

All-Optical Manipulation of Magnetization in Ferromagnetic Thin Films Enhanced by Plasmonic Resonances

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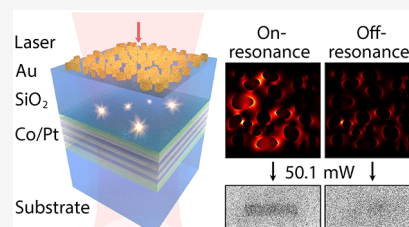
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Supporting Information

ABSTRACT: In this paper, we report all-optical manipulation of magnetization in ferromagnetic Co/Pt thin films enhanced by plasmonic resonances. By annealing a thin Au layer, we fabricate large-area Au nanoislands on top of the Co/Pt magnetic thin films, which show plasmonic resonances around the wavelength of 606 nm. Using a customized magneto-optical Kerr effect setup, we experimentally observe an 18.5% decrease in the minimum laser power required to manipulate the magnetization, comparing the on- and off-resonance conditions. The results are in very good agreement with numerical simulations. Our research findings demonstrate the possibility to achieve an all-optical magnetic recording with low energy consumption, low cost, and high areal density by integrating plasmonic nanostructures with magnetic media.

KEYWORDS: plasmonics, all-optical manipulation, magnetization, ferromagnetic materials



The rapid development of plasmonics over the last two decades has triggered a variety of research areas, such as biochemical sensing,^{1,2} plasmon-enhanced energy harvesting,^{3,4} plasmon-induced hot carrier generation,⁵ plasmonic metamaterials,⁶ magneto-plasmonics, and heat-assisted magnetic recording (HAMR) for data storage. In particular, magneto-plasmonics focus on the enhancement of magneto-optical effect by plasmonic nanostructures, such as nanodisks,^{7–12} nanoholes,^{13–19} and gratings.^{20–28} On the other hand, we can utilize plasmonic nanoantennas,^{29–34} resonant nanocavities,³⁵ or solid-immersion superoscillatory lens³⁶ to enhance the optothermal effect within the magnetic recording medium for HAMR technology. An external magnetic field is then applied to store data within the heated areas. So far, HAMR with an areal density up to petabyte per meter square has been realized.^{30,31} The use of the plasmonic nanostructures has also been proposed in the research field of all-optical magnetization switching.^{37–41} In this scheme, local magnetic fields delivered by a recording head are no longer needed. Instead, magnetization can be directly controlled by ultrafast laser pulses. Therefore, it is expected that the writing speed can be much faster than the current magnetic data storage technologies. In 2015, confined magnetic switching at the scale less than 100 nm was demonstrated in a system that gold antennas were integrated on top of a ferrimagnetic TbFeCo film, while the highly inhomogeneous nature of the switching process was also observed due to the heterogeneity of the TbFeCo film.³⁷ A recent publication reported the layer-selective all-optical magnetic recording assisted by surface plasmon polaritons, which would potentially increase the storage density of an opto-magnetic recording by a factor of at

least two.³⁸ Besides the use of plasmonic nanostructures, researchers have also proposed other methods that can potentially increase the data storage density via azimuthally polarized vortex beams⁴² and 3D light-induced magnetic holography.⁴³

In addition to the areal density and writing speed, energy consumption is an equally important issue for data storage, which has not been explicitly explored in plasmon-mediated all-optical manipulation of magnetization. In this work, we present experimental observations of all-optical magnetization manipulation in ferromagnetic thin films enhanced by plasmonic resonances. Compared with ferrimagnetic materials, ferromagnetic materials generally have larger spin polarization and magnetic anisotropy, which are technically important for real applications, such as integrated magneto-optical memory, data storage, and data processing.⁴⁴ The plasmonic nanostructure adopted in our work is Au nanoislands, which can be easily fabricated with low cost and large area by annealing a thin Au layer under appropriate temperature and time conditions.^{45–47} Such nanostructures have demonstrated applications in surface-enhanced Raman spectroscopy,⁴⁸ plasmon-enhanced photodetection,⁴⁹ and broadband absorbers.⁵⁰ It is shown that, by integrating Au nanoislands on top of magnetic media, we can locally enhance the electric fields

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within the magnetic media, resulting in a lower laser threshold (about 18.5% decrease) to manipulate magnetization at the resonance wavelength of 606 nm in comparison with the off-resonance condition. We have performed full-wave simulations, which show a 17.0% enhancement of the electric field intensity in the Co/Pt thin films when comparing the on- and off-resonance wavelengths. This result quantitatively explains the observed reduction in the threshold. Due to their random geometries and distributions, the sample exhibits uniform plasmonic resonance, which is beneficial for practical applications.

Results and Discussion. Sample Fabrication. As illustrated in Figure 1a, the overall configuration of the sample

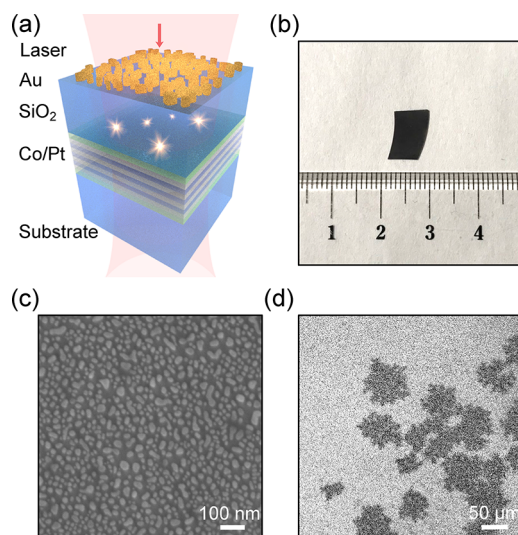


Figure 1. (a) Schematic view of the sample. A laser beams illuminates from the top side consisting of Au nanoislands. The electric field within the magnetic Co/Pt thin films is enhanced thanks to the plasmonic resonance of Au nanoislands, which reduces the minimum laser power to manipulate magnetization. (b) Photograph of the sample along with a ruler that has a unit length of 1 mm. (c) SEM image of the fabricated Au nanoislands. (d) MOKE microscopy image of the Co/Pt sample integrated with Au nanoislands, which clearly shows domains with out-of-plane magnetization.

can be considered as a metal (gold)-insulator (SiO_2)-metal (Co/Pt thin films) structure, which has been widely adopted to achieve perfect light absorption.^{51,52} Figure 1b shows a photograph of the sample, indicating that our fabrication process can easily fabricate plasmonic nanostructures over a large scale yet with low cost, in comparison with conventional nanofabrication methods, such as electron beam lithography and focused ion beam (FIB) milling.^{53–56} The scanning electron microscopy (SEM) image of the resulting Au nanoislands is presented in Figure 1c. The fabricated Au nanoislands are random in shapes and sizes, giving rise to the uniform and relatively broadband plasmonic resonance. The magnetic medium is Co/Pt ferromagnetic thin films, fabricated by DC magnetron sputtering onto a glass substrate at room temperature. The deposition was performed with an Ar pressure 3×10^{-3} Torr and a background pressure 10^{-7} Torr. The Co/Pt multilayer structure is Ta(3)/Pt(0.8)/[Co(0.8)/Pt(0.8)]₃/Ta(3), where the number in the bracket indicates the layer thickness in nanometers, and Ta serves as the seeding and cladding layer. The 20 nm SiO_2 spacing layer was fabricated by atomic layer deposition (ALD), and a thin

layer of 5 nm Au was deposited by DC magnetron sputtering afterward. After fabrication, the sample was annealed in a chamber filled with Ar. The temperature was set to rise from 20 to 550 °C at a speed of 5 °C/min, held at 550 °C for 1 h, and then naturally cooled down to room temperature. Figure 1d presents the magneto-optical Kerr effect (MOKE) microscopy image of the sample. The black and white areas correspond to magnetic domains pointing downward and upward, respectively. When the laser illuminates the sample from the top side with the Au nanoislands, the electric field within the Co/Pt thin films will be enhanced at the plasmonic resonance wavelength of Au nanoislands. As a result, the minimum laser power required to manipulate magnetization in the magnetic medium will be reduced.

Sample Characterization. We have experimentally measured the transmittance spectra of the fabricated samples, including Co/Pt thin films with and without the Au nanoislands, by a spectrometer. The results are plotted in Figure 2a. Compared with the bare Co/Pt thin film,

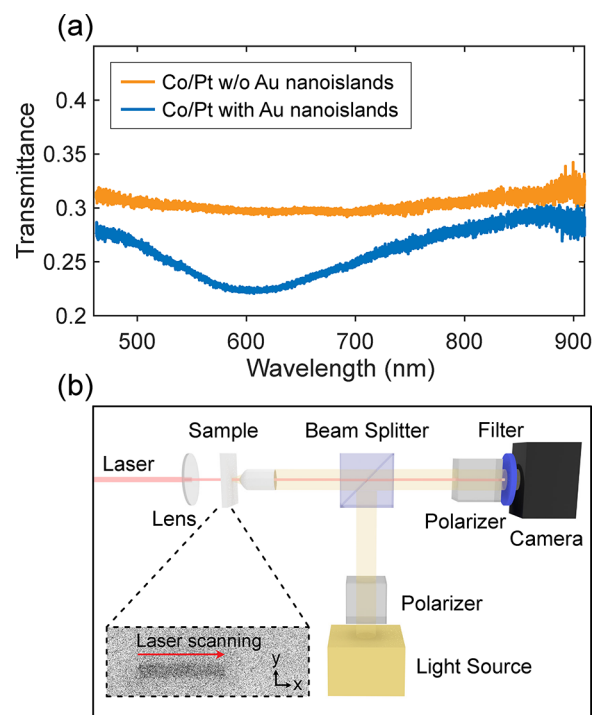


Figure 2. (a) Transmittance spectra of Co/Pt thin films with and without Au nanoislands. The results indicate the plasmonic resonance at 606 nm. (b) Illustration of the experimental setup. Inset: an example of the MOKE image after laser scanning.

film integrated with Au nanoislands shows a transmittance minimum at 606 nm, which indicates the plasmonic resonance at this wavelength. Therefore, we have chosen 606 nm, which is on-resonance, and 505 nm, which is off-resonance, to characterize the performances of all-optical magnetization manipulation.

Figure 2b illustrates the experimental setup. To manipulate the magnetization, we use a Ti:sapphire laser coupled with an optical parametric oscillator (OPO) from Coherent Inc., which allows for the tuning of the output wavelength. The repetition rate of the laser is 80 MHz, and the pulse width is 200 fs. The laser beam is focused on the sample by a lens. The magnetic domains are imaged by the home-built MOKE system, which

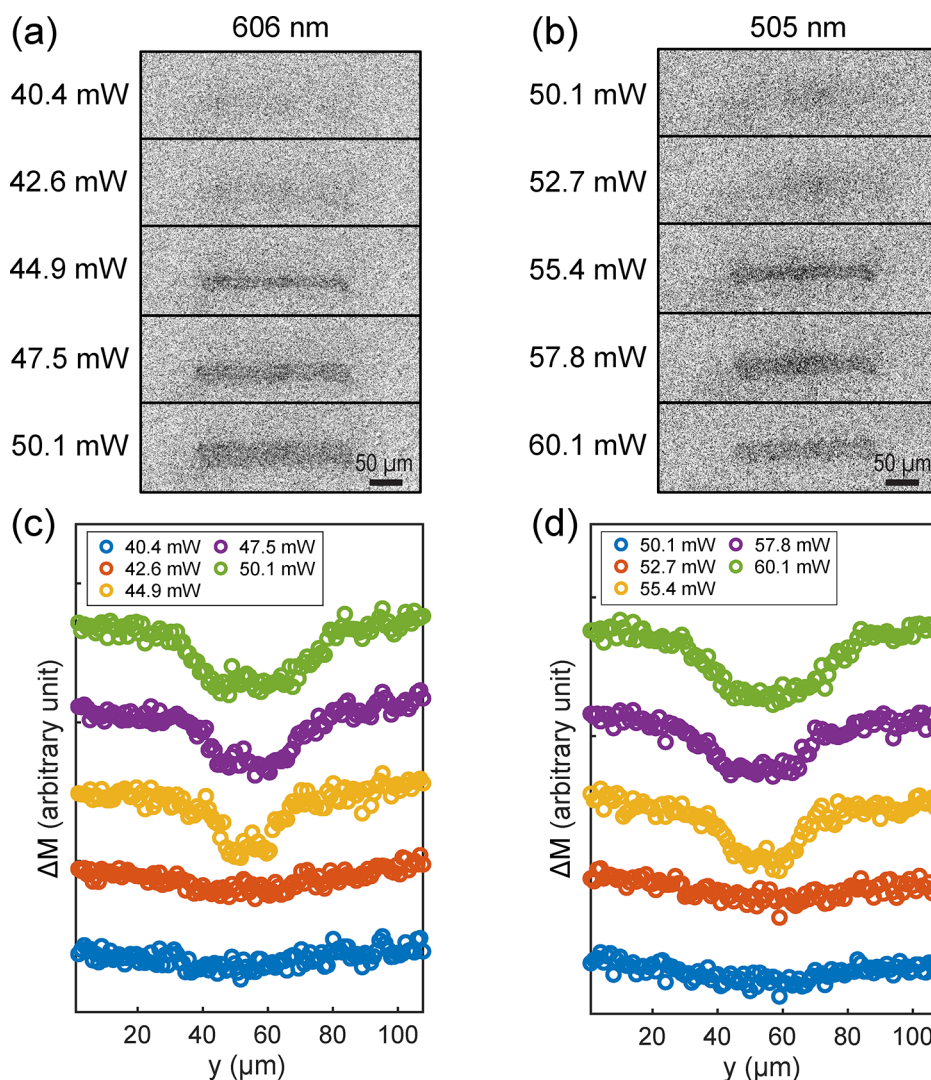


Figure 3. MOKE images of magnetization after laser scanning using different powers at the wavelength of (a) 606 nm and (b) 505 nm. (c, d) Cross-sectional plot of magnetization versus the y -axis at the two wavelengths, respectively.

Table 1. Threshold Power (in the Unit of mW) to Manipulate Magnetization Measured at Six Different Locations

test	1	2	3	4	5	6	mean	SD
606 nm	45.2	45.2	44.9	44.9	45.1	44.9	45.0	0.2
505 nm	55.4	54.8	54.9	55.3	55.4	55.1	55.2	0.3

consists of one white light source, one camera, and two Glan–Taylor polarizers. A short-pass filter with the cutoff wavelength of 500 nm (Thorlabs, FESH0500) is placed before the camera to block the high-intensity laser pulses while transmitting a large portion of the imaging white light. Depending on the magnetization orientation, linearly polarized light exhibits opposite polarization rotation angles that can be analyzed by a polarizer. Therefore, we can optically distinguish magnetizations pointing upward and downward, which show white and black contrasts in the MOKE image, respectively. The inset in Figure 2b shows an example of the MOKE image after scanning the laser on the Co/Pt thin films integrated with Au nanoislands. We define the laser scanning direction as the x -axis and the vertical direction as the y -axis for the subsequent analysis. It should be noted that all of the switched magnetization can be erased by external magnetic fields, unless the laser power is too high to damage the sample locally.

Plasmon-Enhanced Magnetization Manipulation. We have measured the minimum laser power required to manipulate magnetization in the Co/Pt with Au nanoislands at the on- and off-resonance wavelengths. Figure 3a,b presents the MOKE images after each laser scanning event for the two cases when the laser power gradually increases. At a 606 nm wavelength, which is the on-resonance condition, the magnetization manipulation can be achieved at 44.9 mW. While for 505 nm, which is the off-resonance condition, the magnetization manipulation is achieved at 55.4 mW. We estimated that the laser beam diameter at the wavelength of 606 and 505 nm was 28.9 and 24.1 μm , respectively. The fluence can be calculated by $F = \frac{P}{f\pi r^2}$ in which $f = 80$ MHz is the laser repetition rate, P denotes the laser power, and r is the laser beam radius. Therefore, the fluence thresholds are approximately 0.086 mJ/cm^2 and 0.15 mJ/cm^2 at the on-resonance

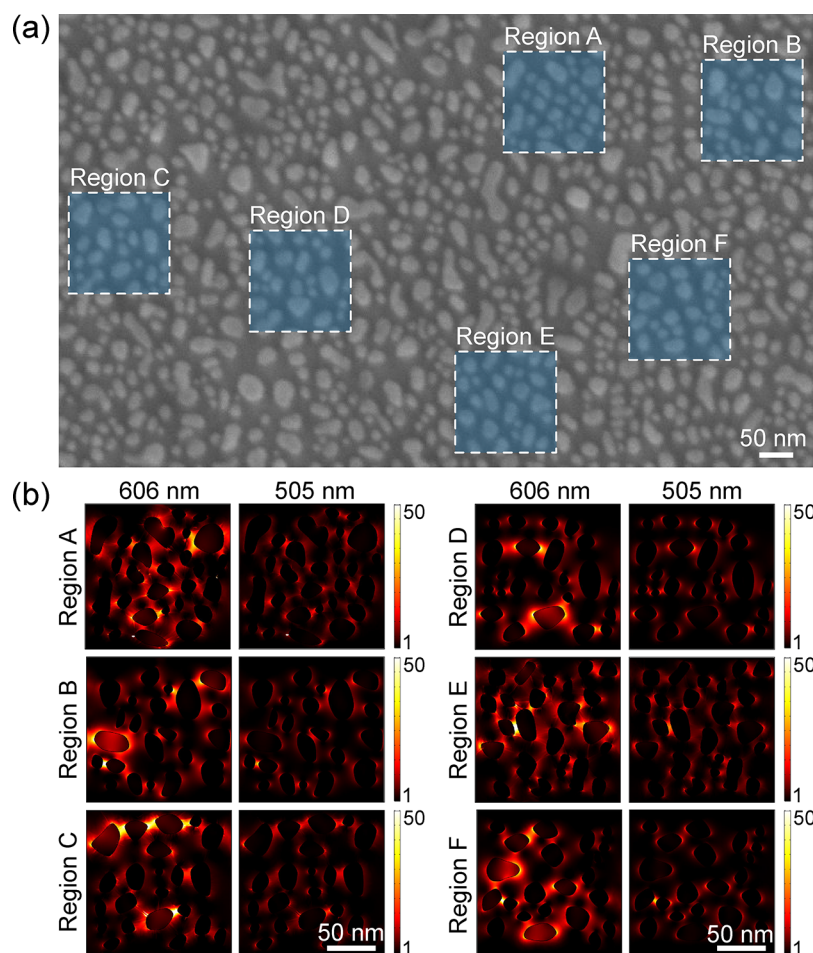


Figure 4. (a) SEM image of Au nanoslands, in which 6 regions (A–F), each in size of $150\text{ nm} \times 150\text{ nm}$, are randomly selected for the simulations. (b) Enhancement of electric field intensity in the middle plane of the Au nanoslands at the wavelength of 606 and 505 nm. A clear enhancement can be observed through the comparison between the on- and off-resonance wavelengths.

wavelength (606 nm) and off-resonance wavelength (505 nm), respectively.

To test the average performance of the sample, we have repeated these measurements at 6 randomly chosen locations on the sample, and similar results were observed. In Table 1, we present the observed threshold to manipulate magnetization for the 6 locations. As can be seen, at 606 nm, the averaged threshold is 45.0 mW with a standard deviation (SD) of 0.2 mW. At 505 nm wavelength, the averaged threshold is 55.2 mW with the standard deviation of 0.3 mW. The small standard deviation values indicate the uniform performances at different locations of our sample. From these experimental results, we calculate the percentage change for the magnetization manipulation threshold power as $(P_{\text{on-resonance}} - P_{\text{off-resonance}})/P_{\text{off-resonance}} = (45\text{ mW} - 55.2\text{ mW})/55.2\text{ mW} = -18.5\%$, which indicates an 18.5% reduction.

We have conducted additional analyses to better qualify the observed phenomena. By integrating the value of pixels along the x -axis within the laser scanning regions, we plot the magnetization versus the y -axis. The cross-section of the magnetization at the wavelengths of 606 and 505 nm are shown in Figure 3c and 3d, respectively. For both wavelengths, no magnetization manipulation can be observed when the laser power is under the threshold value. If we increase the laser power to a value that is larger than the threshold, we observe the resulting magnetization as a trough in the plot, and the

width of the trough becomes larger as the laser power increases. This is due to the Gaussian profile of the focused laser beam. When the laser power increases, there is a larger region that exceeds the threshold, resulting in a wider line after laser scanning events. The laser scanning experiments on bare Co/Pt thin films and the comparison with the samples integrated with Au nanoslands can be found in Sections 1 and 2 in the Supporting Information.

Modeling of the Plasmonic Enhancement Effect. We have simulated the local field distributions for the Co/Pt film integrated with Au nanoslands by commercial electromagnetic solver COMSOL Multiphysics. In order to obtain the averaged performance of the Au nanoslands, we randomly selected 6 regions as marked in Figure 4a, all in the size of $150\text{ nm} \times 150\text{ nm}$. After selection, the SEM images of Au nanoslands were vectorized and imported into COMSOL to reproduce their exact geometries. The thickness of the Au nanoslands was estimated to be 17 nm. The Co/Pt thin films and the Ta seed and capping layers were also included in the modeling. In Figure 4b, we present the enhancement of electric field intensity of the 6 regions in the middle plane of the Au layer at 606 and 505 nm. The enhancement is calculated by normalizing with the intensity of horizontally polarized incident light, which is set as $1\text{ V}^2/\text{m}^2$. Note that all results were plotted with the same color bar so that we can compare the local enhancement directly. We can clearly see that, at the

Table 2. Averaged Electric Field Intensity in the Co/Pt Middle Plane for Regions A–F

region	A	B	C	D	E	F	mean	SD
606 nm	0.161	0.159	0.165	0.176	0.175	0.151	0.165	0.010
505 nm	0.136	0.138	0.141	0.146	0.142	0.140	0.141	0.003

resonance wavelength of 606 nm, the local fields in all of the 6 regions exhibit larger enhancement than those at the off-resonance wavelength. The simulated results show local hot spots generated by the plasmonic nanostructures, which give rise to a pronounced optothermal effect (see Section 3 in the Supporting Information). In addition, they can potentially be applied to achieve high-density magnetic storage.

To estimate the experimentally observed decrease of threshold power for magnetization manipulation, we have quantitatively calculated the averaged electric field intensity in the Co/Pt middle plane, which is normalized to the intensity of incident light for the 6 regions at the wavelength of 606 and 505 nm. The results are summarized in Table 2. We have calculated the averaged normalized electric field intensity for 606 nm, which is 0.165 with the standard deviation of 0.010, while, for 505 nm, it is 0.141 with the standard deviation of 0.003. As a result, we calculate the percentage increase as $(I_{\text{on-resonance}} - I_{\text{off-resonance}})/I_{\text{off-resonance}} = (0.165 - 0.141)/0.141 = 17.0\%$ in the averaged normalized electric field intensity, which is consistent with the 18.5% decrease of the magnetization manipulation threshold observed in our experiment. It is expected that by patterning the plasmonic nanostructures into regular shapes, such as nanodisk and nanohole arrays,^{57–59} we can further reduce the magnetization manipulation threshold at particular wavelengths. Our current simulation results of Au nanodisk arrays show that the maximum local intensity enhancement in the magnetic medium can reach up to 460% (see Section 4 in the Supporting Information).

In conclusion, in this work, we have proposed a method to reduce the energy consumption for magnetization manipulation by using the plasmonic resonance of Au nanoislands. The Au nanoislands are fabricated by annealing a thin Au layer and thus suitable for low-cost and mass production. We have experimentally measured an 18.5% reduction in the power required to manipulate magnetization. Full-wave simulations show an average enhancement of about 17.0% for the electric field intensity within the middle plane of magnetic media, which agrees well with the observed reduction of power threshold. Our results manifest the potential to achieve low-power and high-density all-optical magnetic data storage through the integration of plasmonic nanostructures.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.0c02089>.

Additional data and discussions including the all-optical manipulation of magnetization in the bare Co/Pt thin film and its comparison with the sample integrated with Au nanoislands, the simulation of the optothermal effect, and the design of Au nanodisk arrays to further enhance all-optical manipulation of magnetization (PDF)

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Notes

The authors declare no competing financial interest.

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