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A study of the non-uniform effect on the shape anisotropy in patterned NiFe films of ferromagnetic resonance

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Abstract

A systematic ferromagnetic resonance (FMR) study shows that an in-plane magnetic anisotropy in the patterned submicron rectangular permalloy elements is mainly determined by the element geometry. As the aspect ratio increases, the in-plane shape anisotropy increases, but the contribution from the non-uniform demagnetizing effect decreases. An expression is proposed to represent the in-plane shape anisotropy due to the non-uniform demagnetizing effect. When the magnetic field is applied near the film normal direction, a set of evenly spaced peaks appears on the low-field side of the main FMR peak. The multiple peaks are considered as spin-wave resonance spectra of non-uniformly magnetized elements.

Patterned small magnets and particles have recently attracted increasing interest due to their potential application in magnetic random access memory (MRAM) and ultrahigh-density data storage [1]. They show rather different behaviours from the continuous polycrystalline thin films. For the latter, the in-plane shape magnetic anisotropy is often negligible, but in patterned small elements it is significant due to the element shape and the magnetostatic interactions [2, 3]. Magnetic anisotropy is one of the most fundamental properties, for it influences other properties such as hysteresis loop, coercivity, remanence, and saturation field [4]. Ferromagnetic resonance (FMR) has proven to be a powerful tool for quantifying magnetic anisotropy of thin films due to its high sensitivity and capability of mapping out symmetry [5]. It also provides quantitative information on the magnetization, gyromagnetic ratio and exchange interaction.

In addition, it probes magnetization non-uniformity and excitations such as magnetostatic modes and spin waves in thin films [6–14]. However, FMR study of patterned magnetic structures is scarce. This paper presents our FMR study on patterned arrays of submicron rectangular permalloy elements. Specifically, we focus on the influence of the magnetization non-uniformity on the in-plane anisotropy in the patterned submicron rectangular elements.

Magnetic single-layer films of Ta(5 nm)/NiFe(20 nm)/Ta(5 nm) and Ta(5 nm)/NiFe(30 nm)/Ta(5 nm) were deposited on silicon substrates using ion-beam sputtering. Arrays of patterned elements were fabricated using electron-beam lithography and ion-milling, as reported earlier [15]. Each array consists of approximately 10^6 nominally identical evenly spaced elements, with the inter-element edge-to-edge separation ranging from 1 to 3 μm . All elements are 0.9 μm wide, but the length-to-width aspect-ratio values are 5, 2, 1.5 and 1. The FMR study was performed at room temperature using Bruker ER-200D-SRC ESR equipment with a TE102 rectangular microwave cavity at a microwave frequency of 9.78 GHz.

The resonance field (H_r) in the FMR spectra in the film plane exhibits a well-defined in-plane anisotropy that conforms to the shape of the patterned elements, such as the dots shown in figures 1(a)–(d). For the rectangular elements, the angular dependence of the in-plane resonance field has a dominant uniaxial symmetry due to the in-plane shape anisotropy. H_r reaches a maximum when the steady magnetic field is oriented along the short edge. For the square elements, the in-plane anisotropy is clearly fourfold, and H_r is higher along the diagonal directions than along the square edges. The amplitude of the resonance field oscillations, which is a measure of the anisotropy, decreases from 305 to 138 Oe when the aspect ratio decreases from 5 to 1.5 as shown in figure 1. The film thickness also affects the magnitude of the in-plane anisotropy. It changes to 472 and 245 Oe for the same aspect ratios of 5 and 1.5 respectively when the film thickness increases from 20 to 30 nm. These experimental facts indicate that the in-plane anisotropy in patterned films follows the symmetry of the element shape when their lateral dimensions are reduced to the submicron scale, and the magnitude of the anisotropy depends on both the lateral dimensions and film thickness.

Now let us make a theoretical analysis of the anisotropy and FMR of the patterned thin films. Since the spacing between the patterned elements is larger than 1 μm , we can neglect the magnetostatic interaction between the elements [2, 3]. Using an approximation of a homogeneously magnetized ellipsoid, for a rectangular element the total free energy density F can be written as

$$F = -MH[\cos\theta \cos\theta_H + \sin\theta \sin\theta_H \cos(\phi - \phi_H)] + K_u^\perp \sin^2\theta + \frac{1}{2}N_z M^2 \cos^2\theta + \frac{1}{2}N_x M^2 \sin^2\theta \cos^2\phi + \frac{1}{2}N_y M^2 \sin^2\theta \sin^2\phi. \quad (1)$$

Here (θ, ϕ) , and (θ_H, ϕ_H) are the angles for the magnetization and applied field vectors respectively in spherical coordinates. The various terms are respectively the Zeeman energy, the perpendicular anisotropy energy, the demagnetizing energy normal to the film and the in-plane demagnetizing energy. N_x , N_y and N_z are the demagnetizing factors in three orthogonal directions. The expression (1) can be, up to a constant, written as

$$F = -MH[\cos\theta \cos\theta_H + \sin\theta \sin\theta_H \cos(\phi - \phi_H)] + K_u^\perp \sin^2\theta + \frac{1}{2}(N_z - N_x)M^2 \cos^2\theta + \frac{1}{2}(N_y - N_x)M^2 \sin^2\theta \sin^2\phi. \quad (2)$$

Using the above expression the results of the theoretical fitting shown by dashed curves in figures 1(a)–(d) were not very consist with the data points. In figure 1 we see that the difference between the theoretical curve and experimental data increases with decreasing aspect ratio. For square elements in particular, equation (2) fails to describe the fourfold symmetry. This is because the submicron rectangular thin-film elements are non-ellipsoidal; therefore the demagnetizing field is non-uniform within the elements, which leads to non-uniform

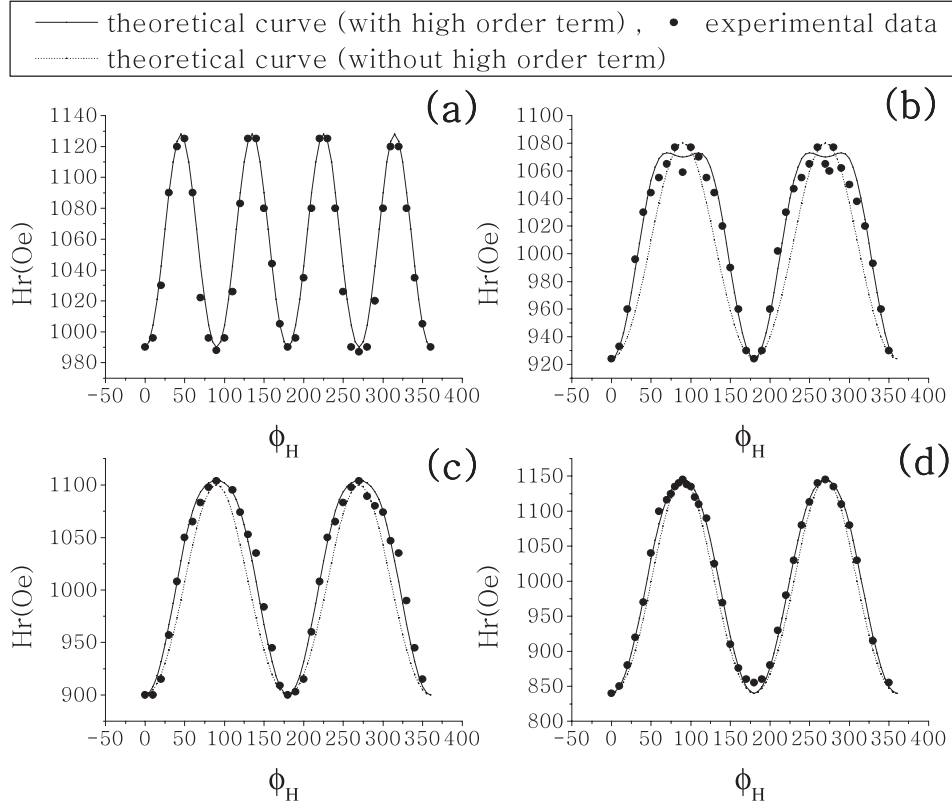


Figure 1. The experimental data points and the theoretical curves representing the evolution of the FMR field as a function of field orientation ϕ_H in the film plane of the patterned rectangular elements with the aspect ratios of 1 (a), 1.5 (b), 2 (c) and 5 (d) and thickness of 20 nm. $\phi_H = 0$ refers to field along the long edge. For (b)–(d), the dashed curves are the fitting with only the second-order terms and the continuous curves agree better with the experimental data on adding the higher-order term.

magnetization. A numerical calculation of the non-uniform magnetization and demagnetizing field using micromagnetics shows that the non-uniformity increases with the decreasing aspect ratio, as will be reported elsewhere [17]. So the in-plane shape anisotropy of small elements cannot be described in the same way as that of a homogeneously magnetized ellipsoid by using the constant demagnetizing factors shown in equations (1) and (2). Following [2], we proposed the following expression representing the energy of non-uniform demagnetizing field and found that it fitted the experimental data rather well. For a rectangular element the total free energy density F is written as

$$F = -MH[\cos\theta\cos\theta_H + \sin\theta\sin\theta_H\cos(\phi - \phi_H)] + K_u^\perp\sin^2\theta + \frac{1}{2}(N_z - N_x)M^2\cos^2\theta + \frac{1}{2}A_2M^2\sin^2\theta\sin^2\phi + \frac{1}{4}cA_2M^2\sin^4\theta\sin^4\phi \quad (3)$$

where the last two terms are respectively the in-plane demagnetizing energy including a second- and a fourth-order term. The second-order term approximates the energy of a quasi-uniform state, which would be the exact demagnetizing energy for ellipsoidal elements. The fourth-order term refers to the shape anisotropy energy due to the non-uniform demagnetizing effect, and the constant c describes the relative importance of the non-uniform demagnetizing effect.

Table 1. The anisotropy fields of patterned films with different thicknesses and different aspect ratios determined by fitting of FMR data.

Thickness of samples d (nm)	Aspect ratio of the element	A_2M (Oe)	H_K due to higher-order effect (Oe)	c
20	5	178	36	-0.2
	2	127	51	-0.4
	1.5	103	52	-0.5
30	5	267	53	-0.2
	2	200	80	-0.4
	1.5	153	77	-0.5

For square elements the second and fourth terms are combined to give rise to a term of dominant fourfold symmetry, $A_s M^2 \sin^2 \theta \sin^2 2\varphi$.

The following general expression for FMR resonance frequency derived from the Landau-Lifshitz equation and the total free energy density minimization with respect to the magnetization orientation (θ, ϕ) [16] was used:

$$\left(\frac{\omega}{\gamma}\right)^2 = \frac{1}{(M \sin \theta)^2} \{F_{\theta\theta} F_{\phi\phi} - F_{\theta\phi}^2\} \quad (4)$$

where F_{ij} are the second-order derivatives of the free energy of equations (2) or (3) with respect to θ and ϕ . Substituting equations (2) or (3) into (4), we can obtain the theoretical FMR frequency or the resonance field as a function of field orientation based on different expressions for free energy. The solid curves in figures 1(a)–(d) are the theoretically fitted curves based on equation (3), which agree very well with the experimental data points. Clearly, they agree with the experimental data much better than the dashed curves.

From the fitting, the anisotropy fields are determined for the patterned films with different thicknesses and different aspect ratios and are shown in table 1.

From the table, the equivalent quasi-uniform anisotropy field, A_2M , decreases with decrease of the aspect ratio. On the other hand, the anisotropy field due to the higher-order effect increases with decrease of the aspect ratio. The absolute value of the non-uniformity parameter c or the ratio of the fourth- to the second-order anisotropy fields is found to decrease as the aspect ratio increases, which implies that the rectangular elements with a higher aspect ratio are magnetized more uniformly. The anisotropy fields of the square elements (A_sM) are 19 and 33 Oe respectively for sample thicknesses of 20 and 30 nm, which are much smaller than those of the rectangular elements. Both the second- and fourth-order terms contribute to the amplitude of the resonance field. The theoretical value of the amplitude of the resonance field is approximately equal to $(2 + c)A_2M$. They are respectively 320, 203 and 154 Oe for the 20 nm thickness and 481, 320 and 229 Oe for the 30 nm thickness for aspect ratios of 5, 2 and 2.5. They do not differ much from the experimental values mentioned previously.

It is interesting to note that for the rectangular elements with an aspect ratio of 1.5 in figure 1(b), a shallow dip appears at 90° and at 270° , showing a non-vanishing fourfold contribution to the anisotropy energy. Only the theoretical curve considering the non-uniform effect reproduces it.

In the in-plane FMR spectra of all samples, we observed the maximum intensity in the resonance peak when the field was directed along the easy axes. It decreased when the field was rotated away from the easy axes, while the linewidth became broadened. The main resonance peak almost disappeared when the field was close to the hard (diagonal) direction in the case of square elements. For the rectangular elements, the main resonance peak of the uniform

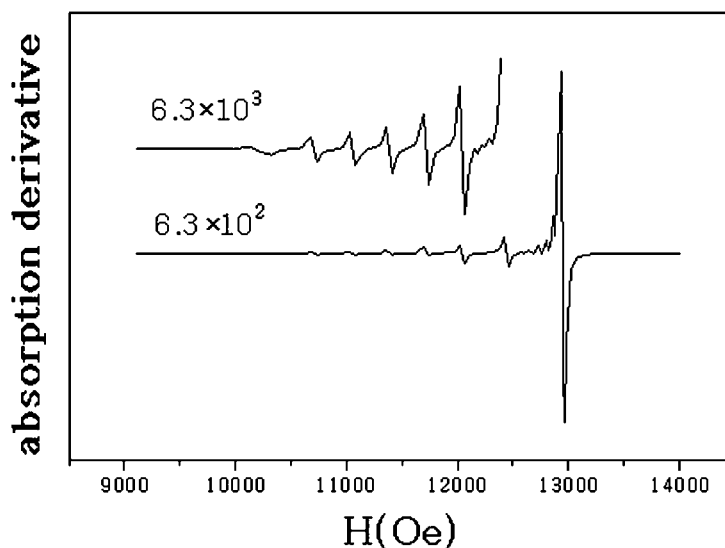


Figure 2. The FMR spectrum of the sample with an aspect ratio of 5 and thickness of 20 nm when the steady magnetic field is perpendicular to the normal of the film plane. A series of peaks (up to eight) appeared on the low-field side of the main peak.

precession mode disappeared even when the field was about 15° – 20° from the hard (short edge) direction. At the hard axis, several small peaks emerged and replaced the main peak. The separation between the adjacent small peaks ranges from 50 to 90 Oe.

When the steady magnetic field was applied near the film normal, a series of peaks (up to eight) appeared on the low-field side of the main peak for all samples as shown in figure 2. The resonance field of the side peaks decreased linearly with the peak number.

The occurrence of the multiple side peaks in FMR is the first report of this for patterned small elements. Quantitative theoretical descriptions are needed to fully account for the effect. However, on the basis of the existing experimental facts we can make the following analyses. The decrease of the main resonance peak intensity and the broadening of the linewidth with the change of the in-plane field direction are indicative of a higher degree of non-uniformity in the internal field and magnetization in those directions. Hence, non-uniform magnetization excitations may be the origin of the multiple small peaks. Nevertheless, it is not clear whether the non-uniform magnetization excitations are magnetostatic modes of some kind [6] or standing spin waves. The multiple peaks in figure 2 resemble the spectra of the spin-wave resonance in thin films studied early in the 1950s by means of FMR [7] and, later, Brillouin light scattering [8, 9]. Spin-wave spectra were observed for continuous NiFe films only when the thickness was much greater than ours, about 100–300 nm, and according to the early theory the separation of peaks is inversely proportional to the square of the film thickness and the wavelength number, which does not fit with our results quantitatively. Our data are similar to the characteristics of the recent spin-wave excitation by Brillouin light scattering [10, 11], in which, however, the static field was in the film plane, while in this paper the field was along the film normal. In addition, the multiple peaks observed in our patterned rectangular elements when the steady field is along the film normal show the characteristic linear relation of the resonance field with the mode number, including the excitations of both odd and even modes. Similar characteristics were found by Portis, Puzskarski and Qian [12–14] for unpatterned films. They

attributed the linear resonance field relation and the excitations of odd and even modes to non-uniform magnetization and asymmetrical boundary pinning. We speculate that our spin-wave-like spectra are related to the non-uniform demagnetizing effect and magnetization in the small rectangular patterned element. In the patterned elements, non-uniform demagnetizing fields and magnetization do exist, which may also cause a pinning effect. Further study on the mechanism of our multiple-peak spectra and its relation with the non-uniform magnetization is being undertaken.

In conclusion, our FMR study has demonstrated a unique capability for pinpointing in-plane magnetic anisotropy of patterned submicron elements. In general, the anisotropy can be approximately separated into quasi-uniform and non-uniform contributions. The relative importance of the non-uniform demagnetizing field and magnetization is a function of the patterned element geometry. The novel excitations of magnetostatic modes and/or the spin-wave spectra could reveal important information about non-uniform magnetization near saturation and the effect of element boundaries. Finally, we should mention that FMR results measure the average effect of the patterned elements in a sample of 2 mm^2 size rather than that of an individual element. They are not sensitive to any difference between the individual elements in an array.

Acknowledgments

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