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Stochastic resonance in photo-switchable spin-crossover solids

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The stochastic kinetic in photo-switchable spin-crossover materials with periodic driving force in the context of stochastic resonance (SR) was studied. The resonance phenomena in spin-crossover system have been analyzed by means of spectral power amplification (SPA) function. The influence of the parameters of harmonic signal (amplitude and frequency) together with changes of noise intensity have been considered. The SPA is characterized by double peak curve with qualitatively different mechanisms of amplification of the peaks and is examined by Fourier analysis.

1. Introduction

Stochastic resonance (SR) as a cooperative, noise-induced phenomenon arises from the interplay between deterministic and random dynamics in a nonlinear system wherein the coherent response to a deterministic monochromatic signal can be enhanced by the presence of an optimal amount of noise [1,2]. Therefore the presence of noise is essential in the nonlinear systems for their optimal performance which is counter-intuitive in comparison to the classical consideration of the role of noise in physical systems [3]. The phenomenon of SR firstly was studied by Benzi and co-workers [4], who introduced the elegant model of earth's climate system wherein the climatic fluctuations and the weak periodic modulations are argued to be the periodic occurrence of such crucial climatic events like ice ages. Many attempts were made to study of SR in various bistable systems, such as lasers, Schmitt triggers, SQUIDs, tunnel diodes, sensory neurons, mammalian neuronal tissue, chemical solutions, communications devices, optomechanical systems etc. [1,2,5–9]. The special role of SR has been reported for sensory neural coding where the processes that occur in the brain are trying to be reproduced [2,10].

The principal tool of the SR theory is the signal-to-noise ratio (SNR) or the ratio between the area under sharp peak at the driving frequency and the noise background that shows a resonance-like curve as a function of the input noise strength. The study of the SR phenomenon by SNR, in the framework of linear-response theory based on the rate equation approach for a two-state model, was developed by McNamara and Wiesenfeld and is presented in paper [11]. However the expressions for the transition probabilities are only valid for very small frequencies and small modulation strengths. For that reason Jung and Hänggi [12] described SR within the framework of stochastic processes and presented numerical results without the restriction to small driving amplitudes or frequencies. Besides this, the specific asymmetric dynamic potential of real systems and different forms of the left and right wells lead to unusual behavior during the SR phenomenon. Such systems may have not only one but several δ-peaks in the power spectrum at the external driving frequency. The signal power amplification (SPA) [1,12–14], that is, the weight of the signal part in the output spectrum, can be employed to investigate SR for such systems. The SNR and SPA show the nonmonotonous behavior as functions of the noise intensity, but since the

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SPA does not depend on the background spectrum it is finer technique for SR characterization. The SPA technique that we have used in the paper is based on a simulation of stochastic ordinary differential equations that describe the evolution of the system.

The problem that we wish to address is cooperative effect caused by noise and harmonic signal on behavior of spin-crossover (SC) systems. The synthesized SC compounds like coordination inorganic complexes with d^4–d^7 electronic configuration of metal ion orbitals, situated in the center of the octahedral ligand field, are prototypic bistable magnetic molecular systems. The most common spin-crossover compound which is actively studied is the one with d^6 configuration. It is characterized by two stable states with different total spin number S: the so-called high spin (HS) paramagnetic state with $S = 2$ and the diamagnetic low spin (LS) one with $S = 0$. The system's states can be changed by means of temperature, pressure, light, magnetic or electrical fields [15–17]. From the point of view of practical applications and possibility of control, the most convenient is interstate transition induced by light irradiation. In this paper the photoexcitation dynamics and characteristic sigmoidal relaxation in SC materials are described by a phenomenological macroscopic evolution equation. The spin-crossover complex is the bistable system with asymmetric potential and may be characterized by two thresholds which correspond to HS and LS states. In other words the spin-crossover complex is an example of bistable multithreshold system, therefore the study of SR in such kind of system is very interesting physical problem. In the same time the experimental realization of SR phenomenon in spin-crossover compound may discover wide range of different practical applications in computing and informational devices. The modern experimental evidence of stochastic resonance in bistable systems has been reported in several recent papers [18–25] where the techniques of its detection are discussed.

The paper is organized as follow: in the Section 2 is discussed the model that we use for studying the phenomenon of SR in nonlinear physical system. The results obtained from numerical simulation are presented in Section 3, where the stochastic dynamics of spin-crossover compound is modeled. Among the obtained results the special attention deserves the dependence of signal enhancement on the frequency of periodic signal. In the Section 4 the summary of the obtained results is provided.

2. Model

For studying the resonance phenomenon in nonlinear magnetic system we considered the photoinduced phenomenological spin-crossover model described in terms of excitation and relaxation processes [7,26,27]. The macroscopic evolution of the system was characterized by dynamics of the fraction of HS molecules n_H following the dimensionless equation

$$\frac{dn_H}{dt} = f_{exc}(n_H) - f_{rel}(n_H) \equiv f(n_H), \tag{1}$$

where photoexcitation and relaxation terms are $f_{exc}(n_H) = \beta(1 - n_H)$ and $f_{rel}(n_H) = n_H \exp(-\alpha n_H)$ respectively. The driven parameter β is determined by intensity of light irradiation and absorption cross section of optically active element. The self-acceleration parameter α determines empirically sigmoidal relaxation in the system. The dimensionless time t is renormalized on the high-temperature asymptotic of the relaxation rate at the end of the process ($n_H \to 0$).

The deterministic master Eq. (1) is valid regarding to extremely long time dynamics (ideally for $t \to \infty$) comparatively to the processes that occur on intramolecular time scale. For this reason more rigorous approach for non-stationary time dynamics requires the accounting of external fluctuations that influence on the stability of system's states. The analytical description of non-stationary evolution of spin-crossover system may be provided through the dynamic Lyapunov potential in framework of Langevin formalism with corresponding Fokker–Planck equation that describes the probability evolution of system's states. The Langevin master equation of stochastic system dynamics takes the form

$$\frac{dn_H}{dt} = -\frac{dU(n_H)}{dn_H} + \xi(t), \tag{2}$$

where $\xi(t)$ is stochastic process describing external fluctuations with zero-mean value and time autocorrelation function $\langle \xi(t)\xi(t') \rangle = 2\varepsilon\delta(t - t')$. Here, the dynamic potential $U(n_H) = -\int f(n_H)dn_H$ describes the system energy at different HS fraction n_H for the deterministic case. Thus the zero-points of first derivative of dynamic potential represent the system's extrema, i.e. stable, metastable and unstable states. About the stability of (meta) stable system states is possible to judge from the analysis of the depth of potential wells which is given in an elegant manner in the work [28].

As it was reported before [7] the enhancement of output signal of non-stationary spin-crossover system is possible if its stochastic behavior is additionally driven by small periodic external force. In the simplest case the periodic driving signal may be described as harmonic oscillation with fixed frequency and follow this assumption the Langevin master equation (2) should be rewritten in the next form

$$\frac{dn_H}{dt} = -\frac{dU(n_H)}{dn_H} + A\cos(\omega t + \phi_0) + \xi(t). \tag{3}$$

Here, A and ω are amplitude and frequency of applied harmonic signal, ϕ_0 is the initial phase (for simplicity it is neglected in our simulation). The steady probability density function obtained as solution of the Fokker–Planck equation for the system