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Demagnetization in photomagnetic films

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ABSTRACT

We present a model for demagnetization in photomagnetic films, and investigate different regimes for the magnetizing process using finite element analysis. It is found that the demagnetizing factor may depend strongly upon the high-spin fraction of the film, and the specifics of the dependence are dictated by the microscopic morphology of the photomagnetic domains. This picture allows for facile interpretation of existing data on photomagnetic films, and can even explain an observed photoinduced decrease in low-field magnetization concurrent with increase in high-spin fraction. As a whole, these results reiterate the need to consider demagnetizing effects in photomagnetic films.

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1. Introduction

Demagnetizing effects are important to consider for magnetic materials with shape anisotropy, and are particularly relevant for films with photoinduced magnetic order. It will be important to understand how demagnetization may alter the response of photomagnetic films as scientists look forward to potential device applications. While a wide variety of photomagnetic materials have been reported, the calculations herein were motivated by long-lived photoinduced magnetism in coordination polymers [1,2]. In such materials, the application of light may give rise to large changes in the bulk magnetization, caused by transitioning ions from diamagnetic to paramagnetic states. Furthermore, technologically relevant film morphologies of photomagnetic coordination polymers have been reported in the literature. These films have shown similar changes in magnetization with light as the bulk when the film planes are oriented parallel to applied magnetic field, but a reduction in the magnetic response when the film planes are oriented perpendicular to applied magnetic fields [3–5]. Moreover, certain preparation methods and experimental conditions can show a decrease in the bulk magnetization during photoirradiation, even while the total number of magnetic electrons is increasing [3,4]. Thin film studies of magnetic anisotropy in structural analogs to photomagnets that do not have a photoeffect have shown demagnetization to play a dominant role in the orientation dependence of the magnetic susceptibility [6,7]. These studies have provided motivation to explain magnetic anisotropy in photomagnetic films with the same underlying physical phenomenon of demagnetization.

In the present work, we seek to use the transparent formalism of demagnetization along with finite element analysis to explain the reported anisotropic response in photomagnetic thin films, emphasizing the importance of film microstructure. Specifically, we show how demagnetization can actually decrease the effective magnetic susceptibility while the amount of magnetic material increases. The results are presented in a general way so as to be applicable to more than just the case studies chosen.

2. Material and methods

All finite element calculations were performed on a standard desktop computer with free, open-source software using Python 2.6.5 with NumPy 1.6.1 and SciPy 0.7.2 libraries for use with the existing finite element package FiPy 2.1.1 [8]. Three dimensional square meshes with appropriate periodic boundary conditions were chosen for their stability, and results were checked to ensure no dependence upon grid density for the reported data. The magnetic potential formalism was used for its computational simplicity [9]. Demagnetizing factors were calculated by taking the average of the normalized internal magnetic field for a given magnetization distribution.

3. Theory/calculation

The internal field of a magnet can cause a decrease in the effective measuring field, as shown by

$$H_{eff} = H_{lab} - NM_{actual}, \quad (1)$$

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where N is the geometrical demagnetizing factor, M_{actual} is the measured magnetization per unit volume, H_{lab} is the applied field in free space within the magnet coils, and H_{eff} is the effective field inside the material taking into account demagnetization. The bare magnetic susceptibility, χ_{bare}, is defined as the relationship between magnetization and effective magnetic field, namely

$$M_{actual} = \chi_{bare} H_{eff}, \tag{2}$$

while the effective magnetic susceptibility, χ_{eff}, is defined as the relationship between the magnetization and the lab magnetic field,

$$M_{actual} = \chi_{eff} H_{lab}. \tag{3}$$

Based upon these equations, a simple formula for the effective magnetic susceptibility is

$$\chi_{eff} = \frac{\chi_{bare}}{1+\chi_{bare} N}. \tag{4}$$

For the photomagnetic systems in question, the volume susceptibility is directly proportional to the amount of material in the magnetic or high-spin state, n_{HS}, such that

$$\chi_{bare} \rightarrow n_{HS}\chi_{bare} \tag{5}$$

and

$$\chi_{eff} = \frac{n_{HS}\chi_{bare}}{1+n_{HS}\chi_{bare} N}. \tag{6}$$

From Eq. (6), the photomagnetic response of a film will depend upon the demagnetizing factor, with the largest response being observed for small demagnetizing factors. By differentiating Eq. (6) with respect to n_{HS}, with only the explicit dependence upon n_{HS} and $\chi_{eff} > 0$, $\chi_{bare} > 0$, n_{HS} between 0 and 1, and N between 0 and 1, it is clear that there is no way to decrease the χ_{eff} while increasing n_{HS}. However, if the demagnetizing factor is allowed to depend upon the amount of magnetic material, the effective susceptibility will decrease if

$$\chi_{bare} \frac{\partial N}{\partial n_{HS}} {n_{HS}}^2 > 1. \tag{7}$$

Therefore, it is important to know how demagnetizing factors of a given system change as a function of the amount of magnetic material, and in the next section, model calculations of experimentally motivated morphologies are presented.

The limit of film thickness being negligible compared to breadth is assumed for all calculations, and this motivates the use of periodic boundary conditions within the plane of the film. For coherent domain growth models, the film is parameterized with CG, the ratio between the height of the film and the separation between magnetic growth nucleation sites. For random growth, the film is parameterized with RG, the ratio between the height of the film and the length of the randomly excited region. Coherent growth with dense site nucleation, $CG > 1$, is modeled using regularly growing cubes with uniform magnetization that are stacked to give integral values of CG. Coherent growth with sparse site nucleation, $CG < 1$, is similarly modeled with uniformly magnetized sheets in cells of varying size. The random growth process only allows for dense, $RG > 1$, configuration.

4. Results

The magnetic fields due to the previously mentioned photomagnetic growth models were calculated, and epitomic field

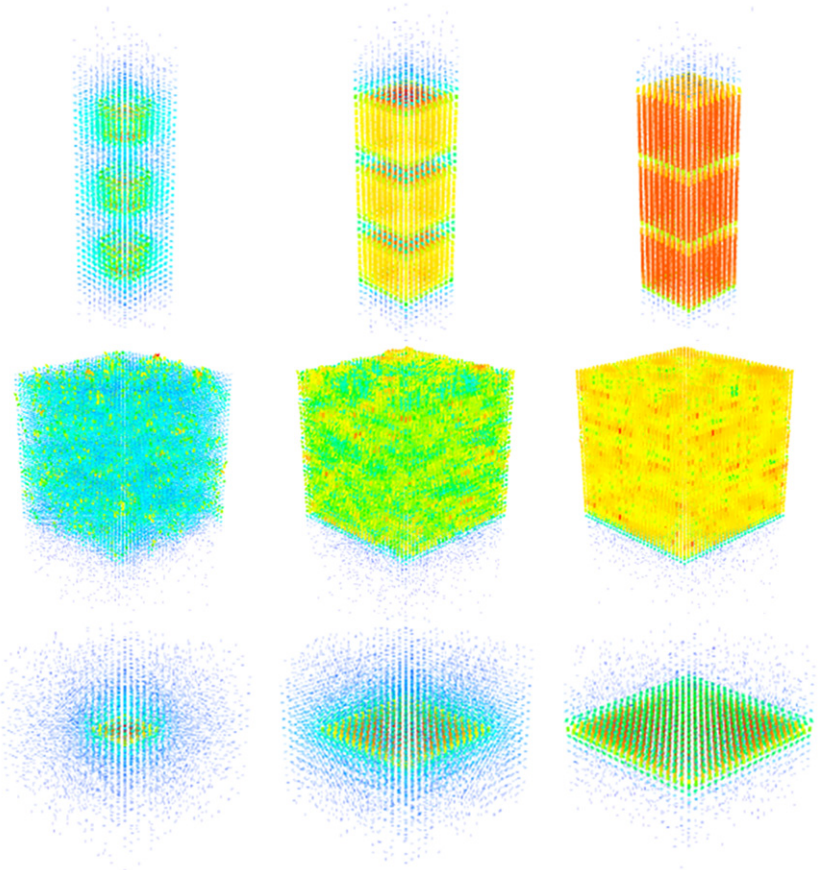

Fig. 1. Magnetic field distributions for photomagnetic growth processes. From left to right, n_{HS}=0.1, 0.5, and 0.9. (top row) Coherent growth with CG=3, (middle row) random growth with RG=50, and (bottom row) coherent growth with CG=0.05.

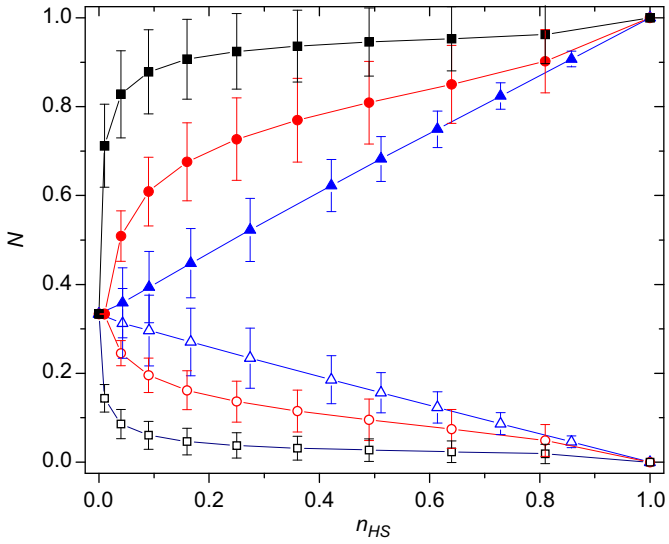

Fig. 2. Demagnetizing factors as a function of high-spin fraction. Parallel (open symbols) and perpendicular (filled symbols) orientations of the plane of the film are shown for CG=0.01 (squares), CG=0.05 (circles), and CG=2 (triangles). Other values of CG=1, 3, 4, 5, and 10 gave similar values as CG=2, but with slightly different variances. Similarly, all values of RG showed simple interpolation between spherical and planar behavior. Uncertainty bars represent one standard deviation from the mean.

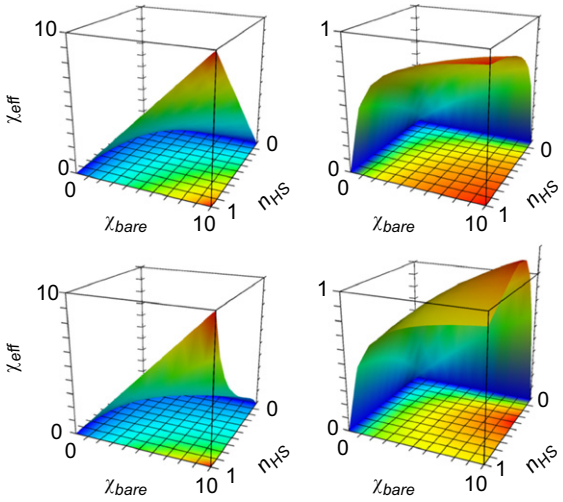

Fig. 3. Effect of demagnetization on susceptibility. (top row) The effective susceptibility, χ_{eff}, as a function of n_{HS} and χ_{bare}, for (left) parallel and (right) perpendicular orientations of the film in the limit that the demagnetizing factor does not change as the high-spin fraction changes, CG→0. (bottom row) The effective susceptibility, χ_{eff}, as a function of n_{HS} and χ_{bare}, for (left) parallel and (right) perpendicular orientations of the film using the calculated values of dependence of the demagnetizing factor upon the high-spin fraction for CG > 1.

distributions are displayed in Fig. 1. The demagnetizing factors as a function of the high-spin fraction, Fig. 2, show a crossover from isolated sphere behavior to infinite film behavior as the amount of magnetic material is increased. The details of the changes in the demagnetizing factor depend upon the way in which magnetic regions grow in the sample. For dense site nucleation and random growth, a simple monotonous interpolation between the two limited behaviors is observed. This is in contrast with sparse site nucleation, which shows a faster approach to film-like behavior, and as a result increasingly smaller changes in the demagnetizing factor for intermediate and large values of the high-spin fraction as CG becomes smaller.

Using the calculated dependence of N upon n_{HS}, the effective magnetic susceptibility can be calculated for different values of n_{HS} and χ_{bare}, and CG→0 and CG > 1 data are shown in Fig. 3. For CG > 1, a decrease in χ_{eff} can be seen with increasing n_{HS} when the film is oriented perpendicular, while CG→0 only shows monotonic increase, and in either regime parallel orientations of the films only give increases in the effective susceptibility.

5. Discussion

The simple theoretical model allied with the simple numerical model shows that the response of photomagnetic films is expected to be modulated by demagnetization, and that demagnetization may actually cause a decrease in the measured magnetization, even as the amount of magnetic material increases in a sample. All calculations showed reduced response in the perpendicular orientation compared to the parallel orientation, which becomes more pronounced with increasing susceptibility. As a photomagnetic device would seek to compare changes before and after a photoexcitation event, it may be desirable to use high measuring fields, which reduce the susceptibility due to saturation effects while the magnitude of the magnetization can be large, or in-plane measuring fields, which circumvent ambiguity in the memory state.

It is illustrative to calculate magnetization versus temperature and compare calculated behavior to experimental data, to see if the proposed models may explain the differences between bulk and photomagnetic films, and two case studies are briefly presented. Experimental probes of Prussian blue analog photomagnets suggest coherent photomagnetic domain growth process. [10,11] Sequentially adsorbed films of rubidium cobalt hexacyanoferrate on a plastic solid support show an increase of magnetization with photoexcitation in parallel orientations for all fields, and a decrease in magnetization with photoexcitation in perpendicular orientations for small fields but an increase in perpendicular orientations for sufficiently large fields. These features can be reproduced with the demagnetizing model, Fig. 4, are suggestive of coherent growth in the dense limit, and the cross-over between photoinduced decrease and increase with increasing field is understood as reduction in the magnetic susceptibility towards saturation. The Langmuir–Blodgett clay films of cobalt hexacyanoferrate show a dramatic reduction of the magnetic response in the perpendicular orientation compared to the parallel orientation, which is suggestive of coherent domain growth in the dilute limit where the demagnetizing factor stays mainly constant and ideal-film-like with changing high-spin fraction, Fig. 4.

It is worth noting that more sophisticated models may take into account magnetic domains within photomagnetic domains, as well as their interactions, in addition to surface roughness, which is expected to decrease the anisotropy of the demagnetizing factor. However, without additional experimental probes to investigate the microstructure of given samples, the specific microscopic parameters to be used are not obvious, but the lack of specificity in the present report does not detract from the overall conclusion. Interestingly, a specific calculation that studied magnetic percolation effects in a two-dimensional calculation platform also showed modification of demagnetizing factors with changing magnetic volume fraction. [12] Also, while we have focused our attention on a particular set of coordination polymers, other systems that display photocontrol of magnetic objects on the nanoscale and microscale may show a similar magnetostatic behavior when constrained to thin films. One additional class of such system is amorphous spin glasses, in which irradiation may induce the so-called bubbles of ordered moments within the spin glass continuum of the solid. [13]

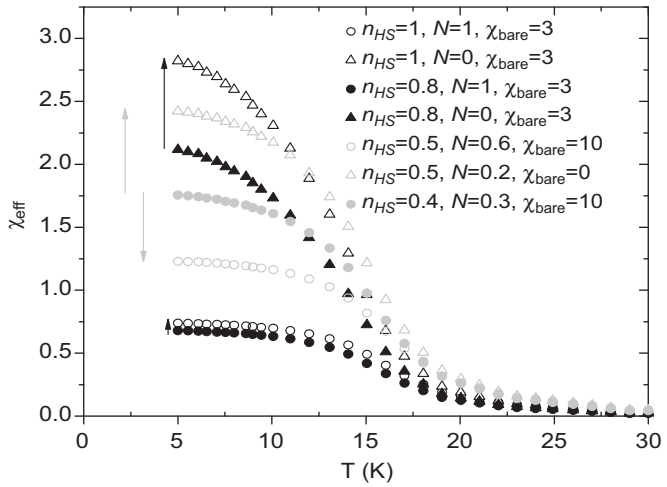

Fig. 4. Effective susceptibility versus temperature. The calculated effective susceptibility as a function of temperature can be made to mimic the results of Park et al. [3] shown as gray symbols and Yamamoto et al. [5] shown as black symbols for different dependence of the demagnetizing factor upon the high-spin fraction. While the precise volume susceptibility and domain structure in photomagnetic films has yet to be experimentally reported, many different values of the parameters may give similar qualitative behavior. Arrows at left draw attention to the expectation of photoinduced increase or decrease.

6. Conclusions

Demagnetization effects are important to consider for photomagnetic systems, especially where there is obvious shape anisotropy of the magnet such as in thin films. Simple demagnetizing models suggest a cross-over from ideal sphere demagnetization to ideal plane demagnetization as the volume fraction of the magnetic material in a thin film increases. In the extreme case, demagnetization can actually cause a decrease in the magnetic response, even while the saturation magnetization of a film increases. These effects may be avoided using films with designer morphologies that show the desired angular dependence of shape, or by only operating in regions with small magnetic susceptibility such as in high magnetic fields. Finally, existing data on photomagnetic films may be phenomenologically modeled, and these results may be further tested experimentally with photomagnetic thin films of additional materials.

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