Enhanced In-plane Anisotropy and Ferromagnetic Resonance Frequency in Permalloy Films with Nitrogen-Doped Tantalum

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Abstract— Soft magnetic materials are used in integrated power magnetic devices (such as micro-inductors and micro-transformers) to achieve reasonable inductances at frequencies over a few megahertz. However, the out-of-plane (perpendicular) anisotropy, observed in thick cores produced by vapour deposition, worsens the soft magnetic properties. This paper reports on the nano-lamination of permalloy ($Ni_{81}Fe_{19}$) thin films with the introduction of nitrogen-doped tantalum in between to eliminate the out-of-plane anisotropy and improve the in-plane anisotropy. This significantly improves the in-plane soft magnetic properties, reducing the coercivity from 1352 A/m to 25.5 A/m and increasing the anisotropy field from 180 A/m to 660 A/m. The high-frequency permeability was uniform up to 500 MHz, and the ferromagnetic resonance (FMR) frequency was increased to 1 GHz. These properties are ideal for high efficiency magnetic applications at high frequencies and an extended FMR is imperative for gigahertz-range devices.

Index Terms— Anisotropy, Ferromagnetic resonance frequency, Magnetic laminations, soft magnetic materials.

1. Introduction

Permalloy (Py) is the most widely used soft magnetic material in energy conversion [Xu 1998, Dastagir 2010], magnetic recording heads [Yu 2011] and magnetic sensor applications [Kwiatkowski 1984]. Soft magnetic properties, such as low coercivity (H_c < 40 A/m), high saturation flux density (~ 1 T), relative permeability (> 2000), deposition flexibility (electrochemical or vapour based) and patterning processes [Anthony 2015, Seet 2005], establish it as an ideal material for power magnetic components. However, its low resistivity (~ 20 μΩ-cm) results in significant eddy current losses and, as a consequence, the effective core thickness is often restricted to the skin depth. A thin core in micro-inductors reduces the inductance density (inductance per unit footprint area of the device) and makes the core prone to saturation. Furthermore, additional conductor turns are needed to achieve the specified inductance [Meere 2009, Park 2002]. Additional turns increase the footprint of the device and the winding losses. Over the years, significant research has been undertaken to overcome eddy current losses by introducing dielectric or high resistivity material between thin magnetic layers to form a "laminated core". This includes introducing thin sputter-deposited dielectric layers [Ochiai 1987, Greve 2006, Yao 2009] or electrochemically laminating magnetic cores with electrophoretic photoresist [Brunet 2006]. The authors have previously reported an electrochemical process to study core/resist lamination technology by depositing Py on an SU-8 photoresist acting as a dielectric layer with a self-assembled monolayer and an electroless deposition method [Anthony 2015, Anthony 2016]. However to achieve multiple laminations, successive photolithography or surface activation steps are needed which is time consuming and adds to the expense of the process. Sputter deposition, on the other hand, has distinct advantages such as batch deposition of alternate magnetic and dielectric films. Moreover, sputtering systems are also compatible with *in-vacuo* surface treatment and *in-situ* analysis.

Sputtered Py films tend to show spin reorientation from in-plane to the perpendicular out-of-plane direction as film thickness increases. The columnar Py film growth (due to self-shadowing effects) produces a characteristic out-of-plane anisotropy property where grains are magnetically decoupled [Pires 2014], resulting in poor in-plane magneto-crystalline anisotropy. Moreover, along with film thickness, growth conditions such as the sputtering pressure and the substrate temperature can also lead to out-of-plane anisotropy. The influence of Ar pressure on anisotropy has been extensively reported previously [Svalov 2010]. As a consequence, poor in-plane soft magnetic properties are observed in thick sputter-deposited magnetic films (> 180 nm [Romera 2011]). In our experiments, we observed out-of-plane anisotropy in Py films that were greater than 500 nm in thickness.

At lower Py ($Ni_{81}Fe_{19}$) thickness values (40 nm – 100 nm) there is a negligible contribution from the magneto-elastic anisotropy (K_{me}) [Ounadjela 1988]. Competition between the magneto-crystalline anisotropy (K_{mc}) and shape anisotropy ($-\frac{\mu_O * M_S^2}{2}$) determines the direction of magnetization since there is no significant contribution from K_{me}. The shape demagnetizing factor favours in-plane anisotropy, while K_{mc} promotes the out-of-plane anisotropy. Apart from film thickness and deposition rate, the stress in the film is largely dependent on the deposition process, substrate type and its temperature during deposition [Weiss 1962]. In the case of tantalum nitride Ta(N)/Py/Ta(N) multilayers, where Ta(N) denotes Ta doped with N, the individual Py thickness is kept below the critical thickness. The planar compressive stress, which could be attributed to lattice mismatch (dependent on deposition conditions) between the films, influences the magneto-elastic anisotropy ($\frac{3}{2}\lambda_s\sigma$) , where $\sigma > 0$ is the compressive stress and $\lambda_s < 0$ [Jungblut 1994, Sommer 1999, Cheng 2004]. Keeping individual Py layer thickness < $t_{critical}$, K_{Vol} is given by $K_{me} + K_{mc} - \frac{\mu_O * M_S^2}{2}$. The reduction in the volume anisotropy with the decrease in K_{me} promotes an in-plane anisotropy.

2. Experimental details

In this work, we report Py-Ta(N) multilayers with 2, 3 and 5 layers of Py separated with ultra-thin Ta(N) spacers. The deposition parameters (power and pressure) for individual Py films were first optimized to achieve soft in-plane magnetic properties (H_c < 40 A/m) and the thickness of each Ta(N) spacer layer is maintained at ~20 nm to ensure pin-hole free deposition. Excellent soft magnetic properties (H_c ~25.5 A/m), and a high ferromagnetic resonance (FMR) frequency are desirable outcomes for high-frequency power magnetics applications.

Sputter deposited films are 2D in nature (monolayers) which transforms into island-like 3D growth at the surface (*Stranski-Krastanov growth*) beyond $t_{critical}$, resulting in poor soft magnetic properties. It is hence important to optimize the Py growth conditions to ascertain $t_{critical}$ before introducing Ta(N) spacers. Figures 1 (a) and (b) show the measured in-plane hysteresis loop for out-of-plane anisotropic and in-plane anisotropic thick Py films, respectively. This two-stepped constricted *hysteresis* loop (transcritical condition) suggests stripe domains in the film and a poor in-plane H_c of ~1350 A/m (Fig. 1 (a)). A Nordiko DC magnetron sputtering system was used to deposit the films onto 1 µm thick thermally oxidized p-type doped (100) silicon substrates of 100 mm diameter: a 20 nm Ti adhesion layer was followed by the 117 nm Py and 20 nm Ta(N) layers; the sputtering conditions were 100 W and 1 mTorr argon for the Py and 0.15 kW and 5.3 mTorr Ar for the Ta(N) at a substrate temperature of about 466 K. There was no externally applied magnetic field to ensure that the anisotropy was not externally induced, except for the magnet assembly below the target in the sputter system which could contribute to the in-plane anisotropy. The final shape has a significant role in anisotropy of the films due to the demagnetization effect [Jamieson 2010, El-Ghazaly 2015]. Hence, all the samples were prepared at wafer scale (100 mm diameter) such that the large size maximizes the magnetostatic demagnetizing forces that promote in-plane magnetization. The optimized soft magnetic H_c measured was ~ 32 A/m (Fig. 1 (b)). Three different samples were prepared with 1, 2 and 4 layers of 20 nm thick Ta(N) sandwiched between 2, 3 and 5 Py layers of 110 nm thickness each (with net thickness of 240 nm, 370 nm and 630 nm, respectively) without breaking the chamber vacuum. The Py films were deposited in optimized sputter conditions described above. A similar approach has been reported with various spacer layers such as Gd sandwiched between Py [Svalov 2012] which showed improved soft magnetic and magneto-impedance responses. Although the coercivity has been significantly reduced by the laminations, the electronic conductivity is too high to isolate the Py layers which is necessary for high frequency applications. Ta(N) films have desirable properties, such as being non-magnetic, electrically resistive with good adhesion, thermally stabile and chemically inert, which are needed to act as a spacer layer. Moreover, the Ta(N) resistivity can be increased in an N_2 ambient to more than 1000 Ω/m^2 during deposition by altering the N_2/Ar partial pressure to achieve stable phases (such as orthorhombic Ta_3N_5, tetragonal Ta_4N_6 or hexagonal Ta_5N_6) [Kim 2005, Kang 2008]. The non-ferromagnetic nature of the spacer layer maintains uniform H_c of the multilayer structures, while the highly resistive nature keeps the Py films electrically isolated. The films were examined using scanning electron microscopy (SEM, FEI Quanta FEG 650). The static magnetic properties of the nanostructures where analysed in-plane at 10 Hz with a BH loop tracer (MESA 200 HF, SHB Instruments, USA) and high frequency permeability response were acquired by wide-band (1 MHz to 9 GHz) permeameter (9MM 9G; Ryowa Electronics, Japan) measurement.

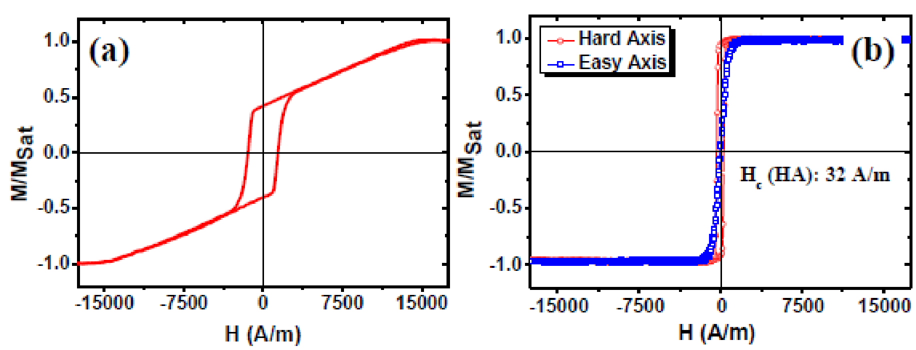

Figure 1. Measured in-plane hysteresis loop of (a) 550 nm thick and (b) optimized 117 nm Py films.

3. Results and Discussion

Figure 2(a) depicts a schematic of the desirable nano-laminates and fig. 2 (b) shows the SEM cross-section of the nano-laminated Py separated by ultra-thin (~ 20 nm) Ta(N) films, respectively. The Py films thickness were uniformly deposited (115 nm - 110 nm) using the optimized sputter conditions described above. The four point probe electrical resistivity measurement suggested a Py resistivity of about 25 μΩ-cm. The electron dispersive spectroscopy (EDS) measurements indicated a Ni: 82% and Fe: 18% Py composition.

The measured Ta(N) resistivity of about 3200 μΩ-cm depends on the Ta(N) composition and the deposition conditions. Measurement of a single laminated design (2 Py layers separated by a single Ta(N) spacer) (figure 3 (a)) had a coercivity of 29.45 A/m (as shown in the inset). However, a substantial increase in anisotropy field (H_k) was observed with additional multilayers. This increase in H_k can be attributed to the magneto-elastic anisotropy with each Ta(N) inter-layer influencing the two adjacent Py layers [Sommer 1999]. The reduction in H_c values diminishes with additional multilayers (Figs. 3(a), 3(b) and 3(c)) while an increase in the H_k (from 180 A/m to 660 A/m was observed).

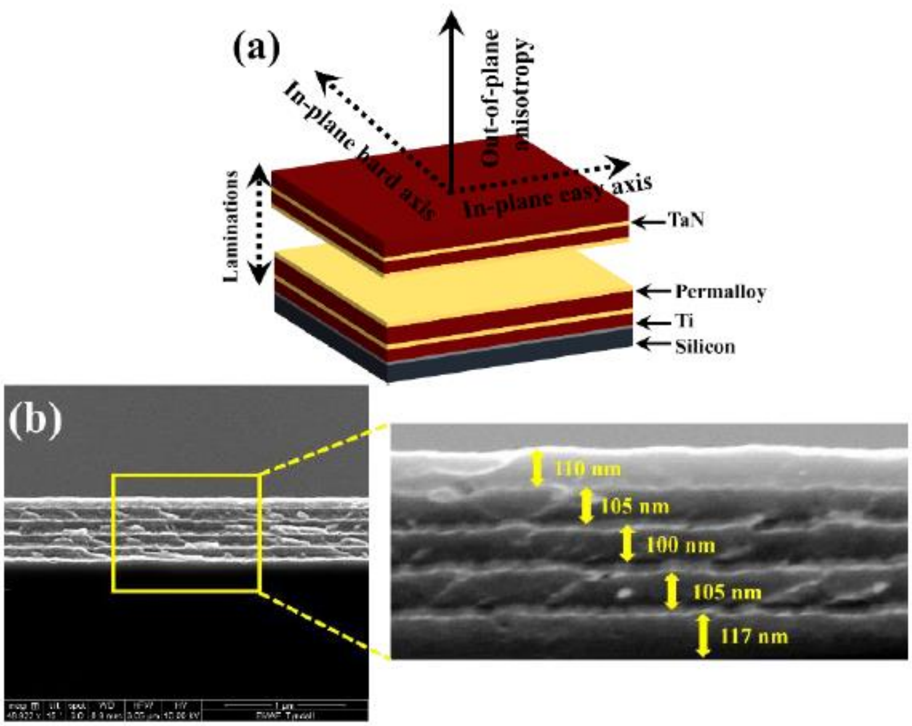

Figure 2. (a) Schematic of TaN/Py multilayers and (b) Scanning electron microscopic images of 5 Py layers separated by thin Ta(N) spacers.

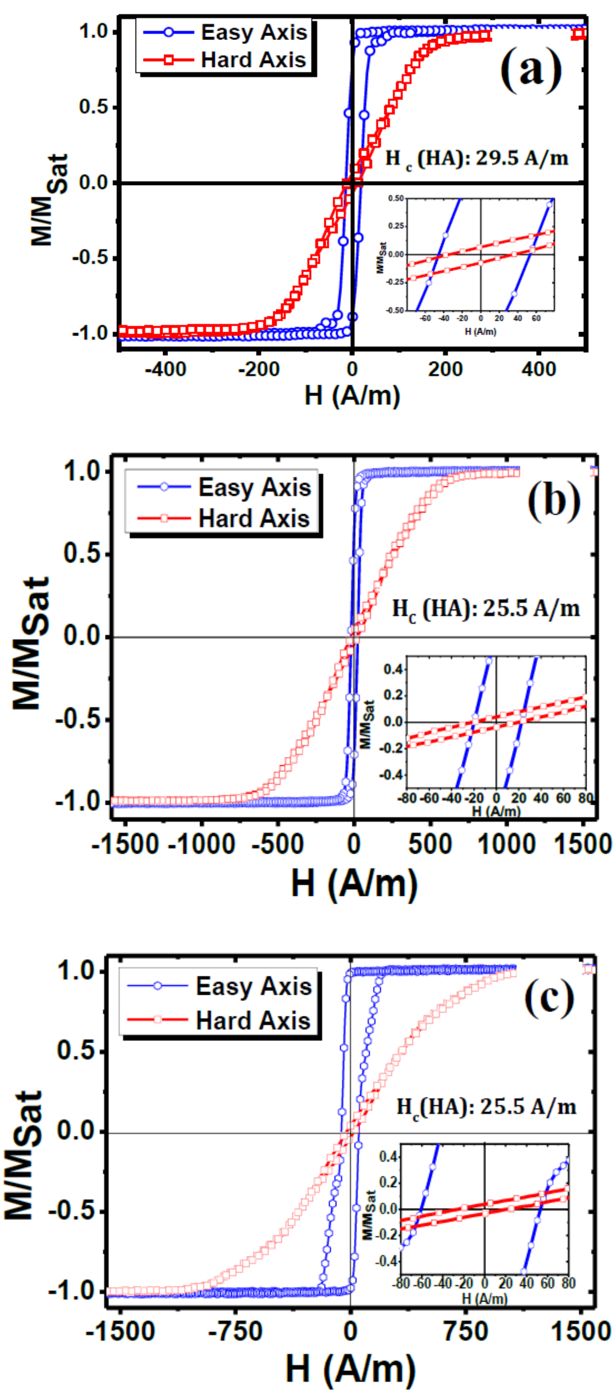

Figure 3. Hysteresis loops of (a) 2 Py layers; (b) 3 Py layers and (c) 5 Py layers separated by Ta(N) films.

The saturation fields and coercivities of the laminated multilayers were measured at different in-plane angles. Figures 4(a) and 4(b) show the saturation field and H_c change with the direction of applied field respectively. It is interesting to note that even though H_k along the HA in the 5 Py layer laminate shows significantly higher H_k (~ 660 A/m) compared to 3 and 2 Py layers, the H_c is almost the same for 3 and 2 Py layers. The 2 Py layer films show that there is significant in-plane anisotropy and HA coercivity. This was also observed with 3 and 5 Py layers, where the inclusion of successive Ta(N) layers increases in-plane anisotropy (fig. 4(a)). Although anisotropy increases with the number of Py films, the hard axis coercivity of all the films shows a negligible reduction (fig. 4(b)).

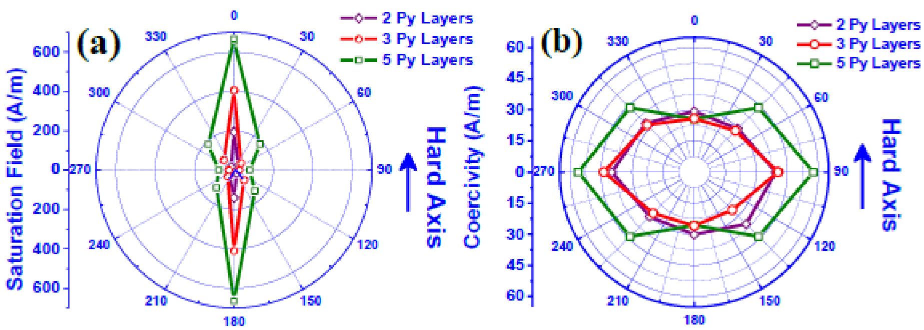

Figure 4. Directional dependence of (a) Saturation field and (b) Coercivity (H_c) measured in-plane for nano-laminated films.

It is known that the permeability response of non-laminated Py films are non-uniform beyond 100 MHz. The imaginary part of the permeability increases due to eddy current losses [Ó'Donnell 2010]. This restricts the use of Py as the magnetic core to applications below 100 MHz. Moreover, the low film resistivity (~ 20 μΩ-cm) limits the FMR frequency to well below 1 GHz as predicted by the Landau-Lifschitz-Gilbert model. The cut-off frequency (f_c) in terms of film permeability is expressed as $\frac{\rho}{t^2 \mu_o \mu_r \pi}$. Where, the film thickness, bulk resistivity and relative permeability are represented as t, ρ and μ_r, respectively [Ó'Donnell 2010].

The inclusion of non-magnetic and highly resistive spacers (such as TaN), increases the bulk resistivity of the multilayer (from 20 μΩ-cm to 105 μΩ-cm). For a constant magnetic film thickness (t) the f_c increases with number of multilayers. Figure 5 shows the relative permeability response of Py thin film to frequency at zero bias. Figures 5 (a) and 5 (b) show the real and imaginary parts of the permeability of the laminated Py films versus frequency.. It is clear that the permeability response is flat to beyond 500 MHz. The laminated films reduce the eddy current losses and increase the FMR frequency to approximately 1 GHz. This can be attributed to the increase in the bulk resistivity with the number of laminations (fig. 5 (c)) which increases the cut-off frequency. However, the capacitive coupling between the Py films in the multilayers influences the eddy current losses, thereby reducing the cut-off frequency above the 'transition frequency' as expected from the cut-off frequency equation [Feng 1977, Yao 2009].. The subsequent increase in the anisotropy with the number of laminations reduces the coercivity and the increase in bulk resistivity also results in higher f_c. As the application of magnetic devices approaches the gigahertz regime, the shape effects in the laminated films should be taken into account to achieve FMR in the gigahertz regime.

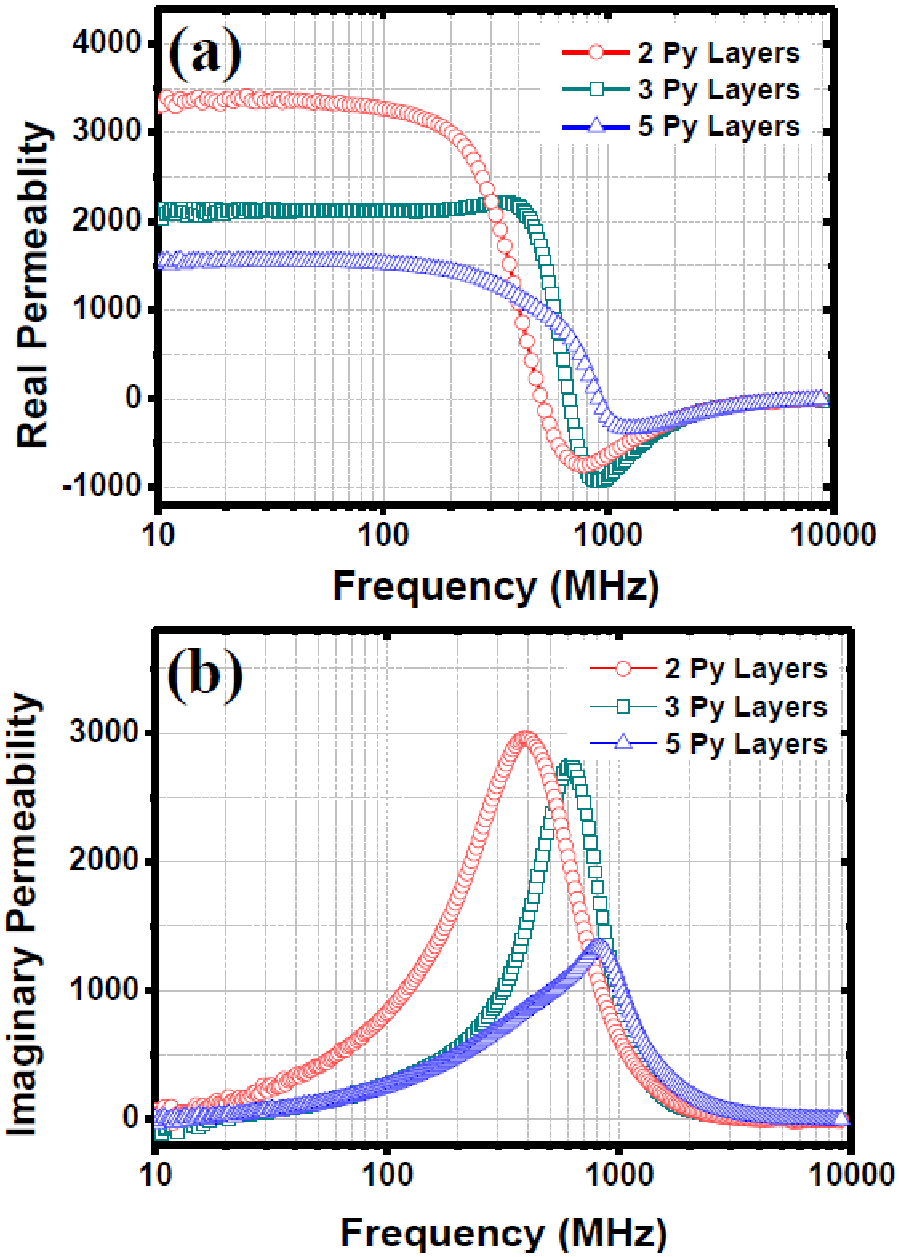

Figure 5. Relative permeability: (a) Real and (b) Imaginary spectra of TaN/Py/Ta(N) films measured at zero bias. (c) Increase in resistivity and cut-off frequency (in-set) with number of laminations.

4. CONCLUSION

In this work we have developed processes for and characterized laminated thin films for high frequency integrated magnetic applications. We reduced the out-of-plane anisotropy component observed in single Py films above the $t_{critical}$ region by introducing thin Ta(N) layers. We found that inclusion of successive Ta(N) spacers between Py layers eliminates the perpendicular (out-of-plane) anisotropy component. Moreover, the H_c could be reduced more than 50-fold by comparison with non-laminated Py films of similar thickness (from 1350 A/m to 25.5 A/m). Unlike single Py films, high frequency magnetic measurements of the laminated materials show uniform permeability over 500 MHz, pushing the FMR frequency above 1 GHz. The inclusion of pin-hole free deposited Ta(N) films (~ 3200 µOhm-cm) separates the Py films and increases the bulk resistivity of the film. Thus the multilayers acting as multiple thin sheets of magnetic core material, can increase the cut-off frequency and the anisotropy without compromising on the hysteresis loss. The sputtered nano-laminations are suitable for use in high frequency integrated magnetics applications.

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