Time-dependent spintronic anisotropy in magnetic molecules

Kacper Wrześniowski* and Ireneusz Weymann
Faculty of Physics, Adam Mickiewicz University, ul. Uniwersytetu Poznańskiego 2, 61-614 Poznań, Poland
(Dated: June 9, 2020)

We theoretically study the quench dynamics of induced anisotropy of a large-spin magnetic molecule coupled to spin-polarized ferromagnetic leads. The real-time evolution is calculated by means of the time-dependent density-matrix numerical renormalization group method implemented within the matrix product states framework, which takes into account all correlations in very accurate manner. We determine the system’s response to a quench in the spin-dependent coupling to ferromagnetic leads. In particular, we focus on the transient dynamics associated with crossing from the weak to the strong coupling regime, where the Kondo correlations become important. The dynamics is examined by calculating the time-dependent expectation values of the spin-quadrupole moment and the associated spin operators. We identify the relevant time scales describing the quench dynamics and determine the influence of the molecule’s effective exchange coupling and leads spin-polarization on the dynamical behavior of the system. Furthermore, the generalization of our predictions for large values of molecule’s spin is considered. Finally, we analyze the effect of finite temperature and show that it gives rise to a reduction of magnetic anisotropy by strong suppression of the time-dependent spin-quadrupole moment due to thermal fluctuations.

I. INTRODUCTION

Molecular magnetism is a rapidly developing area of theoretical and experimental research, providing concepts for novel applications in spintronic devices and quantum technologies [1–10]. Single-molecule magnets (SMM), in particular those of large spin (S ≥ 1), are especially appealing due to their unique magnetic characteristics and a wide perspective of engineering and synthesizing new specimen with sought properties [3]. One prominent feature present in magnetic molecular systems is the uniaxial magnetic anisotropy, which leads to the magnetic bistability and suppression of spin-reversal processes [11–15]. It is a property of crucial importance for the memory storage and information processing applications. Additionally, when transverse anisotropy component is considerable, quantum tunneling of magnetization may occur [16–19]. The transport properties of magnetic molecules have already been extensively studied [20–31], including the influence of the Kondo effect [32–36] in the strong coupling regime [37–43]. However, when the physics of SMM systems incorporates spintronics, new prominent effects are revealed, including switching with spin-polarized currents, Berry-phase blockade or spin-polarized anisotropy among many others [44–52]. In fact, the latter effect is of particular interest, as it allows for generation of magnetic anisotropy in spin-isotropic molecules [53].

Moreover, the dynamics of molecular systems, an important aspect of the ongoing research in molecular magnetism, has recently gained a lot of attention and has been explored both experimentally [54–56] and theoretically [57–60]. Thus, broadening further the knowledge and understanding of transport and dynamical properties of large-spin molecules is important both because of exciting fundamental aspects as well as due to possible applications in modern nanoelectronics, spintronics and quantum information.

Motivated by recent progress within this field, in this paper we investigate the dynamical behavior of a large-spin magnetic molecule attached to spin-polarized leads, with an emphasis on the buildup of quadrupolar exchange field, referred to as spintronic anisotropy. Commonly, the intrinsic magnetic anisotropy arises from the spin-orbit interaction. However, it has been shown that the ferromagnetic proximity effect [61–64] can generate significant magnetic anisotropy in molecular systems in form of an effective exchange field [53]. The advantage of this approach is the possibility to electrically control both the magnitude of the anisotropy and the spin state of the system. When the coupling strength to external contacts is varied, a rapid change in the magnetic properties of the system occurs [53, 65]. In particular, the molecule’s quadrupolar moment is significantly reduced, when the system is tuned from the weak to the strong coupling regime. This rapid change of the moment is due to the Kondo screening of the molecule’s orbital level spin and is a non-trivial many-body effect resulting from the interplay of magnetism and the Kondo physics. It was also shown experimentally that quadrupole interaction in SMM systems has an important influence on tunneling dynamics [66]. In a real setup, tuning between the different coupling regimes can be achieved by electrically shifting the tunnel barriers with respective gates.

Considering all the above, we focus on the theoretical study of the quench dynamics of the spintronic quadrupole moment due to the Kondo correlations. In particular, we identify the universal time scale for the dynamics describing the quench of spintronic anisotropy. Moreover, we examine the influence of the magnitude of
effective exchange coupling, leads spin-polarization and total spin of the molecule on discussed dynamical effects. Lastly, we also analyze the influence of finite temperature, showing that in certain range of temperatures, a strong suppression of magnetic properties is predicted. In pursuance of the precise analysis of the system’s response to the considered quench in the strong coupling regime, we resort to the Wilson’s numerical renormalization group (NRG) method [67–69]. We use the extended implementation allowing for studying the time evolution of the system, namely, the time-dependent numerical renormalization group (tNRG) [70–74]. This method allows for taking into account all the correlations in a fully non-perturbative manner and, thus, generating reliable predictions for the dynamics of the system under investigation.

This paper is structured as follows. Section II consists of the Hamiltonian description of the considered system, the overview of the quench protocol and a summary of the numerical renormalization group method used for calculations of time-dependent expectation values of local observables. In Sec. III we present the numerical results and relevant analysis for the quantum quenches in the coupling strength from the weak to the strong coupling regime. We also present and discuss the effects of finite temperature on dynamical behavior. Finally, the work is concluded in Sec. IV.

II. THEORETICAL FRAMEWORK

A. Hamiltonian

The effective spin Hamiltonian of a molecular magnet expressed only with spin coordinates can be written as

$$H_{\text{eff}} = B S_z + D Q_{zz}. \quad (1)$$

Here, $S_z$ is the $z$-th component of the total spin $S$, $Q_{zz} \equiv S_z^2 - S(S+1)/3$ is the $z$-th diagonal tensor element of the spin-quadrupole moment, $B$ is a dipolar field corresponding to external magnetic field, $D$ is a quadrupolar field related to intrinsic spin-orbit interaction. This approximate approach is well-established for convenient description of the system’s spectrum and is often used to interpret the spectroscopic data [3]. In our considerations, however, the above-introduced quantities are generated purely by the spin-dependent coupling to ferromagnetic leads [53, 62], which allows for tuning of both $B$ and $D$ by electrical means—the property that makes this approach advantageous from the application point of view.

In order to analyze the dynamical behavior of the system with tNRG and, especially, capture all the ferromagnetic-proximity induced effects, we model the entire system in the following way. Magnetic molecule is described by a single molecular level, through which the electronic transport takes place, which is exchange-coupled to magnetic core of the molecule specified by the effective spin $S_{MC}$. Thus, the molecule can be expressed by the Hamiltonian

$$H_{\text{SMM}} = \varepsilon n + U n_+ n_- - J S_{MC} \cdot s. \quad (2)$$

where the level occupation is expressed as $n = n_+ + n_- = d^{\dagger}_\sigma d_{\sigma}$, with $d^{\dagger} (d_{\sigma})$ being the fermionic creation (annihilation) operator for an electron with spin $\sigma$. The molecular level energy is denoted by $\varepsilon$ and the Coulomb correlations are described by $U$. We assume ferromagnetic exchange interaction $J > 0$ between the spin of the electron on the orbital level $s$ and the magnetic core spin $S_{MC}$. The total spin is then expressed as $S = S_{MC} + s$.

The molecule is coupled to left and right spin-polarized ferromagnetic leads [75–78], see Fig. 1. Here, we exploit the correspondence between the coupling to two leads at equilibrium with the magnetic moments arranged in the parallel configuration and the coupling to a single ferromagnetic lead. The equivalence can be shown by carrying out an orthogonal transformation [33], after which the central part of the system couples exclusively to even linear combination of reservoir’s operators with effective coupling strength $\Gamma^\sigma = \Gamma^\sigma_L + \Gamma^\sigma_R$ and spin polarization $p$. Consequently, the leads can be described by an effective reservoir of noninteracting quasiparticles

$$H_{\text{Lead}} = \sum_{k\sigma} \varepsilon_{k\sigma} c^{\dagger}_{k\sigma} c_{k\sigma}, \quad (3)$$

where $c^{\dagger}_{k\sigma} (c_{k\sigma})$ is the creation (annihilation) operator of an electron with momentum $k$, spin $\sigma$ and energy $\varepsilon_{k\sigma}$, which is given by appropriate linear combination of electron operators in the left and right leads. On the other hand, the spin-dependent coupling is specified by the tunneling term

$$H_{\text{Tun}} = \sum_{k\sigma} V \sigma (c^{\dagger}_{k\sigma} d_{\sigma} + \text{H.c.}), \quad (4)$$

where $c^{\dagger}_{k\sigma} (c_{k\sigma})$ is the creation (annihilation) operator of an electron with momentum $k$, spin $\sigma$ and energy $\varepsilon_{k\sigma}$, which is given by appropriate linear combination of electron operators in the left and right leads. On the other hand, the spin-dependent coupling is specified by the tunneling term

![FIG. 1. Schematic of the considered system. A large-spin molecule with a molecular level is coupled to external spin-polarized leads with the spin-dependent coupling strengths $\Gamma^\sigma_L$ and $\Gamma^\sigma_R$, for the left and right lead. The molecular level is exchange-coupled to magnetic core spin of the molecule with strength $J$. Coulomb correlations of the molecule are denoted by $U$.](image-url)
where $V_\sigma$ are the effective tunnel matrix elements, assumed to be momentum independent.

The spin-dependent coupling between the molecule and the effective lead is expressed as, $\Gamma^\sigma = \pi \rho^\sigma |V_\sigma|^2$, with $\rho^\sigma$ being the spin-dependent density of states of ferromagnetic electrodes. By introducing the spin polarization of the leads $p$, the coupling strength can be written in the following manner, $\Gamma^{\uparrow \downarrow} = \Gamma (1 \pm p)$, with $\Gamma^{\uparrow \downarrow}$ denoting the coupling to the spin-up (spin-down) electron band of the ferromagnetic reservoir and $\Gamma = (\Gamma^\uparrow + \Gamma^\downarrow)/2$.

Finally, the full Hamiltonian of the considered system reads

$$H = H_{\text{SMM}} + H_{\text{Lead}} + H_{\text{Tun}}. \quad (5)$$

### B. Quench protocol and NRG implementation

The time-dependent Hamiltonian describing the dynamics after a quantum quench has the following general form

$$H(t) = \theta(-t)H_0 + \theta(t)H, \quad (6)$$

where the Hamiltonian $H_0$ denotes the initial Hamiltonian of the system. On the other hand, $H$ is the Hamiltonian describing the time evolution after the sudden quench at time $t = 0$ and $\theta(t)$ is the Heaviside step function. Both Hamiltonians have a form outlined in Eq. (5) with appropriate parameters modified according to the evaluated quench. The time-dependent expectation value of a given local operator $O(t)$ can be calculated from

$$O(t) \equiv \langle O(t) \rangle = \text{Tr} \{ e^{-iHt} \rho_0 e^{iHt} O \}. \quad (7)$$

Here, $\rho_0$ is the initial density matrix of the system described by the Hamiltonian $H_0$.

Let us now briefly discuss the most important aspects concerning the NRG implementation of the quench calculations [67–69]. The essential part of the NRG procedure is the logarithmic discretization of the conduction band followed by mapping of the discretized Hamiltonian to a one-dimensional tight-binding chain called the Wilson chain [68]. This is performed for both Hamiltonians $H$ and $H_0$. Subsequently, the two Hamiltonians are independently solved in an iterative fashion using the NRG procedure [69]. At each step of iteration, there are states that are used to construct the state-space of the next iteration and the states that are discarded. The discarded states are used to create the full many-body eigenbases [70]

$$\sum_{nse} |nse\rangle_0^D \langle nse|_0 = 1 \quad \text{and} \quad \sum_{nse} |nse\rangle^D \langle nse| = 1, \quad (8)$$

of both Hamiltonians, $H_0$ and $H$, respectively and to construct the full density matrix $\rho_0$ at temperature $T \equiv 1/\beta$ [79]

$$\rho_0 = \sum_{nse} e^{-\beta E_{\text{nmn}}^D} |nse\rangle_0^D \langle nse|_0, \quad (9)$$

where

$$Z \equiv \sum_{nse} e^{-\beta p^D_{\text{nnn}}} \quad (10)$$

is the partition function. Here, $s$ denotes a state at Wilson site $n$, while $e$ corresponds to an environmental state describing the rest of the chain.

The time-dependent expectation value $\langle O(t) \rangle$ of an operator $O$ can be conveniently evaluated in the frequency space and then Fourier-transformed to the time domain [80]. The frequency-dependent expectation value $\langle O(\omega) \rangle$ of a local operator $O$ expressed in the corresponding eigenstates of the two Hamiltonians is given by [81]

$$\langle O(\omega) \rangle = \sum_{n'n''} \sum_{ss'e} X |nse\rangle|w_{n''}\rho_{0n'}|ns'e\rangle^X \cdot X' \langle nse|O|ns'e\rangle \delta(\omega + E_{ns} - E_{ns'}). \quad (11)$$

where $X = K(X = D)$ denotes a kept (discarded) state. Here, $\rho_{0n'}$ is the contribution of the density matrix coming from iteration $n''$ and $w_{n''}$ is the corresponding weight.

We also use NRG to determine the linear-response conductance between the two ferromagnetic leads from the following formula [82]

$$G = \frac{e^2}{\hbar \pi} \int d\omega \left( \frac{\partial f}{\partial \omega} \right) \{ (1 + p)A_1(\omega) + (1 - p)A_1(\omega) \}, \quad (12)$$

where $A_\sigma(\omega)$ is the molecular level's spectral function, defined as $A_\sigma(\omega) = -1/(1/\pi) \text{Im} \langle \langle d_\sigma^\dagger(d_\sigma)^R \rangle \rangle_\omega$, with $\langle \langle d_\sigma^\dagger(d_\sigma)^R \rangle \rangle_\omega$ being the Fourier transform of the retarded Green’s function $\langle \langle d_\sigma|d_\sigma^\dagger \rangle \rangle_1^R = -i\theta(t)\langle \langle d_\sigma(t),d_\sigma^\dagger(0) \rangle \rangle_1$, $A(X, D)$.

For the NRG calculations we used the discretization parameter $2 \leq \Lambda \leq 3$, set the length of the Wilson chain to be $N = 80$ and kept at least $N_K = 4000$ energetically lowest-lying states at each iteration. In order to suppress the band discretization effects, we also used the Oliveira’s z-averaging [83] by performing calculations for $N_z = 4$ different discretizations. More details and technicalities concerning the implementation of calculations can be found in Ref. [81].

### III. RESULTS AND DISCUSSION

#### A. Static properties of the molecule

In order to obtain a better understanding of the magnetic correlations present in the considered system, let us first examine the static properties. As the main focus is put on the quadrupolar field, we tune the system to the particle-hole symmetry point by setting the energy of the orbital level to $\varepsilon = -U/2$. As a result, the dipolar field vanishes [61] and only quadrupolar term is present in the system [second term of Eq. (1)]. This approach allows us to precisely describe the generated uniaxial anisotropy, quantified by the amplitude $D$, see Eq. (1).
orbital level was fully screened by the Kondo correlation values of spin operators influence the value of the spin-quadrupole moment (dark-blue line) is that in the weak-coupling regime, where the spin-quadrupole moment is saturated acquiring \( \langle Q_{zz} \rangle = S(2S - 1)/3 = 1 \), to the strong coupling regime. In the latter case, the Kondo correlations are present and the moment is reduced to \( \langle Q_{zz} \rangle = S_{MC}(2S_{MC} - 1)/3 = 1/3 \).

In order to study the dynamics of this effect, we perform a quantum quench of the initial Hamiltonian \( H_0 \) in the coupling strength from \( \Gamma_0/U = 0.002 \) to various final values of \( \Gamma/U \) in the range of strong coupling where the Kondo effect is well-established. In Fig. 3(b) we present the spin-quadrupole moment \( \langle Q_{zz} \rangle \) (solid lines) and the linear-response conductance \( G \) (dashed lines) plotted versus \( \Gamma \) for different values of exchange coupling \( J \).

**B. Dynamics of quadrupole moment**

The important alteration in the SMM’s magnetic properties is when the coupling strength \( \Gamma \) is switched from the weak coupling regime, where the spin-quadrupole moment is saturated acquiring \( \langle Q_{zz} \rangle = S(2S - 1)/3 = 1 \), to the strong coupling regime. In the latter case, the Kondo correlations are present and the moment is reduced to \( \langle Q_{zz} \rangle = S_{MC}(2S_{MC} - 1)/3 = 1/3 \).

In Fig. 2(a) we present the spin-quadrupole moment \( \langle Q_{zz} \rangle \) (solid lines) and expectation values of the corresponding spin operators, \( \langle S^2 \rangle \) and \( \langle S_z^2 \rangle \), as the coupling strength \( \Gamma \) is varied. The general behavior of spin-quadrupole moment (dark-blue line) is that in the weak-coupling regime \( \Gamma/U \lesssim 10^{-1} \), it acquires the value \( \langle Q_{zz} \rangle = S(2S - 1)/3 = 1 \), while in the strong-coupling regime \( \Gamma/U \gtrsim 10^{-1} \) this value is strongly reduced to \( \langle Q_{zz} \rangle = S_{MC}(2S_{MC} - 1)/3 = 1/3 \). The suppression of the moment in the strong-coupling regime is due to the presence of the Kondo correlations. The Kondo effect is exposed in the conductance dependence (yellow line), as it saturates to unitary value \( G/G_0 = 1 \) in the strong coupling regime. In consequence, the spin of the molecular level is screened, and the total spin of the molecule is reduced from \( S = 3/2 \) to \( S = S_{MC} = 1 \), leading eventually to \( \langle Q_{zz} \rangle = 1/3 \). To clearly show how the expectation values of spin operators influence the value of the spin-quadrupole moment, we also plot \( \langle S^2 \rangle \) and \( \langle S_z^2 \rangle \) as a function of the coupling strength. It is noteworthy that both quantities in the Kondo regime no achieve the value, that would be expected when electron on the orbital level was fully screened by the Kondo correlations. The expected values only approach this limit, i.e. \( \langle S_z^2 \rangle \to 2, \langle S_z^2 \rangle \to 1 \), however, the resulting value of the spin-quadrupole moment is indeed \( \langle Q_{zz} \rangle = 1/3 \).

In Fig. 2(b) we present the spin-quadrupole moment \( \langle Q_{zz} \rangle \) (solid lines) and the linear-response conductance \( G \) (dashed lines) as a function of \( \Gamma \) for different values of the exchange coupling \( J \). As evident, when the magnitude of \( J \) is varied, the spin-quadrupole moment values in the weak and strong coupling regimes are conserved, however, the increase of the exchange coupling extends the transitional range of coupling strength where the crossover between the weak and Kondo regimes develops. Although the considered model is an effective one, we expect that in the case of molecules with strong exchange couplings between localized spins and those of itinerant electrons, the quench in the coupling strength needs to be superior than in the case of systems with small magnitudes of the exchange couplings. The role of magnitude of \( J \) on the dynamics of the spin-quadrupole moment is discussed in more detail in Sec. III C.
nal coupling strength $\Gamma$, the timescale of the considered transition can be varied by an order of magnitude.

To clearly display the above behavior, in Fig. 3(b) we show the time evolution of quadrupolar moment for different values of $\Gamma$ chosen from the range, where the dynamics is most interesting. Finally, in Fig. 3(c) we show the same results in the form of several curves on a rescaled time axis in order to clearly indicate that the relaxation is universally governed by $t \propto 1/\Gamma^2$. The long-time limit is achieved at times $t \approx 2/\Gamma^2$, when one can see that all the curves converge.

C. Role of molecule’s exchange coupling

An important impact on the magnetic and transport properties of SMM systems has the magnitude of exchange coupling $J$ between the effective magnetic core spin $S_{MC}$ and the spin $s$ of electrons occupying the or-

FIG. 3. The time dependence of the spin-quadrupole moment $Q_{zz}(t)$ after quench in the coupling strength from $\Gamma_0/U = 0.002$ to different values of the final coupling strength. (a) presents the density plot of $Q_{zz}(t)$, while (b) and (c) show $Q_{zz}(t)$ as a function of $t \cdot \Gamma$ and $t \cdot \Gamma^2$, respectively. The other parameters are the same as in Fig. 2.

bital level. In Fig. 4 we show the time-dependent spin-quadrupole moment, considering similar quench as in previous section, with $\Gamma_0/U = 0.002$ and $\Gamma/U = 0.4$, as a function of exchange coupling $J$.

It can be clearly seen that the magnitude of $J$ does not have a substantial influence on the short time evolution of spin-quadrupole moment for elapsed time up to $t \cdot \Gamma \approx 10^2$. For $J/U \lesssim 0.002$, the long-time limit $\langle Q_{zz} \rangle = 1/3$ is achieved already around $t \cdot \Gamma \approx 10^1$ and there is no further dynamics as the time elapses. However, when the exchange coupling is increased above $J/U \approx 0.002$, further decrease of spin-quadrupole moment takes place at long times, i.e. for $t \cdot \Gamma \gtrsim 10^2$. The observed reduction is down to values well below $\langle Q_{zz} \rangle = 1/3$, i.e. $Q_{zz}(t \to \infty) \lesssim 0.3$, and the dynamics of this process is strongly dependent on the magnitude of $J$. When the time-dependence for larger values of $J$ is evaluated, the new long-time limit is achieved at earlier times, while the value of spin-quadrupole moment is accordingly further suppressed.

In order to better understand the effect of exchange coupling on the spin-quadrupole moment dynamics, we examine the time-dependence of two spin operators, $S^2_z(t)$ and $S^2(t)/3$, which are the constituent components of the operator $Q_{zz}(t)$. The corresponding plots are shown in Fig. 5. The results were obtained for exchange coupling set to $J/U = 0.01$, where the time dependence reveals two stages of magnetic moment’s reduction.

In Fig. 5(a) we display the time-dependent spin-quadrupole moment after the quench in the coupling strength, while in Fig. 5(b) we present the time-dependent expectation values of spin operators $S^2_z(t)$ and $S^2(t)/3$.
ors. The red background contains short-time dynamics, when a rapid suppression of all expectation values takes places in a similar manner like in the case of weaker $J$ coupling. This dynamics is associated with the change of the ground state and the reduction of the total spin $S$ from $S = 3/2$ to $S = 1$ due to the Kondo screening of the orbital spin. This stage of time evolution ends up at times around $t \cdot \Gamma \approx 10^{3}$. Consequently, the next section is marked with purple background and contains the further dynamics associated with influence of the exchange interaction $J$. For times $t \cdot \Gamma \gtrsim 10^{4}$, the time-dependence of the spin operators clearly shows that the total spin of the system $S$ remains intact and has already achieved the long-time limit. Further ongoing dynamics takes place exclusively for $S^z(t)$ in the form of second stage of slow suppression of the total spin-quadrupole moment to $Q_{zz}(t) \lesssim 1/3$. The total time of this stage evolution greatly depends on the strength of $J$, as shown in Fig. 4, i.e. the system faster reaches equilibrium for higher values of the exchange coupling $J$.

D. Influence of leads’ spin polarization

The ferromagnetism of the leads is of great importance for the considered molecular system. In general, when a finite spin polarization of electrodes is assumed ($p > 0$) and their magnetic moments are aligned, the effective dipolar and quadrupolar exchange fields are generated [53, 84]. The strengths of these fields and their occurrence strongly depend on the degree of spin polarization $p$. In our analysis, we focus on the quadrupolar term by tuning the molecular level to the particle-hole symmetry point $\varepsilon = -U/2$. In such a configuration, the charge fluctuations for both spin directions are equal and independently of the magnitude of $p$, the dipolar field is canceled. Moreover, it was shown that for large-spin molecules, a very small spin polarization ($p = 0.01$) of the leads can give rise to $\langle Q_{zz} \rangle = S(2S-1)/3 = 1$. In equilibrium, a similar dependence to that shown in Fig 2 is predicted for wide range of $p$, with finite temperature suppressing the spin-quadrupole moment in the regime of very weak coupling strengths $[65]$. Therefore, let us now discuss the influence of leads spin polarization $p$ on the dynamics of the spin-quadrupole moment after the quench from the weak to the strong coupling regime ($\Gamma_0/U = 0.002$ and $\Gamma/U = 0.4$). The results presenting $Q_{zz}(t)$ for a wide range of spin polarizations $p$ are shown in Fig. 6(a).

In the range of small and moderate spin polarizations ($p \lesssim 0.4$), the time evolution of spin quadrupole moment has both qualitatively and quantitatively similar dependence, with two stages of $Q_{zz}(t) = 1$ reduction present, as discussed in previous section for the case of significant exchange coupling $J/U = 0.01$. However, when the spin polarization is increased further ($p \gtrsim 0.4$), a new step emerges right before the time $t \cdot \Gamma \approx 1$, when $Q_{zz}(t) \approx 3/4$. This step is elongated as $p$ is increased, reaching times up to $t \cdot \Gamma \approx 10^{4}$ for $p = 0.9$ and eventually not fully relaxing to $Q_{zz}(t) = 1/3$, when $p \rightarrow 1$. This behavior might seem to be counter-intuitive, as one could expect that with increased spin-polarization of the leads, the dynamics of the quench should be faster along with rapid suppression of the spin-quadrupole moment. Here, we predict quite opposite dependence, as the dynamics is faster and more straightforward when the spin-polarization of the leads has low-to-moderate values.

The mechanism responsible for the emergence of an additional step in $Q_{zz}(t)$ in the case of highly spin-polarized electrodes is closely related to an enhanced difference between spin-dependent couplings $\Gamma^\uparrow = \Gamma(1 + p)$ and $\Gamma^\downarrow = \Gamma(1 - p)$ whilst $p$ is increased. The bottleneck of the associated dynamics is govern by the coupling to the minority band, which in result slows down the dynamics of the spin-quadrupole moment. To clearly analyze the discussed effect, we show the spin-quadrupole moment $Q_{zz}(t)$ in Fig. 6(b) and the square of molecular-level magnetization $s_z^2(t)$ in Fig. 6(c) as a functions of $t \cdot \Gamma$. Here, we recall that molecular-level magnetization is defined as $s_z(t) = (n_\uparrow(t) - n_\downarrow(t))/2$. The discussed dynamics revealed for highly spin-polarized lead takes place at times $t \cdot \Gamma \lesssim 1$. The effect is evident for $p = 0.9$, where the dependence of $Q_{zz}(t)$ exposes a short plateau with $Q_{zz}(t) \approx 3/4$. The time-dependence of the local operator $s_z^2(t)$ plays an important role here. As can be seen in the figure, $s_z^2(t)$ is a monotonically decreasing function of time for $p \lesssim 0.7$ in this time regime. Interestingly, further enhancement of spin-polarization generates a subtle oscillation in the time-dependence of $s_z^2(t)$. In that time
molecules of arbitrarily given total spin $S$ different values of magnetic core spin $Q$ in a quantum dot system coupled to ferromagnetic lead [81].

Eventually, the impor-

range, $\Gamma^\dagger = \Gamma(1 + p)$ is responsible for a rapid drop of $s_z^2(t)$ due to decrease of the spin-up component. However, the time evolution is not balanced by the spin-down component due to a significantly slower dynamics governed by $\Gamma^\dagger = \Gamma(1 - p)$. As a result, the molecular level has a minimum in $s_z^2(t)$, which is below the long-time limit thermal value, at time $t \cdot \Gamma \approx 6 \cdot 10^{-1}$, see Fig. 6(c). Subsequently, a small increase takes places leading to eventual relaxation. This non-monotonic dependence of $s_z^2(t)$ is observed as a result of the interplay between the spin-dependent couplings and, in consequence, temporarily pauses the reduction of $Q_{zz}(t)$. A similar dynamical behavior was predicted for the magnetization of a single quantum dot system coupled to ferromagnetic lead [81].

### E. Influence of magnitude of molecule’s spin

In order to generalize our analysis for magnetic molecules of arbitrarily given total spin $S$, we examine the results of tNRG calculations for models with different values of magnetic core spin $S_{MC}$. First of all, we would like to note that in equilibrium, qualitatively a very similar dependence of $\langle Q_{zz} \rangle$ on the coupling strength $\Gamma$ is expected independently of the value of $S_{MC}$, see Fig. 7(a). As the spin of magnetic core is increased, the maximal value of the spin-quadrupole moment in the weak coupling regime is increasing accordingly with $\langle Q_{zz} \rangle = S_z^2 - S(S + 1)/3$. Furthermore, the transition range of coupling strengths preceding the strong coupling regime is similar for all cases, i.e. $5 \cdot 10^{-2} \lesssim \Gamma/U \lesssim 2 \cdot 10^{-1}$. Eventually, the important change influenced by the total spin of the molecule is in the value, by which the spin-quadrupole moment is reduced, when the coupling regime is switched from the weak to the strong one. This difference is enhanced, when the total spin number is increased.

The time-dependent spin-quadrupole moment after the quench as a function of time for several values of $S_{MC}$ is shown in Fig. 7(b). Here, one can see that the dynamics is still governed by the strength of coupling to electrodes, as the reduction of spin-quadrupolar moment starts and achieves long-time limit at similar moments on the time axis rescaled with $\Gamma$. For all considered values of $S_{MC}$,
time-evolutions behave in a similar fashion with the main distinction of initial and final values of $\langle Q_{zz} \rangle$ and the rate of reduction. Similar effects and dependencies due to the exchange interaction or spin-polarization of the leads as discussed in previous sections are predicted also for molecules with even higher total spin number (not shown here). Therefore, our dynamical studies presented in this work have a general character and are valid for broad range of SMM systems, in which the quadrupolar exchange field emerges from ferromagnetic proximity effect.

**F. Finite temperature effects**

Lastly, we examine the influence of finite temperature $T$ on the spin-quadrupole moment and its time evolution following the quench in the coupling. In Fig. 8 we show the time-dependent expectation value of the spin-quadrupole moment as well as the corresponding spin operators for several values of temperatures.

For the regime of very low temperatures, $T/U \lesssim 10^{-8}$, both the initial and final values, as well as the quench dynamics of the spin-quadrupole moment remain similar to the case of zero temperature. Further increase of temperature, however, has a significant impact on the system’s behavior. First of all, the initial value of the spin quadrupole moment in the weak coupling regime is significantly suppressed as the temperature is increased. The inset in Fig. 8(a) shows how the increase of temperature reduces the static spin-quadrupole moment in the weak coupling regime. The vertical dashed line represents the initial coupling strength $\Gamma_0/U = 0.002$, we have used in the evaluation of the quench dynamics. From the above considerations, it is evident that the fine tuning of the coupling strength for the initial state is critical, in particular when the suppression of the maximal value of the moment is expected. Furthermore, the long-time limit value of the time-dependent spin-quadrupole moment is also reduced by the temperature, however, not as strongly as the initial value. In consequence, as the temperature is increased, the whole transient dynamics is exposing more moderate dependence, with almost completely flat one for temperatures $T/U \gtrsim 10^{-6}$. It is also important to note, that all the characteristic time scales discussed in earlier analysis for $T = 0$ are conserved for the finite temperatures, where a considerable suppression of spin-quadrupole moment is predicted. In particular, the rapid reduction associated with the change of the ground state that dominates the dynamics for times $t \cdot \Gamma \lesssim 10^4$ and further slower dynamics influenced by the exchange coupling taking place up to $t \cdot \Gamma \approx 10^5$ are all still noticeable for temperatures up to $T/U \approx 10^{-7}$.

The inspection of the spin operators $S_z^2(t)$ and $S_z(t)$, see Figs. 8(b) and 8(c), brings the conclusion that the temperature influences spin-quadrupole moment only by reduction of the $z$-th component of the SMM’s spin. Meanwhile the square value of the total spin remains unaffected, as it corresponds to the change of the molecule’s ground state. This fact clearly indicates the reduction of the molecule’s magnetic anisotropy due to thermal fluctuations.

**IV. CONCLUSIONS**

We have analyzed the quench dynamics of large-spin magnetic molecules attached to spin-polarized ferromagnetic leads. The study was performed by using the time-dependent numerical renormalization group method. We focused on the dynamics of the spin-quadrupole moment and a quench associated with switching the system from the weak coupling regime to the strong coupling one, in which the Kondo correlations are present and the screening of the molecular level spin develops.

In general, we have shown that the time necessary to achieve the new thermal value in the strong coupling regime is inversely proportional to squared coupling

![ FIG. 8. (a) The spin-quadrupole moment $Q_{zz}(t)$, (b) $S_z^2(t)$ and (c) $S_z(t)$ for several values of temperature $T$ plotted as a function of time elapsed after the quench in $\Gamma$. The inset in (a) shows the influence of temperature on the static value of the spin-quadrupole moment in the weak coupling regime. Dashed vertical line indicates the initial value of the coupling strength. The parameters are the same as in Fig. 5.](image-url)
strength, i.e. $t \propto 1/T^2$. Furthermore, we examined the role of ferromagnetic exchange coupling $J$ and showed that when the magnitude of this interaction is considerable, additional step in the time-dependence emerges, corresponding to slower dynamics mediated by $J$, which is an interesting case of interplay between the Kondo correlations and ferromagnetism exposed in our dynamical studies. The influence of electrodes’ spin-polarization was also discussed, strongly indicating that the quench dynamics is faster and more straightforward for low-to-average values. We also generalized our studies to systems with higher total spin numbers to show that observations and conclusions are valid for a wide class of SMM systems with arbitrary total spin number.

Finally, we studied the influence of finite temperature on the spin-quadrupole moment. A strong suppression of the time-dependent value is predicted in certain range of temperatures, due to reduction of the molecule’s anisotropy. The important role of the coupling strength fine tuning is also indicated, which may have relevance for the experiments.

**ACKNOWLEDGMENTS**

This work was supported by the Polish National Science Centre from funds awarded through the decision No. 2017/27/B/ST3/00621. The computing time at the Poznań Supercomputing and Networking Center is acknowledged.

[1] Roman Boča, *Theoretical Foundations of Molecular Magnetism* (Elsevier, 1999).
[2] J. Tejada, E. M. Chudnovsky, E. del Barco, J. M. Hernandez, and T. P. Spiller, “Magnetic qubits as hardware for quantum computers,” *Nanotechnology* 12, 181–186 (2001).
[3] D. Gatteschi, R. Sessoli, and J. Villain, *Molecular Nanomagnets* (Oxford University Press, 2006).
[4] Lapo Bogani and Wolfgang Wernsdorfer, “Molecular spintronics using single-molecule magnets,” *Nat. Mater.* 7, 179–186 (2008).
[5] Jörg Lehmann, Alejandro Gaita-Arino, Eugenio Coronado, and Daniel Loss, “Quantum computing with molecular spin systems,” *J. Mater. Chem.* 19, 1672–1677 (2009).
[6] Matteo Mannini, Francesco Pineider, Philippe Sainctavit, Chiara Danielli, Edwige Otero, Corrado Sciancalepore, Anna Maria Talarico, Marie-Anne Arrio, Andrea Cornia, Dante Gatteschi, and Roberta Sessoli, “Magnetic memory of a single-molecule quantum magnet wired to a gold surface,” *Nat. Mater.* 8, 194–197 (2009).
[7] Romain Vincent, Svetlana Klyatskaya, Mario Ruben, Wolfgang Wernsdorfer, and Franck Balestro, “Electronic read-out of a single nuclear spin using a molecular spin transistor,” *Nature* 488, 357–360 (2012).
[8] Peter Jacobson, Tobias Herden, Matthias Muenks, Gennadii Laskin, Oleg Brovko, Valeri Stepanyuk, Markus Ternes, and Klaus Kern, “Quantum engineering of spin and anisotropy in magnetic molecular junctions,” *Nat. Commun.* 6, 1–6 (2015).
[9] Roberta Sessoli, “Magnetic molecules back in the race,” *Nature* 548, 400–401 (2017).
[10] Khadijeh Najafi, Aleksander L. Wysocki, Kyungwha Park, Sophia E. Economou, and Edwin Barnes, “Toward Long-Range Entanglement between Electrically Driven Single-Molecule Magnets,” *J. Phys. Chem. Lett.* 10, 7347–7355 (2019).
[11] P. Gambardella, S. Rusponi, M. Veronese, S. S. Dhesi, C. Grazioli, A. Dallmeyer, I. Cabria, R. Zeller, P. H. Dederichs, K. kern, C. Carbone, and H. Brune, “Giant Magnetic Anisotropy of Single Cobalt Atoms and Nanoparticles,” *Science* 300, 1130–1133 (2003).
[12] Cyrus F. Hirjibehedin, Ching-Yuan Lin, Alexander F. Otto, Markus Ternes, Christopher P. Lutz, Barbara A. Jones, and Andreas J. Heinrich, “Large Magnetic Anisotropy of a Single Atomic Spin Embedded in a Surface Molecular Network,” *Science* 317, 1199–1203 (2007).
[13] Pietro Gambardella, Sebastian Stepnow, Alexandre Dmitriev, Jan Honolka, Frank M. F. de Groot, Magalí Lingenfelder, Subhra Sen Gupta, D. D. Sarma, Peter Bencok, Stefan Stanescu, Sylvain Clair, Stéphane Pons, Nian Lin, Ari P. Seistsonen, Harald Brune, Johannes V. Barth, and Klaus Kern, “Supramolecular control of the magnetic anisotropy in two-dimensional high-spin Fe arrays at a metal interface,” *Nat. Mater.* 8, 189–193 (2009).
[14] Lei Xu, Ziba Zangeneh, Ravj Yadav, Stanislaw Avdoshenko, Jeroen van den Brink, Anton Jesche, and Liviu Hozoi, “Spin-reversal energy barriers of 305 K for Fe2+ d6 ions with linear ligand coordination,” *Nanoscale* 9, 10596–10600 (2017).
[15] Aleksander L. Wysocki and Kyungwha Park, “Nature of Hyperfine Interactions in TbFe2 Single-Molecule Magnets: Multiconfigurational Ab Initio Study,” *Inorg. Chem.* 59, 2771–2780 (2020).
[16] Eugene M. Chudnovsky and Javier Tejada, “Macroscopic Quantum Tunneling of the Magnetic Moment,” *Cambridge Core* (1998), 10.1017/CBO9780511524219.
[17] Eman K. Brehin, Colette Boskovic, Wolfgang Wernsdorfer, Jae Yoo, Akira Yamaguchi, E. Carolina Sainudo, Thomas R. Concilioni, Arnold L. Rheingold, Hideliko Ishimoto, David N. Hendrickson, and George Christou, “Quantum Tunneling of Magnetization in a New [Mn18I2+ Single-Molecule Magnet with S = 13],” *J. Am. Chem. Soc.* 124, 9710–9711 (2002).
[18] M. Misiorny and J. Barnaś, “Quantum tunneling of magnetization in single molecular magnets coupled to ferromagnetic reservoirs,” *EPL* 78, 27003 (2007).
[19] M. Mannini, F. Pineider, C. Danieli, F. Totti, L. Sorace, Ph. Sainctavit, M.-A. Arrio, E. Otero, L. Joly, J. C. Cezar, A. Cornia, and R. Sessoli, “Quantum tunneling of the magnetization in a monolayer of oriented single-molecule magnets,” *Nature* 468, 417–421 (2010).
[20] Gwang-Hee Kim and Tae-Suk Kim, “Electronic Transport in Single-Molecule Magnets on Metallic Surfaces,” *Nature* 468, 457–461 (2010).
Phys. Rev. Lett. 92, 137203 (2004).

[21] Carsten Timm and Florian Elste, “Spin amplification, reading, and writing in transport anisotropic magnetic molecules,” Phys. Rev. B 73, 235304 (2006).

[22] Maciej Misiorny, Ireneusz Weymann, and Józef Barnaś, “Spin effects in transport through single-molecule magnets in the sequential and cotunneling regimes,” Phys. Rev. B 79, 224420 (2009).

[23] M. Misiorny, I. Weymann, and J. Barnaś, “Spin diode behavior in transport through single-molecule magnets,” EPL 89, 18003 (2010).

[24] Maciej Misiorny, Ireneusz Weymann, and Józef Barnaś, “Temperature dependence of electronic transport through molecular magnets in the Kondo regime,” Phys. Rev. B 86, 035417 (2012).

[25] M. Misiorny, I. Burzurí, R. Gaudenzi, K. Park, M. Leijnse, M. R. Wegewijs, J. Paaske, A. Cornia, and H. S. J. van der Zant, “Probing transverse magnetic anisotropy by electronic transport through a single-molecule magnet,” Phys. Rev. B 91, 035442 (2015).

[26] Anna Plomińska and Ireneusz Weymann, “Pauli spin blockade in double molecular magnets,” Phys. Rev. B 94, 035422 (2016).

[27] Anna Plomińska, Maciej Misiorny, and Ireneusz Weymann, “Manipulating spins of magnetic molecules: Hysteretic behavior with respect to bias voltage,” EPL 121, 38006 (2018).

[28] F. Pawlicki and I. Weymann, “Andreev transport through single-molecule magnets,” Phys. Rev. B 98, 085411 (2018).

[29] A. Chiesa, E. Macaluso, P. Santini, S. Carretta, and E. Pavarini, “First-principles many-body models for electron transport through molecular nanomagnets,” Phys. Rev. B 99, 235145 (2019).

[30] Silvia Giménez-Santamarina, Salvador Cardona-Serra, and Alejandro Gaita-Ariño, “Theoretical insights on the importance of anchoring vs molecular geometry in magnetic molecules acting as junctions,” J. Magn. Magn. Mater. 485, 212–216 (2019).

[31] J. de Bruijckere, P. Gehring, M. Palacios-Corella, M. Clemente-León, E. Coronado, J. Paaske, P. Hedegård, and H. S. J. van der Zant, “Ground-State Spin Blockade in a Single-Molecule Junction,” Phys. Rev. Lett. 122, 197701 (2019).

[32] Jun Kondo, “Resistance minimum in dilute magnetic alloys,” Progress of Theoretical Physics 32, 37–49 (1964).

[33] L. I. Glazman and M.E. Raikh, “Resonant kondo transparency of a barrier with quasilocal impurity states,” JETP Lett. 47, 452 (1988).

[34] A. C. Hewson, The Kondo problem to heavy fermions (Cambridge University Press, 1997).

[35] D. Goldhaber-Gordon, Hadas Shtrikman, D. Mahalu, David Abusch-Magder, U. Meirav, and M. A. Kastner, “Kondo effect in a single-electron transistor,” Nature 391, 156 EP – (1998).

[36] Sara M. Cronenwett, Tjerk H. Oosterkamp, and Leo P. Kouwenhoven, “A tunable kondo effect in quantum dots,” Science 281, 540–544 (1998), http://science.sciencemag.org/content/281/5376/540.full.pdf.

[37] V. Madhavan, W. Chen, T. Jammeala, M. F. Crommie, and N. S. Wingreen, “Tunneling into a Single Magnetic Atom: Spectroscopic Evidence of the Kondo Resonance,” Science 280, 567–569 (1998).

[38] C. Romeike, M. R. Wegewijs, and H. Schoeller, “Spin Quantum Tunneling in Single Molecular Magnets: Fingerprints in Transport Spectroscopy of Current and Noise,” Phys. Rev. Lett. 96, 196805 (2006).

[39] Alexander F. Otte, Markus Ternes, Kirsten von Bergmann, Sebastian Loth, Harald Brune, Christopher P. Lutz, Cyrus F. Hirjibehedin, and Andreas J. Heinrich, “The role of magnetic anisotropy in the Kondo effect,” Nat. Phys. 4, 847–850 (2008).

[40] J. J. Parks, A. R. Champagne, T. A. Costi, W. W. Shum, A. N. Pasupathy, E. Neuscamman, S. Flores-Torres, P. S. Cornaglia, A. A. Aligia, C. A. Balseiro, G. K.-L. Chan, H. D. Abrüna, and D. C. Ralph, “Mechanical Control of Spin States in Spin-1 Molecules and the Underscreened Kondo Effect,” Science 328, 1370–1373 (2010).

[41] Maciej Misiorny, Ireneusz Weymann, and Józef Barnaś, “Interplay of the Kondo Effect and Spin-Polarized Transport in Magnetic Molecules, Adatoms, and Quantum Dots,” Phys. Rev. Lett. 106, 126602 (2011).

[42] Maciej Misiorny, Ireneusz Weymann, and Józef Barnaś, “Underscreened Kondo effect in $S = 1$ magnetic quantum dots: Exchange, anisotropy, and temperature effects,” Phys. Rev. B 86, 245415 (2012).

[43] Peter Zalam, Joeri de Bruijckere, Rocco Gaudenzi, Herre S. J. van der Zant, Tomáš Novotný, and Richard Kortyr, “Magnetically Tuned Kondo Effect in a Molecular Double Quantum Dot: Role of the Anisotropic Exchange,” J. Phys. Chem. C 123, 11917–11925 (2019).

[44] Maciej Misiorny and Józef Barnaś, “Magnetic switching of a single molecular magnet due to spin-polarized current,” Phys. Rev. B 75, 134425 (2007).

[45] Gabriel González and Michael N. Leuenberger, “Berry-Phase Blockade in Single-Molecule Magnets,” Phys. Rev. Lett. 98, 256804 (2007).

[46] Maciej Misiorny and Józef Barnaś, “Effects of intrinsic spin-relaxation in molecular magnets on current-induced magnetic switching,” Phys. Rev. B 77, 172414 (2008).

[47] F. Delgado, J. J. Palacios, and J. Fernández-Rossier, “Spin-Transfer Torque on a Single Magnetic Adatom,” Phys. Rev. Lett. 104, 026601 (2010).

[48] Maciej Misiorny and Józef Barnaś, “Effects of Transverse Magnetic Anisotropy on Current-Induced Spin Switching,” Phys. Rev. Lett. 111, 046603 (2013).

[49] Maciej Misiorny and Ireneusz Weymann, “Transverse anisotropy effects on spin-resolved transport through large-spin molecules,” Phys. Rev. B 90, 235409 (2014).

[50] Pawan Tyagi and Edward Friebe, “Large resistance change on magnetic tunnel junction based molecular spintronics devices,” J. Magn. Magn. Mater. 453, 186–192 (2018).

[51] Emil Sierda, Micha Elsebach, Roland Wiesendanger, and Maciej Bazarnik, “Probing Weakly Hybridized Magnetic Molecules by Single-Atom Magnetometry,” Nano Lett. 19, 9013–9018 (2019).

[52] B. Verlhac, N. Bachelier, L. Garnier, M. Ormaza, P. Abufager, R. Robles, M.-L. Bocquet, M. Ternes, N. Lorente, and L. Limot, “Atomic-scale spin sensing with a single molecule at the apex of a scanning tunneling microscope,” Science 366, 623–627 (2019).

[53] Maciej Misiorny, Michael Hell, and Maarten R. Wegewijs, “Spintronic magnetic anisotropy,” Nat. Phys. 9, 801–805 (2013).

[54] J. O. Johansson, J-W. Kim, E. Allwright, D. M. Rogers, N. Robertson, and J.-Y. Bigot, “Directly probing spin dynamics in a molecular magnet with femtosecond time-
resolution,” Chem. Sci. 7, 7061–7067 (2016).

[55] Keiichi Katoh, Satoshi Yamashita, Nobuhiro Yasuda, Yasutaka Kitagawa, Brian K. Breedlove, Yasuhiro Nakazawa, and Masahiro Yamashita, “Control of the Spin Dynamics of Single-Molecule Magnets by using a Quasi One-Dimensional Arrangement,” Angew. Chem. Int. Ed. 57, 9262–9267 (2018).

[56] Gheorghe Taran, Edgar Bonet, and Wolfgang Wernsdorfer, “Decoherence measurements in crystals of molecular magnets,” Phys. Rev. B 99, 180408(R) (2019).

[57] David Roosen, Maarten R. Wegewijs, and Walter Hofstetter, “Nonequilibrium dynamics of anisotropic large spins in the kondo regime: Time-dependent numerical renormalization group analysis,” Phys. Rev. Lett. 100, 087201 (2008).

[58] Anna Plomińska, Maciej Misiorny, and Ireneusz Weymann, “Spin-resolved dynamical conductance of a correlated large-spin magnetic molecule,” Phys. Rev. B 95, 155446 (2017).

[59] H. Hammar and J. Fransson, “Transient spin dynamics in a single-molecule magnet,” Phys. Rev. B 96, 214401 (2017).

[60] Anna Plomińska, Ireneusz Weymann, and Maciej Misiorny, “Dynamical spin accumulation in large-spin magnetic molecules,” Phys. Rev. B 97, 035415 (2018).

[61] J. Martinek, Y. Utsumi, H. Imamura, J. Barnaś, S. Maekawa, J. König, and G. Schön, “Kondo Effect in Quantum Dots Coupled to Ferromagnetic Leads,” Phys. Rev. Lett. 91, 127203 (2003).

[62] J. Martinek, M. Sindel, L. Borda, J. Barnaś, R. Bulla, J. König, G. Schön, S. Maekawa, and J. von Delft, “Gate-controlled spin splitting in quantum dots with ferromagnetic leads in the Kondo regime,” Phys. Rev. B 72, 121302(R) (2005).

[63] J. R. Hauptmann, J. Paaske, and P. E. Lindelof, “Electric-field-controlled spin reversal in a quantum dot with ferromagnetic contacts,” Nat. Phys. 4, 373–376 (2008).

[64] M. Gaass, A. K. Hüttel, K. Kang, I. Weymann, J. von Delft, and Ch. Strunk, “Universality of the Kondo Effect in Quantum Dots with Ferromagnetic Leads,” Phys. Rev. Lett. 107, 176808 (2011).

[65] Krzysztof P. Wójcik, Maciej Misiorny, and Ireneusz Weymann, “Giant superconducting proximity effect on spintronic anisotropy,” Phys. Rev. B 100, 045401 (2019).

[66] Gheorghe Taran, Edgar Bonet, and Wolfgang Wernsdorfer, “The role of the quadrupolar interaction in the tunneling dynamics of lanthanide molecular magnets,” J. Appl. Phys. 125, 142903 (2019).

[67] Kenneth G. Wilson, “The renormalization group: Critical phenomena and the kondo problem,” Rev. Mod. Phys. 47, 773–840 (1975).

[68] Ralf Bulla, Theo A. Costi, and Thomas Pruschke, “Numerical renormalization group method for quantum impurity systems,” Rev. Mod. Phys. 80, 395–450 (2008).

[69] We used the open-access Budapest Flexible DM-NRG code, http://www.phy.bme.hu/~dmnrg/; O. Legeza, C. P. Moca, A. I. Tóth, I. Weymann, G. Zaránt, arXiv:0809.3143 (2008) (unpublished).

[70] Frithjof B. Anders and Avraham Schiller, “Real-time dynamics in quantum-impurity systems: A time-dependent numerical renormalization-group approach,” Phys. Rev. Lett. 95, 196801 (2005).

[71] Frithjof B. Anders and Avraham Schiller, “Spin precession and real-time dynamics in the kondo model: Time-dependent numerical renormalization-group study,” Phys. Rev. B 74, 245113 (2006).

[72] H. T. M. Nghiem and T. A. Costi, “Generalization of the time-dependent numerical renormalization group method to finite temperatures and general pulses,” Phys. Rev. B 89, 075118 (2014).

[73] H. T. M. Nghiem and T. A. Costi, “Time-dependent numerical renormalization group method for multiple quenches: Application to general pulses and periodic driving,” Phys. Rev. B 90, 035129 (2014).

[74] H. T. M. Nghiem and T. A. Costi, “Time-dependent numerical renormalization group method for multiple quenches: towards exact results for the long time limit of thermodynamic observables and spectral functions,” Phys. Rev. B 98, 155107 (2018).

[75] J. Martinek, Y. Utsumi, H. Imamura, J. Barnaś, S. Maekawa, J. König, and G. Schön, “Kondo effect in quantum dots coupled to ferromagnetic leads,” Phys. Rev. Lett. 91, 127203 (2003).

[76] Ohn-Soo Choi, David Sánchez, and Rosa López, “Kondo effect in a quantum dot coupled to ferromagnetic leads: A numerical renormalization group analysis,” Phys. Rev. Lett. 92, 056601 (2004).

[77] J. Martinek, M. Sindel, L. Borda, J. Barnaś, R. Bulla, J. König, G. Schön, S. Maekawa, and J. von Delft, “Gate-controlled spin splitting in quantum dots with ferromagnetic leads in the kondo regime,” Phys. Rev. B 72, 121302 (2005).

[78] M. Sindel, L. Borda, J. Martinek, R. Bulla, J. König, G. Schön, S. Maekawa, and J. von Delft, “Kondo quantum dot coupled to ferromagnetic leads: Numerical renormalization group study,” Phys. Rev. B 76, 045321 (2007).

[79] Andreas Weichselbaum and Jan van Delft, “Sum-rule conserving spectral functions from the numerical renormalization group,” Phys. Rev. Lett. 99, 076402 (2007).

[80] Andreas Weichselbaum, “Tensor networks and the numerical renormalization group,” Phys. Rev. B 86, 245124 (2012).

[81] Kacper Wrześniowski and Ireneusz Weymann, “Quench dynamics of spin in quantum dots coupled to spin-polarized leads,” Phys. Rev. B 100, 035404 (2019).

[82] Yigal Meir and Ned S. Wingreen, “Landauer formula for the current through an interacting electron region,” Phys. Rev. Lett. 68, 2512–2515 (1992).

[83] M. Yoshida, M. A. Whitaker, and L. N. Oliveira, “Renormalization-group calculation of excitation properties for impurity models,” Phys. Rev. B 41, 9403–9414 (1990).

[84] J. Martinek, M. Sindel, L. Borda, J. Barnaś, R. Bulla, J. König, G. Schön, S. Maekawa, and J. von Delft, “Gate-controlled spin splitting in quantum dots with ferromagnetic leads in the kondo regime,” Phys. Rev. B 72, 121302 (2005).