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Implications of a temperature-dependent magnetic anisotropy for superparamagnetic switching

Martin Stier [a], Alexander Neumann [b], André Philippi-Kobs [b,c], Hans Peter Oepen [b], Michael Thorwart [a,*]

[a] I. Institut für Theoretische Physik, Universität Hamburg, Jungiusstraße 9, 20355 Hamburg, Germany
[b] Institut für Nanostruktur- und Festkörperphysik, Universität Hamburg, Jungiusstraße 11, 20355 Hamburg, Germany
[c] Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany

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ABSTRACT

The macroscopic magnetic moment of a superparamagnetic system has to overcome an energy barrier in order to switch its direction. This barrier is formed by magnetic anisotropies in the material and may be surmounted typically after $10^9 - 10^{12}$ attempts per second by thermal fluctuations. In a first step, the associated switching rate may be described by a Néel-Brown-Arrhenius law, in which the energy barrier is assumed as constant for a given temperature. Yet, magnetic anisotropies in general depend on temperature themselves which is known to modify the Néel-Brown-Arrhenius law. We illustrate quantitatively the implications of a temperature-dependent anisotropy on the switching rate and in particular for the interpretation of the prefactor as an attempt frequency. In particular, we show that realistic numbers for the attempt frequency are obtained when the temperature dependence of the anisotropy is taken into account.

1. Introduction

The development of devices for magnetic storage media faces several challenges in the ongoing miniaturization of information units. One of the fundamental physical problems is the so-called superparamagnetic limit. In the small scale limit, the alignment of the macroscopic magnetic moment of the ferromagnetic particle with a direction along a preferred axis is no longer stable, but permanently affected by thermal fluctuations. Despite similarities between para- and superparamagnetism, distinct differences exist. Quantum mechanics allows us to describe paramagnetism rigorously with the magnetic field treated as a small perturbation. In superparamagnetism, the spins of a nanostructure are coupled by the exchange interaction which causes a quasiclassical behavior of a collective magnetic moment with a large variety of possible energetically continuous states. Due to the large number of coupled spins which form the macroscopic magnetic moment, smaller contributions of the single spins to the total energy may become important. They can be rooted in, e.g., dipolar and/or spin–orbit interaction effects. The collection of such secondary effects is commonly summarized to form the anisotropies. The energy scale of a nanomagnet with its small dimensions can become comparable with the thermal energy. The total energy of the nanomagnet with its collectively formed magnetic moment reveals remarkable differences compared to the case of a single moment. In particular, due to the competition with thermal energies, important collective features strongly change as function of temperature. For example, the blocking of the macrospin along a certain direction (easy axis of magnetization) is substantially influenced. Here, two energetically degenerate states are oriented parallel to the easy axis and are separated by an energy barrier ΔE. The latter suppresses thermal switching. In the case of a uniaxial system, the barrier height scales with the volume V and strength of the anisotropy K. At high enough temperature, the barrier can be overcome and the collective magnetic moment can flip back and forth, resulting in a vanishing time averaged magnetic moment. In superparamagnetic systems, a problem becomes immediately obvious since temperature determines both the collective magnetization as well as the switching rate. This is in the focus of the present work.

Superparamagnetic behavior of nanomagnets is usually analyzed in terms of the switching of the magnetization [1,2]. To this end, a switching frequency $f(T)$ is determined as a function of temperature T which commonly reveals an Arrhenius-like temperature dependence following

$$f(T) = f_0 e^{-\frac{KV}{k_B T}}, \tag{1}$$

with f_0 being the so-called attempt frequency and k_B the Boltzmann constant. To obtain an Arrhenius law, a temperature-independent anisotropy energy has to be assumed, such that a separation of time

* Corresponding author.
E-mail address: michael.thorwart@physik.uni-hamburg.de (M. Thorwart).

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scales is possible [5]. Thus, the switching events have to be rare compared to the frequent attempts made until switching occurs, meaning $f(T) \ll f_0$ or $KV \gg k_B T$, respectively. When the logarithm of the temperature-dependent switching frequency is plotted versus the inverse temperature, a straight line results, with the slope yielding the anisotropy energy and the intercept yielding the attempt frequency. Such an analysis is common to many experiments and is appealing due to its simplicity, albeit detailed investigations whether a clear separation of time scales is really given are not properly made and are sometimes not even possible due to experimental constraints.

Even though the magnitude and the dependence on temperature of the attempt frequency have been modeled on the basis of detailed assumptions about the reversal mechanisms [3–7], large deviations from these predictions by several orders of magnitude are reported in the literature [8–10]. Several possible physical explanations for these substantial discrepancies have been offered since then [1,11,12].

In this work, we readdress these discrepancies by analyzing results from such an Arrhenius fit. Certainly, the Arrhenius plot itself is a powerful tool and its validity is confirmed by innumerable successful applications. The mere numbers that come out of such an analysis are correct on their own, but point out that the assignment to specific material properties has to be done with care. In particular, precise knowledge about the temperature dependence of the exponent in Eq. (1) is necessary for reliable extractions of material parameters. Likewise, a straightforward extrapolation of the Arrhenius plot to zero temperature in order to derive the prefactor f_0 and interpret it as a constant "attempt frequency" has to be done with caution.

In this context, it was shown that the use of the free energy instead of the total energy entering for the energy barrier generates a temperature dependent contribution in connection with the entropy [13,14]. This additional contribution to the activation energy can reduce the prefactor [15] compared to Brown's result [3] and plays an important role for large statistical ensembles with a broad distribution of activation energies and high-dimensional energy landscapes [15].

Of more importance is the explicit dependence of the anisotropy on temperature, as addressed in the present work in greater detail. This has been investigated mostly in the blocked regime where switching is largely suppressed [16–19], but has also been mentioned to influence the superparamagnetic switching [16,20].

There are several origins of a temperature dependence of the total anisotropy. At first, the magnetic anisotropy can vary with temperature due to slight changes of the structure and stress in the material. This is well known for bulk materials like Co. Second, the effect can be due to the shape anisotropy which is often the origin of the uniaxial anisotropy of nanoparticles. As this part is determined by the saturation magnetization, it changes with varying temperature. The third influence originates from the scaling property of the magneto-crystalline anisotropy with the saturation magnetization in a power-law like characteristics [21–26]. The temperature dependence of the effective anisotropies in magnetic nanoparticles with different shapes with cubic or uniaxial bulk anisotropy and Néel surface anisotropy has been calculated by using a constrained Monte Carlo approach [27]. The impact of thermal magnon excitations on coercivity has been investigated in Ref. [28].

The effect of the temperature dependence of the shape anisotropy on the coercivity for aligned Stoner-Wohlfarth systems has been considered in Ref. [19]. The standard Néel-Brown formula for the coercive field has been extended to include the temperature dependence of the magnetization, leading to an effective temperature dependent anisoptropy barrier. The role of a temperature dependent magnetocrystalline anisotropy on the coercivity of nanostructured materials was investigated theoretically in Ref. [16]. Furthermore, it was shown in Ref. [29] that the temperature dependence of the magnetic anisotropy also needs to be carefully taken into account when the magnetic remanence is considered.

Experimental results on Co-Fe magnetic nanoparticles [30] show that the temperature dependence of the anisotropy has to be taken into account for a matching with a Néel-Brown-Arrhenius law. In this case, an empirical Brükhatov-Kirensky ansatz was used for the temperature dependence of the anisotropy. Such an approach reproduces realistic zero-temperature values $K(0)$ for the bulk anisotropies as well as realistic times for the inverse attempt frequencies.

The purpose of this work is not to address the details of the physical effects, which contribute to the temperature dependence of the anisotropy. Instead, our focus is on its general impact on the analysis of the superparamagnetic behavior via its switching characteristics. We demonstrate that the temperature interval (which is usually somewhat limited in experiments) in which a finite number of data points are fitted to an Arrhenius law has to be selected with care. It may determine very sensitively the resulting parameters of the Arrhenius plot, being the slope (anisotropy) and the intercept (attempt frequency). In general, besides the blocking temperature T_B in superparamagnetism, a second temperature scale becomes relevant in the macrospin description, i.e., the Curie temperature T_C, being decisive for the magnetic ordering. Thus, experimentally determined prefactors [8,31–33] have to be carefully interpreted, particularly, but not exclusively, when the temperature range, in which the switching measurement are performed, is comparable to the magnetic ordering temperature [16]. We illustrate quantitatively that naively assuming the applicability of the Arrhenius law may give rise to unintended misinterpretations if a temperature-independent height of the energy barrier is presumed.

2. Theoretical models for a temperature-dependent anisotropy

A generic superparamagnetic system can be described by the Heisenberg model

$$H = -J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + H_{\text{aniso}}, \tag{2}$$

where the spins $\mathbf{S}_i$ tend to align parallel due to the exchange interaction of strength J in order to form a macroscopic magnetic moment. The exchange interaction itself is rotationally invariant and thus does not favor a certain direction for the resulting magnetic moment. It basically determines the temperature dependence of the bulk magnetic system. However, superparamagnetic systems typically possess either intrinsic material symmetries or external ones such as a shape anisotropy. Then, distinct directions of the magnetic moment are preferred. To model this feature, a generic anisotropy term H_{aniso} is introduced in Eq. (2). The specific form of the anisotropy has to be defined for the particular experimental situation. It is important to realize that the anisotropy part in small systems does not only define easy or hard axes, but also influences the temperature dependence of the magnetization $M(T)$ itself. Thus, $M(T)$ in small systems can be very different from comparable bulk systems. The theoretical determination of $M(T)$ in those small samples is a non-trivial task [13] and depends on the details of the Heisenberg model. As a matter of fact, we focus on general consequences of a temperature dependence of the magnetization and we do not aim to calculate $M(T)$ from first principles. For the purpose of this work, it is sufficient to assign some generic behavior to it. First, we assume the magnetic system to be large enough to form a collective macroscopic magnetic moment but still sufficiently small to avoid a separation into multiple magnetic domains.