Supplementary Information

Control of magnetic contrast with nonlinear magneto-plasmonics

Wei Zheng\textsuperscript{1}, Aubrey T. Hanbicki\textsuperscript{2}, Berend T. Jonker\textsuperscript{2}, and Gunter Lüpke\textsuperscript{1}

\textsuperscript{1}Department of Applied Science, College of William & Mary, Williamsburg, VA 23187, USA
\textsuperscript{2}Materials Science & Technology Division, Naval Research Laboratory, Washington, D.C. 20375, USA
Experimental Methods

In this study, we employ MSHG and MOKE measurements with P-polarized fundamental fields coupled by a prism into a single-crystalline Fe film (Fig. 2a). A picosecond pulsed beam at 800-nm wavelength from a Ti:Sapphire amplifier laser system is used as the source for MSHG, MOKE, and SP. A data gathering system including chopper, boxcar, lock-in amplifier, photon multiplier tube (PMT), and polarizers with 1:10000 extinction ratios enable us to detect the small MSHG signal. A GPIB controlled electromagnet provides an external magnetic field H in longitudinal or transverse direction. The single crystal iron sample is critical because the crystal structure is the main source for the cubic magnetic anisotropy, which results in the two-jump switching process. It has been shown previously that the Fe/MgO system possesses an additional small uniaxial magnetic anisotropy term. We conducted MSHG measurements under ATR conditions on the Fe/MgO samples for the two in-plane [110] and [-110] directions which show similar hysteresis loops indicating that the uniaxial anisotropy term is very small and can be neglected.

Sample Growth and Characterization

The 10-nm thick Fe film samples were grown by molecular beam epitaxy at room temperature on <100> MgO substrates. The substrates were cleaned with isopropyl alcohol and annealed in-situ to 823 K for 5 minutes prior to growth. Reflection high-energy electron diffraction (RHEED) measurements indicate single-crystal Fe growth. There is no protective overlayer. The Fe films are stable for several years (at least 4-5 years) as checked periodically with MOKE and vibrating sample magnetometer (VSM) measurements. According to our previous measurements, the native oxide layer is only approximately 1-nm thick, because the single crystal nature of the Fe film limits the oxidation rate. Therefore, the effect of the oxide layer on the surface plasmon and the field enhancement is very small according to our calculations shown in Figure S1. For
this study, MSHG and MOKE measurements have been carried over the duration of one year after film deposition and the hysteresis loops from the Fe films did not change during this time period.

Figure S1. Effect of oxide on surface plasmon. The oxide layer has a very small effect on the field strength and the attenuated total reflection because of the very small thickness.

Magnetic contrast of two-jump hysteresis loop

The magnetic contrast for one-jump hysteresis loop can be expressed as:

$$C = \frac{I(+M) - I(-M)}{I(+M) + I(-M)},$$  (4)
where \( I(+M) \) and \( I(-M) \) are intensities for the two magnetization states. In the same way, we define the T- and L- magnetic contrasts for two-jump loop as:

\[
C_T = \frac{(I(M_{III}) - I(M_{II})) + (I(M_{IV}) - I(M_I))}{I(M_I) + I(M_{II}) + I(M_{III}) + I(M_{IV})}
\]

and

\[
C_L = \frac{(I(M_{III}) - I(M_{II})) + (I(M_{IV}) - I(M_I))}{I(M_I) + I(M_{II}) + I(M_{III}) + I(M_{IV})},
\]

where \( M_I, M_{II}, M_{III} \) and \( M_{IV} \) are depicted in Fig. 1. Next, we derive expressions for the MSHG intensities \( I^{2\omega}(M_i), i=I,..,IV \). The total MSHG response (polarization) from a magnetic material can be simplified to

\[
P(2\omega) = p^{\text{even}} \pm p_T^{\text{odd}} \pm p_L^{\text{odd}},
\]

where \( p^{\text{even}} \) is the second-order polarization from the crystal, or the non-magnetic contribution, \( p_T^{\text{odd}} \) and \( p_L^{\text{odd}} \) are the magnetization-induced polarizations for L- and T- magnetization components, respectively, which change sign as magnetization reverses. Since these polarization contributions are complex quantities, involving fundamental fields, corresponding effective susceptibility tensors and polarization angles, the total (MSHG) signal is thus given by

\[
I^{2\omega}(M_I) = (p^{\text{even}})^2 + (p_T^{\text{odd}})^2 + (p_L^{\text{odd}})^2 \\
\pm 2 \cdot p^{\text{even}} \cdot p_T^{\text{odd}} \cdot \cos \phi_T \pm 2 \cdot p^{\text{even}} \cdot p_L^{\text{odd}} \cdot \cos \phi_L \pm 2 \cdot p_T^{\text{odd}} \cdot p_L^{\text{odd}} \cdot \cos \phi
\]

Here \( \phi_T \) is the phase difference between \( p^{\text{even}} \) and \( p_T^{\text{odd}} \), \( \phi_L \) is the phase difference between \( p^{\text{even}} \) and \( p_L^{\text{odd}} \), and \( \phi \) is the phase difference between \( p_T^{\text{odd}} \) and \( p_L^{\text{odd}} \). Taking into consideration of the polarization angle \( \alpha \), \( I^{2\omega}(M_I) \) is thus given by
\[ I^{2\alpha}(M) = (p_{\text{even}} \cdot \cos \alpha)^2 + (p_{T}^{\text{odd}} \cdot \cos \alpha)^2 + (p_{L}^{\text{odd}} \cdot \sin \alpha)^2 \]
\[ \pm 2 \cdot p_{\text{even}} \cdot p_{T}^{\text{odd}} \cdot \cos \alpha \cdot \cos \alpha \cdot \cos \varphi_T \pm 2 \cdot p_{\text{even}} \cdot p_{L}^{\text{odd}} \cdot \cos \alpha \cdot \sin \alpha \cdot \cos \varphi_L \pm 2 \cdot p_{L}^{\text{odd}} \cdot p_{T}^{\text{odd}} \cdot \sin \alpha \cdot \sin \alpha \cdot \cos \varphi \]  

(8)

where \( \alpha \) equals to 0° when the analyzer is set to P polarization, and 90° when the analyzer is set to S polarization. This is the general expression for the MSHG intensity. More specifically,

\[ I^{2\alpha}(M_{I}) = (p_{\text{even}} \cdot \cos \alpha)^2 + (p_{T}^{\text{odd}} \cdot \cos \alpha)^2 + (p_{L}^{\text{odd}} \cdot \sin \alpha)^2 \]
\[ -2 \cdot p_{\text{even}} \cdot p_{T}^{\text{odd}} \cdot \cos \alpha \cdot \cos \alpha \cdot \cos \varphi_T - 2 \cdot p_{\text{even}} \cdot p_{L}^{\text{odd}} \cdot \cos \alpha \cdot \sin \alpha \cdot \cos \varphi_L + 2 \cdot p_{L}^{\text{odd}} \cdot p_{T}^{\text{odd}} \cdot \sin \alpha \cdot \sin \alpha \cdot \cos \varphi \]  

(9)

\[ I^{2\alpha}(M_{II}) = (p_{\text{even}} \cdot \cos \alpha)^2 + (p_{T}^{\text{odd}} \cdot \cos \alpha)^2 + (p_{L}^{\text{odd}} \cdot \sin \alpha)^2 \]
\[ -2 \cdot p_{\text{even}} \cdot p_{T}^{\text{odd}} \cdot \cos \alpha \cdot \cos \alpha \cdot \cos \varphi_T + 2 \cdot p_{\text{even}} \cdot p_{L}^{\text{odd}} \cdot \cos \alpha \cdot \sin \alpha \cdot \cos \varphi_L - 2 \cdot p_{L}^{\text{odd}} \cdot p_{T}^{\text{odd}} \cdot \sin \alpha \cdot \sin \alpha \cdot \cos \varphi \]  

(10)

\[ I^{2\alpha}(M_{III}) = (p_{\text{even}} \cdot \cos \alpha)^2 + (p_{T}^{\text{odd}} \cdot \cos \alpha)^2 + (p_{L}^{\text{odd}} \cdot \sin \alpha)^2 \]
\[ + 2 \cdot p_{\text{even}} \cdot p_{T}^{\text{odd}} \cdot \cos \alpha \cdot \cos \alpha \cdot \cos \varphi_T + 2 \cdot p_{\text{even}} \cdot p_{L}^{\text{odd}} \cdot \cos \alpha \cdot \sin \alpha \cdot \cos \varphi_L + 2 \cdot p_{L}^{\text{odd}} \cdot p_{T}^{\text{odd}} \cdot \sin \alpha \cdot \sin \alpha \cdot \cos \varphi \]  

(11)

\[ I^{2\alpha}(M_{IV}) = (p_{\text{even}} \cdot \cos \alpha)^2 + (p_{T}^{\text{odd}} \cdot \cos \alpha)^2 + (p_{L}^{\text{odd}} \cdot \sin \alpha)^2 \]
\[ + 2 \cdot p_{\text{even}} \cdot p_{T}^{\text{odd}} \cdot \cos \alpha \cdot \cos \alpha \cdot \cos \varphi_T - 2 \cdot p_{\text{even}} \cdot p_{L}^{\text{odd}} \cdot \cos \alpha \cdot \sin \alpha \cdot \cos \varphi_L - 2 \cdot p_{L}^{\text{odd}} \cdot p_{T}^{\text{odd}} \cdot \sin \alpha \cdot \sin \alpha \cdot \cos \varphi \]  

(12)

Applying Eqs. (6) - (9) to Eq. (2), we obtain

\[ C_L = \frac{2 \cdot k_L \cdot \tan(\alpha) \cdot \cos(\varphi_L)}{1 + k_T^2 + k_L^2 \cdot \tan^2(\alpha)} \]  

and,

\[ C_T = \frac{2 \cdot k_T \cdot \tan(\alpha) \cdot \cos(\varphi_T)}{1 + k_T^2 + k_L^2 \cdot \tan^2(\alpha)} \]  

where \( k_L = \frac{p_{L}^{\text{odd}}}{p_{\text{even}}} \) and \( k_T = \frac{p_{T}^{\text{odd}}}{p_{\text{even}}} \).
Equation (10) provides the L- and T-magnetic contrasts in a two-jump system as a function of polarization angle $\alpha$ including the ratios of magnitude and relative phases between non-magnetic and magnetic MSHG components.

For each incident angle, two-jump hysteresis loops are taken with different analyzer angles. The contrast ratios for the T- and L- components are obtained using equation (2). By fitting $C_T$ and $C_L$ as a function of analyzer angle to equation (3), $\phi_T$, $\phi_L$, $k_T$ and $k_L$ are obtained. The same process is applied to different incident angles over the ATR range, and the results are summarized in Figs. 4a and 4b. Figures S2a and S2b are the fitting curves when the incident angle is 43.5°. The fitting curves agree very well with the experimental data.

Figure S2.  Fittings to determine the phase difference and ratio of magnitude as a function of incident angle. a, L-MSHG. b, T-MSHG. The incident angle is 43.5°. The external magnetic field H is applied along the transverse direction, i.e. the hard axis [110].

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