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Relaxation of a coherent, magnetic s–p model system coupled to one and two thermal baths and a laser pulse

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ABSTRACT

We study an s–p model magnetic system with a triplet ground state coupled to two temperature baths. By varying the temperatures we both generate non-thermal electronic distributions and create additional coherences in the density matrix of the system. Thus the thermally-induced magnetic response goes beyond the simple picture of majority-minority population dynamics. Furthermore, we discuss the influence of temperature induced relaxation effects on the dynamics induced by an optical perturbation for this quantum system.

1. Introduction

In today's understanding energy dissipation is a rather macroscopic phenomenon, a fact that needs to be widely reconsidered on a fundamental scale since electronic devices continuously become smaller and thus the microscopic nature of the quantum effects can no longer be neglected. At the same time the microreversibility of closed quantum systems fundamentally conflicts with macro-irreversibility. The spin Seebeck effect was experimentally observed by Uchida et al. for the first time using a technique that involves the spin Hall effect [1]. This allows passing a pure spin current without electric currents, thereby opening the door to spin caloritronics [2–4]. So far, these mesoscopic effects have not been investigated on a microscopic scale [5]. In fact, the research on spintronic devices is currently a booming field and many realizations already show high potential in the field of information processing technology of the future [6–11]. Here, especially relaxation processes are of utmost interest. Atxitia *et* al. [12] showed in a two-temperature model using the Landau-Lifshitz-Bloch equation that the demagnetization as well as the magnetization recovery time slow down as the laser pump fluence is increased.

Although, the influence of a phononic coupling for bulk systems has been investigated [12,13], the effect of relaxation on the electronic and the spin scale [14] for molecular quantum systems has not been exhaustively discussed so far. For such systems two interesting characteristics have been proposed. The first one is the thermalization to a Boltzmann, Bose–Einstein or Fermi–Dirac distribution of a bounded quantum system with eigenfunctions obeying Berry's conjecture even if it is isolated from the environment [15]. The second one states that a closed quantum–mechanical system with finite but very small perturbation and a large number of degrees of freedom behaves in a Markovian way [16]. These two characteristics are the building blocks of the eigenstate thermalization hypothesis. Furthermore, for a closed Heisenberg-type spin system with 12–20 spins the Markovian description is applicable, too [17]. Schowalter et al. [18] found that laser-cooled trapped ions show a bifurcating behavior different from the classical picture of thermodynamics and provide an important contribution to the studies of arbitrarily driven nonequilibrium systems, such as quantum heat engines (QHEs). In fact, the extraction of work with QHEs has become an emerging field as well [19–21]. Not only theoretical, but also experimental realizations of QHEs have been proposed lately [22–24]. For some of them magnetism is an essential ingredient [25–27]. Considering the recent interest in this topic, it has so far been established that nonequilibrium baths as well as the spin degree of freedom can enhance their efficiency and work output [28–30]. Whereas the latter one is achieved by including spin–orbit coupling, the former one is a consequence of an optical perturbation via a laser pulse. Motivated by

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these findings we investigate thermodynamic effects on both an unperturbed and an optically perturbed magnetic model by considering both, the spin degree of freedom as well as the nonequilibrium baths simultaneously.

Here we study a minimal Λ model consisting of s and p states, to which spin–orbit coupling and an external magnetic field are added, and which is propagated in time under the influence of a laser pulse and two heat reservoirs (baths). Experimentally, the coherent optical control of similar systems has been studied for a long time, since it is relevant to at least two research areas: (i) isolated atoms in Stimulated Raman Adiabatic Passage (STIRAP) [31–33], and (ii) the underlying microscopic explanation of the ultrafast laser-induced demagnetization and all-optical switching in extended systems [14,34,35]. Additionally our system is relevant, e.g., for the study of thermoelectric effects of quantum dots attached to ferromagnetic leads [36–38].

2. Theory

In our model system we consider triplet states with the zero-field splitting due to spin–orbit coupling (SOC). Furthermore, a Zeeman splitting distinguishes the spin-up and the spin-down states, which are typically involved as the initial and final states of an optically induced, spin-flipping Λ-process [39]. Mathematically, this is achieved by diagonalizing the Hamiltonian

$$\widehat{H}' = \widehat{H}_0 + \widehat{H}_1, \tag{1}$$

where $\widehat{H}_0$ is the unperturbed part and $\widehat{H}_1$ is the perturbation due to SOC and an additional external magnetic field. It reads

$$\widehat{H}_1 = \sum_{i=1}^{N_{\mathrm{el}}} \frac{Z_a^{\mathrm{eff}}}{2c^2 R_i^3} \hat{\mathbf{L}} \cdot \hat{\mathbf{S}} + \mu_l \sum_{i=1}^{N_{\mathrm{el}}} \hat{\mathbf{L}} \cdot \mathbf{B}_{\mathrm{stat}} + \mu_s \sum_{i=1}^{N_{\mathrm{el}}} \hat{\mathbf{S}} \cdot \mathbf{B}_{\mathrm{stat}}. \tag{2}$$

Here, N_{el} is the number of electrons, c is the speed of light, R_i is the distance of electron i from the nucleus. $\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$ are the operators of angular momentum and spin, respectively, with μ_s being the gyromagnetic moment and $\mu_l = \mu_s/2$. $\mathbf{B}_{\mathrm{stat}}$ is the external, static, magnetic field. This field is necessary in order to distinguish between spin-up and spin-down states (Zeeman effect). Without it the relaxation of the populations would still follow similar patterns, but no spin dynamics would result, as the Kramers degeneracy theorem states. In real systems the role of an effective field can also be played by the magnetocrystalline and exchange anisotropies. For the description of the dynamics we use the density matrix formalism. The thermal exchange of the quantum system with the environment is described by the Lindblad superoperator [40] in the Kraus representation [41,42], where the (unknown) bath-part of the density matrix is traced out. This is possible since we assume a weak coupling. Assuming further a Markovian system, which has no memory, the Lindblad superoperator $\mathcal{L}(\cdot)$ is given by

$$\mathcal{L}(\rho) = \sum_n \gamma_n \left(\hat{c}_n \rho \hat{c}_n^\dagger - \frac{1}{2}\{\hat{c}_n^\dagger \hat{c}_n, \rho\} \right) + \sum_n (\gamma_n + 1) \left(\hat{c}_n^\dagger \rho \hat{c}_n - \frac{1}{2}\{\hat{c}_n \hat{c}_n^\dagger, \rho\} \right) \tag{3}$$

and describes the effect of the bath on the system (non-unitary part of the evolution). It preserves the trace and ensures the hermiticity of ρ. Here $\{\cdot,\cdot\}$ denotes the anticommutator and $\hat{c}_n$ and $\hat{c}_n^\dagger$ are the annihilation and creation operators. We assume that the system-bath interaction results from photon exchange between the system and the bath, therefore $\hat{c}_n$ and $\hat{c}_n^\dagger$ describe only electric-dipole transitions. However, as long as all states can even indirectly couple to all other states, full relaxation to a Boltzmann distribution results for infinitely long times [43]. $\hat{c}_n$ and $\hat{c}_n^\dagger$ are derived from the dipole matrices of the underlying system. The coupling parameters γ_n depend on the strength of the coupling between the system and the bath, the temperature of the bath and the energies of the states coupled by $\hat{c}_n$ and $\hat{c}_n^\dagger$. The two sums in the Lindblad superoperator correspond to thermally induced excitations and deexcitations and the difference +1 of the two respective prefactors results from the two different densities of states of the baths at the relevant energies. Assuming that the bath has a continuous energy spectrum in the energetic range of the quantum system, the corresponding constants are given by the continuous Boltzmann distribution as

$$\gamma_n = \kappa \frac{1}{e^{\beta \Delta E_n} - 1}, \tag{4}$$

with $\beta = \frac{1}{k_B T}$ being the inverse temperature, and κ the constant of the coupling strength between the bath and the quantum system. ΔE_n is the energy difference between initial and final state of the thermally induced excitations and deexcitations. For a coupling to a single bath $\mathcal{L}$ is responsible for two effects: The population relaxation (change of the diagonal elements of ρ) and the dephasing (gradual disappearance of the off-diagonal elements of ρ).

The existence of a bath coupled to the system causes an additional perturbation in the system described by the Lamb-shift term $H_{\mathrm{Lamb}} = \sum_{m,n} \Delta_{m,n} \hat{c}_n^\dagger \hat{c}_m$. This term is responsible for the creation of coherence due to thermal fluctuations. The constants $\Delta_{m,n}$ express the strength of the coupling between the states within the system due to the difference of the thermal population of a single bath at the energies of the coupled system states referred to as Lamb shift [44]. They are given by

$$\Delta_{m,n} = i\delta_{E_m,E_n} \sum_l \frac{1}{1 + e^{\beta}(|E_l - \widetilde{E}_{nm}|)} \mathbf{D}_{ml} \mathbf{D}_{ln} \kappa_{\mathrm{Lamb}}, \tag{5}$$

where $\widetilde{E}_{nm} = |\frac{E_m + E_n}{2}|$, E_m the energy of state $|m\rangle$, κ_{Lamb} is the Lamb-shift constant; for $\kappa = \kappa_{\mathrm{Lamb}}$ the effect is barely observable. $\delta_{E_m,E_n} = \frac{1}{1+(E_m - E_n)^2}$ is a Delta function with a Lorentz broadening. $\mathbf{D}_{ml}$ are the electric-dipole transition matrix elements between states $|m\rangle$ and $|l\rangle$.

Since we furthermore want to study the thermal effect on the optical perturbations, the Hamiltonian

$$\widehat{H}_{\mathrm{laser}}(t) = \hat{\mathbf{D}} \cdot \mathbf{E}_{\mathrm{laser}}(t) \tag{6}$$

is taken into account. Here $\mathbf{E}_{\mathrm{laser}}(t)$ is the time dependent amplitude of the laser, i.e., the electric field.

The Liouville-von Neumann equation together with the Lindblad superoperator and the Lamb-shift now yields the following master equation of the time-dependent density matrix for the quantum system

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}\left[\widehat{H}' + \widehat{H}_{\mathrm{Lamb}} + \widehat{H}_{\mathrm{laser}}, \rho\right] + \mathcal{L}(\rho), \tag{7}$$

where $[\cdot,\cdot]$ denotes the commutator and is responsible for the unitary part of the system's time evolution and $\widehat{H}'$ is the Hamiltonian given in Eq. (1).

The effects of the coupling of a molecular system to two different temperature baths due to the spatial localization of the virtual excitations has already been investigated [45]. In the present work we investigate the effects of the Lindblad superoperator $\mathcal{L}$ and $\widehat{H}_{\mathrm{Lamb}}$ on a model system. Therefore we pick a system with a triplet ground state with a considerably large energy gap to the remaining higher lying states. This is helpful for the investigation of the thermal effects on the triplet state, only. For the following optically induced Rabi oscillations higher lying states are necessary, as it is also for example in the Λ-process.