Controlling the Influence of Elastic Eigenmodes on Nanomagnet Dynamics Through Pattern Geometry

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ABSTRACT

The effect of the nanoscale array geometry on the interaction between optically generated surface acoustic waves (SAWs) and nanomagnet dynamics is investigated using Time-Resolved Magneto-Optical Kerr Effect Microscopy (TR-MOKE). It is demonstrated that altering the nanomagnet geometry from a periodic to a randomized aperiodic pattern effectively removes the magneto-elastic effect of SAWs on the magnetization dynamics. The efficiency of this method depends on the extent of any residual spatial correlations and is quantified by spatial Fourier analysis of the two structures. Randomization allows observation and extraction of intrinsic magnetic parameters such as spin wave frequencies and damping to be resolvable using all-optical methods, enabling the conclusion that the fabrication process does not affect the damping.

I. INTRODUCTION

Understanding nanomagnet dynamics in densely packed arrays is important due to their potential applications in next generation spintronic devices. A number of experiments can be employed for this purpose including ferromagnetic resonance [1], Brillouin light scattering [2], x-ray spectroscopy [3], and the time-resolved magneto-optic Kerr effect (TR-MOKE) [4-8]. Each has their own benefits and drawbacks, and involves various techniques such as rf magnetic fields [1], spin polarized currents [9,10] and picosecond acoustic pulses [11] for instigating magnetization precession in order to gain insight into the underlying mechanisms governing the motion of the spins. In all-optical TR-MOKE the precession is initiated by optical excitation with femtosecond pump pulses that are absorbed by the magnetic material. This method provides high spatial and temporal resolution. While the intrinsic magnetic response of a nanomagnet is most naturally studied using a single element [4], measurements on arrays are valuable as they may be the only way to generate large enough magneto-optical signals to observe the dynamics of a given system and because they provide a complementary set of information that helps identify extrinsic effects due to inter-element variations and interactions. However, a side effect in regularly patterned arrays is the simultaneous generation of surface acoustic waves (SAWs) due to the thermal expansion of the elements upon irradiation with the pump pulse. These SAWs exist at specific frequencies and have been extensively investigated recently due to the emerging field of phononic bandgap materials. In these systems, the phononic band structure is a function of material properties as well as the position and size of elements in arrays which create stopbands in the spin-wave spectrum [12-14]. Time-resolved studies on periodic arrays of Al nanoelements revealed that the SAW frequencies depend inversely on the array pitch [15]. The interplay of mechanical and magnetic oscillations in arrays has been investigated as well [16].

Specifically, optically generated SAWs can drive the magnetization dynamics in nanostructured arrays via magneto-elastic coupling. This causes the spin precession resonances to be pinned at the SAW frequencies over an extended applied field range around the point where the two resonances are degenerate. While this opens up the possibility for utilizing SAWs as an extra degree of freedom for investigating nanomagnet arrays, the SAW induced magnetization dynamics complicate the extraction of the intrinsic magnetic response. Here, we demonstrate a method for suppressing the laser-induced SAW influence on magnetization dynamics. By altering the array geometry from a periodic to a randomized pattern, the effect of the SAWs on the magnetization dynamics can be mitigated, thereby restoring the intrinsic magnetic response. Using spatial Fourier analysis, we quantify the efficacy of the approach and show that it is limited by residual correlations in structures with a finite filling factor. The approach is powerful enough to permit the observation of the field-dependent Kittel mode and the extraction of the effective damping in densely packed nano-elements using all-optical pump-probe methods.

II. EXPERIMENTAL DETAILS

Fig. 1(a) shows SEM images of the two samples investigated, exhibiting a periodic (top) and an aperiodic (bottom) pattern, respectively. They were fabricated using electron beam lithography on a (100) Si substrate capped by a 110 nm thick hafnium oxide antireflection coating as previously reported [16-18]. They are both comprised of 30nm thick 156nm wide polycrystalline Ni squares. In the periodic array the elements have a pitch of 330nm corresponding to a fill factor of 0.22. This same fill factor is used in the randomized pattern so that the only difference between the two is the arrangement of the Ni squares. The elements have an easy axis in the sample plane with an H_K value of 1807 Oe.

The magnetization dynamics of these samples were studied using a two-color TR-MOKE setup. 165-fs pulses (repetition rate 76 MHz) are split into pump (400nm, FWHM 10.6 μ m) and probe (800nm, FWHM 7.1 μ m) pulses and are focused onto the sample using a 60x microscope objective with a pump fluence of 0.22 mJ/cm². The probe beam with a fluence of 0.03 mJ/cm² travels through an optical delay line to provide sub-picosecond temporal resolution. The changes in the probe beam polarization and the total reflected power were measured simultaneously using a balanced photodetector configuration, where the sum and difference signals yield the non-magnetic and magnetic responses, respectively [19]. A variable magnetic field was applied along the x-direction (Fig.1) at 30° with respect to the surface normal. At each field value the time-dependent signal from the magnetic and non-magnetic channels are converted into the frequency domain using a FFT algorithm. These frequency spectra will be referred to as the TR-MOKE Fourier spectra, in order to distinguish them from the spatial Fourier Transforms of the sample patterns.

The heating from the pump pulse has two important effects on the array. Firstly, the anisotropy is rapidly modified causing the magnetization to precess around the effective field [19]. Secondly, the heating causes a ubiquitous thermal expansion of all the elements. The subsequent mechanical oscillations of the irradiated elements all have the same eigen-frequencies determined by

$$\rho \frac{\partial^2 u_i}{\partial t^2} = \sum_{j=1}^3 \frac{\partial \sigma_{ij}}{\partial x_j} \quad (1)$$

where $i, j = 1, 2, 3$ denote the Cartesian coordinates, ρ is the density, u_i is the displacement, and σ_{ij} is the Cauchy stress tensor [16].

Each element acts as a node which launches these acoustic oscillations into the substrate which then interfere constructively to create surface acoustic waves (SAWs) at frequencies

determined by the array geometry and the substrate material [15]. These SAWs modify the elastic strain in the elements $\varepsilon_{ij}(\vec{r}, t) = (\partial u_i / \partial x_j + \partial u_j / \partial x_i) / 2$ which magneto-elastically couple to the magnetic system. This interaction manifests itself as an additional energy term in the free energy

$$U_{mel} = \frac{B_1}{M_S^2} \sum_i M_S^2 \varepsilon_{ii} + \frac{B_2}{M_S^2} \sum_i \sum_{j \neq i} M_i M_j \varepsilon_{ij} \quad (2)$$

where M_S is the saturation magnetization, $M_{i,j}$ are the magnetization components along axes i, j , and B_1, B_2 are the magneto elastic constants [16]. This additional energy term dynamically modifies the frequency and the characteristic lifetime of the magnetic relaxation.

FIG. 1. (a) SEM images of the periodic array (top) and the random aperiodic pattern (bottom). (b) 2D Spatial Fourier Transforms of the patterns. The dashed line in the Random 2D Fourier Transform corresponds to the k -vector with the largest average Fourier amplitude, and the solid lines to the limits of SAW influence as defined in the text. (c) Average radial Fourier amplitude as a function of k -vector magnitude (k_r). The periodic array displays

discrete k-vectors while the randomized pattern has a continuous spread in k-vectors. The dashed line is a Gaussian fit to this distribution and the solid line the e^{-1} cutoff. The arrow is the maximum average radial Fourier amplitude.

III. RESULTS AND DISCUSSION

In order to understand the influence of the nanomagnet array geometry on the generation of SAWs, it is instructive to analyze the array geometry in Fourier space. Fig. 1(b) shows the two-dimensional spatial Fourier transforms of the two samples under investigation. In the periodic array, the regular arrangement of elements in particular directions manifests itself as discrete points in Fourier space. The spacing of these points is a function of the array pitch. Each point corresponds to a different SAW eigenmode launched in that direction. For small mass loading, the eigen-frequencies of the SAWs can be approximated as

$$f_{ij} = \frac{v\sqrt{i^2+j^2}}{p} \quad (3)$$

[15], where v is the velocity of the SAW in the substrate, p is the pitch of the array, and (i, j) the indices of the k-vector. The observed frequencies (Fig. 2) agree well with equation (3) using $v = 2673 \text{ m/s}$ - predominantly determined by the elastic properties of the AR coating layer, where the majority of the elastic energy is confined [16]. In Fig. 1(c), the average spatial Fourier amplitude vs. k-vectors of constant magnitude i.e. $k_r = \sqrt{k_x^2 + k_y^2}$ is plotted. This is instructive since the (1,0) and (0,1) SAW modes are propagating in orthogonal directions, yet have the same magnitude in k-space for this square array, and therefore the same frequencies. This means they are both responsible for modifying the magnetization precession at that frequency. The same is true with higher order SAWs as well.

In contrast, the randomized pattern exhibits a smeared isotropic ring in k-space. The spread of the ring corresponds to a distribution of distances of neighboring elements, and the

radial symmetry emphasizes that this distribution exists in all directions. The average spatial Fourier amplitude for k -vectors of different magnitude is more continuous in nature as seen in Fig. 1(c), indicating a spread in the radial periodicity. By fitting a Gaussian distribution to this average we can estimate e^{-1} limits of the spread in radial pitches to be 196nm – 694nm, where $p = 1/k$. This distribution of pitches causes a spread in the launched SAWs. Assuming the same speed of sound in the substrate as in the periodic array ($\sim 2700\text{m/s}$), SAWs may exist across a broad band of frequencies between 3.8-13.8 GHz. These limits are displayed in Fig. 1(b) and 1(c) as solid lines. The k -vector with the maximum radial averaged Fourier amplitude corresponds to a pitch of 305nm and is displayed as a dashed line in Fig. 1(b). The SAW that corresponds to this pitch has a frequency of 8.8 GHz. From this analysis, it can be expected that the SAW influence on the magnetization dynamics is substantially reduced, while not being completely eliminated.

FIG. 2. (a) Measured TR-MOKE Fourier spectra of the periodic array. The non-magnetic channel displays well-defined field-independent SAW resonances. The magnetic response is pinned at these SAW frequencies. Dotted black line is the simulated Kittel mode. (b) In the randomized array, the non-magnetic channel shows a drastically reduced signal from the SAWs, indicating that their dominant influence has been suppressed. In the magnetic channel the intrinsic Kittel mode has been restored. There is still slight driving of the magnetization from SAWs at 9.6 GHz and 12.7 GHz. Dotted black line is the simulated Kittel mode.

Fig. 2 shows the magnetic and non-magnetic response of the two samples represented as TR-MOKE Fourier spectra maps at each external applied field value, calculated from the background-corrected time traces of the magneto-optic signal [16]. The signals were normalized to the intensity of the highest Fourier peak in each figure. The following results are all independent of laser spot location. In the periodic array, the non-magnetic channel displays a field-independent response with constant frequencies. These frequencies correspond to the discrete SAW eigenmodes given by equation (3). The resonance near 15 GHz corresponds to the surface skimming longitudinal wave which has an in-plane displacement and higher phase velocity than the SAW Rayleigh waves [16,20].

The magnetic channel shows a much more complex response. Two field-dependent center and edge Kittel modes are present at low applied fields, but as the H-field is increased, the intrinsic modes and the SAW frequencies approach degeneracy. As this occurs, the mechanical oscillations of the SAWs magneto-elastically couple to the magnetization. This drives the magnetic precession and pins it at a fixed frequency across a large field range. The TR-MOKE Fourier amplitude reaches a maximum at degeneracy [16]. The coupling of the elastic and magnetic degrees of freedom occur at all SAW eigenmodes in the array and makes observation of the intrinsic Kittel modes nearly impossible, especially at high fields—above 4kOe. Furthermore, it is impossible to initiate magnetic precession without SAWs by reducing the laser fluence. As the pump fluence is lowered SAWs continue to be excited even after the Kittel mode signal disappears.

Fig. 2(b) shows the same plots for the aperiodic random pattern. In the non-magnetic channel, the field-independent SAW resonances are no longer present. A comparison of the TR-

MOKE frequency spectra of the non-magnetic plots obtained from the two samples demonstrates how effective altering the pattern geometry is on the suppression of the SAWs. A similar effect was recently observed in CoFeB films that were vertically stacked in a periodic and aperiodic fashion [21]. In the aperiodic pattern's magnetic channel the intrinsic Kittel modes have been restored. The dotted black line is the simulated Kittel Mode for these elements. This is one of the principal results of this Letter. It shows that the intrinsic magnetic response can be measured all-optically by altering the array geometry. A slight enhancement of the TR-MOKE Fourier Amplitude through weaker residual radial SAWs is observed. This is a result of the fact that the spatial frequency content of the randomized array is not perfectly flat due to constraints from element size and fill factor as discussed above. The strongest residual pinning occurs at 9.6 GHz, close to the predicted value of 8.8 GHz. A second, weaker pinning site at 12.7 GHz also falls within the e^{-1} range specified previously. This deviation in the maximum pinning frequency can be explained by a slightly different SAW propagation speed in this array. Another source of this discrepancy could arise from a larger periodicity in a specific direction that is averaged out in the radial average. Apart from these slight enhancements, the SAW influence on the magnetization dynamics has been removed. We further corroborate this conclusion by directly comparing the Fourier spectra at selected fields as shown in Fig. 3. At low fields the Kittel Mode in the Periodic array is still visible. However, at high fields we see that the spectra in the periodic array is dominated by the SAW modes at the frequencies extracted from the non-magnetic channel (dashed lines) which do not shift with applied field. In contrast, the spectra for the randomized array show a single dominant peak at the frequency of the Kittel mode which does show the expected field dependence. The absolute value of the Fourier amplitude at the Kittel resonance (red arrows) is $\sim 2\times$ larger in the randomized array at 4.5kOe, clearly showing that the intrinsic

magnetic response has been restored. We note that the Kittel mode amplitudes cannot be directly compared at 6kOe because the SAW dominates the response at that field.

FIG. 3. Side-by-side comparison of absolute Fourier spectra at different applied fields for (a) periodic and (b) randomized array. The periodic spectra are dominated by field-independent modes at the SAW frequencies (dashed lines), while the randomized array shows a dominant, strongly field-dependent peak at the Kittel resonance (red arrows). At 4.5 kOe, the absolute Fourier amplitude of the restored Kittel mode is $\sim 2\times$ larger than the amplitude at that frequency in the periodic array.

The question arises whether this degree of randomization is sufficient to enable a meaningful analysis of the magnetic properties of the sample. In order to answer this question, we analyzed the effective damping, an important parameter in the characterization of magnetic materials. The effective damping, α_{eff} , was extracted from the TR-MOKE time traces by fitting exponentially decaying sinusoidal oscillations to all the resonances visible in the Fourier spectrum using

$$M(t) = \sum_i^n M(0) e^{-\frac{t}{\tau_i}} \sin(\omega_i t + \varphi_i) \quad (4)$$

where ω_i is the Kittel frequency and τ_i is the decay time of oscillation i , which are related to the effective damping through $\tau_i = 1/\omega_i \alpha_{eff,i}$ [18]. At each field at which the Kittel mode was resolvable $\alpha_{eff,i}$ corresponding to the Kittel mode was extracted. This was not always possible in the periodic array because the precession was pinned at the SAW frequencies, often times with multiple pinning frequencies occurring at the same field. In these cases, the dominant resonance, defined as the frequency with the largest TR-MOKE Fourier Amplitude in the vicinity of the Kittel mode, was analyzed. Fig. 4 shows the effective damping at each field value in the periodic and random arrays along with the 95% confidence intervals of the fit. In an unpatterned film, as the applied field is increased, the effective damping gradually decreases, approaching the intrinsic Gilbert damping at high fields [22,23] (dashed line). However, the effective damping in the periodic array follows this trend only at low fields at which the Kittel mode could still be observed. Once pinning to SAWs occurs, the sample displays a much lower apparent damping than the film damping because the SAWs driving the magnetization precession are less damped. There are a few intermediate points where the Kittel mode could be resolved. However, the SAW frequencies dominate the signal which leads to very large uncertainty in the Kittel mode fit. Therefore, the intrinsic damping cannot be reliably extracted from the periodic array.

FIG. 4. (a) Example of random array TR-MOKE time trace ($H = 6$ kOe) and fit to Eqn. (2) ($R^2=0.88$); (b) Effective damping of the Kittel mode or dominant mode in the periodic array along with 95% confidence intervals of the fit. (c) The effective damping in the random pattern shows a restoration of the intrinsic relaxation with high confidence especially at higher fields. In the intermediate region there is still some driving due to weaker radial SAWs.

In the random pattern, on the other hand, the intrinsic response has been restored, and the extracted effective damping matches that of the unpatterned film over the entire field range (Fig. 4c). In particular, α_{eff} approaches the same value as the film at high fields (~ 0.035) [18,21] with high confidence. This indicates that the damping is the same in both the film and the nanoelements, suggesting that the fabrication process of the nanomagnets does not have an effect on the intrinsic damping in these Ni squares.

We note that α_{eff} is slightly lower than the film values at various intermediate points due to the residual effects from SAWs in the frequency range ~ 4 -14 GHz as discussed above. Additionally, the high-field damping value is slightly higher than intrinsic values reported on Ni. This can be attributed to extrinsic damping effects such as variations in the local anisotropy field and two-magnon scattering [24].

IV. CONCLUSION

In conclusion, we have demonstrated a method for drastically reducing the influence of SAWs on the magnetization dynamics in densely packed arrays. By randomizing the array geometry, the constructive mechanical interaction between individual elements can be reduced which leads to a near-complete elimination of the magneto-elastic interactions within the elements. This allows for the determination of intrinsic material parameters such as the Kittel mode and the effective damping in densely packed arrays.

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