challenging due to the employed approximations (e.g., in the implementation of the impurity solvers) and numerical error propagation. Thus we explicitly verify current conservation in all our calculations, as detailed in the Appendix, finding generally satisfactory results.

The current can then be expressed as the sum of two contributions, which we refer to as the coherent and incoherent parts,

$$I^{\sigma} = I_C^{\sigma} + I_{IC}^{\sigma}. \quad (58)$$

This decomposition follows from Eq. (4), together with the expressions for the lesser and greater components of the lead and many-body self-energies given in Eqs. (8) and (11). The coherent contribution reads

$$I_C^{\sigma} = \frac{e}{h} \int_{-\infty}^{\infty} dE T^{\sigma}(E) [f_L(E) - f_R(E)], \quad (59)$$

where $T^{\sigma}(E)$ denotes the transmission coefficient, given by

$$T^{\sigma}(E) = \frac{1}{\Omega_k} \int_{\text{BZ}} dk \text{Tr}[\Gamma_L^{\sigma}(E, \mathbf{k}) G^{\sigma r}(E, \mathbf{k}) \times \Gamma_R^{\sigma}(E, \mathbf{k}) G^{\sigma r}(E, \mathbf{k})^{\dagger}]. \quad (60)$$

Equation (59) is the generalization of the Landauer-Büttiker formula to finite bias (see, for example, reference [20,27,32]). The incoherent contribution, instead, is

$$I_{IC}^{\sigma} = \frac{e}{2h \Omega_k} \int_{\text{BZ}} dk \int dE \text{Tr}\{[f_L(E) - F_{\text{MB}}^{\sigma}(E, \mathbf{k})] \Gamma_{\text{MB}}^{\sigma}(E) \times G^{\sigma r}(E, \mathbf{k})^{\dagger} \Gamma_L^{\sigma}(E, \mathbf{k}) G^{\sigma r}(E, \mathbf{k})\}$$

Fe/MgO and Co/Cu junctions, where KS DFT + U Green's functions were instead used, as well as with available experimental data, as discussed in Sec. IV. The remaining differences mainly correspond to modest changes in the spectral width and do not affect the conclusions relevant for the present work.

E. Charge current

The expression for the charge current flowing from the noninteracting left-hand side (right-hand side) lead into the CR of a two-terminal device in steady-state conditions was derived by Meir and Wingreen in their seminal paper [100], and is given by

$$- \text{Tr}\{[f_R(E) - F_{\text{MB}}^{\sigma}(E, \mathbf{k})] \Gamma_{\text{MB}}^{\sigma}(E) G^{\sigma r}(E, \mathbf{k})^{\dagger} \times \Gamma_R^{\sigma}(E, \mathbf{k}) G^{\sigma r}(E, \mathbf{k})\}. \quad (61)$$

In DFT + NEGF, there is no many-body self-energy, and thus the incoherent contribution vanishes. The current is therefore solely given by Eq. (59), with $T^{\sigma}(E)$ evaluated using the KS Green's function, $g^{\sigma r}(E, \mathbf{k})$. Accordingly, charge transport is described as the coherent transmission of electrons through the central region and is determined by the occupation of the incoming and out-coming states, f_L and f_R . The integrand in Eq. (59) is significantly different from zero only within an energy range approximately spanning from $E_F - eV/2$ to $E_F + eV/2$, referred to as the bias window. Thus, in practice, the current is often approximated by integrating $T(E)$ over this window.

Within DFT + NEGF + DMFT, both the coherent and incoherent contributions to the current may become significant. The coherent part, I_C , still corresponds to the current arising from transmission through the central region, but is now evaluated using the many-body retarded Green's function, and therefore reflects a modified electronic structure compared to the DFT KS one. Importantly, due to the imaginary part of the many-body self-energy, the electronic states are not only shifted in energy but also acquire a finite lifetime. This already partially breaks the notion of perfectly coherent transport, despite the conventional terminology. A similar effect has been reported within the Kubo formalism under the “bare-bubble” approximation implemented with DMFT Green's functions [131]. In our calculations based on Eq. (59), this finite lifetime generally leads to a reduction of the transmission coefficient [36,74], and consequently, of the current.

In contrast, the properly defined incoherent contribution to the current captures the impact of inelastic scattering on transport. Notably, Eq. (61) resembles Eq. (59), but with the out-of-equilibrium electron distribution of the central region, F_{MB}^{σ} , and the many-body broadening matrix, $\Gamma_{\text{MB}}^{\sigma}$, replacing f_R (f_L) and Γ_R^{σ} (Γ_L^{σ}) in the first (second) term of the integrand. This suggests an intuitive picture in which the effect of electron-electron interactions can be represented by an

additional “virtual electrode,” characterized by the many-body self-energy and an effective distribution function $F_{\text{MB}}(E, \mathbf{k})$. Then, I_{IC} can be interpreted as the current flowing from the left-hand side lead into this virtual electrode and subsequently from there into the right-hand side lead. Crucially, once electrons enter the virtual electrode, they may re-emerge with modified energy and phase, thereby mimicking inelastic scattering events arising from electron-electron interaction. This picture provides a natural analogy to the de-phasing Büttiker probe concept [55], wherein phase-breaking and energy-dissipating processes are captured through a fictitious, current-conserving voltage probe [132]. In contrast, DMFT provides a quantitative, atomistic, and theoretically grounded framework to address this behavior.

The incoherent contribution to the current has often been neglected in literature. However, in some systems, it can have a significant impact, leading to a substantial increase in the total current with bias [65]. In this work, we generally compute the total current using the original Meir-Wingreen formula and estimate I_{IC} by subtracting I_{C} from it, rather than applying Eq. (61). This approach requires only the Green’s function and avoids the need to store the many-body self-energy or to decompose it into Γ_{MB} and F_{MB} at each energy and momentum point, thereby reducing memory usage.

III. COMPUTATIONAL DETAILS

The DFT + NEGF calculations are performed using the LSDA [12,13] for the exchange-correlation functional. The leads’ self-energies are calculated using the semianalytic algorithm from Ref. [103]. Norm-conserving Troullier-Martins pseudopotentials [133] are employed to treat the core electrons. Valence states are expanded using a numerical atomic orbital basis set, which includes multiple ζ and polarized functions [97]. The electronic temperature is set to 300 K, and the real space mesh is determined by an equivalent energy cutoff of 400 Ry.

At zero-bias, the DFT density matrix, given by Eq. (6), is computed self-consistently using a 20×20 transverse $\mathbf{k}$ -point mesh. The integration over the energy is performed using a semicircular contour in the complex energy plane [32]. In this approach, 16 poles are used to represent the Fermi distribution, while 16 energy points are sampled along both the semicircular arc and the imaginary line that forms the contour. The converged density matrix is read as an input for a non-self-consistent DFT calculation using a 200×200 transverse $\mathbf{k}$ mesh to obtain the zero-bias DFT DOS and transmission coefficient. This is enough for resolving the transmission function over the transverse BZ in the case of tunneling. All energy values are shifted to align the Fermi level at 0 eV.

Finite-bias calculations are performed non-self-consistently, using the Hamiltonian of the central region calculated at zero-bias with an additional ramp potential [134] that reproduces the linear voltage drop across the MgO insulating barrier (vacuum gap) in Fe/MgO/Fe MTJ (Cu/Co/vacuum/Cu), namely $V_e(z) = -eVz/l + eV/2$ where $z = 0$ ($z = l$) corresponds to the beginning (end) of the barrier (gap), as in Fig. 1. Similar to the equilibrium case, a 200×200 transverse $\mathbf{k}$ -mesh is used to obtain the finite bias DFT DOS and transmission coefficient.

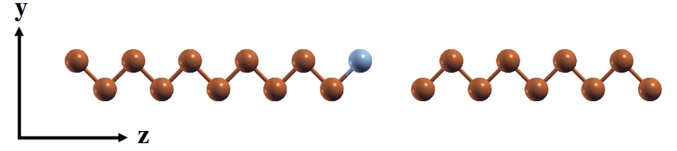


FIG. 4. The Cu/Co device investigated in this work. A Co layer is attached to a left Cu lead and separated from a right Cu lead by a 4 Å vacuum gap. Periodic boundary conditions are applied in the plane transverse to the stack.

DMFT calculations are performed assuming general orbital-dependent screened Coulomb interaction parameters for the $3d$ orbitals of Co and Fe, as mentioned in Sec. IID 1. The local Green’s function is calculated by summing the retarded Green’s function over 20×20 $\mathbf{k}$ points, whereas a 50×50 $\mathbf{k}$ mesh is considered to plot the DMFT DOS and transmission coefficient. The second-order contribution to the self-energy in the impurity solver is computed on an energy grid comprising 4000 (6000) points and extending from -10 to 10 eV (-25 to 15 eV) at low bias for the Cu/Co/vacuum/Cu system (Fe/MgO/Fe MTJ).

All calculations were performed on a 2022-era HPC system using a single compute node equipped with 44 Intel Xeon E5-2699 v4 (2.20 GHz) CPU cores and 256 GB of RAM.

The NEGF and projection algorithms are parallelized over energy points. Although linear-scaling ($O(N)$) NEGF implementations are available in our code and allow the treatment of very large systems [135], these were not employed here. Instead, matrix operations scale with the standard $O(N^3)$ complexity, where N is the number of basis orbitals, which in the present calculations is on the order of a few hundred.

For all systems, a single DFT + NEGF iteration requires on the order of one minute of wall-clock time and negligible memory. The projection step typically requires about 20 minutes of wall-clock time but is memory-intensive, requiring approximately 20 GB for the Cu/Co/vacuum/Cu junction (one correlated atom) and about 180 GB for the Fe/MgO junction (six correlated atoms). A quite computationally demanding part of the calculation is the evaluation of the impurity self-energy, which is executed in serial mode because it involves energy convolutions.

For the Fe/MgO/Fe junction, each iteration of the nonequilibrium DMFT self-consistency loop requires approximately 6 hours of wall-clock time (corresponding to about 264 CPU-hours). In contrast, for the Cu/Co/vacuum/Cu system, a single iteration requires approximately 1 hour of wall-clock time. The DMFT self-consistent loop typically converges within a few iterations, to tolerances of order 10^{-3} , defined as the maximum difference, over all energies, between the self-energy at two successive iterations.

IV. RESULTS

A. Cu/Co/vacuum/Cu

The first system investigated comprises a Co monolayer attached to a left-hand side Cu lead and separated from a right-hand side Cu lead by a 4 Å-long vacuum gap, as shown in Fig. 4(a). At finite bias, the potential drops within this gap. The setup mimics STM studies of magnetic surfaces.

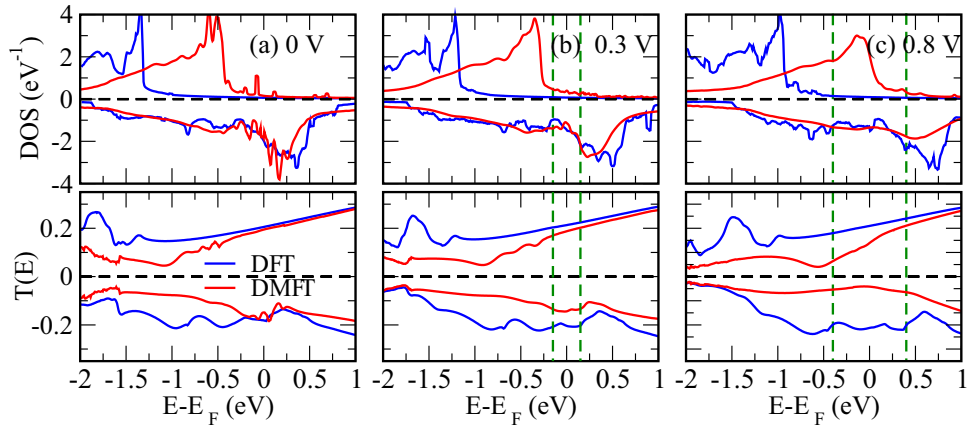


FIG. 5. DFT and DMFT $3d$ -Co PDOS and transmission coefficient at (a) $V = 0$, (b) 0.3 , and (c) 0.8 V. Spin-up (down) values are shown as positive (negative). The green dashed lines delimit the bias window.

While a fully quantitative STM simulation would require the use of a large supercell with the right lead attached to a tip [83]—whose geometry and electronic structure may be nontrivial (e.g., Refs. [134,136])—the essential nonequilibrium physics related to the potential drop and charge current flowing through the central region is expected to be well captured already by our simplified system. The correlated subspace, $\mathcal{C}$ is spanned only by the Co $3d$ orbitals, whereas the Cu $3d$ orbitals are fully occupied and therefore play a negligible role over the correlation effects. We employ an average on-site Coulomb and exchange interaction parameters of $U = 1.5$ eV and $J = 0.5$ eV, respectively. These values are found to reproduce well the experimentally observed energy positions of the $3d$ -Co states, as discussed below. A detailed analysis of the DOS, albeit limited to the zero-bias regime, for similar Co/Cu systems as a function of the interaction parameters is presented in Ref. [74], and is therefore not repeated here.

1. Zero-bias electronic structure

The $3d$ -Co PDOS calculated with DFT and DMFT at zero bias are shown in Fig. 5(a), illustrating the effect of static versus dynamical correlations. The DFT result (blue curve) exhibits a pronounced spin-splitting: the spin-up states (majority states) are almost entirely occupied, with a main peak at approximately -1.3 eV, whereas the spin-down states (minority states) extend from low energies across E_F up to about 0.7 eV.

DMFT leads to a redistribution of the PDOS (red curve), resulting in a reduced spin-splitting—qualitatively similar to what has been reported in many DMFT studies of transition metal ferromagnets [47,48,63–65,74]. Notably, our DMFT PDOS is in very good agreement with photoemission experiments for Co/Cu(001) surfaces, where the majority-spin peak is observed at $E - E_F \approx -0.5$ eV [137].

The energy shift of the $3d$ -Co states and the corresponding reduction of the spin splitting is due to real part of the retarded DMFT self-energy, shown in Fig. 6 (averaged over all five $3d$ orbitals). In the spin-up channel, $\text{Re}[\Sigma_{\text{avg}}^{\uparrow}(E)]$ is large and positive, pushing the PDOS upward in energy toward E_F . Conversely, in the spin-down channel, $\text{Re}[\Sigma_{\text{avg}}^{\downarrow}(E)]$

becomes negative at ~ 0.2 eV, shifting the high-energy part of the PDOS downward toward E_F and thereby sharpening its main peak.

The imaginary part of the retarded DMFT self-energy vanishes at E_F and exhibits the typical behavior for a Fermi liquid, namely $\text{Im}[\Sigma_{\text{avg}}^{\sigma}(E)] \propto -(E - E_F)^2$ at low energies. Furthermore, it is related to the lesser DMFT self-energy via the fluctuation-dissipation relation in Eq. (11), reflecting the occupation of electronic states according to the Fermi-Dirac distribution in equilibrium.

2. Finite-bias electronic structure

Upon the application of a finite bias voltage V , the $3d$ -Co PDOS shifts upward almost rigidly by $eV/2$, reflecting the strong coupling of the Co layer to the left lead. This effect is clearly visible in both the DFT and DMFT results shown in Figs. 5(b) for $V = 0.3$ V and 5(c) for 0.8 V, where the green dashed lines delimit the bias window. However, besides this somewhat trivial shift, the most interesting finding is that the imaginary part of the DMFT self-energy deviates from the Fermi-liquid behavior. Specifically, $\text{Im}[\Sigma_{\text{avg}}^{\uparrow}(E_F)]$ becomes finite and increases with bias (Fig. 6), indicating a loss of coherence due to electronic excitations. This displays in the progressive broadening and smoothing of the DMFT PDOS [Figs. 5(b) and 5(c)], particularly in the spin-down channel—an effect that is, of course, absent in the effective single-particle DFT picture.

The departure from the Fermi-liquid regime at finite bias is also evident in the breakdown of the fluctuation-dissipation theorem. Specifically, the lesser DMFT self-energy $\Sigma_{\text{avg}}^{\sigma}(E)$, shown in Figs. 6(e) and 6(f), displays a strong deviation from Eq. (11) for energies within the bias window. This signals that the electronic distribution function departs from a simple Fermi function. To highlight this aspect, in Figs. 7(b) and 7(c), we plot $\delta^{\sigma} = f(E, \mu_L) - \bar{F}_{\text{avg,MB}}^{\sigma}(E)$, where we take $f(E, \mu_L)$ [see Fig. 7(a)] as our reference equilibrium distribution, since the Co layer is much more strongly hybridized with the left-hand side lead, and $\bar{F}_{\text{avg,MB}}^{\sigma}(E)$ is the occupation of the correlated subspace averaged over the five $3d$ orbitals. The result shows that δ is positive for energies below $\mu_L = eV/2$, where $f = 1$, whereas it becomes negative for energies above

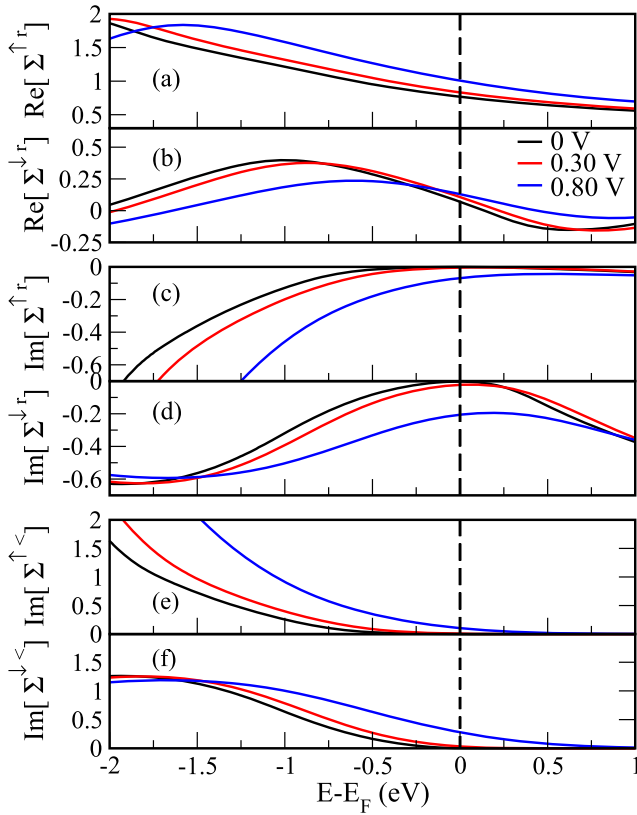


FIG. 6. DMFT self-energy for Co, averaged over all the 3d orbitals, at $V = 0, 0.3$, and 0.8 V. (Top) Real part of the retarded component. (Middle) Imaginary part of the retarded component. (Bottom) Lesser component.

μ_L , where $f = 0$. This indicates that, away from equilibrium, the electronic distribution is further smeared out. In practice, electrons are excited from occupied states to higher-energy empty states, leaving behind holes. This effect becomes significant in particular for $V \gtrsim 0.4$, a threshold that coincides with the entry of the spin-up DOS peak into the bias window, and directly reflects in the calculated current, as discussed in the following section.

3. Current

The charge current as a function of bias is plotted in Fig. 8(a). The DFT and DMFT results (blue and red lines) coincide at low V but begin to diverge for $V \gtrsim 0.4$. Although the deviation remains relatively small, the underlying transport mechanisms differ fundamentally. In DFT, the current is fully coherent, whereas in DMFT, the incoherent contribution (black line) increasingly dominates with bias, particularly in the spin-down channel.

The DFT current, obtained from the Landauer-Büttiker formula [Eq. (59)] as the integral of the transmission coefficient over the bias window, increases linearly with V , since $T(E)$ is smooth and nearly constant across a wide energy range (see Fig. 5). In the spin-up channel, transport is dominated by delocalized 4s states, which give a large transmission. In contrast, the 3d-Co states lie well below the bias window for all V considered, as seen in the PDOS in Figs. 5(b) and 5(c), and thus they are irrelevant for transport. In the spin-down

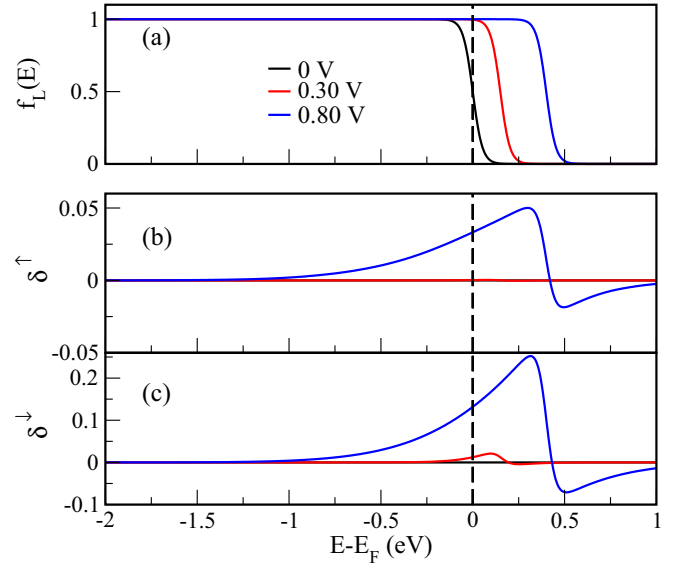


FIG. 7. (a) Fermi function of the Cu left lead attached to the Co layer at $V = 0, 0.3$, and 0.8 V. (b) and (c) show the difference $\delta^\sigma = f(E, \mu_L) - F_{MB}^\sigma(E)$, averaged over all 3d orbitals, for spin-up and spin-down channels, respectively.

channel, the 3d-Co states play instead a more important role as they are located at energies near E_F . However, their strong hybridization with the Cu left-hand side electrode causes a substantial spectral broadening, yielding a transmission coefficient that is equally large in magnitude and smooth in energy. Interestingly, this leads to spin-up and spin-down currents,

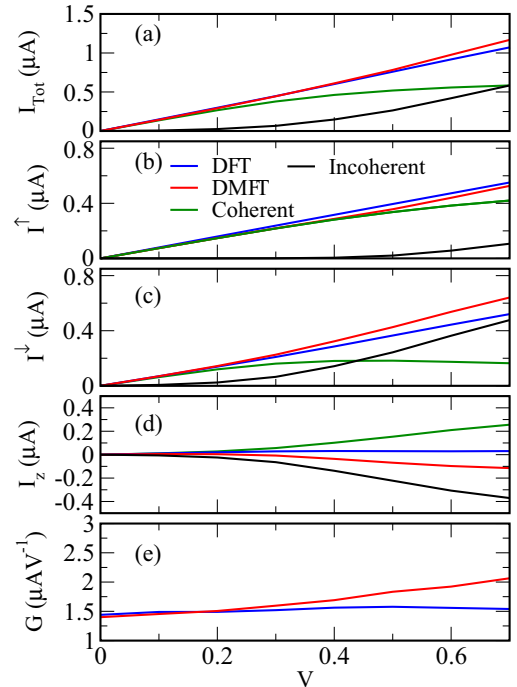


FIG. 8. (a) Total, (b) spin up, (c) spin down, (d) spin current, and (e) conductance within DFT and DMFT, decomposed into its coherent and incoherent contributions, as functions of the bias.

which are nearly identical: namely, within the DFT picture, a single Co layer cannot spin-polarize the current in our set-up.

When the calculation is performed at the DMFT level, the coherent current (green line in Fig. 8) dominates at low bias and, as in DFT, increases linearly with V . At higher bias, however, it saturates, plateauing around 0.6 V, while the incoherent current rises sharply. This results in a crossover between coherent and incoherent transport at $V \sim 0.7$ V. Furthermore, the crossover is accompanied by the emergence of a negative spin current, $I^z = I^\uparrow - I^\downarrow$ [see Fig. 8(d)], indicating that bias-driven correlation effects induce spin-filtering.

The behavior of the DMFT coherent current arises from the reshaping of the transmission coefficient due to dynamical correlations [Fig. 5], most notably the ~ 0.8 eV upward shift of the spin-up $3d$ states, which brings them into the bias window already at ~ 0.4 V. At the same time, the transmission through the Co $3d$ states is suppressed relative to the DFT result because of the damping and coherence loss of the electronic excitations (see the red curves in the bottom panels of Fig. 5). This effect is encoded in the imaginary part of the DMFT self-energy. As V increases and the system is driven further away from equilibrium and from a Fermi-liquid behavior, this suppression counterbalances the widening of the bias window, and eventually results in the coherent current plateau. The effect is stronger in the spin-down channel, where $3d$ states lie in the bias window even at low V , whereas the spin-up channel contributes only once the bias exceeds ~ 0.4 V.

At a microscopic level, the suppression of coherent transport originates from bias-driven inelastic scattering processes, which conversely generate incoherent current. An electron entering the Co layer from the right-hand side lead can scatter off another electron, creating an electron-hole pair excitation and transferring part of its energy, before being collected in an empty state of the right-hand side lead. The electron-hole pair excitations directly manifest in the nonequilibrium distribution function as already observed in Fig. 7. Scattering between electrons of opposite spin occurs with the full interaction strength U , whereas same-spin scattering is weaker, occurring with an effective interaction $U - J < U$, reduced because of the Hund's coupling. As a result, opposite-spin scattering processes dominate and produce the strongest effect on transport.

Several scattering processes can be specifically identified, as shown in Fig. 9. A spin-down electron can create a spin-down electron-hole pair excitation [panel (b)]. Since the spin-down $3d$ states span the Fermi level, this process can occur even at low bias, resulting in a nonzero spin-down incoherent current already at small voltages, as seen in Fig. 8(c). As V increases and the spin-up $3d$ states enter the bias window (approximately 0.35 eV), virtual transfer of spin-up electrons into the right lead becomes possible, leaving behind spin-up particle-hole pairs that can scatter with spin-down electrons [panel (c) in Fig. 9]. Since this process involves opposite-spin interactions, it is stronger and leads to the sharper upturn in the spin-down incoherent current at $V \sim 0.3$ V seen in Fig. 8(c). Finally, at higher bias ($V \gtrsim 0.4$ V), once many spin-up states become available inside bias window, spin-up electrons likewise start contributing to incoherent transport by transferring energy to generate electron-hole pair excitations of both same

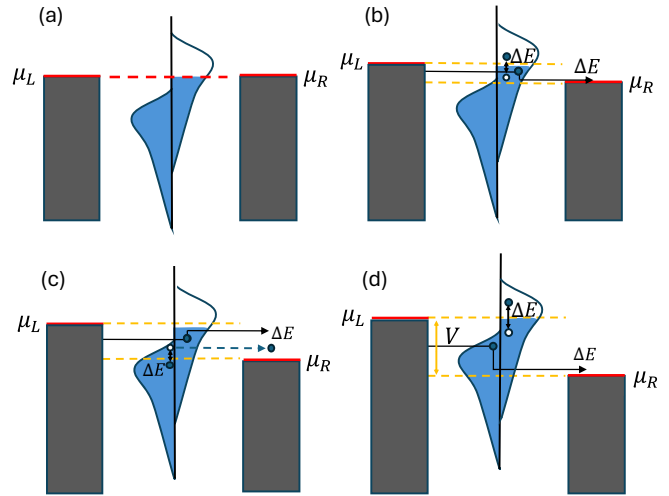


FIG. 9. Dominant scattering processes contributing to the incoherent current. Dark gray rectangles represent the Fermi boxes of the L and R leads, displaced by $\mu_L - \mu_R = V$, with the schematic Co $3d$ PDOS shown in between. The bias window is delimited by yellow dashed lines. Electrons (holes) are represented as filled (empty) circles. (a) Zero-bias energy-level alignment. (b) A spin-down electron creates a spin-down electron-hole pair excitation and scatters into the right lead. (c) As the bias V increases and the spin-up $3d$ states approach the bias window, virtual transfer of spin-up electrons into the right lead becomes possible (dashed blue arrow), leaving behind spin-up particle-hole pairs that can scatter with spin-down electrons. (d) Once the bias window is large enough to include a significant portion of the spin-up PDOS, spin-up electrons contribute directly to incoherent transport by exciting opposite- (or same-) spin electron-hole pairs and scattering inelastically into the right lead.

and opposite spin [panel (d) in Fig. 9]. This is reflected in the rise of the spin-up incoherent current in Fig. 8(b).

Notably, the incoherent current contribution gives an increase in the differential conductance, dI/dV at $V \sim 0.25$ eV, as seen in Fig. 8(e). This feature is consistent with STS measurements at positive bias on Co, though observed not in extended layers but at the centers of Co nanoislands on Cu [138,139]. While STS results are often interpreted in terms of coherent transport through the spin-up $3d$ Co bands within a DFT-based Tersoff-Hamann picture [140], our results indicate that this interpretation is incomplete. DMFT is crucial both to correctly position the spin-up $3d$ states, as DFT places them too low in energy, and to capture the relevant scattering processes.

While such processes have been included within the NEGF formalism for s - d exchange models [141,142] and in *ab initio* approaches combining NEGF with time-dependent DFT for describing magnetic adatoms [143,144], they are usually neglected for surfaces. Our calculations demonstrate how these limitations can be overcome.

In summary, our analysis demonstrates that nonequilibrium correlations, captured by DMFT, qualitatively change the nature of the system, reshaping its electronic structure. The crossover between coherent and incoherent contributions under bias provides a natural explanation for experimental conductance features, calling for a revision of the commonly

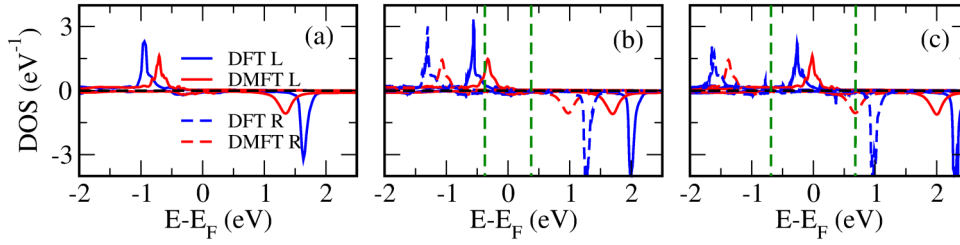


FIG. 10. The DFT and DMFT PDOS of the Δ_1 orbitals of the Fe layer to the left (L) and to the right (R) of the MgO barrier in Fe/MgO/Fe MTJ. Results are presented at (a) $V = 0$, (b) 0.75, and (c) 1.375 V. Spin-up (down) values are shown as positive (negative).

used single-particle pictures, and highlights the importance of many-body scattering in transport through magnetic layers.

B. Fe/MgO/Fe

The second system investigated is the prototypical Fe/MgO/Fe MTJ, which is modelled here by using the same set-up as in reference [65], shown in Fig. 2. The calculations presented in Ref. [65] were based on the rigid-shift approximation of Sec. IID 4, which was assumed valid *a priori*. In contrast, here we perform fully nonequilibrium calculations with our improved framework to explicitly test the validity of that approximation.

The CR of our MTJ consists of three Fe layers on each side of a six-layer MgO barrier and is connected to generic simple metallic leads, which are computationally simulated as a *bcc* lattice of Au atoms using only the 6s orbitals as a basis set. This effectively implements a wide-band approximation. The layers on the left-hand side of MgO can have a magnetization vector either parallel or antiparallel to that of the right-hand side layer, thereby realizing the two magnetic configurations of the MTJ. In this work, we focus exclusively on the parallel configuration and refer the reader to Ref. [65] for a discussion of the antiparallel case and the TMR. The correlated subspace $\mathcal{C}$ comprises the Fe 3d orbitals, whereas the MgO barrier is treated within KS DFT, since it is weakly correlated. In fact, its conduction and valence bands have predominantly Mg-3s and O-2p character [145], respectively. The DMFT calculations use the average screened interaction parameters $U = 2.0$ eV and $J = 0.5$ eV, which were found appropriate to describe Fe surfaces in previous studies using our implementation of DFT + DMFT [64].

1. Electronic structure at zero bias

Tunneling through Fe/MgO/Fe MTJs is dominated by spin-split evanescent states with Δ_1 symmetry, which have the slowest decay rate inside the MgO barrier [3,146]. Within our reference frame, where the transport direction is aligned along the z axis, and for a cubic space group, the Δ_1 states are those that transform as a linear combination of s , p_z , and d_{z^2} atomic orbitals. The zero-bias Δ_1 -PDOS of the Fe atom at the Fe/MgO interface are presented in Fig. 10(a) for DFT and DMFT, respectively. By symmetry, the results are identical for the two Fe layers at opposite sides of the junction.

Within DFT, sharp spin-split peaks appear at $E - E_F \approx -0.9$ and ≈ 1.6 eV for the spin-up and spin-down channels, respectively. In contrast, DMFT yields a substantial shift in energy of these peaks, resulting in a reduction of the spin-

splitting, as observed in the Cu/Co system. This is due to the real part of the self-energy, and it is accompanied by a pronounced broadening caused by the imaginary part, which vanishes at E_F as a consequence of its Fermi-liquid nature. Our results reproduce exactly those reported in Ref. [65], except for small quantitative differences in the peak positions that can be attributed to the different treatment of the double-counting correction (see discussion in Sec. IID 6). Overall, our results provide a detailed account of correlation effects at the Fe/MgO interface.

2. Electronic structure at finite bias

At finite bias, the electrostatic potential drops within the MgO barrier. The Fe Δ_1^σ state at the left-hand side Fe/MgO interface, $\Delta_{1(L)}^\sigma$, shifts upward in energy by $+eV/2$, while the corresponding state at the right-hand side, $\Delta_{1(R)}^\sigma$, shifts downward by $-eV/2$, as can be seen in Figs. 10(b) and 10(c). Differently from the Co/Cu case, where the self-energy undergoes significant changes at large bias, in the Fe/MgO/Fe MTJ the imaginary part of the self-energy [Figs. 11(c) and 11(d)] just shifts with energy, essentially retaining its Fermi-liquid character, to vanish quadratically at $\mu_L = E_F + eV/2$ ($\mu_R = E_F - eV/2$) for the Fe atoms at the left-hand (right-hand) side interface.

Since the correlated Fe atoms are strongly coupled to one lead and separated from the opposite lead by a sufficiently thick MgO barrier—so that their interaction with the latter is negligible—they can be considered in equilibrium with the lead which they are coupled to. Indeed, we find that the retarded and lesser self-energies satisfy the fluctuation-dissipation relation, Eq. (11), at all V , as shown in Fig. 11. In other words, the electronic distribution of each Fe atom remains equal to the Fermi function of the lead to which it is coupled, in contrast to the behavior observed for the Cu/Co/vacuum/Cu system shown in Fig. 7. This directly demonstrates that the rigid-shift approximation holds for this system and confirms *a posteriori* its application in Ref. [65]. As a result, the electronic structure at finite bias essentially follows from the zero-bias one, with genuine nonequilibrium correlation effects being negligible.

3. Current

The charge current as a function of the bias is shown in Fig. 12. At low biases, the DFT and DMFT results nearly coincide, with a fully coherent current dominated by the spin-up component $I^\uparrow$. This is because transport is mostly through the highly conductive s states forming the Fe Δ_1 band. As

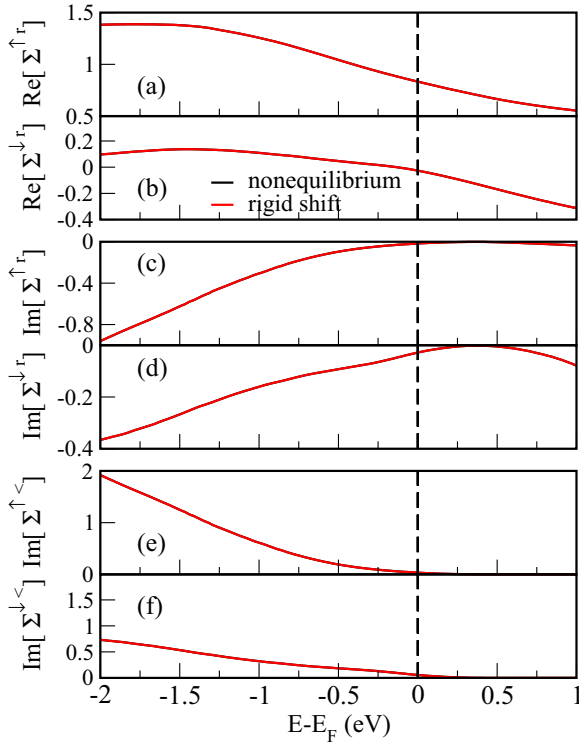


FIG. 11. Comparison of the DMFT self-energy with and without the rigid-shift approximation at $V = 0.75$ V for the Fe monolayer closest to the left-hand side of the MgO barrier in the Fe/MgO/Fe MTJ. The results are averaged over the d orbitals. Top panels (a) and (b) show the real part of the retarded component for the spin-up and spin-down channels, respectively. Middle panels (c) and (d) show the imaginary part of the retarded component for the spin-up and spin-down channels, respectively. Bottom panels (e) and (f) show the lesser component for the spin-up and spin-down channels, respectively. The results obtained with the rigid-shift approximation (red curves) are indistinguishable from those without it (black curves) and therefore completely overlap.

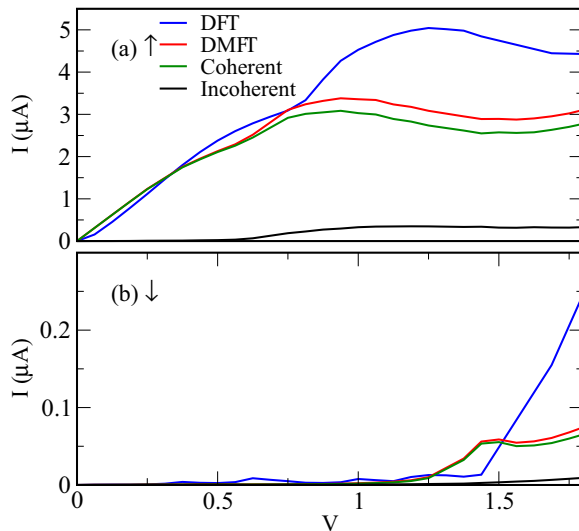


FIG. 12. Spin (a) up and (b) down currents for the Fe/MgO/Fe MTJ calculated with DFT and DMFT, and decomposed into coherent and incoherent contributions as a function of bias.

the bias increases, however, the spin-up Δ_1 peak—dominated by d_{z^2} states from the left Fe layers—enters the bias window [Fig. 10(b) and 10(c)], and $I^\uparrow$ saturates into a plateau. The crossover from the linear regime to the plateau occurs in DMFT at a V lower than in DFT, reflecting the different energy positions of the spin-up d_{z^2} states predicted by the two methods. More interestingly, at this crossover bias, DMFT also reveals the onset of an incoherent contribution to the current, similar to what we found in the Cu/Co junction. In this case, however, the incoherent component accounts for only about 10% of the total spin-up current. The underlying scattering process involves an incoming spin-up electron interacting with an electron-hole pair of the same spin, which is activated when the spin-up Δ_1 state of the left layers enters the bias window at $V \approx 0.7$ V.

In the spin-down channel, the current remains negligible as long as the spin-down Δ_1 state is outside the bias window. Once this state enters the window, the spin-down current suddenly increases, with the threshold in DMFT being at lower energies than in DFT. This, again, reflects the DMFT shift in the energy of the Δ_1 state.

In summary, although the applied bias has only a weak effect on Fe, bias-induced renormalization of the Δ_1 energy position and scattering from dynamical correlations remain important in Fe/MgO/Fe. This is consistent with the findings of Ref. [65], where their experimental relevance is discussed in detail. DMFT thus offers a better description than DFT, even for this technologically relevant system.

V. CONCLUSION

We have introduced a framework that combines DFT, DMFT and the NEGF method to study steady-state transport in two-terminal devices beyond the linear response, and including dynamical electron-correlation effects. By employing second-order perturbation theory as an impurity solver, our approach is well-suited for moderately correlated $3d$ transition metals. However, the method can in principle be extended to more strongly correlated materials, provided that reliable multiorbital nonequilibrium solvers become available—an outstanding challenge at present.

As a first application, we have investigated a single Co monolayer sandwiched between Cu electrodes, where a vacuum gap is introduced at one side to enable a potential drop under bias. This is a configuration that resembles that encountered in an STM/STS experiment. Our results show that nonequilibrium dynamical correlation in Co drives a qualitative transition from Fermi-liquid to non-Fermi-liquid behavior, originating from inelastic scattering of electrons with collective electron-hole excitations. This re-shapes the electronic structure and generates incoherent contributions to the current, accompanied by a sharp increase in conductance around $V \sim 0.4$ V, where the spin-up Co states enter the bias window. These findings provide a natural explanation for STS results, calling for a revision of commonly used single-particle models, and underscore the critical role of many-body scattering in transport through magnetic layers.

We have then applied the framework to an Fe/MgO/Fe MTJ. We find that the Fe layers remain close to equilibrium, showing that the rigid-shift approximation of Ref. [65] holds

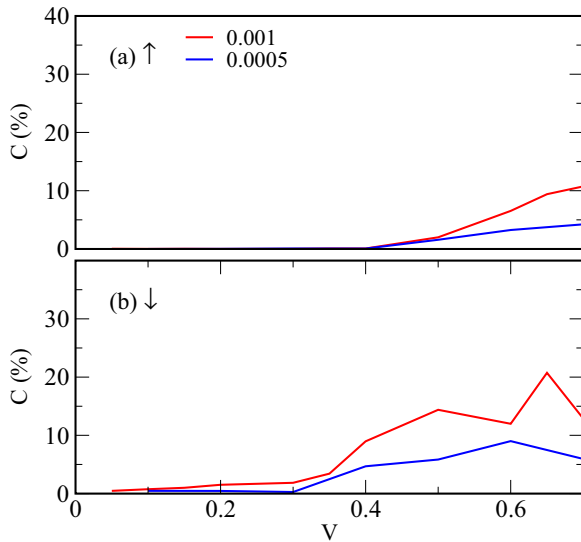


FIG. 13. The current conservation for the (a) spin up and (b) spin down channel for the Cu/Co/vacuum/Cu system.

for this system. As a result, the electronic structure at finite bias essentially follows that at zero bias, with genuine nonequilibrium correlation effects being negligible. Nonetheless, we still observe the emergence of a partially incoherent current as the Fe $3d$ states enter the bias window, in agreement with previous results [65], demonstrating that dynamical correlation affects transport even in this technologically relevant system.

Overall, our framework provides a pathway toward modeling spintronic devices with explicit many-electron effects at finite bias in far-from-equilibrium conditions. Looking ahead, this framework could further be extended beyond DMFT to compute other transport characteristics such as shot noise—expressed as a current-current correlator—and therefore by exploring generalized nonequilibrium vertex corrections, as developed in the context of disorder averaging [147]. Moreover, our current DMFT implementation can be readily generalized to compute spin currents in noncollinear and

spin-orbit-coupled systems by building on the computational modules already employed in standard DFT + NEGF calculations [28,148]. This will make it possible to study how scattering processes, such as those identified here, influence spin-transfer torque [41,42] and spin-orbit torque [149,150], offering new insights into current-induced spin dynamics beyond the state-of-the-art *ab initio* single-particle treatment.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: CURRENT CONSERVATION

Charge conservation in DFT + NEGF + DMFT is quantified as $C = (2|\frac{I_L - I_R}{I_L + I_R}|) \times 100\%$. Figure 13 shows C for the Cu/Co/vacuum/Cu system out-of-equilibrium, with the DMFT self-energies converged in the self-consistent loop of Fig. 3 to tolerances of 10^{-3} and 5×10^{-4} . Reducing the tolerance improves current conservation across the bias range. The data in Fig. 8 use the 5×10^{-4} tolerance, yielding a maximum deviation of 10%. This demonstrates that our DFT + NEGF + DMFT framework provides qualitatively—and potentially quantitatively—reliable predictions for transport in realistic heterostructures.

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