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Ultrafast Magnetic Switching of GdFeCo with Electronic Heat Currents

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Abstract

We report the magnetic response of Au/GdFeCo bilayers to optical irradiation of the Au surface. For bilayers with Au thickness greater than 50 nm, the great majority of energy is absorbed by the Au electrons, creating an initial temperature differential of thousands of Kelvin between the Au and GdFeCo layers. The resulting electronic heat currents between the Au and GdFeCo layers last for several picoseconds with energy flux in excess of 2 TW m^{-2}, and provide sufficient heating to the GdFeCo electrons to induce deterministic reversal of the magnetic moment.

Introduction

Ultrafast reversal of the magnetic moment can be optically induced in metals that possess two antiferromagnetically coupled sublattices, e.g. Gd and FeCo [1]. This phenomena, commonly referred to as all optical switching (AOS), was first observed by Stanciu *et al.* in 2007 [2]. In all prior AOS experiments, GdFeCo electrons are optically excited with an ultrafast laser pulse to eV energies above the Fermi level [3]. Subsequently, the FeCo sublattice demagnetizes within a few hundred femtoseconds [4]. The Gd sublattice also loses magnetic order, but at a slower rate [4]. The differing rates of demagnetization, together with the transfer of angular momentum from the Gd to FeCo sublattice, enables reversal of the magnetic moment on ps time-scales [1,3,5]. While initial studies credited the ultrafast reversal of the magnetization to a helicity-dependent light-matter interaction [2], subsequent investigations with linearly polarized lasers show the reversal is driven solely by energy absorption [1,6].

While the phenomenology of AOS is well documented [3,7-9], many of the basic mechanisms underpinning ultrafast demagnetization and AOS remain poorly understood. For example, the role of thermal vs. nonthermal electrons in ultrafast demagnetization and subsequent magnetization reversal remains controversial [10-17]. In the first hundred femtoseconds following laser irradiation, electrons are non-thermal, i.e. Fermi-Dirac statistics provides a poor description of the excitation energies [18]. Highly excited nonthermal electrons could allow for magnetization quenching via the generation of Stoner excitations [16,18]. Nonthermal distributions can also enable nonlocal superdiffusive transport of energy and

angular momentum [11,12,14]. Superdiffusive spin currents are posited to be important in ultrafast demagnetization of ferromagnetic metals [14] and could also play an important role in AOS [10,11]. Potentially supporting the hypothesis that superdiffusive transport assists AOS are ultrafast diffraction measurements of large nonlocal spin currents flowing between Gd and Fe rich regions of a compositionally inhomogeneous GdFeCo film in the ps following laser irradiation [10]. Other models for AOS completely ignore the initially non-thermal electron distribution. Atomistic simulations based on the Landau Lifshitz Gilbert equation predict ultrafast switching of ferrimagnets by coupling spin degrees of freedom to a thermalized distribution of hot electrons with a Langevin random field term added to the dynamic equation [1,19]. Models based on Boltzmann rate equations that describe AOS via scattering between electronic and spin degrees-of-freedom also neglect the initially nonthermal electron distribution [20,21].

In this letter, we demonstrate direct laser irradiation of GdFeCo is not necessary for deterministic reversal of the magnetization. Purely electronic heat currents are also effective at switching. We report the magnetic response of Au/GdFeCo bilayers to optical irradiation of the Au surface (Fig. 1). By varying the Au thickness from 5 to 91 nm, we control the ratio of laser energy absorbed by the GdFeCo to that absorbed by the Au. The total absorbed fluence required for switching increases by only a factor of three when the Au thickness is increased from 5 to 91 nm, despite a factor of twenty decrease in the amount of energy directly absorbed by the GdFeCo (Fig 2). Our results suggest thermal currents can reverse the magnetization more efficiently than is possible by direct optical heating.

Results

We focus our study on six Au/GdFeCo bilayer samples prepared via magnetron sputter deposition on sapphire substrates. The Au film thicknesses for the six samples are 5, 10, 37, 58, 73 and 91 nm. The GdFeCo film thickness is ~11.5 nm in all six samples. Further details regarding the growth and sample characterization are in Ref. [22].

We use an amplified Ti:sapphire laser with 810 nm center wavelength in our experiments (Coherent RegA 9050). The laser pulse duration full-width at half maximum (FWHM) is tunable from 55 fs to 25 ps by adjusting the final pulse compressor in the chirped pulse amplifier [9]. We run the laser amplifier at a repetition rate of 250 kHz for time-resolved pump/probe measurements, or instead eject single laser pulses. In the majority of experiments, the pump laser is incident on the Au surface of the bilayer, while the probe laser is incident on the GdFeCo film through the sapphire substrate.

We use a magneto-optic Kerr effect (MOKE) microscope for monitoring the GdFeCo magnetization after laser irradiation (Fig. 1b). The MOKE microscope focuses on the GdFeCo film through the sapphire substrate. In these experiments, an external magnetic field $H \approx \pm 100$ Oe saturates the magnetization of the sample out-of-plane. Following removal of the external field, a single linearly polarized laser pulse irradiates the Au surface. As shown in Fig. 1 for the Au (58 nm) / GdFeCo bilayer, if a laser pulse of sufficient energy irradiates the Au film surface, the magnetization in a small region reliably toggles between white (up) and black (down).

The goal of our study is to examine systematically how heating GdFeCo electrons via thermal currents instead of optical absorption influences energy requirements for magnetization reversal. For the Au/GdFeCo bilayers with Au layers thicker than 50 nm, electronic heat currents flowing from the adjacent Au layer deposit the majority of heat in the GdFeCo layer. In Fig. 2, we report the total fluence, F_T , the bilayer must absorb to induce magnetization reversal of the GdFeCo. Irradiation of the Au surface with sufficient fluence results in reversal of the GdFeCo magnetization in all samples, despite the GdFeCo directly absorbing a negligible amount of laser energy in the bilayers with thick Au films. As a control experiment, we also perform measurements on a Pt (5 nm)/Au(75 nm)/MgO(3 nm)/Au(5 nm)/GdFeCo(10 nm) sample. For this sample, the 5 nm of Pt absorbs most of the optical pulse, and the insulating MgO layer prevents any electronic currents from reaching the GdFeCo layer. No magnetization switching is observed in this sample at any fluence.

To interpret our experimental data, we use a thermal model to predict the temperature responses of the electrons and phonons in the bilayers (see Fig. 1). Details of the thermal model are in Ref. [22]. Following optical heating of the Au electrons, the Au electrons reach a temperature in excess of 2000 K in bilayers where the majority of energy is absorbed in the Au (Fig. 1a). The high diffusivity of the Au electrons allows rapid diffusion of heat [23], resulting in TW m^{-2} heat currents into the GdFeCo layer (Fig. 3a).

The Au/GdFeCo bilayers with thicker Au layers require more absorbed fluence to induce magnetization reversal (Fig. 2). The increase in F_T with

increasing Au thickness is due to only a fraction of the energy absorbed by the Au reaching the GdFeCo electrons via diffusion. In parallel to energy transfer from the hot Au electrons to the GdFeCo electrons, significant energy is transferred to the Au phonons via electron-phonon scattering [23]. The characteristic length-scale over which the electronic heat can diffuse before the hot Au electrons transfer most of their energy to the phonons is $d_{ep} \approx \sqrt{\Lambda_{e,Au} / g_{ep,Au}} \approx 100 \text{ nm}$, where $\Lambda_{e,Au} \approx 250 \text{ W m}^{-1} \text{ K}^{-1}$ is the thermal conductivity of the Au electrons and $g_{ep,Au} \approx 2.8 \times 10^{16} \text{ W m}^{-3} \text{ K}^{-1}$ is the electron-phonon coupling constant of Au [24].

Figure 3 shows our thermal model's estimate for the total fluence transferred between the Au film and the GdFeCo layer for each experiment. Figure 3a shows the optical and electronic heat currents into the GdFeCo for the Au (38 nm)/GdFeCo and Au (91 nm) / GdFeCo bilayers. Integrating both the optical and electronic heat-currents over the time interval of the experiment yields the total fluence absorbed by the GdFeCo electrons, F_{GFC} , as a function of Au thickness (Fig. 3b). We observe that a total fluence between 5 and 6 J m^{-2} must be absorbed by the GdFeCo from electronic and/or optical heat currents for magnetization reversal to occur in all samples.

The weak dependence of F_{GFC} on the Au thickness is surprising for two reasons. First, the time-scale for energy delivery varies by nearly an order of magnitude for optical vs. electronic heat current (Fig. 3a). Several prior studies have observed that for some alloy compositions, longer pulse durations can cause an increase in the absorbed fluence necessary for magnetization reversal [8,9,25].

Second, the optical absorption of laser energy excites a nonthermal distribution of electrons, while heating of the GdFeCo electrons via electronic heat currents from the Au layer will excite a thermal distribution. The average initial energy per optically excited electron is roughly half the optical photon energy, or ~800 meV. Alternatively, for 6 J m^{-2} distributed across the excitation energies predicted by Fermi-Dirac statistics at 2000 K, the average energy per excitation is ~ 200 meV. If nonthermal electron distributions affect magnetization dynamics [11,16], F_{GFC} should depend on whether the initially excited distribution is thermal or nonthermal.

We credit the weak dependence of F_{GFC} on Au thickness (Fig. 3b) to the GdFeCo magnetization dynamics being sensitive to *both* heating duration and whether a thermal vs. nonthermal distribution of electrons is initially excited in the GdFeCo. Picosecond heating is less efficient at inducing magnetization reversal than femtosecond heating. However, electronic heat currents are more effective at inducing magnetization reversal than direct optical absorption. To show that F_{GFC} is sensitive to both the duration and thermal/non-thermal character of the heating, we first examine the dependence of F_{GFC} on pulse duration in experiments where the GdFeCo is heated nonthermally via optical absorption. Then we compare these results to the results in Fig. 3b for the thickest Au/GdFeCo bilayers, where heating of GdFeCo is primarily due to electronic heat currents.

We measure the dependence of F_T on laser pulse duration in three experiments where GdFeCo is heated nonthermally via direct optical absorption. Figure 4 reports F_T for pulse durations between 55 fs and 0.7 ps in the Au (5 nm) / GdFeCo and Au (10 nm) / GdFeCo samples. Additionally, Fig. 4 reports the

dependence of F_T on laser pulse duration of the Au (58 nm) / GdFeCo sample, but with the pump laser irradiating the GdFeCo surface instead of the Au surface. In all nonthermal heating experiments, we do not observe magnetization reversal at any laser fluence for pulses longer than 0.7 ps.

Figure 4 also includes values from Fig. 3b for F_{GFC} of the Au (73 nm) / GdFeCo and Au (91 nm) / GdFeCo bilayers. These two thick Au samples were irradiated from the Au side, therefore F_{GFC} is primarily from electronic heat currents with duration of 1 and 3 ps FWHM. In contrast to our experiments where the GdFeCo is optically heated, heat currents from an adjacent Au layer reverse the magnetization despite ps durations and lower peak currents (Fig. 3a). Furthermore, the optical fluence needed to switch the Au (5 nm) / GdFeCo bilayer with a 0.5 ps pulse is ~ 9 J m^{-2}. Absorption of only 5 J m^{-2} by the GdFeCo is needed to cause magnetization reversal in the Au (91 nm) / GdFeCo sample (Fig 3b and 4).

In our analysis above, we assume that exciting the GdFeCo electrons via heat-currents across the Au/GdFeCo interface will generate a thermal distribution. Prior to heating the GdFeCo electrons, energy must travel across the Au film. In the time it takes the heat to travel from the Au surface, the electron-electron and electron-phonon scattering in the Au will move the initially nonequilibrium distribution towards a thermal distribution. The time-scale for energy to diffuse across a 73 nm thick Au film via hot electrons is $\tau \approx 0.6$ ps. Alternatively, the time-scale for energy to ballistically travel across the film is given by $\tau \approx h_{Au}/v_F \approx 50$ fs, where $v_F \approx 1.4\times10^6$ m/s is the Fermi velocity of Au.

To test the time-scale for energy transport across the Au film, we prepared three new samples for time-resolved pump/probe measurements. Pump/probe measurements of the Au/GdFeCo bilayers were not possible because the low optical absorption of Au requires a high incident laser fluence to induce magnetization dynamics. The high incident fluence causes sample damage when we operate the laser at a repetition rate of 250 kHz instead of the single shot mode we use in the experiments described above. The geometries of the three new samples are Au (10nm) /MgO (170 nm) /Ta (2nm)/ GdFeCo (10 nm) / Au (h_{Au})/ Pt (5 nm), with h_{Au} = 0, 30 and 75 nm. The addition of Pt to the stack lowers the incident fluence necessary to induce magnetization dynamics by a factor of five, and eliminates the sample damage problem. The addition of the Au/MgO bilayer adjacent to the GdFeCo enhances the MOKE contrast [Qureshi et al., APL 85, 3 (2004)]. Figure. 5 shows pump/probe measurements on these three samples. The insertion of a 30 and 75 nm thick Au layer between the Pt and GdFeCo causes a delay of ~0.1 and ~0.7 ps in the demagnetization of the GdFeCo layer. These sub-picosecond time-scales are longer than the 20 and 50 fs time-scales we expect for ballistic travel. The consistency of zero delay time between samples is better than 30 fs, and is primarily determined by the 1.6 μm depth of focus of the objective lens. Therefore, our pump/probe measurements provide strong support for our thermal modelling.

In conclusion, by adding a Au layer adjacent to GdFeCo to serve as an optical absorber, we examine how exciting GdFeCo with electronic thermal currents differs from direct optical excitation. Prior theories have focused on the

role nonthermal distributions may play in enhancing ultrafast magnetization dynamics [11,13,14]. Here, we observe that nonthermal excitation of GdFeCo electrons on picosecond time-scales is actually less efficient than excitation via thermal currents at causing magnetization reversal of GdFeCo in Au/GdFeCo bilayers. The discovery that purely thermal currents are effective in magnetization reversal of GdFeCo signals new opportunities for potential device applications of ultrafast magnetization switching.

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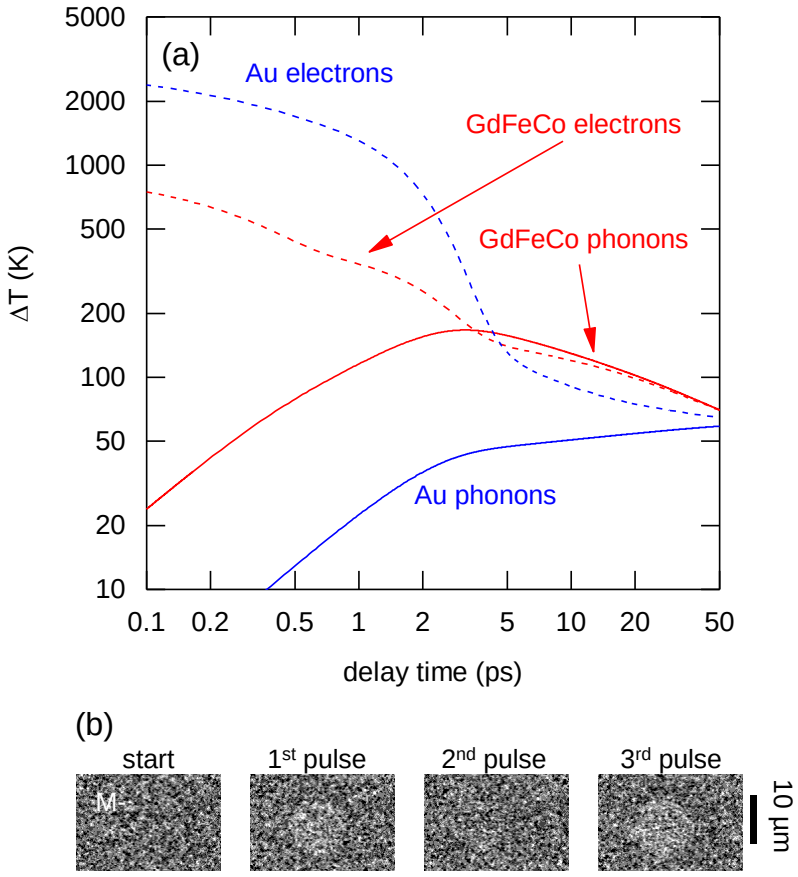

Fig. 1. (a) Temperature response of a Au (57 nm) / GdFeCo bilayer after the Au electrons absorb 11 J m^{-2} from a 55 fs laser. Each curve represents an average temperature across the layer. The large temperature difference between the Au electrons and GdFeCo electrons for the first few picoseconds following irradiation generates electronic heat currents in excess of 2 TW m^{-2}. The large electronic heat currents are sufficient to reverse the GdFeCo magnetization. (b) MOKE micrographs of the GdFeCo magnetization in a 91 nm Au / 11.5 nm GdFeCo bilayer after the Au surface is successively irradiated with linearly polarized laser pulses. The sample's initial magnetization is down (M−).

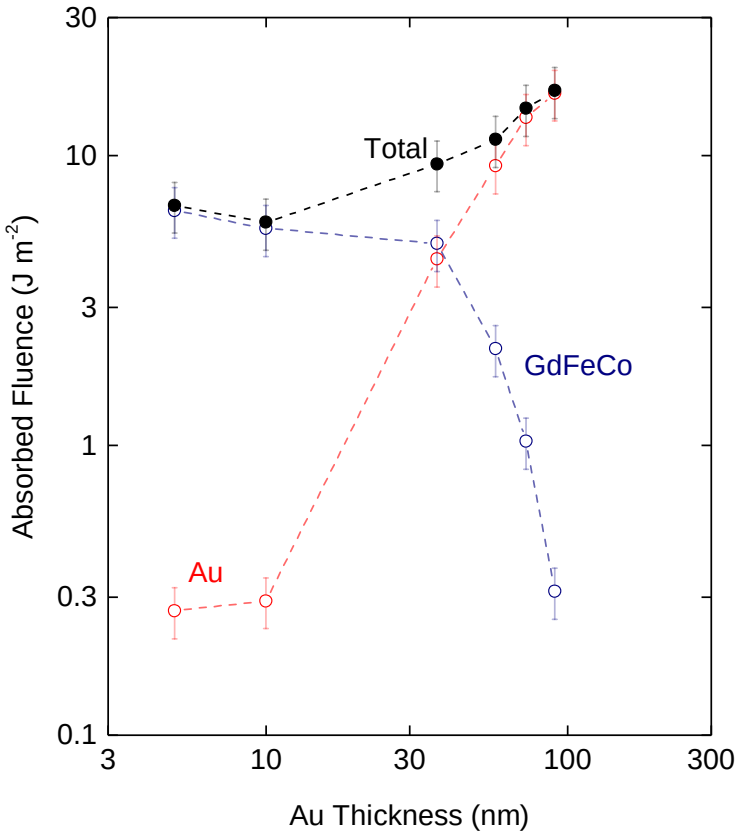

Fig. 2. Dependence of the absorbed laser fluence, F_T, needed to reverse the magnetization of the GdFeCo as function of the thickness of the Au capping layer. Lines are to guide the eye. A multilayer reflectivity calculation with n = 0.2 + 4.9*i* for Au and n = 3.2 + 3.5*i* for GdFeCo determines the amount of fluence absorbed in the GdFeCo and Au layer at each Au thickness.

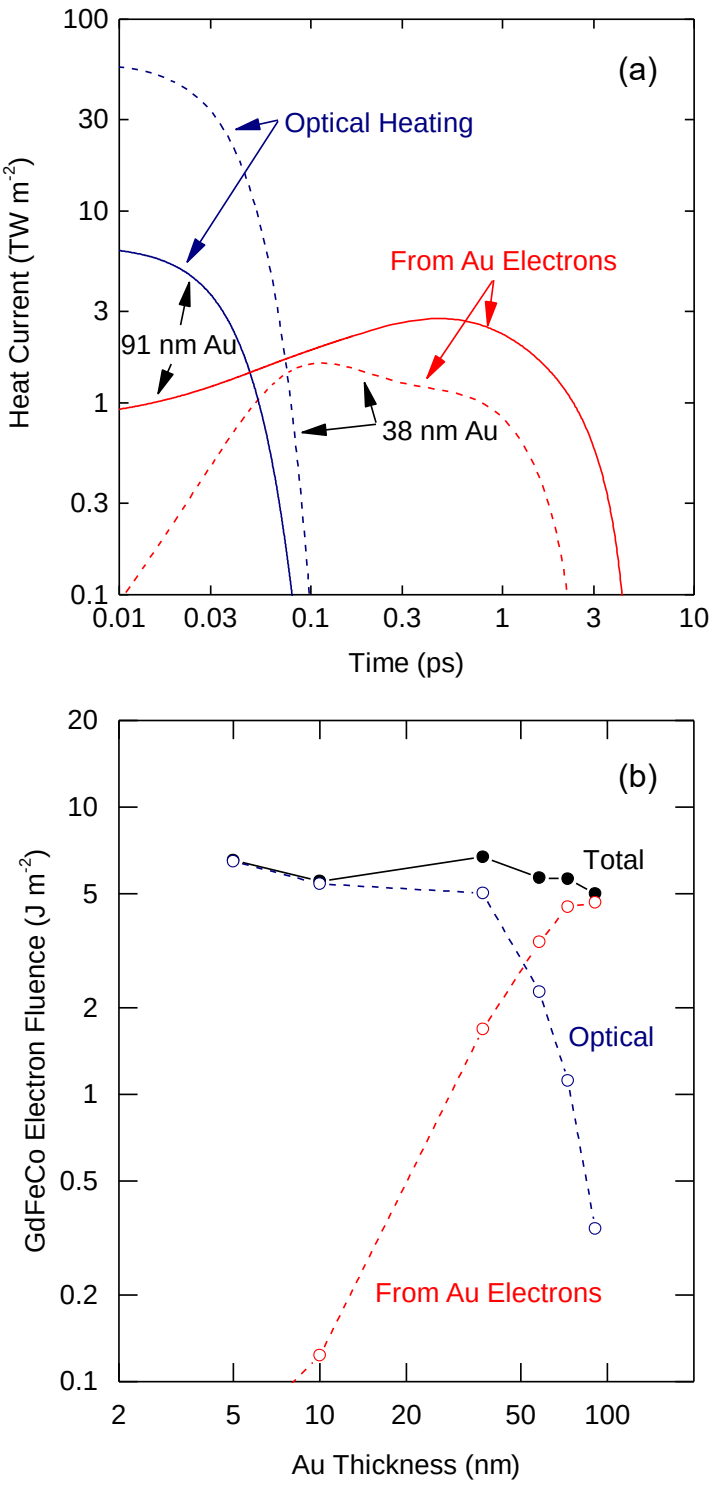

Figure 3. (a) Heat currents into the GdFeCo electrons from both direct optical absorption and from the adjacent Au film. Solid and dashed lines are calculations for the Au (38 nm) and Au (91 nm) bilayers with an absorbed fluence of 9.4 and 17

J m^{-2}, respectively. Red curves represent electronic heat-currents from the adjacent Au layer, while blue curves are heat-currents from direct optical absorption of the laser energy by the GdFeCo electrons. (b) Fluence absorbed by GdFeCo electrons vs. Au film thickness. The red open circles demark the fluence from heat currents via the Au electrons, the blue open circles represent the fluence from direct optical absorption, and the filled black circles represent the total fluence absorbed by the GdFeCo electrons from all sources. Lines are to guide the eye.

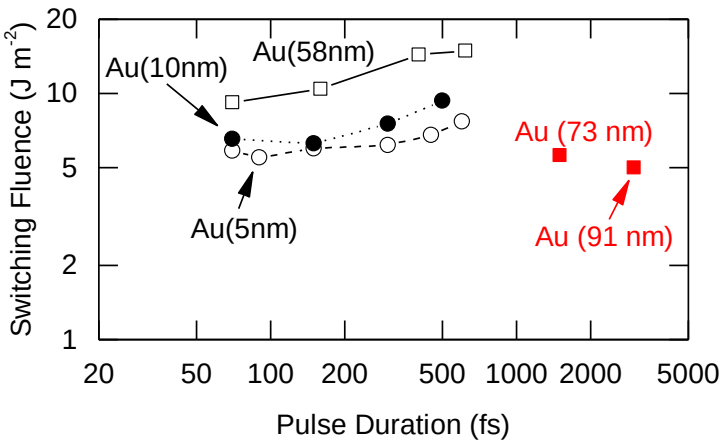

Fig. 4. Switching current vs. pulse duration. Black open and closed circles represent the minimum absorbed fluence needed to switch the Au (5 nm) / GdFeCo and Au (10 nm) / GdFeCo samples, respectively. Open black squares represent the fluence needed to switch the Au (58 nm)/GdFeCo sample with the laser incident on the GdFeCo. Included for comparison are values from Fig. 3b for F_{GFC} of the Au (73 nm) /GdFeCo and Au (91 nm)/GdFeCo samples (red squares). The pulse duration values for the black markers are the FWHM of the laser pulse irradiating the sample. Alternatively, the pulse duration value for the red squares are the thermal model predictions of the FWHM of the heat-current across the Au/GdFeCo interface.

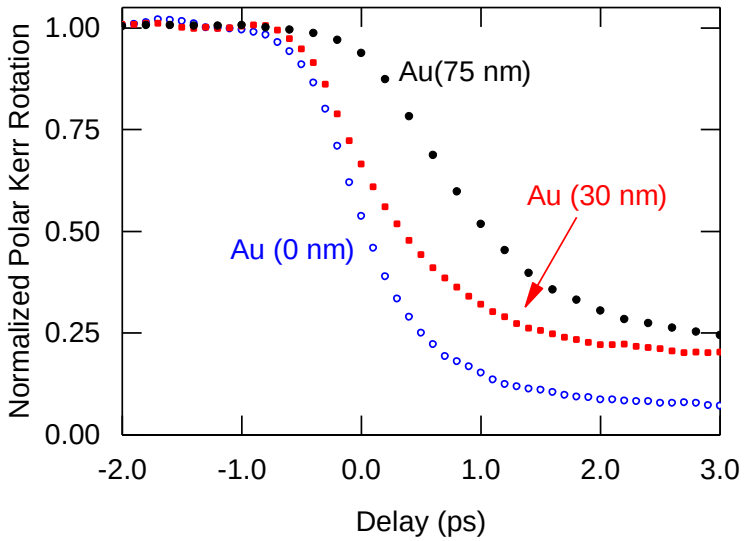

Fig. 5. Pump probe data of Pt / Au / GdFeCo samples. The pump laser is incident on the Pt. The 0.1 and 0.7 ps delay in demagnetization of the samples with 30 nm Au and 75 nm Au between the Pt absorber and GdFeCo layer is consistent with diffusive heat transfer by hot Au electrons.

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