

# Ultrafast non-reciprocal spin resonances in frustrated plasmonic metasurfaces

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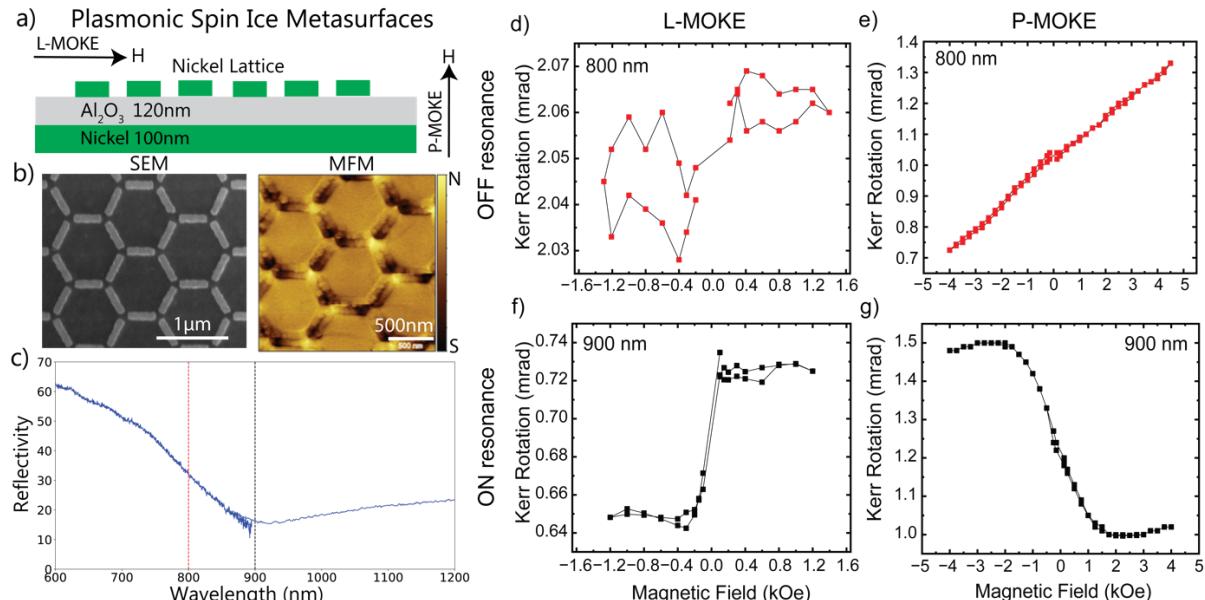
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**Abstract:** We demonstrate non-reciprocal spin resonance evolution on picosecond time scales via the time resolved magneto-optic Kerr effect at the plasmonic (metal-insulator-metal) resonant wavelength of a frustrated Kagome-type Nickel spin-ice metasurface. © 2024 The Author(s)

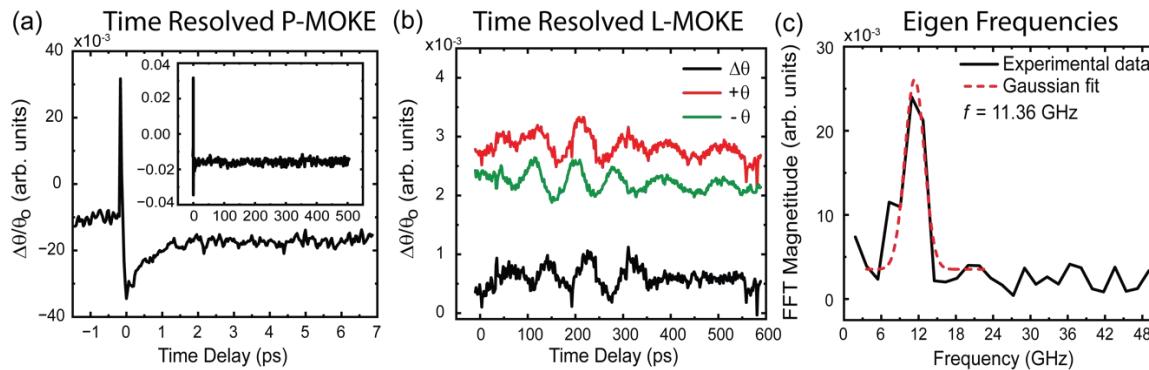
Artificial spin-ice (ASI) systems based on nanomagnet arrays have emerged as a novel platform to realize emergent many-body and condensed matter phenomena [1] with compelling applications in microelectronics and artificial intelligence. Indeed, the intrinsic reconfigurability of these systems has recently been leveraged to perform stochastic computing and learning [1]. Nevertheless, at present, conventional routes for ASI control result in a limited number of input/output channels and operational frequencies that are constrained to the radio-frequency regime. Here, we demonstrate the potential of a plasmonically resonant frustrated spin-ice metasurface, made up of a sub-wavelength array of magnetic Ni meta-atoms in a Kagome-type lattice, to overcome these limitations via the all-optical spatiotemporal control of spin states. Here, plasmonic resonances are realized using a [Ni-ASI]-Al<sub>2</sub>O<sub>3</sub>-[Ni-thin-film] architecture (Fig. 1a), which enhance the electric field intensity in the near-field of the nano-magnets [2] and subsequently increasing their responses in magneto-optical measurements. This, in turn, enables us to demonstrate the time-evolution of a non-reciprocal spin resonance at room temperature.



**Figure 1.** (a) Schematic of the plasmonic spin ice metasurface (MS) made as a metal (Ni 100nm)-insulator (Al<sub>2</sub>O<sub>3</sub>-120nm)-metal (Ni lattice 80nm) which forms the artificial spin-ice Kagome-type lattice on an insulating silicon substrate. We note the orientation of the applied magnetic field (H) for longitudinal (L-) and polar (P-) magneto-optic Kerr effect (MOKE) as horizontal and vertical arrows on the schematic. (b) Scanning electron microscopy (SEM) and magnetic force microscopy (MFM) of the top-down view of the metasurface. The bright regions on the MFM images represent the north (N) polarity and dark regions represent the (S) polarity on the color bar. (c) Reflection spectra of the plasmonic metasurface showing a resonant dip at 900nm. The vertical dashed lines (red at 800nm & the black at 900nm) represents the wavelengths at which MOKE measurements were similar performed on and off the resonance wavelength.

Plasmonic ASI metasurfaces resonant at 900 nm were designed using finite difference time domain (FDTD) simulations and nano-fabricated using electron-beam lithography followed by a lift-off process to realize the sub-wavelength Kagome lattice of Ni nanomagnets (Fig. 1b). The Ni nanomagnet array was deposited on a thin film stack of Al<sub>2</sub>O<sub>3</sub> (120 nm thick) and Ni (100 nm thick) on an insulating silicon substrate. Magnetic force microscopy

measurements were done at room temperature to identify that the plasmonic ASI metasurface showed in-plane magnetic frustration due to the Kagome lattice structure (Fig 1b). In addition, polarized reflection spectroscopy was also used to confirm that the plasmonic resonance was located at the 900 nm design wavelength (Fig 1c).



**Figure 2.** (a) Time-resolved MOKE (trMOKE) results for the Ni metasurface taken in the P-MOKE configuration. The inset in (a) shows the longer time scan of the trMOKE signal. (b) Time resolved L-MOKE signal for the Ni metasurface in the positive field ( $+H[+θ]$ , red trace), negative field ( $-H[-θ]$ , green trace) configurations, and their difference ( $Δθ$ , black trace). (c) The fast Fourier transform (FFT) magnitude of oscillations observed in (b) revealing an oscillatory mode at 11.36 GHz, fit with a Gaussian function (red dashed line).

Static MOKE measurements were then performed using a 1 MHz amplified ultrafast laser system producing optical pulses with central wavelengths between 800-900 nm and nominal pulse widths of ~60 femtoseconds (fs). The polarization rotation of the laser beam, following reflection from the metasurface, was interrogated using phase-sensitive lock-in detection enabled by a photo-elastic modulator. We observe saturation behavior from the metasurface in both static longitudinal-MOKE (L-MOKE) and polar-MOKE (P-MOKE) measurements only when the laser wavelength is tuned to coincide with the plasmonic resonance (Fig. 1f,g). In contrast, measurements made away from the resonance at 800 nm are either weak (Fig. 1d) or lack saturation effects (Fig. 1e). We therefore conclude that the near-field enhancement of the electric field afforded by the plasmonic resonance enables us to resolve the in-plane magnetization of the spin-ice lattice [3]. Subsequently, we measured the trMOKE signal at the plasmonic resonance wavelength by splitting the laser beam into a degenerate pump (9mW, TE pol) and probe (1mW, TM pol), both of which were focused onto the sample at near normal incidence using an achromatic lens. Figure 2a shows the trMOKE trace in the P-MOKE configuration where we observe a rapid demagnetization followed by a 1-1.5 ps recovery. As seen in the inset of Fig. 2(a), no additional dynamics are present, even on the several hundred-picosecond time scale. This is in stark contrast to the trMOKE dynamics in the L-MOKE configuration. Figure 2(b) shows the response of the metasurface with positive (red trace) and negative (green trace) in-plane magnetic fields applied across the device; in both cases, we observe strong damped coherent oscillations. Moreover, the oscillators are phase shifted, evidenced by their persistence in the difference trace ( $Δθ$ , black trace). By taking the Fourier transform of the difference trace, we observe an Eigen mode at 11.36 GHz (Fig. 2c). The phase shift between the oscillations (i.e., the oscillations in the  $Δθ$  trace) is evidence of the non-reciprocal behavior of the Ni spin-ice lattice under the influence of an external magnetic field and enhanced by the plasmonic resonance.

This plasmonic spin-ice metasurface architecture offers an ideal platform to exploit all optical reconfiguration and readout of the spin states necessary to realize emergent or collective interactions within the device, which may enable new computing and microelectronic applications. Furthermore, we expect to leverage the non-reciprocal nature of such plasmonic spin-ice metasurfaces to realize the next generation of the stochastic learning architectures including directional graph networks and deep reservoirs, which are beyond the scope of current computing architectures.

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- [1] Gartside, J. C., Stenning, K. D., Vanstone, A., Holder, H. H., Arroo, D. M., Dion, T., ... & Branford, W. R. (2022). Reconfigurable training and reservoir computing in an artificial spin-vortex ice via spin-wave fingerprinting. *Nature Nanotechnology*, 17(5), 460-469.
- [2] Schuller, J. A., Barnard, E. S., Cai, W., Jun, Y. C., White, J. S., & Brongersma, M. L. (2010). Plasmonics for extreme light concentration and manipulation. *Nature materials*, 9(3), 193-204.
- [3] Maksymov, I. S. (2016). Magneto-plasmonic nanoantennas: Basics and applications. *Reviews in Physics*, 1, 36-51.
- [4] Beaini, D., Passaro, S., Létourneau, V., Hamilton, W., Corso, G., & Liò, P. (2021, July). Directional graph networks. In International Conference on Machine Learning (pp. 748-758). PMLR.