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Magnetization-induced second-harmonic generation of light by exchange-coupled magnetic layers

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A longitudinal magneto-optical Kerr effect and magnetization-induced second-harmonic generation (MSHG) of light (at 2ω) have been measured in a SiO2/Fe96Si4/Dy30Fe58Co12/glass exchange-coupled magnetic bilayer system with competitive anisotropies. Theoretical MSHG predictions in this structure that give rise to an effect proportional to magnetization components and that are allowed by an electric dipole mechanism are reported and discussed. The magnitude of the MSHG effect depends on the electric field of the incoming radiation at each interface and on the corresponding incoming (at ω) and outgoing (at 2ω) Fresnel coefficients. It is demonstrated that transverse pp MSHG selectively probes the magnetization of the first SiO2/Fe96Si4 interface, while transverse sp and longitudinal pp MSHG is sensitive, but less selectively, to the Fe96Si4/Dy30Fe58Co12 interface that supports a planar magnetic domain wall. (p and s are the usual parallel and perpendicular polarizations to the plane of incidence.) The contribution to MSHG by gradient magnetization terms is negligible. © 2005 Optical Society of America

1. INTRODUCTION

The magneto-optical Kerr effect (MOKE) has been used to probe the magnetic properties of thin-layered and multilayered structures.1 It gives information on the magnetic state of the structure over the light’s penetration depth, i.e., a few tens of nanometers in metals. Magnetic surface properties are now well probed by electron techniques such as spin-polarized scanning-electron microscopy,2 spin-polarized low-energy electron microscopy,3 and spin-polarized scanning tunneling microscopy.4 So most unsolved problems concern buried interface magnetism in multilayer structures. There are few available techniques to check interface magnetism, and the usefulness of magnetization-induced second-harmonic generation (MSHG) for this purpose has still to be confirmed. For electric dipole mechanisms, which are often favored in centrosymmetric thin metallic film structures, MSHG is predicted to come only from the surface and interfaces as a consequence of local symmetry breaking. This interface-induced effect has been predicted theoretically, and light-polarization selection rules have been deduced for all-optical configurations.5,6 Whereas it has been demonstrated experimentally that MSHG undoubtedly comes from surface magnetism,7–10 there have been few studies of real multilayer structures involving several types of buried interface.11–15

Our aim in the present study is to check theoretical predictions about MSHG in a simple exchange-coupled layer structure built from two magnetic layers in contact that have perpendicular and in-plane anisotropy, respectively. MSHG is then assumed to check partially the magnetization state at their interface, on which is centered a planar domain wall. A search for MSHG contributions that are due to magnetization gradients16 can be also initiated in that case.

Such a type of structure is commonly used in the design of magneto-optical and magnetic recording media and for achieving new generations of magnetic memories. In thermally assisted writing of the information, such a structure has been proposed to improve the direct overwriting process17 and to design a recording–readout stack of layers for achieving magnetic superresolution.18 In perpendicular recording, a capping layer with in-plane anisotropy favors magnetic flux closure and thus lowers the write–erase field.19 For coupled ferrimagnetic–ferromagnetic bilayers, as studied here, an exchange bias field acts on the soft ferromagnetic layer with in-plane anisotropy.20 This kind of exchange-coupled bilayer...
structure is also needed for stabilizing the magnetization in the hard layer of magnetic random-access memories.

2. EXPERIMENT

A ternary rare-earth transition metal Dy$_{30}$Fe$_{58}$Co$_{12}$ layer was grown on a 1-mm-thick glass substrate by sputtering of the elements at an Ar pressure of 1.5 × 10$^{-3}$ Torr at a deposition rate of 9.3 nm/min. Above this layer a Fe$_{96}$Si$_4$ layer was subsequently grown by sputtering at 7.5 × 10$^{-3}$-Torr Ar pressure at a deposition rate of 4 nm/min. Finally, a 10-nm-thick SiO$_2$ layer was deposited on the top of this structure to prevent its oxidation. This structure (Fig. 1) can serve as a magnetic or magneto-optic recording medium. Two samples were prepared. In each case the Dy$_{30}$Fe$_{58}$Co$_{12}$ layer thickness was fixed at 30 nm, but the FeSi layer was either 5 or 10 nm thick. X-ray analyses have confirmed the amorphous character of both the Dy$_{30}$Fe$_{58}$Co$_{12}$ and the Fe$_{96}$Si$_4$ layers. We also prepared two other Dy$_{30}$Fe$_{58}$Co$_{12}$ or Fe$_{96}$Si$_4$ single-layer samples to check independently their magnetic and magneto-optical properties.

The composition of the ferrimagnetic Dy$_{30}$Fe$_{58}$Co$_{12}$ layer was selected to produce a large uniaxial anisotropy constant $K_1$ (equal to 1.7 10$^8$ erg/cm$^3$ at room temperature) favoring an out-of-plane orientation of both rare-earth and transition metal sublattice magnetization. As we shall see below, the temperature of the sample was raised to 80 °C under focused laser irradiation. So the Dy$_{30}$Fe$_{58}$Co$_{12}$ layer composition was also chosen to produce a compensation temperature ($T_{\text{comp}} = 75^\circ C$) close to that reached during measurements. This layer also exhibits a low Curie temperature, $T_c = 180^\circ C$. Under these conditions the saturated magnetization of the illuminated Dy$_{30}$Fe$_{58}$Co$_{12}$ layer becomes much lower than its room-temperature value. Thus the interface exchange interaction is reduced sufficiently to permit the planar wall to be located easily in the vicinity of the Dy$_{30}$Fe$_{58}$Co$_{12}$/Fe$_{96}$Si$_4$ interface. The composition of the ferromagnetic Fe$_{96}$Si$_4$ layer was selected to produce a weak in-plane anisotropy and a small coercive field. The room-temperature out-of-plane and in-plane coercivity of the Dy$_{30}$Fe$_{58}$Co$_{12}$/Fe$_{96}$Si$_4$ interface explain why the room-temperature LMOKE hysteresis loop has square shape.

In the exchange coupled layer structure the competition between in-plane and out-of-plane anisotropies is efficient mainly at the Dy$_{30}$Fe$_{58}$Co$_{12}$/Fe$_{96}$Si$_4$ interface. The Dy$_{30}$Fe$_{58}$Co$_{12}$ layer still shows a square loop in a perpendicular field but with a smaller coercivity ($H_c = 1.3$ kOe at room temperature) than for the same isolated film. An exchange bias effect is then expected between the ferrimagnetic and ferromagnetic layers, but for our sample cooled in zero field through the Curie temperature of Dy$_{30}$Fe$_{58}$Co$_{12}$ there is no exchange bias effect. So, under that condition, the absence of any shift in the hysteresis loop above room temperature can be checked by the LMOKE. The magnetic coercivity of the Fe$_{96}$Si$_4$ layer, however, is nearly isotropic in the film plane. As a consequence of the exchange interaction between these magnetic layers, the room-temperature coercivity of Fe$_{96}$Si$_4$ is larger [$H_c = 40$ Oe for Fe$_{96}$Si$_4$ (5 nm) and 52 Oe for Fe$_{96}$Si$_4$ (10 nm)] than that found for the single layer ($H_c = 3$ Oe). The amorphous nature of the Fe$_{96}$Si$_4$ layer and the presence of a planar domain wall at the Dy$_{30}$Fe$_{58}$Co$_{12}$/Fe$_{96}$Si$_4$ interface explain why the room-temperature LMOKE hysteresis loop has square shape.

The competition between anisotropy orientations induces a variable deviation angle $\Phi$ with respect to the film normal, of the magnetization through the film thickness. A numerical simulation of $\Phi$ versus incident position $z$ from the Dy$_{30}$Fe$_{58}$Co$_{12}$/glass interface was simulated by the OOMMF software program. The following parameters were used: $M_s = 8$ emu/cm$^3$ for the Fe$_{96}$Si$_4$ layer; $K = 0$ and $K_1 = 1.7 \times 10^8$ erg/cm$^2$ for the Fe$_{96}$Si$_4$ and Dy$_{30}$Fe$_{58}$Co$_{12}$ layers, respectively; $J = 12 \times 10^{-12}$ erg/cm$^2$ and $J' = 13 \times 10^{-11}$ erg/cm$^2$ for the exchange interaction in the Dy$_{30}$Fe$_{58}$Co$_{12}$ and the Fe$_{96}$Si$_4$ layers, respectively; and $J_{\text{int}} = J'/2$ for exchange interaction $J_{\text{int}}$ between the two magnetic layers. Whereas $\Phi$ vanishes at the Dy$_{30}$Fe$_{58}$Co$_{12}$/glass interface, it increases near the Dy$_{30}$Fe$_{58}$Co$_{12}$/Fe$_{96}$Si$_4$ (5-nm) interface (Fig. 2). This means that the planar domain wall spreads inside the two magnetic layers. As a consequence, the magnetic domain wall’s width is much greater than that of the in-
termixed structural region at the interface. Thus the symmetry breaking at the interface is essentially structural in origin. The origin of MSHG at interfaces is consequently associated with efficient nonmagnetic nonlinear susceptibility terms. In a zero in-plane magnetic field the value of $\Phi$ inside the Fe$_{96}$Si$_4$ layer rapidly becomes nearly constant. In spite of the in-plane anisotropy character of the Fe$_{96}$Si$_4$ top layer, $\Phi$ does not reach $90^\circ$ at the Fe$_{96}$Si$_4$/SiO$_2$ interface. The variation of $\Phi$ with the depth position for the Fe$_{96}$Si$_4$ (10-nm) film is quite similar to that represented in Fig. 2 for the Fe$_{96}$Si$_4$ (5-nm) film. In the presence of an in-plane applied field the value of $\Phi$ is increased by $\sim 15^\circ$ and $\sim 16^\circ$ in the Fe$_{96}$Si$_4$ layer between $H = 0$ and 200 Oe for the 5- and 10-nm-thick Fe$_{96}$Si$_4$ layers, respectively.

Our MSHG setup was described previously.$^{15,23}$ It allows experiments to be performed in $pp$, $ss$, $ps$, and $sp$ optical configurations$^6$ to check the components of the magnetization in the film plane ($M_x$ and $M_y$ in the transverse or longitudinal case, i.e., for $H\parallel x$ or $H\parallel y$, respectively). The light source was a mode-locked Ti:sapphire laser that provided 100-fs pulses with 800-nm wavelength, and the MSHG signal was detected at the first-harmonic frequency, i.e., at 400 nm. The average light power on the sample surface was 25 mW, and the beam was focused over a 30-40-$\mu$m wide spot. The LMOKE was measured with exactly the same incident light-beam conditions, but detection was achieved for light reflected at the fundamental frequency (at 800 nm). Thus, all reported LMOKE and MSHG experiments were performed with the same laser beam focused at a given position on the film surface, which allowed us to compare all the data.

### 3. MSHG AND FRENSNEL ELEMENT CALCULATIONS

As was discussed earlier,$^{24}$ one can use either of two equivalent models for treating MSHG emission from uniformly magnetized surfaces and buried interfaces.$^{24,25}$ To be consistent with our previous research we prefer to analyze our MSHG data by using the electric point dipole model.$^{24}$ The present analysis does not consider higher-order terms in the expression of the dipole moment, such as that which is quadratic in magnetization, or magnetization gradients. Let us recall briefly the steps in the MSHG calculations:

#### (i) Electric field $E_{\nu}^{(2\omega)}$ at fundamental frequency $\omega$, on each interface $\nu$, is expressed in matrix form:

$$E_{\nu}^{(2\omega)} = X_{\nu}^{(2\omega)} J_{0}^{(\omega)},$$

where $X_{\nu}^{(2\omega)}$ is the matrix of the incoming generalized Fresnel coefficients and $J_{0}^{(\omega)}$ gives the components of the incident electric field. The $z$ axis is oriented along the normal of the film plane, and $y$ lies in the planes both of the film and of incidence.

#### (ii) Electric field $E_{\nu}^{(2\omega)}$ gives rise to electric point dipoles oscillating on interfaces at frequency $\omega$, with moment amplitude$^{24}$

$$\mu_{\nu}^{(2\omega)} = X_{\nu} \otimes E_{\nu}^{(\omega)} E_{\nu}^{(\omega)}, \tag{2}$$

where $X_{\nu}$ is a third-rank nonlinear susceptibility tensor.

The elements of $X_{ij\nu}$ can be classified as nonmagnetic, i.e., $X_{ij\nu}^{(nm)}(M) = X_{ij\nu}^{(nm)}(-M)$, or magnetic, i.e., $X_{ij\nu}^{(m)}(M) = -X_{ij\nu}^{(m)}(-M)$, susceptibility tensor elements. Similarly, the electric dipole moments can be separated into $\mu_{\nu}^{(2\omega)(nm)}$ and $\mu_{\nu}^{(2\omega)(m)}$ magnetic and nonmagnetic emitted electric dipole contributions, respectively, which change or do not change their sign on reversal of the magnetic field. Selection rules for electric dipoles emitting at interfaces as a function of magnetization components are listed in Table 1. Unfortunately, even considering symmetry arguments, the too-large number of nonzero susceptibility tensor elements $X_{ij\nu}$ (see, e.g., Ref. 6) often prevents their experimental determination without crude assumptions. Other MSHG contributions can come from magnetization gradients$^{16}$ or from antiferromagnetic order.$^6$ The relevance of the two last-named contributions is discussed below.

Another limitation in the interpretation of MSHG data comes from difficulty in estimating the characteristic efficient thickness of the second-harmonic generation in the vicinity of a given interface from which second-harmonic light is emitted and that for all values of $X_{ij\nu}$. This effect has been discussed for particular situations.$^{26,27}$

#### (iii) Radiating point dipoles $\mu_{\nu}^{(2\omega)}$ on the $\nu$th interface imply modified boundary conditions of the electric and magnetic fields at interfaces.$^{24,28}$ The relationship between $\mu_{\nu}^{(2\omega)}$ and outgoing electric field amplitudes $E_{\nu}^{(2\omega)}$ at $2\omega$ frequency (with $II = \{s, p\}$ polarization states) is written as

### Table 1. Selection Rules for Magnetic and Nonmagnetic Terms$^a$

| Configuration | Polar $M||z$ | Longitudinal $M||y$ | Transverse $M||x$ | Nonmagnetic |
|---------------|-------------|---------------------|-----------------|-------------|
| $P_{ss}P_{ss}$ | $\mu_x = X_{zzz}^{(m)} E_x^2$ | $\mu_x = X_{zzz}^{(m)} E_x^2 + X_{zzx}^{(m)} E^2$ | $\mu_y = X_{yy}^{(m)} E_y^2$ | $\mu_z = X_{zxy}^{(nm)} E_x E_y$ |
| $P_{ss}s_{ss}$ | $\mu_x = X_{zzz}^{(m)} E_x^2$ | $\mu_x = X_{zzz}^{(m)} E_x^2 + X_{zzx}^{(m)} E^2$ | $\mu_y = X_{yy}^{(m)} E_y^2$ | $\mu_z = X_{zxy}^{(nm)} E_x E_y$ |
| $s_{ss}s_{ss}$ | $\mu_x = X_{zzz}^{(m)} E_x^2$ | $\mu_x = X_{zzz}^{(m)} E_x^2 + X_{zzx}^{(m)} E^2$ | $\mu_y = X_{yy}^{(m)} E_y^2$ | $\mu_z = X_{zxy}^{(nm)} E_x E_y$ |

$^a$Components of the SHG radiated dipole moment $\mu_{\nu}^{(2\omega)}$ generated by electric field $E_{\nu}^{(\omega)}$ at the $\nu$th interface for various MSHG configurations and interface magnetization components. To simplify the notation, we skip the superscripts $2\omega$ for $\mu_{\nu}^{(2\omega)}$ and $\omega$ for $E_{\nu}^{(\omega)}$. The underlined contributions are dominant.
\[
\begin{bmatrix}
\varepsilon_{x,v}^{(2\omega)} \\
\varepsilon_{p,v}^{(2\omega)}
\end{bmatrix} = \begin{bmatrix}
Z_{sx,v}^{(2\omega)} & 0 & 0 \\
0 & Z_{py,v}^{(2\omega)} & Z_{pz,v}^{(2\omega)}
\end{bmatrix} \begin{bmatrix}
\mu_{x,v}^{(2\omega)} \\
\mu_{y,v}^{(2\omega)} \\
\mu_{z,v}^{(2\omega)}
\end{bmatrix},
\]

where \(Z_{sx,v}^{(2\omega)}\) stands for the matrix of generalized outgoing Fresnel elements.

(iv) The resultant outgoing electric field amplitudes, \(\varepsilon_{I,v}^{(2\omega)}\), of the entire multilayer structure (denoted by the subscript tot) are then determined first, by integration over all radiating dipoles located on each interface, and second, by summarizing all interface contributions. When the magnetization and \(\chi_e\) are uniform on each interface, \(\varepsilon_{I,v}^{(2\omega)}\) is simply expressed by a sum over \(\varepsilon_{I,v}^{(2\omega)}\) originating from one point dipole on a given interface:

\[
\varepsilon_{I,v}^{(2\omega)} = \sum_{\mu} \varepsilon_{I,v}^{(\mu,2\omega)}.
\]

(v) The measured far-field second-harmonic generation radiated intensity is then given by

\[
I_{tot}^{(2\omega)} \sim |N_{2}^{(2\omega)}|^{2} |\varepsilon_{x,v}^{(2\omega)}|^{2} + |\varepsilon_{p,v}^{(2\omega)}|^{2},
\]

where \(N_{2}^{(2\omega)} = [(N^{(2\omega)})\sin \phi]^{2}\), where \(\phi\) is the incidence angle. \(N^{(\omega)}\) and \(N^{(2\omega)}\) are the refractive indices of the air at frequencies \(\omega\) and \(2\omega\), respectively.

For different optical configurations the total magnetic emitted light intensity, \(I_{tot}^{(2\omega)(m)}\), that is linear in magnetization is

\[
I_{tot}^{(2\omega)(m)} = 2|N_{2}^{(2\omega)}| |\varepsilon_{I,v}^{(2\omega)(m)}|^{2} \sum_{\mu} \varepsilon_{I,v}^{(\mu,2\omega)(m)}|^{2},
\]

where the summation runs over all interfaces \(\mu\) and \(\bar{\mu}\) and is a complex-conjugate symbol. \(\varepsilon_{I,v}^{(2\omega)(m)}\) expresses the total nonmagnetic electric amplitude emitted by all interfaces.

Relation (6) shows that the magnetic part of the total radiated light intensity, \(I_{tot}^{(2\omega)(m)}\), is given simply by a summation over contributions from all interfaces. Note that the contribution that is due to the \(\nu\)th interface, and proportional to the magnetization, is determined as the product of the magnetic part of the electric field radiated at the \(\nu\)th interface and the total nonmagnetic part of the electrical field originating from all interfaces. As follows from relation (6), \(I_{tot}^{(2\omega)(m)}\) is related to products of nonlinear nonmagnetic and magnetic susceptibility elements \(\chi_{v}^{(m)}(n,m)\). The components of the radiating dipole \(\mu_{2\omega}^{(\mu)}\) are dependent only on particular components of the electric field, \(E_{v}^{(\mu,\omega)}\) (Table 1).

In this section we have neglected the MSHG that is due to \(\chi_{v}^{(m)}(n,m)\) products. This is justified because no quadratic contribution to magnetization has been evidenced in our case. As was mentioned previously, another possible mechanism can be associated with a nonuniform magnetization in-depth profile associated with the presence of a planar domain wall expanding inside the Dy30Fe58Co12 and Fe96Si4 layers. This can theoretically give rise to MSHG through a term proportional to the gradient of the magnetization that may be nonzero inside a centrosymmetric medium, as in the layers themselves. Another contribution can come from the presence of antiferromagnetically aligned moments at the Dy30Fe58Co12/Fe98Si4 interface.

The in-depth profile of the electric field inside the Dy30Fe58Co12/Fe98Si4 structure at frequency \(\omega\) is presented in Fig. 3 for the Fe98Si4 (5-nm) film. The profile of the tangential electric field components, \(E_{x}^{(\omega)}\) and \(E_{z}^{(\omega)}\), decreases monotonically through the multilayer structure. In a counterpart, the profile of the normal component, \(E_{z}^{(\omega)}\), exhibits a steplike variation at interfaces. As discussed previously, the step amplitude is huge for metal–dielectric interfaces because they exhibit different diagonal permittivities, whereas it is rather small for the Dy30Fe58Co12/Fe98Si4 interface.

The calculated values of the generalized incoming Fresnel elements \(X_{sx,v}^{(\omega)}\), \(X_{px,v}^{(\omega)}\), and \(X_{px,v}^{(\omega)}\) are represented in polar form in Fig. 4. This presentation allows representation of the phase information on \(E_{v}^{(\omega)}\) at each interface. Assuming an incident field with normalized amplitude, the electric field modulus at the interfaces is given by \(|E_{x,v}^{(\omega)}| = |X_{sx,v}^{(\omega)}|\), \(|E_{y,v}^{(\omega)}| = |X_{py,v}^{(\omega)}|\), and \(|E_{z,v}^{(\omega)}| = |X_{pz,v}^{(\omega)}|\). In all cases \(E_{z,v}^{(\omega)}\) is discontinuous at each interface, we define it as the average of \(E_{z,v}^{(\omega)}\) values at both sides of the interface. Penetrating further into the film causes the modules and the phases of \(X_{sx,v}^{(\omega)}\) and \(X_{px,v}^{(\omega)}\) and \(X_{px,v}^{(\omega)}\) at the interfaces to decrease progressively. The largest value of \(X_{px,v}^{(\omega)}\) is obviously obtained at the air–SiO2 interface, but it becomes negligible for the deeper Fe98Si4/Dy30Fe58Co12 and Dy30Fe58Co12/glass interfaces.

The generalized outgoing Fresnel elements \(Z_{sx,v}^{(2\omega)}\), \(Z_{px,v}^{(2\omega)}\), and \(Z_{pz,v}^{(2\omega)}\) give access to the radiation emitted in the air by dipoles located on an interface \(v\). They are represented in polar coordinates in Fig. 5 for our Fe98Si4 (5-nm) magnetic bilayer structure. The relation between the radiated intensity (in arbitrary units) and \(Z_{sx,v}^{(2\omega)}\) coefficients is written as \(I_{2} = N_{0}^{(2\omega)}|N_{z,v}^{(2\omega)}|^{2} |Z_{sx,v}^{(2\omega)}|^{2} \sin^{2} \phi\), where \(I_{2} = N_{0}^{(2\omega)}|N_{z,v}^{(2\omega)}|^{2} \sin^{2} \phi\) is the radiative index of air at frequency \(2\omega\) and \(N_{z,v}^{(2\omega)}\) is the z component of the radiated field.
duced wave vector. These relations justify the representations in Fig. 5 of products such as \( N_{z,0}^{(2\omega)}Z_{\text{III},v}^{(2\omega)} \) rather than \( Z_{\text{III},v}^{(2\omega)} \) alone. Both the modulus and the phase of \( Z_{sx,v}^{(2\omega)} \) and \( Z_{py,v}^{(2\omega)} \) elements decrease continuously with the in-depth location of the interface. The situation is not so straightforward for the \( Z_{pz,v}^{(2\omega)} \) elements [i.e., related to the \( \mu_{z,v}^{(2\omega)} \) dipole term], for which all contributions from the first three interfaces give nearly the same modulus and phase. The \( Z_{pz,v}^{(2\omega)} \) element decreases more slowly with depth than do the \( Z_{sx,v}^{(2\omega)} \) and \( Z_{py,v}^{(2\omega)} \) elements.


**Fig. 4.** Polar representation of the incoming Fresnel elements \( X_{sx,v}^{(\omega)} \), \( X_{sy,v}^{(\omega)} \), and \( X_{zp,v}^{(\omega)} \) for each interface \( v \) for the \( \text{SiO}_2 \) (10-nm)/Fe\text{Si}(5-nm)/Dy\text{FeCo}(30-nm)/glass structure at a photon wavelength of 800 nm and for an incidence angle of 45°. The plotted quantity is \( N_{z,0}^{(2\omega)} \). Note that absolute values of \( X \) are identical to those of the electric fields that are present on each interface as represented in Fig. 3.

**Fig. 5.** Polar representation of the outgoing Fresnel elements \( Z_{sx,v}^{(2\omega)} \), \( Z_{sy,v}^{(2\omega)} \), and \( Z_{zp,v}^{(2\omega)} \) coming from each interface \( v \) for the \( \text{SiO}_2 \) (10-nm)/Fe\text{Si}(5-nm)/Dy\text{FeCo}(30-nm)/glass structure at a photon wavelength of 400 nm and for light radiated at an angle of 45°. The light intensity is proportional to \( |N_{z,0}^{(2\omega)}Z_{ij,v}^{(2\omega)}| \), i.e., the modulus of the quantity plotted along the \( x \) axis.
4. EXPERIMENTAL RESULTS AND DISCUSSION

The LMOKE hysteresis loop, measured in the low field with light penetrating from the top film side (Fig. 1), is essentially sensitive to the in-plane magnetic component of the Fe$_{96}$Si$_4$ layer. However, small and nearly reversible effects of a progressive rotation of Dy$_{30}$Fe$_{58}$Co$_{12}$ and Fe$_{96}$Si$_4$ magnetization toward the in-plane field direction can be superimposed. LMOKE hysteresis loops, measured at 45° of incidence, for Fe$_{96}$Si$_4$ (5- or 10-nm) films are shown in Figs. 6(a) and 7(a). The coercivity of the Fe$_{96}$Si$_4$ (5- or 10-nm) layers as measured with our MSHG setup is, respectively, $H_c = 19$ and $H_c = 25$ Oe. These values are reduced by a factor of ~2 as compared with those of coercivity measured by the LMOKE at room temperature under weak light. This effect is unambiguously due to local heating of the film by the rather intense laser beam spot. Independently knowing the variation of $H_c$ with temperature, we deduced that heating corresponds, in our case, to an elevation in temperature of 60 °C. This

![Graphs showing LMOKE and MSHG loops for Fe$_{96}$Si$_4$ (5- or 10-nm) films.](image)

Fig. 6. MOKE and MSHG loops of the Fe$_{96}$Si$_4$ (5-nm)/Dy$_{30}$Fe$_{58}$Co$_{12}$ structure measured in the same thermal conditions: (a) in the LMOKE, (b) in transverse pp MSHG, (c) in transverse sp MSHG, and (d) in longitudinal ps MSHG.

Fig. 7. MOKE and MSHG loops of the Fe$_{96}$Si$_4$ (10-nm)/Dy$_{30}$Fe$_{58}$Co$_{12}$ structure measured in the same thermal conditions: (a) in the LMOKE, (b) in transverse pp MSHG, and (c) in longitudinal ps MSHG.
result is expected because the film is deposited on glass, which is an insulating material with low thermal conductivity. So, as claimed, inside the laser spot area the local temperature is raised to ~80 °C, i.e., close to the compensation temperature of Dy$_{30}$Fe$_{58}$Co$_{12}$. Thus the Dy$_{30}$Fe$_{58}$Co$_{12}$ layer exhibits a huge out-of-plane coercivity (>20 kOe) at this temperature.

So the effective temperature of all MSHG measurements is ~80 °C. Transverse $pp$ and $sp$ MSHG and longitudinal $ps$ MSHG hysteresis loops are still measured for light at 45° incidence and for light penetrating the film structure from the top side (Fig. 1). Hysteresis loops for Fe$_{96}$Si$_4$ (5- or 10-nm) films are measured under a small in-plane magnetic field [Figs. 6(b)–6(d), 7(b), and 7(c)]. For the two samples, the hysteresis loop obtained in transverse $pp$ MSHG is much more nearly square and exhibits a lower coercivity than the LMOKE, transverse $sp$ MSHG, or longitudinal $ps$ MSHG. This is true whatever the direction of the magnetization with respect to an in-plane sample rotation. Moreover, LMOKE, longitudinal $ps$, and transverse $sp$ MSHG loop shapes are fairly different. This proves that all these effects have different in-depth sensitivities for probing the Fe$_{96}$Si$_4$ layer or interfaces.

Let us compare first the transverse $(\mathbf{H} \cdot \mathbf{x})$ $pp$ and $sp$ MSHG hysteresis loops for the Fe$_{96}$Si$_4$ (5-nm) film, measured at exactly the same place on the film surface [Figs. 6(b) and 6(c)]. The coercive field found in $pp$ configuration (10 Oe) is significantly smaller than that determined in $sp$ configuration (21 Oe); the latter value is close to that found in the LMOKE. Moreover, the $pp$ MSHG loop is much more nearly square. The difference in coercivity is explained as follows: $pp$ MSHG is presumably related more to spins located at the SiO$_2$/Fe$_{96}$Si$_4$ interface, which are less closely coupled to any harder in-plane spin direction in the Dy$_{30}$Fe$_{58}$Co$_{12}$ layer. Thus, during sweeping of the in-plane magnetic field, the spins begin to rotate at the top of the Fe$_{96}$Si$_4$ layer. This rotation generates a planar spiraling spin structure from the top to the inside of the Fe$_{96}$Si$_4$ layer. This expected magnetic behavior, coupled to our hysteresis loop observations, leads to the following points of experimental evidence:

(i) The transverse $pp$ MSHG is usually limited to probing the magnetization of the top SiO$_2$/Fe$_{96}$Si$_4$ interface.

(ii) The transverse $sp$ MSHG is sensitive to both the first SiO$_2$/Fe$_{96}$Si$_4$ and the second Fe$_{96}$Si$_4$/Dy$_{30}$Fe$_{58}$Co$_{12}$ interfaces and certainly probes a region that spreads more widely about the interfaces.

Point (i) can be explained by the selection rules (Table 1) and by the estimation of the strength of the electric field at interfaces in $pp$ configuration. The $p$-polarized field at fundamental frequency $\omega$ contains both $E_y^{(\omega)}$ and $E_z^{(\omega)}$ components. The selectivity of $pp$ MSHG to the top SiO$_2$/Fe$_{96}$Si$_4$ interface is associated with the large and abrupt jump of $E_z^{(\omega)}$ at this interface compared with smaller changes at other magnetic interfaces (Fig. 3). The transverse $pp$ MSHG signal (Table 1) is related to $\mu_{xy}^{(2\omega)} = \chi_{xyy}^{\parallel}[E_y^{(\omega)}]^2 + \chi_{xxz} E_x^{(\omega)} E_z^{(\omega)}$ and $\mu_{xz}^{(2\omega)} = \chi_{zxy} E_x^{(\omega)} E_z^{(\omega)}$. Hence the dominant term must be that related to $E_z^{(\omega)}$, so $\chi_{zxy}$ becomes negligible compared with $\chi_{xxz}$ and $\chi_{zzy}$. Moreover, from a comparison of $pp$ and $sp$ MSHG loops, as we show below, the contribution of $\chi_{zxy}$ has to be small compared to that of $\chi_{xxz}$. In that picture the transverse $pp$ MSHG signal comes mainly from the $\mu_{xy}^{(2\omega)} = \chi_{zxy} E_x^{(\omega)} E_z^{(\omega)}$ term. This conclusion is supported by the fact that the dipole radiation emitted by the $\mu_{xy}^{(2\omega)}$ component (and thus from $\chi_{zxy}$) is approximately three times more efficient than that which arises from $\mu_{xz}^{(2\omega)}$ (Fig. 3). Furthermore, as was pointed out by Petukhov and Liebsch, who extended the results of $ab$ initio calculations obtained for Al(111) to our case, $\mu_{xz}^{(2\omega)}$ must originate from a much thinner interface region (~0.1 nm) than the two other components, $\mu_{xy}^{(2\omega)}$ and $\mu_{zz}^{(2\omega)}$ (~1 nm). This result supports the highly selective nature of the transverse $pp$ MSHG to the upper SiO$_2$/Fe$_{96}$Si$_4$ interface, where there is a large contrast between diagonal permittivities. Similar results were indirectly found by Gudde et al. for ultrathin Co and Ni films deposited on Cu(001).

Point (ii) is consistent with the characteristic properties of the transverse $sp$ MSHG. The incoming $s$-polarized field is associated with the $E_x^{(\omega)}$ in-depth profile that keeps nearly the same value at both sides of the SiO$_2$/Fe$_{96}$Si$_4$ or the Fe$_{96}$Si$_4$/Dy$_{30}$Fe$_{58}$Co$_{12}$ interface (Fig. 3). The magnetic $sp$ MSHG signal is then related to $\mu_{yy}^{(2\omega)} = \chi_{yyx}^{\parallel}[E_y^{(\omega)}]^2$ (Table 1). The intensity of the light radiated by the $\mu_{yy}^{(2\omega)}$ dipole component is stronger by a factor 1.6 for the upper SiO$_2$/Fe$_{96}$Si$_4$ interface than for the second interface (Fig. 5). Thus, if one assumes that $\chi_{yyx}$ is similar for the two interfaces, the two contributions must be efficient. This result is even better supported by the fact that $\mu_{zz}^{(2\omega)}$ probes a thicker region than $\mu_{xy}^{(2\omega)}$.

As in transverse $sp$ MSHG, the longitudinal $ps$ MSHG hysteresis loop exhibits a rather large coercivity, which means again that the second Fe$_{96}$Si$_4$ interface contributes more. Hence, the dominant term should not contain in-depth selective $E_z^{(\omega)}$ terms. As the term responsible for the magnetic signal in longitudinal $ps$ MSHG is written as $\mu_{yy}^{(2\omega)} = \chi_{yyx}^{\parallel}[E_y^{(\omega)}]^2 + \chi_{zzy} E_x^{(\omega)} E_z^{(\omega)}$ (Table 1), the contribution of $\chi_{zzy} E_x^{(\omega)} E_z^{(\omega)}$ must be smaller than that which comes from $\chi_{yyx}^{\parallel}[E_y^{(\omega)}]^2$. This statement, associated with the symmetry relation, $\chi_{zzy} = -\chi_{yyx}$, was assumed in the discussion above of point (i), in which $\mu_{xy}^{(2\omega)} = \chi_{zxy} E_x^{(\omega)} E_z^{(\omega)}$ was the dominant term in the $pp$ configuration.

As was mentioned above, another MSHG contribution can come from antiferromagnetically aligned moments in Dy$_{30}$Fe$_{58}$Co$_{12}$. Related MSHG effects have been predicted theoretically but in our system they do not change sign with the reversal of the applied field. Considering the symmetry with respect to zero of recorded loops, we can exclude such a contribution.

Because of the nonuniform magnetization that exists inside the planar domain wall lying in the vicinity of the Fe$_{96}$Si$_4$/Dy$_{30}$Fe$_{58}$Co$_{12}$ interface, one can expect a MSHG contribution related to magnetization gradients. Considering the symmetry of MSHG hysteresis loops, the related effect must reverse with the applied field. Thus, when $\mathbf{H} \cdot \mathbf{x}$, the Fe$_{96}$Si$_4$ magnetization has components along the $x$ or $y$ axis with $\Phi$-dependent amplitudes. Nearly no gradient MSHG terms are expected to come
from the Fe\textsubscript{96}Si\textsubscript{4}/SiO\textsubscript{2} interface because of the flat variation of the magnetization orientation in its vicinity. In counterpart, a gradient MSHG term can be generated in the neighborhood of the Fe\textsubscript{96}Si\textsubscript{4}/Dy\textsubscript{30}Fe\textsubscript{58}Co\textsubscript{12} interface.

In that case, considering the selection rules for MSHG gradient terms for an isotropic medium that are given in Table 1 of Ref. 16, only the local magnetization gradient in $\delta M_x/\delta z$ is found to be active in pp and sp polarization. It can give rise to several nonlinear susceptibility terms: $X_{xyz}X_{xy}$, $X_{xyz}X_{xzy}$, and $X_{xy}X$ in pp polarization and $X_{xyz}$ in sp polarization. Again, as pp MSHG shows a rather square hysteresis loop and a low coercivity, we can exclude here any gradient MSHG coming from the Fe\textsubscript{96}Si\textsubscript{4}/Dy\textsubscript{30}Fe\textsubscript{58}Co\textsubscript{12} interface. The sp MSHG can be generated by a gradient term, but its magnitude relative to the usual MSHG term, in proportion to the magnetization, cannot be determined.

MOKE and MSHG data obtained for Fe\textsubscript{96}Si\textsubscript{4} 10- or 5-nm film show rather similar magnetic behavior. This confirms all the above interpretations and proves that the optical phase changes introduced by different Fe\textsubscript{96}Si\textsubscript{4} film thicknesses in the nanometer range do not have a large effect on the main features of the MSHG hysteresis loop.

5. CONCLUSIONS

We have presented evidence of highly selective pp MSHG of SiO\textsubscript{2}/Fe\textsubscript{96}Si\textsubscript{4} and Fe\textsubscript{96}Si\textsubscript{4}/Dy\textsubscript{30}Fe\textsubscript{58}Co\textsubscript{12} exchange coupled bilayer structures and of transverse sp MSHG and longitudinal ps MSHG probing of the two Fe\textsubscript{96}Si\textsubscript{4} interfaces with a greater selectivity. This explains why the two last hysteresis loops are comparable to those obtained in the LMOKE, which is known to probe in-depth magnetization more nearly uniformly. However, as expected, the loop becomes increasingly more rounded when one goes from transverse pp to transverse sp longitudinal ps MSHG and LMOKE. One can interpret this result qualitatively by saying that sensitivity and selectivity to interfaces becomes correspondingly less pertinent.

As we know, the MSHG signal comes from the interference between nonlinear nonmagnetic and magnetic susceptibilities. In our case, when magnetization varies with depth in the structure essentially in the vicinity of the Fe\textsubscript{96}Si\textsubscript{4}/Dy\textsubscript{30}Fe\textsubscript{58}Co\textsubscript{12} interface, we have demonstrated that only breaking of the nonmagnetic symmetry will reveal large MSHG signals. The most efficient factor that controls the MSHG in-depth selectivity is the step-like profile of the $z$ component of the electric field that becomes large at metal–insulator interfaces owing to the abrupt change of the diagonal permittivity tensor elements. Thus one can say that the magnitude of MSHG is related to the permittivity gradient at interfaces over the effective depth probed by second-harmonic generation. The most efficient terms that are responsible for a MSHG signal have been determined in each normal optical configuration. From hysteresis loop investigations it is unfortunately not possible to determine the magnitude of MSHG gradient terms related to the planar wall.

Finally, our results confirm again, as for the CoO/NiFe/NiO/Co/Cu structure,\textsuperscript{5} that the $\mu_2^{(2\omega)}$ dipole component is emitted from a thinner region at interfaces than for $\mu_2^{(2\omega)}$ and $\mu_1^{(2\omega)}$. For this structure we have already proved that pp MSHG is more selective at interfaces but in this case is generated from the third buried interface and not from the first one as for Fe\textsubscript{96}Si\textsubscript{4}/Dy\textsubscript{30}Fe\textsubscript{58}Co\textsubscript{12}. But, again, the metal–dielectric interface has to be considered the most efficient radiation source in the pp configuration. As MSHG should be of great interest for testing tunnel junction interfaces in giant magnetoresistive devices, more experiments in simple multilayer structures need to be performed to confirm simple predictions of selectivity.

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