Exploiting the magnetomechanical interaction for cooling magnetic molecular junctions by spin-polarized currents

J Brüggemann, S Weiss, P Nalbach and M Thorwart

I. Institut für Theoretische Physik, Universität Hamburg, Jungiusstraße 9, D-20355 Hamburg, Germany
2. Theoretische Physik, Universität Duisburg-Essen & CENIDE, D-47048 Duisburg, Germany
3. Westfälische Hochschule, Münsterstr. 265, D-46397 Bocholt, Germany
4. Author to whom any correspondence should be addressed.
E-mail: thorwart@physik.uni-hamburg.de

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Abstract
We present a scheme for cooling a vibrational mode of a magnetic molecular nanojunction by a spin-polarized charge current upon exploiting the interaction between its magnetic moment and the vibration. The spin-polarized charge current polarizes the magnetic moment of the nanoisland, thereby lowering its energy. A small but finite coupling between the vibration and the magnetic moment permits a direct exchange of energy such that vibrational energy can be transferred into the magnetic state. For positive bias voltages, this generates an effective cooling of the molecular vibrational mode. We determine parameter regimes for the cooling of the vibration to be optimal.

Although the flowing charge current inevitably heats up the vibrational mode via Ohmic energy losses, we show that due to the magnetomechanical coupling, the vibrational energy (i.e., the effective phonon temperature) can be lowered below 50\% of its initial value, when the two leads are polarized anti-parallel. In contrast to the cooling effect for positive bias voltages, net heating of the vibrational mode occurs for negative bias voltages. The cooling effect is enhanced for a stronger anti-parallel magnetic polarization of the leads, while the heating is stronger for a larger parallel polarization. Yet, dynamical cooling is also possible with parallel lead alignments when the two tunneling barriers are asymmetric.

1. Introduction

The continuous efforts of miniaturizing electronic devices lead to several novel challenges in technological applications. One of these challenges is the requirement to develop efficient cooling schemes for micro- and nanoelectronics. To date, most electronic devices utilize passive cooling techniques \[1\]. Thermal contact with the supporting structure provides the possibility to use a colder environment as a passive heat sink. Active or dynamical cooling at the nanoscale has received little attention, although passive thermal transport is inefficient at these length scales. Furthermore, most of the recently developed electronic and magnetic nanodevices operate at low temperatures only. The importance of a dynamical cooling mechanism on the nanoscale can therefore not be overestimated. Dynamical nanorefrigerators would also open the possibility for new experiments and devices. Most existing dynamical cooling methods on the macroscale require spacious equipment and mechanical movement of pumping equipment, for example, thus making them impractical for an application on the nanoscale. Manipulating heat transfer at the nanoscale would then not only drastically decrease the size of the cooling equipment, but also yield the possibility to create scalable arrays of cooled nanodevices.

A directly accessible degree of freedom of electronically conducting molecular nanojunctions is the electronic charge in combination with the electron spin. By applying external bias voltages in ferromagnetic (FM) leads, it is routinely possible to control charge currents and also the polarization of the spins of the current carrying electrons. There are proposals for nanorefrigerators in which heat is carried by an electronic charge current \[2\] without involving spin. In a recent Letter \[3\], we have introduced a cooling scheme based on the interplay of an applied spin-polarized current, the dot magnetic moment and its vibrational degree of freedom.
In the present work, we provide an extended analysis of the underlying mechanism in terms of a sequence of elementary processes involving tunneling, flip-flop processes of spin and magnetic moment and vibrational transitions. This interplay leads to a net cooling effect for an anti-parallel polarization of the FM source and drain leads and a positive bias voltage. In extension to our previous work, we show that also net heating can be generated by inverting the sign of the bias voltage to negative values. Moreover, we show that the cooling effect for the anti-parallel polarization of the leads is stronger for strong polarization. In the parallel configuration, the heating is enhanced for more pronounced parallel polarization. Furthermore, we show that dynamical cooling is also possible with parallel lead alignments, if the two tunneling barriers are not equal. In addition to our previous work [3], we illustrate here that the nanocooling mechanism is a nanoscale adaption of the ubiquitously used magnetocaloric demagnetization cooling [1] which operates on the macroscale by exploiting magnetic degrees of freedom of bulk materials.

A similar approach has recently and independently been proposed in [4, 5] for the particular case of a suspended carbon nanotube quantum dot in contact with two FM leads and with the dot’s magnetic moment coupled to the flexural mechanical modes. A key component of both approaches is an effective magnetomechanical coupling between the magnetic and vibrational degrees of freedom of the molecule. There are several examples of physical systems in the literature in which such a coupling is significant. Theoretical suggestions to realize a sizable magnetomechanical coupling include interactions between a nanomechanical cantilever with a FM tip [8], a nitrogen-vacancy (NV) impurity in diamond which couples to the magnetic tip of a nanomechanical cantilever [9], single molecular magnets or nanoparticles sitting on doubly clamped nanobeams [10], or a single electron spin which couples to a flexural mode of a suspended carbon nanotube [11]. Experimental investigations of such systems were able to identify magnetomechanical interactions as well. For example, the coupling of the magnetic tip of a cantilever to an individual electron spin has been detected [12]. A clear sign of a magnetomechanical coupling has also been reported for a single NV center coupled to a SiC cantilever [13, 14]. Furthermore, a strong coupling between the magnetic moment of a single molecular magnet and a suspended carbon nanotube was demonstrated recently in [15].

On the macroscopic scale, a scheme of cooling a sample using the magnetic degree of freedom has first been suggested in 1927 by Peter Debye and William F Giauque [6]. The mechanism is widely known as demagnetization cooling and lead to the development of the first magnetic refrigerators with the ability to cool samples to temperatures below 0.3 K. These magnetic refrigerators utilize adiabatically changing magnetic fields, in which a magnetic sample is placed, in order to invoke a change in temperature of the sample. The fundamental principle of the demagnetization cooling is the transformation of thermal energy into magnetic energy in an isolated sample which occurs during an adiabatic reorientation process of the magnetic moments of the sample. The generic principle is idealized in a thermodynamic cycle, which is illustrated in figure 1. It consists of four steps:

(I) Adiabatic magnetization: A bulk magnet is placed in an increased external magnetic field $H$, thereby being isolated from any thermal environment. This causes the magnetic moments of the material to align. Since the substance is thermally isolated, both the entropy and the energy are conserved. Correspondingly, this
leads to an increase in the temperature \((T \rightarrow T + \Delta T)\) to compensate for the decrease in the magnetic entropy in the closed system.

(II) Isomagnetic enthalpic transfer: In order to remove the excess heat, it is necessary to bring the material in thermal contact with a coolant. The magnetic field is held constant in this step in order to prevent the magnetic moments from reabsorbing the heat. Once the magnetic solid is cooled down to its initial temperature \((T + \Delta T \rightarrow T)\), the coolant can be removed, and the magnetic material is again isolated.

(III) Adiabatic demagnetization: In the third step, the magnetic field is (adiabatically) decreased, such that the magnetic moments can stochastically reorientate themselves. Since both energy and entropy are once again conserved in the isolated system, the relaxation of the magnetic moments leads to a decrease of the temperature \((T \rightarrow T - \Delta T)\) in the material.

(IV) Isomagnetic entropic transfer: In the final step, the magnetic field is held constant and the material is brought into thermal contact with the sample which is to be cooled. The resulting heat exchange cools the sample, while bringing the material back to its original temperature \((T - \Delta T \rightarrow T)\). Once thermal equilibrium is reached, the cycle can be restarted.

While being successfully applied at the macroscale, a conceptual difficulty of the magnetic refrigeration cycle for miniaturization is the opening and closing of heat links. In a macroscopic setup, the transfer of the excess heat to a coolant (step (II)) requires either to pump the coolant or to mechanically move the magnetic material. In the same way, a thermal contact between the material and the sample (step (IV)) has to be established. These technical constraints of the cooling cycle render the transfer of the concept to the nanoscale difficult. A mechanical move of the setup or large cooling equipment proves largely impractical for most applications to nanodevices. An example would be the development of microchips on the nanoscale where they are to be used in large numbers as part of electronic hardware. Movement of individual microchips or even arrays of microchips as part of a cooling concept will interfere with their constraints in a possible technological application. Additionally, the use of changing external fields for magnetization hinders an application of such a cooling scheme for magnetic memory devices.

Nonetheless, the nanocooling scheme proposed in this work is also based on the exchange of energy between magnetic and phononic degrees of freedom. The core of our concept is the use of spin-polarized currents in order to steer the local magnetic moment. Contrary to the macroscopic cooling cycle which relies on equilibrium thermodynamics, we use quantum transport properties of nanodevices to develop a cooling scheme under nonequilibrium conditions. Using spin-polarized currents for affecting the local magnetic moment yields the advantage of being able to control the contact between the sample and the environment all-electronically by using bias voltages. The environment is represented by FM electronic leads which act as reservoirs of spin-polarized electron. Applying a bias voltage to the leads generates the flow of a spin-polarized current through the nanodevice. The polarized spins can interact with the local magnetic moment of the nanodevice via exchange coupling. This replaces the function of the external magnetic field \(H\) of the macroscopic setup. It is therefore possible to generate and control the magnetic moment of the sample directly by using its charge transport properties. Additionally, the contact to the environment allows the transfer of entropy and energy, thereby reducing the heat generated by the magnetization process (step (I)). The polarization of the local magnetic moment of the nanodevice by the current forces the device dominantly into the spin ground state, thereby lowering its energy. Due to a magnetomechanical coupling between the local magnetic moment and the vibronic degrees of freedom of the device, it is possible for the spins to relax again by reducing the vibrational energy and thus cooling the device (step (III)). In a nonequilibrium set-up, a flowing electron current, however, gives rise to Ohmic heating effects which might counteract the initial goal of cooling the device. Put differently, the spin-polarized current thus induces two effects: a wanted reduction of the magnetic energy of the device, and, an unwanted heating due to Ohmic losses. An effective cooling protocol can only be established if the cooling effect due to magnetization exceeds the heating generated by Ohmic losses. It is a major purpose of this work to illustrate that this is possible and to identify the regimes of the parameters which allow for a realization of this cooling concept.

2. Model

In order to determine the nonequilibrium spin and vibrational dynamics of the magnetic nanorefrigerator, we employ a minimal model for a proof-of-principle. Its minimal ingredients consist of a magnetic quantum dot with a single electronic level, a local magnetic moment \(J\) and a single vibrational mode as sketched in figure 2. The dot is weakly coupled to FM leads via tunneling thus generating spin and charge transport.
More specifically, the quantum dot is modeled by a single electronic level with energy $\epsilon_0$. The Coulomb repulsion of electrons on the dot defines the charging energy of the dot. For small dots, the local charging energy is assumed to exceed all other energies, and a two-electron dot occupancy is energetically forbidden. Again, this assumption is not crucial but is invoked only to keep the minimal model simple. The Hamiltonian of the dot can then be written as (we set $\hbar = 1$)

$$H_d = \epsilon_0(a_\uparrow a_\uparrow + a_\downarrow a_\downarrow) + g \mu_B B s_z,$$

with the electron annihilation and creation operators $a_\sigma$ and $a_\sigma^\dagger$, the g-factor $g$, and the Bohr magneton $\mu_B$. The spin projection quantum number is $\sigma$ and $s_z$ the $z$-component of the electron spin $s = \frac{1}{2} \sum_{\sigma, \sigma'} a_{\sigma}^\dagger \sigma_{\sigma'} a_{\sigma'}$. An external global magnetic field $B$ splits the spin states along the quantization axis. A spin-1/2 impurity represents the minimal model for the local magnetic moment of the quantum dot. A generalization to higher spin values is straightforward such as, for example, in [15]. We denote with $J_z = \pm 1/2$ the projection of the local magnetic moment onto the quantization axis. The corresponding Hamiltonian for the dot’s magnetic degree of freedom is given by

$$H_{J_z} = g \mu_B B J_z + q (s \cdot J),$$

where the electronic spin and the local magnetic moment $J$ are coupled by an exchange interaction of strength $q$.

For simplicity, the exchange coupling is assumed to be isotropic. A single vibrational mode (or, phonon) of (angular) frequency $\omega$ is coupled to the quantum dot in order to be able to study the dynamical heating and cooling of the device. With the bosonic ladder operators $b$ and $b^\dagger$, the phonon Hamiltonian reads

$$H_{ph} = \omega b^\dagger b + \lambda (b + b^\dagger) \sum_\sigma \sigma_{\sigma}^\dagger a_\sigma,$$

Due to the linear coupling of the electronic occupation to the oscillator displacement, it is possible to excite or relax the vibrational mode with each electronic tunneling process which alters the electron population of the dot. These excitations describe the inevitable heating of the nanodevice due to the charge current whereas relaxation processes of the vibrational mode describe a cooling effect. A decrease of the vibrational energy of the phonon mode is possible if its energy exceeds the thermal energy of the FM leads. The latter is the main mechanism used for passive heat sinks via the environment.

Energy exchange between the vibrational mode and the local magnetization is enabled by the magnetomechanical coupling given by

$$H_{J_z^- ph} = \xi (b + b^\dagger)(f_+ + f_-),$$

where $f_\pm = (f_e \pm i f_h)/2$ are spin-1/2 ladder operators. They induce transitions between the magnetic states with a simultaneous relaxation or excitation of the vibrational mode. It should be realized that equation (4) describes an effective coupling. Interactions between a vibrational mode and a spin can be the result of spin–orbit effects. For instance, it is shown in [16] that a combination of electron–phonon and spin–orbit interaction leads to an effective coupling between spin and phonons. A projection of the Hamiltonian onto the low energy sector yields an effective spin–phonon coupling. Consequently, contributions from higher lying orbital states are projected out. In that way the coupling constant $\xi$ can then be derived as the ground-state expectation value of a combination of electron–phonon operators and the generator of the transformation. Notice that the resulting coupling Hamiltonian as presented in equation(4) breaks time-reversal symmetry which could be restored by taking into account the higher lying orbital states for the electron. In this work, we will only focus on the effect of the magnetomechanical coupling in the context of a possible nanocooling mechanism. Hence, we...
use an effective description and choose both the electron–phonon coupling \( \lambda \) and magnetomechanical coupling \( \xi \) independently.

A comparison with existing experimental setups yields parameter values for both coupling strengths.

Ganzhorn et al [15] have realized a set-up with a single molecular magnet covalently bound to a carbon nanotube suspended between two leads. The molecule has a magnetic ground state of \( |J\rangle = 6 \) and the ground state doublet \( |L\rangle = \pm 6 \) is separated from the excited states by several hundreds of Kelvin. A relaxation or excitation of the spin state therefore dominantly occurs via quantum spin tunneling between those two states. The Ising-like spin flip can therefore be described effectively by a spin-1/2 impurity. The spin flip is accompanied by a transition in the vibrational mode and one finds the parameters [15] \( \omega = 34 \) GHz, \( \xi = 1.5 \) MHz, and \( \lambda = \omega \sqrt{g} = 26 \) GHz for \( g = 0.6 \), implying that \( \xi / \omega \sim 4 \times 10^{-2} \) and \( \lambda / \omega \sim 0.76 \). Likewise, an estimate of realistic parameters can be obtained for the aforementioned NV centers [13].

Experimental results yield a vibrational frequency of about \( \omega = 2 \pi \times 625 \) kHz and a coupling strength of \( \xi = \mu_n B \sqrt{1/(2 m \omega)} \sim 172 \) Hz, implying that \( \xi / \omega \sim 4 \times 10^{-6} \). Another realization [14] yields \( \omega = 2 \pi \times 80 \) kHz and \( \xi \sim 8 \) Hz, such that \( \xi / \omega \sim 10^{-4} \). For the theoretical investigation of this work, we choose the strength of the magnetomechanical coupling \( \xi / \omega \) to be comparable to the electron–phonon coupling \( \lambda / \omega \).

The nanodevice is coupled to two FM leads via tunnel coupling. In general, the magnetization directions of the leads can be noncollinear. They are modeled as noninteracting electron reservoirs

\[
H_{\text{leads}} = \sum_{k \alpha} (\epsilon_{k \alpha} - \mu_n) c_{k \alpha}^\dagger c_{k \alpha},
\]

(5)

where \( c_{k \alpha \pm} \) represents the annihilation operator for an electron with the wave number \( k \) and the majority/minority spin in the lead \( \alpha = L, R \) and \( \mu_{L/R} = \pm eV / 2 \) is the chemical potential of the leads shifted by the applied bias voltage \( V \). FM materials exhibit a different density of states at the Fermi energy for different spin species. If we define majority and minority spin carriers along the quantization axis of the quantum dot, we can quantify the polarization \( p_{\alpha} = (u_{\alpha+} - u_{\alpha-}) / (u_{\alpha+} + u_{\alpha-}) \) of lead \( \alpha \) by the relative difference in the density of states \( u_{\alpha \pm} \) for majority/minority spins at the Fermi energy. In this work, we use \( p_L = p_R = p \) and a generalization is straightforward. All energies are measured relative to the Fermi energy at zero polarization. The FM leads induce an exchange magnetic field on the dot [17, 18] which depends on the relative angle of the magnetization directions of the leads. Spin dynamical effects, which in turn affect the vibrational dynamics, are influenced by this field. Since we aim to cool the device by temporarily increasing the occupation of the spin ground state [3], the source–lead polarization is chosen to be antiparallel to the externally applied magnetic field.

In the following, we consider three set-ups with the drain polarization parallel (\( \uparrow \)), perpendicular (\( \downarrow \)) or antiparallel (\( \downarrow \)) to the source. To have an overall quantization axis, the tunneling Hamiltonian depends explicitly on spin rotation matrices according to

\[
H_{\text{tun}} = \sum_{k \alpha \gamma} [\eta_{k \alpha} A_{\gamma \mu} \Lambda_{\mu \nu} \Lambda_{\nu \gamma}^\dagger c_{k \gamma \alpha}^\dagger + \text{h.c.}],
\]

(6)

with \( A = (a_+, a_-), C_{\gamma \alpha} = (c_{k \gamma \alpha}^\dagger, c_{k \alpha}^\dagger) \) and \( N^{\uparrow \downarrow} = N^{R, L, \downarrow \uparrow} = 1, N^{R, L, \uparrow \uparrow} = \sigma_3 \) and \( N^{R, L, \uparrow \downarrow} = (1 - i \sigma_3) / \sqrt{2} \) for the three setups. The hybridization with the dot state in the wide-band limit is given by \( \Gamma_\alpha = 2 \pi |t_{\alpha a}|^2 \langle u_{\alpha+} + u_{\alpha-} \rangle \). The total Hamiltonian is given by \( H = H_{\text{sys}} + H_{\text{leads}} + H_{\text{tun}} \). Below, we will work in the interaction picture and the tunneling term represents the interaction, while \( H_0 = H - H_{\text{tun}} \).

3. Energy spectrum

The spin and vibrational dynamics of the system follow from the spectrum of the system Hamiltonian

\[
H_{\text{sys}} = H_d + H_f + H_{ph} + H_{\text{ph}}. \]

For vanishing magnetomechanical coupling \( \xi = 0 \), a polaron transformation

\[
\mathcal{U} = e^{-\frac{1}{2} \lambda b^\dagger b} \sum_{n} a_n^\dagger a_n,
\]

(7)

yields the rotated system Hamiltonian

\[
\mathcal{U} H_{\text{sys}}^0 \mathcal{U}^\dagger = \left( \epsilon_0 - \frac{\lambda^2}{\omega} \right) (a_+^\dagger a_+ + a_-^\dagger a_-) + g \mu_B B (s_z + I_z) + q (s \cdot J) + \omega b^\dagger b,
\]

(8)

where \( H_d \) remains unaffected by this transformation. Consequently, we are left with an off-diagonal component of the Hamiltonian due to the exchange coupling \( g \). In the absence of tunneling, we can find an eigenbasis formed by the singlet and triplet states of the two spin-1/2 moments of the electrons and the magnetic moment [19]. The eigenstates and -energies are given in table 1. Especially, the bare energy of the electronic level is shifted by the polaron energy \( \lambda^2 / \omega \). In addition, the coupling between electronic and vibrational degrees of freedom is shifted to the tunneling Hamiltonian. Typical terms within the rotated tunneling Hamiltonian involve
Table 1. Eigenstates and eigenenergies of the Hamiltonian of the electron-impurity subsystem for vanishing magnetomechanical coupling $\xi = 0$.

| Eigenstate | Energy |
|------------|--------|
| $|0\uparrow\rangle$ | $\epsilon_0 = B/2$ |
| $|0\downarrow\rangle$ | $\epsilon_0 = -B/2$ |
| $|T^+\rangle = |\uparrow\downarrow\rangle$ | $\epsilon_{T^+} = \epsilon_0 - \lambda'\omega + q/4 + B$ |
| $|T^-\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ | $\epsilon_{T^-} = \epsilon_0 - \lambda'\omega + q/4$ |
| $|T^-\rangle = |\downarrow\downarrow\rangle$ | $\epsilon_{T^-} = \epsilon_0 - \lambda'\omega + q/4 - B$ |
| $|S\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$ | $\epsilon_S = \epsilon_0 - \lambda'\omega - 3q/4$ |

Each tunneling event is accompanied by an excitation or relaxation of the vibrational mode. The probability for such a process is given as the negative exponential of the ratio $\lambda/\omega$. The spin state with lowest energy of the system for a given order of phonon transitions can now be identified depending on the values for the magnetic field $B$ and the exchange coupling $q$, see table 1. With $B, q > 0$, the state with the lowest energy is the empty dot and with magnetic moment polarized downwards, the downwards polarized triplet state or the singlet. In the regime $q > 4\epsilon_0/3 + 3B/4$ the lowest-energy spin state is formed by the singlet $|S\rangle$. If the magnetic field is increased such that $B > 2\epsilon_0 + q/2$ the triplet state $|T^-\rangle$ has the lowest energy.

We note that a diagonalization of the system Hamiltonian for a finite magnetomechanical coupling $\xi \neq 0$ can be done numerically. The numerical effort for solving the eigenvalue problem depends mainly on the dimension of the Hamiltonian, i.e., on the maximal number $K$ of possible phonon states that are taken into account. In this work, we want to utilize the vibrational mode as a means to monitor the effective temperature of the nanodevice and, therefore, it is necessary to keep all information about the state of the phonon mode. The results shown in this work are convergent with typically $K = 6$ phonon states. Calculations with a larger number of phonon states have shown corrections of less than one percent to the results presented in this work.

4. Nonequilibrium quantum dynamics

We use the quantum master equation whose derivation starts with the Liouville–von-Neumann equation for the full density operator $\rho^I(t)$ in the interaction picture

$$\dot{\rho}^I(t) = -i[H^I_{\text{tot}}(t), \rho^I(t)].$$

The superscript $I$ denotes operators in the interaction picture defined as $O^I(t) = e^{itH_o(t-t_0)}O(t)e^{-itH_o(t-t_0)}$ with $H_0 = H - H_{\text{tot}}$. The formal solution of this equation can be found by integration in the form

$$\rho^I(t) = \rho_0^I - i \int_{t_0}^{t} dt' [H^I_{\text{tot}}(t'), \rho^I(t')].$$

Reinserting this solution back into equation (10) and iterating the equation yields

$$\dot{\rho}^I(t) = -i[H^I_{\text{tot}}(t), \rho_0^I] - \int_{t_0}^{t} dt' [H^I_{\text{tot}}(t'), [H^I_{\text{tot}}(t'), \rho^I(t')]].$$

We consider the usual limit when the dynamics is Markovian, such that the memory time $\tau$ for time correlations of the bath and system is much shorter than any system or bath time scale. Furthermore, we restrict ourselves to the regime of sequential tunneling. If the contact between the system and the reservoirs is sufficiently weak, it is reasonable to assume that the electron tunneling occurs sequentially. Carrying out the trace over reservoir degrees of freedom and using factorizing initial conditions between bath and system at initial time $t = t_0$ yields the kinetic equation for the reduced density matrix

$$\dot{\rho}_{ab}(t) = -i(\epsilon_a - \epsilon_b)\rho_{ab}(t) - \sum_{cd} R_{abcd}\rho_{cd}(t),$$

where indices $a, b, c, d$ refer to the energy eigenstates of the reduced system. Furthermore, $R_{abcd}$ is the Redfield tensor containing the rates for transferring the system from one state to another by electron tunneling from $a$ into the leads. Those are obtained by evaluating the second term on the right-hand side of equation (12), thereby exploiting the Born–Markovian approximation. In presence of a coupling of the electronic level to vibrational and magnetic degrees of freedom, it is important to keep also track of the off-diagonal elements of $\rho_{ab}$ which carry information about the quantum coherence in the system. In this work, we solve the kinetic equation for $\rho$ numerically, and the stationary solution of equation (13) is given by
The zeroth order term describes the coherent dynamics of the quantum system and is absorbed in the modified Redfield tensor \( \hat{R} \). To determine the time evolution of \( \rho_{\text{lab}}(t) \), we expand the formal solution \( \rho(t) = e^{\hat{H}t} \rho(0) e^{-\hat{H}t} \) in terms of the left and right eigenvectors of \( \hat{R} \), such that

\[
\rho(t) = \sum_{m} \psi_{ml}^{\dagger} \psi_{lr} \rho \psi_{lr}^{\dagger} \psi_{ml} = \sum_{m} \psi_{ml}^{\dagger} \rho(t_0) e^{-i \Gamma_m t} \psi_{lr}.
\]

Here, \( \psi_{ml}^{\dagger} \) and \( \psi_{lr} \) denote the \( m \)th left/right eigenvector and \( \Gamma_m \) the eigenvalues of the modified Redfield tensor. Knowledge of the time evolution of the reduced density matrix then allows us to calculate expectation values of any desired operator, such as average spin projections, the number of electrons on the dot or the occupation of vibrational states. Accordingly, it is also possible to determine the charge current flowing through the device by counting the change of charges in lead \( \alpha \), i.e.

\[
I_{\alpha} = -\epsilon \left\{ \frac{\partial n_{\alpha}}{\partial t} \right\} = -\epsilon i [H_{\text{int}}, n_{\alpha}].
\]

Often we use the symmetrized expression

\[
I = \langle I_L + I_R \rangle,
\]

where expectation values are calculated by means of the knowledge of \( \rho_{\text{lab}} \) given in equation (13).

5. Principle mechanism

Before discussing the numerical results in more detail, we provide a qualitative picture of the cooling scheme by explaining the principle mechanism under some idealized limiting conditions. For the sake of this discussion, we will assume fully polarized leads in an anti-parallel configuration. The dot is initially empty, i.e., the number of electrons on the dot is zero, and the local magnetic moment \( J \) is in an excited state and aligned parallel to an external magnetic field \( B \) along the dot quantization axis. The FM leads generate an effective local magnetic field on the dot. In order to obtain an interaction between the electronic spin and the local magnetic moment, either a noncollinear orientation of the effective field and the external field is necessary, or—as we choose here—a finite exchange coupling \( g \) between the local magnetic moment and the electronic spin is required. Due to the thermal contact between the leads and the system, the vibrational mode is assumed to be initially in a thermal state with the same temperature \( T \) as the FM leads. A perfect polarization of the source lead anti-parallel to the dot quantization axis provides only spin-down electrons while the drain lead is polarized anti-parallel to the source along the quantization axis. Therefore, it can accept only spin-up electrons. The cooling mechanism can then be understood in terms of the following elementary processes (they are sketched in figure 3):

(I) Applying a finite bias voltage leads to spin-down electrons tunneling onto the initially empty quantum dot. The dot magnetic moment is polarized upwards.

(II) Due to the exchange coupling between the electronic spin and the local magnetic moment, a simultaneous flip of both is possible. The flip lowers the energy of the local magnetic moment.

(III) With the drain being polarized antiparallel to the source, the electron can tunnel out of the dot only after such a spin flip. This restriction lowers the overall current. Due to the electron–phonon coupling of strength \( \lambda \), any tunneling event between dot and leads can lead to an excitation of the vibrational mode. These excitations constitute Ohmic heating and increase the average energy of the vibrational mode above the thermal energy of the leads by a temperature \( \Delta T_e \). The energy of the vibrational mode, however, can be lowered due to the magnetomechanical coupling \( \xi \).

(IV) The vibration can relax when the magnetization is flipped from a low-energy (spin down) into the high-energy (spin up) state parallel to the external field, thereby extracting the necessary energy from the vibrational mode. This reduces the vibrational energy by an amount equivalent to an effective temperature \( \Delta T_v \). The energy stored in the magnetic moment can then again be removed via the interaction with the spin of the next tunneling electron as symbolized in (I). This leads to a net cooling effect in form of a reduced energy in the vibrational mode.

A useful quantifier for the cooling effect is the ratio between the Ohmic heating \( \Delta T_e \) induced by the electronic current and the removal of vibrational energy \( \Delta T_v \) due to spin flips. Furthermore, the polarization of the local magnetic moment by the electronic spin plays an important role. The efficiency of the cooling protocol may be quantified in form of the rate constant which governs the time evolution of the processes. In particular, it may in general depend on the lead polarization, the polarization directions, the electron–phonon coupling and...
magnetomechanical coupling strengths, and the energy difference between the high- and the low-energy spin state induced by the external magnetic field.

6. Initial preparation

In order to quantify a cooling effect, it is necessary both to define an effective temperature of the dot via the vibrational degree of freedom of the system as well as to prepare the system in a meaningful initial state. The stationary solution of the Markovian quantum master equation does not depend on the initial state of the system. We are, however, interested in the spin and the vibrational dynamics of our system. The time evolution of the density matrix explicitly depends on the initial state. While choosing an arbitrary preparation similar to that in the preceding section might yield a meaningful time evolution, it might very well lead to misleading results with respect to the cooling efficiency. Hence, we amend this with two extreme examples. Imagine the
device to be prepared in the ground state of the vibrational mode. In this case, the effective temperature is at a
minimum which makes further cooling of the vibrational degrees of freedom impossible. Similarly, if we prepare
the system in a highly excited state, we start with a high effective temperature. If the initial effective temperature
is higher than the base temperature of the FM leads, a cooling effect will occur due to the thermal contact
between the device and the environment. This cooling effect however does not differ from the mechanism used
in passive cooling devices as described above. Our aim is to investigate the viability of the dynamical
demagnetization cooling on the nanoscale via spin-polarized currents. For this, a realistic preparation is to
initially place the nanodevice in thermal equilibrium with the leads. In this way, passive cooling effects are
maximally suppressed and a decrease of the effective temperature below this initial preparation will show the
viability of a dynamical cooling scheme. Such a preparation can be realized numerically by calculating the
stationary solution of the quantum master equation for the system being in contact with the FM leads but
without a bias voltage, i.e., $eV = 0$.

7. Effective temperature

For a quantitative analysis of the cooling mechanism, we need to define the effective temperature of the system.
The model system includes a single vibrational mode to represent the thermal state of the system. Relaxation and
excitation of the vibrational mode out of its initial state reflects cooling and heating processes and are possible via
the electron-vibrational coupling and the magnetomechanical coupling. The average energy of the vibrational
mode can therefore serve as a measure of the temperature of the nanorefrigerator according to

$$T_{\text{eff}} = \langle H_{\text{ph}} \rangle.$$  

(18)

In principle, the way to define an effective temperature via the vibrational energy is not unique. Phonon operators
appear in the magnetomechanical coupling term, the electron–phonon coupling and the Hamiltonian for the
harmonic oscillator. A numerical comparison between different definitions of the phonon temperature including
and excluding both couplings $\lambda$ and $\xi$ shows no significant differences in the investigated regime of parameters. To
be specific, we choose to include the electron–phonon coupling into the definition while we exclude the
magnetomechanical coupling. The initial effective temperature will be denoted by $T_{\text{ini}} = T_{\text{eff}} (t = 0)$.

In figure 4, we depict the initial effective temperature as a function of the given lead temperature. We find
that, as expected, the effective temperature is not equal to the temperature of the FM leads. Instead it matches the
mean thermal energy of a harmonic oscillator with frequency $\omega$ given by

$$\langle E_{\text{harm}} \rangle = \frac{\sum_{k=0}^{K} k\omega e^{-k\omega/T}}{\sum_{k=0}^{K} e^{-k\omega/T}},$$  

(19)

at the lead temperature $T$. This is marked by the red data points in figure 4. The agreement between the two
curves indicates that our definition of the effective vibrational temperature is a good measure for the average
energy of the phonon mode even in the presence of the FM leads.

8. Effective cooling

The choice of suitable parameters for an optimal cooling protocol is guided by the requirement of optimal
efficiency (apart from numerical convenience in this model study). Within the sequential tunneling
approximation in the quantum master equation, we are limited to weak system-reservoir coupling. This is not a principle limitation and can be straightforwardly released if necessary. The tunnel coupling $\Gamma$ will therefore be the smallest energy of the problem. The energy of the electronic level serves as a scaling parameter for all other energies. Temperature $T$, the phonon frequency $\omega$ and the bias voltage $eV$ can be chosen on the same order of magnitude as the electronic level. Furthermore, we will choose a rather strong magnetic field $B$ of the similar magnitude in order to generate a large splitting between the magnetic ground and excited state. This energy splitting corresponds to the amount of energy we can transfer between the magnetic and the vibrational degrees of freedom via a spin flip induced by the magnetomechanical coupling $\xi$. The coupling strengths $\lambda$ and $\xi$ are chosen to be one or two orders of magnitude smaller. The electron–phonon coupling $\lambda$ has to be small enough to allow the numerical calculations to reach convergence within a reasonable amount of time. On the other hand, it still has to be sufficiently strong to observe signatures of the electron–phonon interaction and heating effects. The magnetomechanical coupling $\xi$ should typically be smaller or at most of the same order of magnitude as is suggested by the experimental values listed in section 2. Here, we choose on purpose a rather large $\xi$ to demonstrate the cooling effect.

Another important set of parameters refers to the lead polarizations and alignments of field directions. Both the magnitude as well as the direction of the lead polarization will have a significant effect on the cooling efficiency since they directly control the spin and the lifetime of electrons on the quantum dot [18]. Consequently, they affect the local magnetic moment via the exchange coupling. To develop an efficient cooling protocol, we compare different setups for the FM leads. We investigate the spin accumulation, the charge current, the number of electrons on the dot, and the effective phonon temperature for the following three different lead setups: a parallel alignment ($\downarrow\downarrow$) of the source and drain polarization, a perpendicular arrangement ($\downarrow\rightarrow$), and, an anti-parallel alignment ($\downarrow\uparrow$).

In the following, we discuss the cooling effect along three different quantities: figure 5 shows the time-dependent probability $P_\uparrow(t)$ to find an electron on the quantum dot. Figure 6 depicts the associated time-dependent average $z$-component $\langle J_z \rangle$ of the magnetic moment of the quantum dot. Moreover, in figure 7, we show the time-dependent charge current $\langle I \rangle$, and in figure 8, the effective time-dependent temperature $T_{eff}$ is shown. Next, we address the three different lead setups in more detail.

8.1. Parallel setup
The parallel setup ($\downarrow\downarrow$) is marked by the black dashed lines in figures 5–8. In this constellation, electrons which carry a majority spin are able to pass through the quantum dot without the necessity for a spin flip. Thus, no spin blockade occurs and the electrons tunnel through the quantum dot mostly without interacting with the local magnetic moment $J$. We therefore expect an average occupation number of the quantum dot of about $1/2$ and a weakly polarized local magnetic moment being antiparallel to the dot quantization axis. This is confirmed in figures 5 and 6, respectively. Even though the average occupation $P_\uparrow$ of the quantum dot is close to that of the other setups, the individual lifetime of each electron on the dot is rather short. Proof of this can be seen in the charge current depicted in figure 7. As a result of the rather large current in the steady state, we expect a relatively large influence of Ohmic heating effects. Each transfer of an electron has a finite probability of exciting the vibrational mode. Almost free charge flow through the quantum dot can therefore be expected to generate fast heating of the vibrational mode as compared to a regime where the current is blocked. Simultaneously, due to the weak interaction with the magnetic moment, spin flips are unlikely to happen, thus reducing the efficiency of the cooling mechanism. Consequently, the effective temperature of the nanodevice slightly increases with time and no cooling effect can be observed (see figure 8).
8.2. Perpendicular setup
The red dotted–dashed lines in figures 5–8 show the results for perpendicularly aligned lead polarizations (\(\downarrow\rightarrow\)). The different density of states in the source and drain lead for majority and minority spin carriers causes a suppression of the current as shown in figure 7. A substantial spin blockade occurs. The electrons are trapped on the quantum dot and need to precess or exchange angular momentum with the local magnetic moment in order to tunnel into the drain lead. Signatures of spin–spin interactions can be found in the polarization of the local moment \(J_z\) depicted in figure 6. A significant increase in the polarization and therefore a decrease of the magnetic energy can be observed. At the same time the average occupation of the quantum dot decreases with the magnetic moment (see figure 5). The combination of these effects leads to a sizable decrease in the effective temperature of the vibrational mode shown in figure 8.

8.3. Antiparallel setup
The blue solid lines in figures 5–8 depict the antiparallel configuration (\(\downarrow\uparrow\)). Compared to the other setups the highest polarization of the local magnetic moment can be found here. The antiparallel setup shows a larger
average occupation of the quantum dot as compared to the other two setups. Due to the opposite spin carrier distributions in the leads, a strong spin blockade is formed trapping the electron on the device. Figure 5 shows the mean occupation number. The spin blockade generates a strong polarization of the local magnetic moment (see figure 6). The trapped electrons can only leave the quantum dot by flipping their spin simultaneously with the local moment. These spin flip-flops lead to a decrease of the vibrational temperature and provide a strong cooling effect as shown in figure 8.

The \( \downarrow \uparrow \)-setup is shown to be optimal in two respects. First, the polarization of the magnetic moment is enhanced by the long lifetime of the electrons on the dot. This results in a low magnetic energy of \( J \) as compared to the other two setups. Second, the spin blockade forces the electrons to exchange angular moment with the local magnetic moment. The vibrational energy decreases in the process. In a fully polarized case, every transmitted electron will contribute to the cooling process. Additionally, the Ohmic heating effects are directly connected to the charge current and therefore suppressed. This significantly improves the cooling efficiency. In figure 7, we see that the charge current is significantly lower than in the other two setups. We can conclude that the \( \downarrow \uparrow \) configuration is optimal for the purpose of our cooling procedure. For the chosen parameters we can report a decrease of the effective temperature as compared to its initial value for both the antiparallel as well as the perpendicular setup. This completes the proof of principle for the nanocooling scheme initially proposed.

8.4. Dependence on the bias voltage: cooling versus heating

A directly accessible parameter in the experiment is the bias voltage. In this section, we discuss the dependence of the ratio of \( T_{\text{eff}} \) and the initial temperature \( T_{\text{init}} \) on the bias voltage. Figure 9 shows the steady state limit of \( T_{\text{eff}} / T_{\text{init}} \) as function of the bias voltage \( eV \) for the three lead setups. The results are consistent with the results of our investigation of the spin and phonon dynamics in figures 5–8. For the perpendicular and antiparallel lead setup, we observe decreasing temperatures for positive bias voltages and increasing temperatures for larger, negative bias voltages. This effect is a result of the asymmetric polarizations of the FM leads. For negative bias voltages, electrons with spins are injected into the quantum dot from the drain lead and not the source lead. The localized magnetic moment of the quantum dot is therefore polarized in a different direction compared to positive bias voltages, leading to an increase of the effective temperature of the vibrational mode. For the parallel lead setup, no such effect can be observed since both source and drain leads are polarized in the same direction. Thus, the net cooling effect for \( V > 0 \) can be turned into heating by inverting the bias voltage to \( V < 0 \).

In the following, we focus on the antiparallel alignment to further investigate the cooling effect in the region \( V > 0 \).

8.5. Role of the electron–phonon and magnetomechanical coupling

The cooling ratio \( T_{\text{eff}} / T_{\text{init}} \) as a function of the magnetomechanical coupling \( \xi \) is displayed in figure 10. In the whole regime of parameters, a net cooling effect can be observed, since \( T_{\text{eff}} / T_{\text{init}} < 1 \) for the chosen values of the electron–vibration coupling \( \lambda \). Surprisingly, for a fixed \( \lambda \), a nonmonotonic dependence of the cooling as a function of \( \xi \) can be observed. For small couplings, \( T_{\text{eff}} \) steadily decreases with increasing \( \xi \). However, a minimum is reached where cooling is optimal. For further increasing \( \xi \), the effective temperature increases again. The energy spectrum of the system without spin–phonon interactions is given in section 3. Finite values for \( \xi \) will lead to corrections of the calculated eigenenergies and can lead to a change of the magnetic ground state. Since the cooling scheme relies on a polarization of the local magnetic moment to the magnetic ground state, large values for \( \xi \) can therefore decrease the cooling ratio.
Moreover, as may be intuitively expected, an increase of the electron–phonon coupling $\lambda$ leads to an increase of the effective temperature as the result of Ohmic heating. This is depicted in figure 11 for varying $\lambda$ for three values of the magnetomechanical coupling $\xi$. The heating effect is, however, outmatched by the magnetic cooling as long as the strength of the magnetomechanical coupling $\xi$ is of the same order of magnitude as the electron–phonon coupling.

A complete picture of the ratio $T_{\text{eff}} / T_{\text{init}}$ as a function of the electron-vibration coupling $\lambda$ and the magnetomechanical coupling $\xi$ is displayed in figure 3 of [3]. A cooling effect is achieved in the full parameter regime depicted.

### 8.6. Influence of the magnetic field and exchange coupling

In figure 12, the steady state limit of $T_{\text{eff}} / T_{\text{init}}$ is shown as a function of the magnetic field $g\mu_B B$ and the magnetic exchange coupling $q$ for a fixed value of $\xi$. As discussed in our previous work [3], the cooling effect is more pronounced for higher magnetic field since the energy gain due to the spin polarization is proportional to the magnetic field. A strong exchange coupling changes the magnetic ground state similar to the previously discussed effect for the magnetomechanical coupling. The efficiency of the cooling protocol, however, directly depends on the energy necessary for a spin flip. A change of the magnetic ground state can lower this energy and therefore decrease the amount of heat extracted from the vibrational mode with each spin flip. In addition to our previous work [3], we address the additional fine structures in figure 12. They are due to resonances between spin flips and vibrational transitions. Since the magnetomechanical coupling is small ($\xi = 0.06\varepsilon_0$), we can give an analytic approximation for these resonances in the noninteracting limit ($\xi \to 0$). The dashed lines in figure 12 indicate these resonances using the eigenenergies given in table 1. The states $|T^+\rangle$ and $|T^-\rangle$ are split by an energy difference $\Delta \varepsilon = 2B$. For the states $|T^0\rangle$ and $|S\rangle$, the energy difference equals $\Delta \varepsilon = q$. A resonance occurs whenever these energy differences are equal to the frequency of the vibrational mode. These two cases are indicated by the horizontal and vertical dashed lines. Finally, when $B = q$ we find that the states $|T^-\rangle$ and $|S\rangle$ are degenerate. This case is indicated by the third dashed line in figure 12. Naturally, the relative level splitting between singlet and triplet states is determined by all coupling constants ($\xi$, $q$, $\lambda$) of the system Hamiltonian and the magnetic field $B$. This interplay gives rise to the nonmonotoneous behavior of $T_{\text{eff}}$ as a function of $\lambda$. 

![Figure 10. Cooling ratio $T_{\text{eff}} / T_{\text{init}}$ in the stationary limit versus the magnetomechanical coupling $\xi$ for fixed values of the electron–phonon coupling $\lambda$. The remaining parameters are the same as in figure 5.](image1)

![Figure 11. Cooling ratio $T_{\text{eff}} / T_{\text{init}}$ as a function of the electron–phonon coupling $\lambda$. The remaining parameters are the same as in figure 5.](image2)
increasing $B$ and $q$ at fixed $\xi$. In particular, the system’s ground state could change which results in a modified cooling ratio in a nontrivial manner.

To further demonstrate the cooling effect, we show the cooling ratio $T_{\text{eff}}/T_{\text{init}}$ as a function of the lead temperature $T$ in figure 13. In addition, the scaled values of $T_{\text{eff}}/\epsilon_0$ and $T_{\text{init}}/\epsilon_0$ are shown. We observe that for all investigated values of the lead temperature $T$, the effective temperature in the steady state limit is lower than its initial value. A cooling effect can be maintained for a range of at least one order of magnitude of the lead temperature. As shown in figure 5(b) in our previous work [3], the initial temperature is not directly proportional to the lead temperature. This is the result of the preparation of the setup and the definition of the effective temperature, as can be seen in figure 4, where we compare it with the average energy of a harmonic oscillator.

8.7. Cooling rate
A useful measure for the efficiency of a cooling protocol is not only the maximally achievable cooling effect, but also the speed at which the temperature can be lowered. We can define an effective temperature as in equation (18) at all times. Comparing the time evolution of the effective temperature in the anti-parallel lead setup for different values of the electron–phonon coupling, we find an exponential approach to the steady state which has been reported in our previous work, see inset of figure 3 in [3]. Since the effective temperature (see equation (18)) is defined by the average energy of the vibrational mode, it depends directly on the relaxation dynamics of the vibrational states. We have seen that the quantum master equation can be solved by an exponential function. The exponent is given by a combination of the Redfield rates and the coherent oscillations of the system. The long-time relaxation dynamics is described by the real part of the smallest eigenvalue, which is composed of Redfield rates. An effective cooling rate $\Gamma_{\text{cool}}$ can therefore be extracted by fitting to an exponential. A typical result of $\Gamma_{\text{cool}}$ as a function of the magnetomechanical coupling is shown in figure 5(a) in [3]. An initial strong increase of the cooling rate with increasing $\xi$ can be observed. However, for $\xi/\epsilon_0 \gtrsim 0.1$, the cooling rate saturates. This result is in agreement with our remarks made in the discussion of figure 12.
8.8. Dependence on the lead polarization

Naturally, the efficiency of the cooling scheme depends on the degree of polarization of the FM leads. In figure 14, we show the effective temperature of the device compared to its initial value as a function of the lead polarization $p$. In figure 14 (a), we depict the results for the antiparallel setup ($\downarrow\uparrow$) which we have used so far to inject spin-down electrons into the nanodevice. The cooling effect scales linearly with $p$ as long as the magnetomechanical coupling is finite. As expected, there is no change in the effective temperature for vanishing coupling. As a side remark, we note that by reversing the polarization of the leads, it is also possible to transform the nanorefrigerator into a nanoheater as shown in figure 14 (b). The source lead then provides spin-up electrons which polarize the local magnetic moment to form an excited state. The magnetomechanical interaction can then only induce spin flips which lower the magnetic energy. The excess energy of such a process is then transferred into the vibrational mode which leads to an increase of the effective temperature.

8.9. Asymmetric tunneling barriers

Finally, we can manipulate the effective temperature by modifying the tunneling couplings. So far, we have considered symmetric tunnel couplings $\Gamma_L = \Gamma_R = \Gamma$. The rate for transitions between the dot and the FM leads to first order in the system-lead interaction are essentially given by Fermi’s Golden rule and therefore directly depend on $\Gamma$. Reducing the tunnel coupling between the nanodevice and the drain lead will thus lead to an overall increase of the lifetime of electrons on the dot. This in turn leads to an increased polarization of the local magnetic moment which leads to an increase of the cooling effect. Figure 15 shows the effective temperature as a function of time for the parallel and the anti-parallel lead setup with asymmetric lead couplings. Compared to figure 8, we do not observe major differences for the anti-parallel setup since the life time of electrons on the dot is mainly determined by the spin blockade. For the parallel setup, however, we do observe a decrease of the effective temperature for times smaller than $t \Gamma \approx 100$. Previously, with symmetric lead couplings, we could not observe any cooling when the lead polarizations were aligned parallel to each other. Reducing $\Gamma$, however, traps the electrons on the quantum dot and yields a decrease of the effective temperature. Establishing a dynamical cooling protocol is therefore possible even with parallel lead alignments, however, under the condition of asymmetric tunnel barriers.

9. Conclusions

We have proposed various extensions of a dynamical cooling mechanism for a magnetic nanodevice based on the interplay of applied spin-polarized currents and magnetomechanical interactions. By establishing a simple model using FM leads and a magnetic quantum dot, we are able to show the viability of the proposed cooling...
scheme in a wide range of parameters. The principle mechanism of the scheme relies on an antiparallel alignment of the source and drain lead polarizations which results in a spin-blockade and therefore in an accumulation of spin on the nanodevice. The resulting charge current and the strong polarization of the magnetic quantum dot yield a cooling of a vibrational mode. The cooling induced by the magnetomechanical coupling overcompensates Ohmic heating effects due to electron–phonon interactions. We find a sizable cooling effect for a wide range of parameters yielding a decrease of the effective temperature of up to 50% below the initial temperature. Surprisingly, a decrease of the cooling effect can be observed for stronger magnetomechanical couplings. In contrast to the cooling effect for positive bias voltages, net heating can occur by inverting the sign of the bias voltage. Clearly, cooling is more pronounced for a stronger anti-parallel magnetic polarization of the leads, while heating is stronger for a larger parallel polarization. Interestingly enough, dynamical cooling is also possible for the setup with parallel lead magnetizations, provided that the two tunneling barriers are unequal.

Future work could extend the minimal model studied here. For instance, the role of higher order tunneling processes for the cooling efficiency is interesting. Moreover, an interesting question is also to study a distribution of phonon modes instead of a single localized mode as considered here. In any case, the proposed effect should also be observable with present day experimental set-ups.

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Figure 15. Effective temperature as a function of time for the anti-parallel (blue line) and parallel (red line) lead setup. The system-lead couplings are chosen to be asymmetric with $\Gamma _L = 0.005\Gamma _0 = 100\Omega _L$. The remaining parameters are the same as in figure 5.