Onset-Time Control of THz Transients Generated by Spintronic Emitters

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***Abstract*—We have generated intense electromagnetic transients by femtosecond laser pulse illumination of ferromagnet/metal (F/M) nanobilayers, in the presence of an external magnetic field. Fourier analysis revealed that the frequency content of these transients extended up to ~5 THz. We have also observed that upon the increase of the magnetic field, the entire THz transient shifts towards earlier times by up to 110 fs. We ascribe this magnetically tunable onset-time shift to extra acceleration of photoelectrons induced due to the Lorenz force.**

I. INTRODUCTION

Optical pulses impinging on a ferromagnet can generate superdiffusive spin currents with spin current density $\boldsymbol{J_S}$ in the direction of light propagation [1]. In ultrathin ferromagnet/metal bilayers, $\boldsymbol{J_S}$ can carry a spin *across* the interface from the ferromagnet (F) into the metal (M), where spin–orbit interaction leads to the generation of a transient, transverse charge current density $\boldsymbol{J}_C \sim \boldsymbol{J}_S \times \boldsymbol{\sigma}$, what is known as the inverse spin Hall effect (ISHE) ($\boldsymbol{\sigma}$ is spin polarization) [2]. The transient $\boldsymbol{J}_C$ propagates along the surface of the metallic film, broadcasting an electromagnetic transient into the free space. If the optical excitation is in a form of a femtosecond pulse, the generated transient has a spectral content extending well into a THz region. Earlier we have shown that the THz transient amplitude, as well as its polarization direction can be tuned by a magnetic field [3]. Here we discuss the magnetic-field tunability of the THz pulse onset-time in the femtosecond time range.

II. RESULTS

Figure 1 shows THz transients generated at a $Ni_{80}Fe_{20}$/Pt nanobilayer under the same optical excitation, but at two different magnetic field intensities, H_1 ~ 8 kA/m and H_2 ~ 72 kA/m, red and black trace, respectively. The magnetic field is applied in the plane of the sample. We note that the black THz transient in Fig. 1 is clearly shifted towards earlier onset time with respect to the red one, and we confirmed that the FFT spectra of the transients recorded at different magnetic field were identical. This shift is magnetically tunable (the maximal value ~110 fs) and occurs for all magnetic materials tested. The observation indicates that besides the ISHE, an additional physical process, or processes, contribute to the THz transient generation. It is safe to assume that moving electrons, triggered by the laser pulse, experience an additional force affecting their trajectory. Here, we consider a physical mechanism affecting the spin propagation in a F/M bilayer, namely the *Lorentz force* acting on the optically triggered electrons. Moving charges in a magnetic field experience the Lorenz force ($\boldsymbol{F}_{LF}$). If we consider that the $\boldsymbol{F}_{LF}$ can act on the spin current $\boldsymbol{J}_S$, we can write $\boldsymbol{F}_{LF} \sim \boldsymbol{J}_S \times \boldsymbol{B}$, where $\boldsymbol{B} = \boldsymbol{M} + \boldsymbol{H}$ is the sum of the sample magnetization $\boldsymbol{M}$ and the external field intensity $\boldsymbol{H}$. In this case, $\boldsymbol{F}_{LF}$ induces an extra acceleration of $\boldsymbol{J}_C$ in the direction of $\boldsymbol{J}_C$, and for high $\boldsymbol{B}$ leads to an onset in the THz transient arrival time.

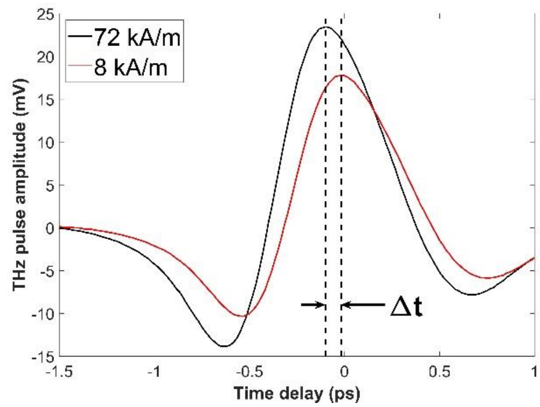

Fig. 1 THz transients generated by laser illumination of a $Ni_{80}Fe_{20}$/Pt bilayer using a train of 100 fs laser pulses, recorded at two external magnetic field intensities. The THz transient recorded at a higher magnetic field (black trace) shows a clear time shift by Δt ~ 110 fs with respect to the transient recorded at low field (red trace). Both transients were measured at a fixed detector position and the same intensity of optical excitation.

Table I. List of tested samples

Sample name	Layers sequence and thicknesses	Coercive field H_C (kA/m)	Magnetic type	Remnant magnetization (emu/cm^3)
Py/Pt	MgO/Ni80Fe20 (2 nm)/Pt (2 nm)	~0.8	Soft	2
Cr/CoPd/Pd	MgO/Cr (1 nm)/$Co_{35}Pd_{65}$ (35 nm)/Pd (2 nm)	~33	Intermediate	59
FeCo/Ir	MgO/$Fe_{60}Co_{40}$ (3 nm)/Ir (3 nm)	~10.5	Hard	430

To confirm our assumption, three different nanobilayer samples, magnetic soft (Py/Pt), intermediate (Cr/CoPd/Pd), and hard (FeCo/Ir) samples, as shown in Table I, were investigated. The Cr layer for Cr/CoPd/Pd sample was acting as an adhesive layer. We measured the magnetization $\mu(\boldsymbol{H})$ hysteresis behavior using a Physical Properties Measurement System (PPMS). The values of the coercive field and the remnant magnetization for all samples are compiled in the Table I. As expected, the measured THz maximum amplitude dependence of external magnetic field $\boldsymbol{H}$ is in perfect agrees with the magnetization hysteresis curve measured from PPMS [3], [4].

The time shift has been measured for all three samples, as shown in Figure 2. The value of Δt is negative, which means that regardless of the direction of magnetization to north or south, the THz transient is always shifts to an earlier time at higher external fields. Moreover, the Δt dependences on $\boldsymbol{B}$ for all three samples are linear, and they are identical, except for the saturation of Δt around 400 mT for the FeCo/Ir sample. The direction-independent, linear, and identical dependency of all samples shown in Figure 2 agrees very well with the Lorentz force assumption.

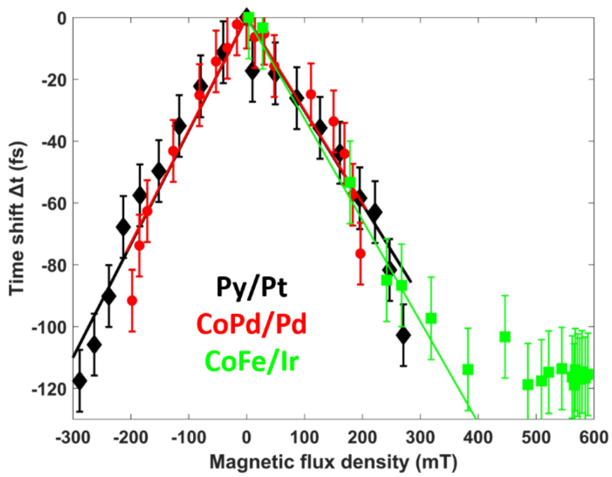

Fig. 2 Time shift Δt dependence on the magnetic flux density $\boldsymbol{B}$ for Py/Pt (black dots), CoPd/Pd (red dots) and CoFe/Ir (green dots) nanobilayers. The error bars correspond to the time delay due to a single step of the delay stage. The solid lines are linear fits of the measurement data.

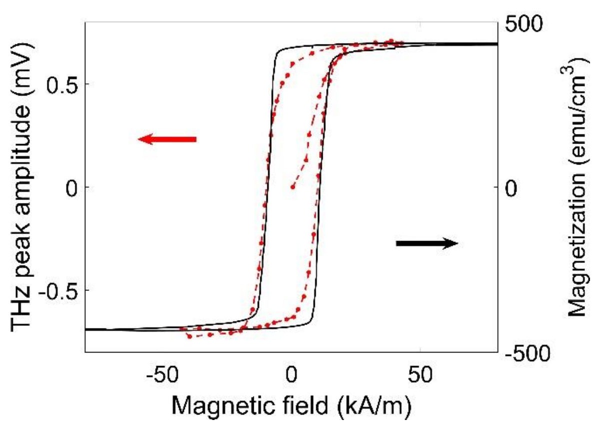

Fig. 3 Magnetic hysteresis loop $\mu(\boldsymbol{H})$ (black slid lines) of a FeCo/Ir bilayer and magnetic field dependence of the THz peak amplitude starting from the demagnetized state (red symbols). By recording the THz transient from a demagnetized FeCo/Ir bilayer, the initial virgin magnetization hysteresis curve can be extracted.

Whereas the time shift feature is easily found for the magnetic soft (NiFe/Pt) and intermediate (CoPd/Pd) nanobilayers, it is not observable for the magnetically hard FeCo/Ir nanobilayer, unless the measurements start from the demagnetized state ($\boldsymbol{M} = 0$). The PPMS magnetic hysteresis data (black solid lines) in the Figure 3 show for a magnetized FeCo/Ir nanobilayer that the magnetization of the sample switches sharply between saturation in south or north direction. Therefore, the magnitude of the magnetization component along the field axis remained unchanged during the measurements, and it was much bigger (430 emu/cm^3 corresponding to 430 kA/m or 0.54 T) than the value of external magnetic field $\boldsymbol{H}$, so that the sum $\boldsymbol{B} = \boldsymbol{H} + \boldsymbol{M}$ only changed slightly during the field sweep measurements. Remember that regardless of the direction of the magnetic flux density, north or south, the THz transient always shifts to an earlier time. In fact, from a magnetized FeCo/Ir nanobilayer, we recorded a shifted THz transient at any external magnetic field. However, we succeeded to observe the time shift feature from THz transients recorded for the initial virgin hysteresis curve starting from an unmagnetized FeCo/Ir sample as shown by the red curve in Figure 3. The time shifts of the transients on the virgin curve are shown in Figure 2 by green symbols.

This data provides further strong evidence that the time shift feature is due to a Lorentz force-induced extra acceleration of the photoelectrons, which is determined by both the external magnetic field and the magnetization of the ferromagnetic material. Bigger magnetic flux density will create a stronger Lorentz force that drives electrons harder and results in THz transients reaching the detector earlier.

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