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Thermal conductance of interfaces with amorphous SiO₂ measured by time-resolved magneto-optic Kerr-effect thermometry

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Abstract

We use time-resolved magneto-optic Kerr effect (TR-MOKE) and ultrathin Co/Pt transducer films to perform thermal transport experiments with higher sensitivity and greater time resolution than typically available in studies of interfacial thermal transport by time-domain thermoreflectance (TDTR). We measure the interface conductance between Pt and amorphous SiO₂ using Pt/Co/Pt ferromagnetic transducer films with thicknesses between 4.2 nm and 8.2 nm and find an average value of $G_{\rm Pt} \approx 0.3$ GW m⁻² K⁻¹. This result demonstrates that interfaces between metals and amorphous dielectrics can have a conductance corresponding to Kapitza lengths of the order of 4 nm, and are thus of relevance when engineering nanoscale devices. For thin SiO₂ layers our method also provides sensitivity to the interface conductance between SiO₂ and Si and we find $G_{\rm Si} \geq 0.6$ GW m⁻² K⁻¹ as the lower limit.

PACS numbers:

I. INTRODUCTION

Thermal transport across interfaces plays a key role in the thermal management and engineering of nanocomposites, nanoelectronics, and the next generation of magnetic data storage^{1,2}. However, the challenges associated with measuring heat flows and temperature have hampered the field of thermal science at the nanoscale from achieving the same level of understanding and control as exists in electronics and photonics^{3,4}.

The interface thermal conductance G is a linear transport coefficient that describes the heat flux J crossing an interface for a given temperature difference ΔT between the distributions of heat carriers incident on the two sides of the interface, $J = G\Delta T^5$. G has been studied for interfaces between metals and dielectric or metallic crystals and for metal-liquid interfaces^{3,6}. For interfaces between crystals, the value of G is typically discussed in the context of scattering of well-defined phononic or electronic modes at the interface governed by interfacial bonding, disorder, and differences in the elastic constants or densities of states⁷. For interfaces with liquids, G is usually explained by molecular interactions at the interface and the overlap of densities of states.

In amorphous materials, only phonons near the zone center have well-defined wavevectors and the majority of the vibrational states that are important for heat transport lack coherence. The conduction of heat in an amorphous material can be described by a random walk of vibrational energy^{8,9}. This fundamental difference in the heat carriers in amorphous and crystalline materials raises the question if conventional models for the interfacial thermal conductance can be applied reliably to boundaries with amorphous materials. Due to limited experimental accessibility, thermal transport at a boundary between a metal and an amorphous dielectric has not received much attention.

Well-established methods for studying thermal properties of interfaces are time-domain thermoreflectance $(TDTR)^{3,10}$ and frequency-domain thermoreflectance $(FDTR)^{11,12}$. They are pump-probe techniques that measure the temperature-response of a metal transducer to pulsed laser heating. Changes in the intensity of reflected laser probe pulses contain information about the thermal properties of the sample underneath the transducer. The sensitivity of a TDTR measurement to the interface conductance depends on the thermal mass (product of thickness h and volumetric heat capacity C) of the transducer. To separate the conductance G of an interface from the bulk thermal conductivity Λ of a sample underneath the transducer, h should not be large compared to the Kapitza length $L_K = \Lambda/G$. For SiO₂ and a typical conductance value of G = 150 MW m⁻² K⁻¹, this means a transducer should not be significantly thicker than 10 nm. However, TDTR and FDTR require optically opaque transducers to avoid spurious thermoreflectance signals generated by the temperature field and changes in optical constants in the sample under the transducer¹³. In most cases, the requirement of an optically opaque metal film transducer limits the transducer thickness to h > 50 nm and the sensitivity of TDTR or FDTR to the conductance Gof interfaces with low thermal conductivity materials is typically small, on the order of $\frac{\Lambda}{hG}$.

Nevertheless, there are a few prior reports of conductance values for metal/SiO₂ interfaces measured by TDTR or FDTR. O'Brien et al.¹⁴ studied bonding-induced thermal conductance enhancement for Cu/SiO₂ interfaces using TDTR and Cu transducers with thicknesses between 38 nm and 98 nm. They report a conductance value of $G_{\rm Cu}=90\pm15~{\rm MW}~{\rm m}^{-2}~{\rm K}^{-1}$ for untreated interfaces, and an up to fivefold enhancement of the conductance after chemical functionalization. For Al/SiO_2 interfaces conductance values reported range from 100 MW m⁻² K⁻¹ to 150 MWm⁻²K⁻¹ measured with 100 nm thick Al transducers^{12,15}. In the later publications no systematic uncertainties are given. All these experiments do not provide sufficient sensitivity to quantify G with low uncertainties. Reason for this is that the thermal mass of an optically thick transducer in combination with the low thermal conductivity of amorphous SiO₂ samples results in a negligible temperature drop across the interface. At low frequencies or for thin SiO₂ layers on Si, the effective thermal conductivity of the sample is reduced. This results in an increase of the sensitivity with respect to G. However, as uncertainties add in quadrature and additional sample parameters (thickness of the oxide layer, conductance of the SiO_2/Si interface, and thermal conductivity of Si) have to be considered, the effective uncertainty does not actually decrease.

To gain better sensitivity than provided by conventional TDTR experiments, we replace the optically thick metal transducer layer by a magnetic thin film with greatly reduced thermal mass. Instead of thermoreflectance, we rely on the transient polar Kerr rotation to selectively probe the temperature of the magnetic transducer and avoid artifacts created by the temperature distribution in the sample underneath.

In an earlier publication we used a 20 nm thick Co/Pt multilayer transducer on 300 nm thick SiO₂ on Si and reported the value $G_{\rm Pt} = 180 \pm 20$ MW m⁻² K⁻¹¹⁶. In this study, both G and Λ were treated as free parameters and adjusted simultaneously. This approach

lead to an underestimation of the error in G due to the dominance of the sensitivity to Λ . Assuming 5 % uncertainty in the heat capacity of the transducer film and 5 % uncertainty in the thermal conductivity of the oxide the results should read $G_{\rm Pt} = 180 \pm 100$ MW m⁻² K⁻¹.

Here, we refine this approach by using Co/Pt transducers as thin as 4.2 nm and determine the conductance G_{Pt} between Pt and SiO₂ with a significantly reduced uncertainty.

In principle, our TR-MOKE method also provides access to the conductance $G_{\rm Si}$ of SiO₂/Si interfaces for which similar sensitivity considerations apply as for metal/SiO₂ interfaces. $G_{\rm Si}$ values reported to date vary by orders of magnitude. For example, values of 12 – 20 MW m⁻² K⁻¹ were measured by TDTR and FDTR using 100 nm thick Al transducers^{12,15}, while Hurley et al. estimate a conductance of 0.4 GW m⁻² K⁻¹ based on time resolved thermal wave microscopy¹⁷. Thus, we also measure the conductance $G_{\rm Si}$ of the SiO₂/Si interface on a sample with a 26 nm thin SiO₂ layer on Si.

Sections II A and II B provide information about the samples and our experimental setup. Thermal modeling and data analysis are presented in Sec. II C. A detailed discussion of sensitivities of TDTR and TR-MOKE experiments is given in Sec. II D. Section III presents our experimental results including reference measurements on sapphire.

II. METHOD

A. Sample details

Amorphous SiO₂ films with thicknesses of 26 nm and 440 nm were grown on p-type Si $\langle 001 \rangle$ wafers in a furnace at 1000°C in ambient atmosphere. SiO₂ thicknesses were measured by ellipsometry.

Co/Pt thin film transducers with total thicknesses of 4.2 nm, 5.5 nm and 8.2 nm were deposited at room temperature at a base pressure below $2 \cdot 10^{-9}$ mbar: Pt seed layers with thicknesses between 0.8 nm and 3.5 nm were grown via ion-beam sputtering utilizing an electron cyclotron resonance (ECR) source. Subsequently, in-situ dc magnetron sputtering was employed to deposit approximately 1 nm of Pt, a 0.7 nm thick Co layer, and a 1.7–3 nm thick Pt capping layer to prevent oxidation^{18,19}. For reference measurements a sapphire (0001) substrate was added to the deposition of 4.2 nm Co/Pt. Furthermore, a sapphire (0001) substrate was coated with 43 nm of Pt by ECR sputtering for TDTR.

Co/Pt and Pt film thicknesses were determined by X-ray reflectivity. Some samples were also characterized by Rutherford backscattering spectrometry to confirm thicknesses and composition. Details for individual samples are listed in Table II and Table III. Figure 1(a) depicts a schematic of our samples. Pt forms a smooth interface with the underlying SiO₂ substrate. The roughness is approximately 0.2 nm, so on the order of one monolayer, the intermixing region is approximately 0.4 nm¹⁹. All Co/Pt films have perpendicular magnetic anisotropy, a remanence of one, and coercive fields between 6 mT and 18 mT, compare Fig. 1(b).



FIG. 1: (a) Schematic of Co/Pt transducer films on SiO₂/Si samples. (b) Hysteresis of Co/Pt thin films with total thicknesses of 4.2 nm (red), 5.5 nm (green), and 8.2 nm (blue) on 440 nm SiO₂ measured by polar magneto-optic Kerr effect at a wavelength of $\lambda = 635$ nm. The magnetic field was applied along the easy axis perpendicular to the sample plane.

B. Setup

We use an experimental setup with two detection schemes for TDTR and TR-MOKE measurements, see Fig. 2. The laser emits pulses with a FWHM spectral width of approximately 10 nm centered at 783 nm. Duration of correlated pump and probe pulses is approximately 1.2 ps. Pump and probe beams are split by a polarizing beam splitter (PBS) and separated spectrally using ultra steep edge filters, namely a long pass filter in the pump path and short-pass filters in the probe path²⁰. The pump beam is modulated at a frequency f of approximately 11 MHz by an electro-optic modulator (EOM) and the probe beam is modulated at 200 Hz by a mechanical chopper to suppress background signals at 10 MHz, e.g. signals originating from coherent pick up. Time delay between pump and probe pulses is varied from -25 ps to 3.6 ns using a mechanical delay stage. Both beams are focused on

the sample by an objective lens to a $1/e^2$ radius of 11 μ m. We use low fluences of approximately 0.07 J/m² for both pump and probe beam, resulting in transient per-pulse heating on the order of 10 K. The reflected beams are split by a non-polarizing beam splitter (BS) to detect both TDTR and TR-MOKE signals. For TDTR detection the reflected pump light is removed by a PBS and a short-pass filter identical to the one in the probe path. The probe beam is focused on a Si photo diode (PD). A low-pass filter removes higher harmonics before the output of the PD is measured by a radio-frequency lock-in amplifier at 11 MHz and subsequently by an audio-frequency lock-in at 200 Hz. For TR-MOKE detection the reflected pump light is blocked by a ultra-steep edge short-pass filter before the probe beam passes through a $\lambda/2$ plate and is split into orthogonally polarized components by a Wollaston prism. The $\lambda/2$ plate is adjusted such that both beam components focused onto a balanced photo detector (BPD) have approximately the same intensity. Transient changes in the polarization of the probe beam are proportional to changes in the relative intensities detected by the BPD. The radio-frequency output of the BPD is measured the same way as TDTR using a band-pass filter after the BPD.



FIG. 2: Experimental setup. Pump and probe beam paths are split by polarizing beam splitters (PBS) and spectrally using short-pass (blue) and long-pass (orange) filters. Time-domain thermoreflectance (TDTR) is measured with a Si photo diode. Part of the beams reflected from the sample is redirected by a beam splitter (BS) towards a balanced photo detector to measure the time-dependent changes in Kerr rotation (TR-MOKE).

In case of non-perfect balancing with the $\lambda/2$ plate, thermoreflectance signals overlap the transient Kerr rotation. To isolate the TR-MOKE signal we subtract the in-phase and out-of-phase signals recorded for oppositely aligned magnetization states of the Co/Pt transducers before analyzing the ratio signal $-V_{\rm in}/V_{\rm out} = -(V_{\rm in}^+ - V_{\rm in}^-)/(V_{\rm out}^+ - V_{\rm out}^-)$ as illustrated in Ref.¹⁶. Given the magnetic properties of our samples (compare Fig. 1) the magnetic field of approximately 400 mT provided by a NdFeB permanent magnet is sufficient to invert the direction of the magnetization of the Co/Pt transducer. Our 'triple-modulation' approach (in addition to modulating pump and probe beam we modulate the magnetic state of the sample) makes TR-MOKE thermometry less prone to error as it ensures getting rid of offsets which can significantly affect TDTR experiments.

All measurements were done at remanence and room temperature.

C. Thermal model and data analysis

We analyze the ratio $R_{\text{data}} = -V_{\text{in}}/V_{\text{out}}$ using a multilayer heat diffusion model²¹. Analyzing the ratio instead of V_{in} alone corrects for non-idealities in the experiment such as laser power fluctuations and variations in pump-probe overlap with delay time²². A thermal model is fitted to the data in the delay-time range of highest sensitivity with respect to the interface thermal conductance (30–500 ps for G_{Pt} , compare Fig. 4(b)) by minimizing the sum of variances $\sigma = \sum ((R_{\text{model}} - R_{\text{data}})/R_{\text{data}})^2$.

TABLE I: Parameters for model calculations and fitting of experimental data. h: layer thickness, A: thermal conductivity, C: volumetric heat capacity. Pt/Co/Pt films are treated as one layer.

	$h(10^{-9} \mathrm{m})$	$\Lambda({\rm W~m^{-1}~K^{-1}})$	$C(10^{6} \mathrm{J m^{-3} K^{-1}})$		
Adsorbate	1	2	2.8 ± 0.6		
$\mathrm{Co/Pt}$	$4.2/5.5/8.2\pm5\%$	20	2.97/2.94/2.90		
SiO_2	$26\pm1/440\pm10$	1.31 ± 0.06	1.65		
Si	∞	130 ± 7	1.61		
Pt	43 ± 3	32	2.82		
Al ₂ O ₃	∞	35.5 ± 2	3.1		

Table I summarizes all parameters including assumptions about the uncertainties that are used as inputs to the thermal model. Each layer of the sample is described by three parameters: thermal conductivity Λ , volumetric heat capacity C, and thickness h. Interfaces are modeled by layers with small heat capacity and thickness.

Given the small thermal masses of our Co/Pt transducers, adsorbates on the sample surface can notably add to the effective heat capacity of the transducer and have to be considered in our thermal model. Under normal environmental conditions as they were present during our experiment, hydrocarbons and water get adsorbed on the surface of solids²³. The thickness of the water layer is known to be in the nanometer range and depends on humidity of the ambient air and the conditions of the surface as oxidation state and carbonaceous contamination^{24,25}.

We empirically define C_{ad} as the heat capacity of adsorbed water and hydrocarbons, and accordingly add a transparent layer on top of the Co/Pt transducer. It does not matter how exactly we model this layer, as our thermal model only depends on the total heat capacity per unit area. We assign it a thickness of 1 nm and adjust the volumetric heat capacity.

 $C_{\rm ad}$ is estimated by simultaneously fitting the volumetric heat capacity of this layer and the conductance $G_{\rm Pt}$ between Co/Pt transducer and SiO₂. Figure 3 shows contour plots obtained for 8.2 nm (blue), 5.5 nm (green), and 4.2 nm (red) Co/Pt on 440 nm SiO₂, as well as 4.2 nm Co/Pt on 26 nm SiO₂ (orange). Lines include all combinations of the free parameters $G_{\rm Pt}$ and $C_{\rm ad}$ for which $\sigma \leq 2\sigma_{\rm min}$, where $\sigma_{\rm min}$ is the minimum of the sums of standard deviations $\sigma = \sum ((R_{\rm model} - R_{\rm data})/R_{\rm data})^2$.

Considering the centers of the contours which mark the best fits, we find an average of $C_{\rm ad} = 2.8 \text{ mJ m}^{-2} \text{ K}^{-1}$ and use this value for the analysis of all Co/Pt and Pt samples. The standard deviation in the best-fit values is only 15 %. However, we assume an uncertainty of 20 % in $C_{\rm ad}$ to account for the uncertainty due to noise in the data which is reflected in the width of the contours.

The approach of considering an additional heat capacity to account for surface properties is not uncommon. Using Al transducers requires the addition of a heat capacity of $C_{\rm Al} =$ 7.3 mJ m⁻² K⁻¹ to obtain the right answer in TDTR experiments. The smaller value $C_{\rm ad} = 2.8 \text{ mJ m}^{-2} \text{ K}^{-1}$ we find for Pt seems reasonable considering the different nature of the additional heat capacities. Al oxidizes, while Pt is comparably inert and we do not expect oxidation given our experimental conditions. Differences in surface oxidation lead to different degrees of contamination with hydrocarbons and water. Heating our samples to 100° C in a vacuum of $4 \cdot 10^{-4}$ mbar did not significantly alter C_{ad} . The inevitable presence of carbonaceous contamination leads to a certain degree of chemisorption of water that is hard to remove²³. Gradual chemisorption would also explain why we do not see a significant increase of C_{ad} after six months, independent of how the samples were stored (N atmosphere or ambient conditions). Mass gain by carbonaceous contamination is diffusion limited and does not saturate²⁴. This is the reason for the mass increase of kg-mass standards over the course of years, and in analogy an eventual increase in C_{ad} has to be expected.

Volumetric heat capacities of Co/Pt layers are calculated based on composition assuming $C_{\rm Co} = 3.73 \cdot 10^6 \text{ Jm}^{-3} \text{ K}^{-1}$ and $C_{\rm Pt} = 2.82 \cdot 10^6 \text{ Jm}^{-3} \text{ K}^{-1}$. For the cross-plane thermal conductivity of Co/Pt we assume an approximate value of 20 W m⁻¹ K⁻¹ which is not of relevance for the results reported here. For the 43 nm Pt film we determine an in-plane electronic contribution of 25 W m⁻¹ K⁻¹ from sheet resistance measurements using the Wiedemann-Franz law and add 7 W m⁻¹ K⁻¹ to account for the theoretical contribution from the lattice²⁶.

In our thermal model, heat is deposited in the center of the Co/Pt transducer layer (or at a depth of 11 nm in the Pt film) to account for the fact that heat is deposited throughout the penetration depth of the laser light, and then spreads bi-directionally, down into the SiO_2/Si (Al₂O₃) substrate and up into the top layer that represents adsorbates.

Our thermal model does not require quantitative knowledge of the optical absorption in the sample. However, we model the absorption in our samples to estimate temperature excursions in our experiments. Furthermore, understanding how the thicknesses of the Co/Pt and SiO₂ layers determine absorption A, reflectance R, and magneto-optic Kerr rotation $\theta_{\rm K}$ helps to optimize the sample design, see Appendix V B. The laser power was selected such that the average heating of the transducer by a single optical pulse is between 10 K and 18 K. As all our Co/Pt transducers are not opaque, part of the laser light is absorbed in the Si substrates. However, the optical penetration depth in Si is large, and even if the entire laser power was absorbed by the Si substrate the temperature rise would be below 0.1 K and can thus be neglected in the thermal modeling.

We measured the static Kerr rotation $\theta_{\rm K}$ between room temperature and 100°C to confirm that the change in Kerr rotation is, to a good approximation, proportional to the change in temperature. Knowing the temperature excursion by a pump optical pulse we can also



FIG. 3: Contour intervals marking the best agreement between TR-MOKE data and thermal model simultaneously adjusting conductance G_{Pt} between transducer and sample and heat capacity C_{ad} of adsorbates. For the 4.2 nm Co/Pt film on 26 nm SiO₂/Si (orange) the contour is much wider compared to the Co/Pt films on 440 nm thick oxide (others). Reason for this is the increased noise in the data arising from the smaller out-of-phase signal due to the higher thermal conductivity of the substrate.

determine the temperature dependence of the magneto-optic Kerr rotation from the in-phase signal of a TR-MOKE measurement. For the 4.2 nm Co/Pt on 440 nm SiO₂ sample we find a constant value of $d\theta_{\rm K}/dT = -1.2 \pm 0.2 \cdot 10^{-5}$ rad K⁻¹ at 30, 70, and 100°C and conclude that the linearity of the TR-MOKE signal required for thermometry is given for the transducers under investigation. Table III summarizes the averaged $d\theta_{\rm K}/dT$ and $\theta_{\rm K}$ values at room temperature for all samples studied. $d\theta_{\rm K}/dT$ is, to a good approximation, proportional to $\theta_{\rm K}$. We find an average ratio of $\frac{\theta_{\rm K}}{d\theta_{\rm K}/dT} = 485 \pm 25$ K.

D. Sensitivities in TDTR and TR-MOKE thermometry

The thermal penetration depth $d = \sqrt{\frac{\Lambda}{C\pi f}}$ in a material with volumetric heat capacity C and thermal conductivity Λ is determined by the modulation frequency f of the EOM. We refer to a sample as 'thermally thick' if its thickness is larger than d. For amorphous SiO_2 , $d \approx 270$ nm at f = 11 MHz. This means that the thermal conductance of the $\operatorname{SiO}_2/\operatorname{Si}$ interface and the thermal conductivity of Si are not important in experiments using 440 nm SiO_2 .

To describe the sensitivity of the measurement signal to properties of the materials at depths shorter than the thermal penetration depth, we define sensitivity coefficients S_{α} as

the logarithmic derivative of the negative of the ratio signal:

$$S_{\alpha} = \frac{\partial \ln \left(-V_{\rm in}/V_{\rm out}\right)}{\partial \ln \alpha} \tag{1}$$

where α is a parameter in the thermal model²⁷.

Figure 4(a) depicts the time dependence of S with respect to the transducer thickness h_{Pt} (blue), the conductance $G_{\text{Pt}} = 0.25 \text{ GW m}^{-2} \text{ K}^{-1}$ (red), as well as the thermal conductivity Λ_{SiO_2} (orange) for a 5 nm thin Pt transducer on thermally thick SiO₂. As in a conventional TDTR measurement, the sensitivities of the ratio signal to the thickness (S_h) and to the volumetric heat capacity $(S_C, \text{ not shown, identical to } S_h)$ of the transducer are large, close to unity at short delay times.

Figure 4(b) illustrates how the sensitivities with respect to $G_{\rm Pt}$, $h_{\rm Pt}$, and $\Lambda_{\rm SiO_2}$ depend on the thickness $h_{\rm Pt}$ of the transducer film. As S changes with delay time between pump and probe pulses, we plot the peak sensitivities $S_{\rm max}$ versus $h_{\rm Pt}$. The black line indicates the delay times $t_{\rm max}$ at which the sensitivity to the conductance $G_{\rm Pt}$ reaches the maximum value $S_{\rm max}$.

A characteristic time-scale for the measurement is the time required for heat to diffuse a distance in SiO₂ that is equal to the Kapitza length, $\tau_{\rm D} = L_K^2 C_{\rm SiO2} / \Lambda_{\rm SiO2}$. The sensitivity to $G_{\rm Pt}$ is at a maximum for delay times comparable to $\tau_{\rm D}$ although the exact position of the maximum is also a function of $h_{\rm Pt}$ relative to $L_{\rm K}$.



FIG. 4: (a) Sensitivity coefficients with respect to interface conductance $G_{\rm Pt}$ (red), transducer thickness $h_{\rm Pt}$ (blue), and thermal conductivity $\Lambda_{\rm SiO_2}$ (orange) versus delay time t for a 5 nm thick Pt transducer on thermally thick oxide. Simulation parameters are listed in Table I, $G_{\rm Pt} =$ $0.25 \,\rm GW \,m^{-2} \,K^{-1}$. (b) Maximum sensitivity coefficients $S_{\rm max}$ at delay time of maximum sensitivity $t_{\rm max}$ calculated for various thicknesses $h_{\rm Pt}$.

For a typical, 90 nm optically thick Pt transducer, the peak sensitivity to $G_{\rm Pt}$ is $S_{\rm max} \approx -0.01$ and occurs at a delay time of $t_{\rm max} = 2$ ns. This means a 100 % variation in G would change the ratio signal $-V_{\rm in}(t_{\rm max})/V_{\rm out}(t_{\rm max})$ by 1 %. Reduction of the transducer thickness leads to a significant increase in $S_{\rm max}$, as the temperature change and thus the signal change for a given amount of heat diffusion across the Pt/SiO₂ interface is bigger the smaller the thermal mass of the transducer. To gain reasonable sensitivity of $|S| \ge 0.1$ the transducer thickness has to be on the order of the Kapitza length $L_{\rm K} = \Lambda_{\rm SiO2}/G_{\rm Pt}$ in SiO₂, which is approximately 5 nm for a conductance of $G_{\rm Pt} = 250$ MW m⁻² K⁻¹.

An uncertainty u_{α} in a model parameter α propagates into the uncertainty u_{β} of a free parameter β according to $u_{\beta} = u_{\alpha} \frac{S_{\alpha}}{S_{\beta}}$. Uncertainties arising from different model parameters add in quadrature. The experimental uncertainty in G_{Pt} based on 5 % uncertainty in both h_{Pt} and Λ_{SiO_2} is 80 % using a 90 nm thick transducer film, 12 % for a 10 nm transducer, and 6 % for a 5 nm thin transducer. Here, we only considered the S_{max} values. For the analysis of our data we consider the maximum of each sensitivity coefficient within the entire fitted time range as well as additional sources of uncertainty as the heat capacity of water and hydrocarbons adsorbed on the surface of the sample.

III. RESULTS AND DISCUSSION

A. Comparison of TDTR and TR-MOKE data for sapphire samples

To test our experimental approach, we compare TDTR and TR-MOKE measurements done with a 4.2 nm thin Co/Pt film on sapphire, as well as a reference TDTR measurement using a 43 nm thick Pt film on the same Al_2O_3 substrate. Figure 5(a) displays normalized in-phase signals for short time delays t between pump and probe laser pulses. The difference in TDTR traces (open squares) arises from the fact that the electronic contribution to the thermoreflectance, which leads to a small peak in the Pt data (black) at delay times around 1 ps, is negligible for the 4.2 nm thin Co/Pt transducer (blue).

TR-MOKE (full blue circles) probes the magnon temperature. We see the characteristic ultrafast demagnetization peak at ≈ 1 ps, which looks the same for all Co/Pt samples studied. Magnons, electrons, and phonons are thermalized at delay times > 2 ps due to strong electron-magnon coupling, strong electron-phonon coupling, and the small heat



FIG. 5: Data measured by time-domain thermoreflectance (TDTR, open squares) with 4.2 nm Co/Pt (blue) and with 43 nm Pt (black) transducer, and by time-resolved magneto-optic Kerr effect (TR-MOKE, blue circles) with 4.2 nm Co/Pt on Al₂O₃. (a) Normalized in-phase signals. (b) Ratio signals, blue squares are multiplied by -1 as ratio is negative. Red lines are the best fits obtained using the interface conductance G between Pt and sapphire as only free parameter. Model parameters are listed in Table I. $C_{\rm ad} = 2.8 \,\mathrm{mJ}\,\mathrm{m}^{-2}\,\mathrm{K}^{-1}$ was considered for both samples.

capacity of the magnons at temperatures far away from the Curie point²⁸.

The time it takes for a transducer to reach equilibrium determines the shortest time delay at which our thermal model can be fitted to the experimental data. In TR-MOKE experiments using ultra-thin magnetic films as transducers it can be limited by either the thermalization times between the thermal reservoirs or by the time it takes to establish a homogeneous temperature distribution across the thickness of the transducer. Close to the Curie temperature $T_{\rm C}$ the magnetic heat capacity increases. This slows down the demagnetization process and can lead to thermalization times of up to 100 ps²⁸. Due to the magnetic properties of Co/Pt films and the small transient heating by our laser pulses (≈ 10 K), our experiments are conducted well below $T_{\rm C}$.

Red lines in Fig. 5(b) are the fits obtained adjusting the interfacial thermal conductance G between Pt and Al₂O₃ as only free parameter in our thermal model. The results are in agreement within the uncertainty of our measurement. We find $G = 170 \pm 10$ MW m⁻² K⁻¹ analyzing TR-MOKE data for 4.2 nm Co/Pt (blue circles) and $G = 145 \pm 15$ MW m⁻² K⁻¹ analyzing TDTR data for 43 nm Pt (black squares). The shortest delay times fitted are 10 ps and 100 ps, respectively. For both samples an adsorbed layer with $C_{\rm ad} = 2.8 \pm 0.6 \text{mJm}^{-2} \text{K}^{-1}$ was considered in the thermal modeling.

TR-MOKE selectively probes the magnon temperature of a ferromagnetic transducer layer. TDTR, on the other hand, probes the phonon and electron temperatures of all regions within the penetration depth of the laser light. Even in case of a transparent sample as Al_2O_3 , which does not contribute much to the thermoreflectance signal, an opaque transducer is essential in TDTR experiments. To demonstrate this we also plot the TDTR ratio signal measured with the 4.2 nm thin Co/Pt film, see blue squares in Fig. 5(b). The ratio is negative which is not possible if the thermoreflectance signal is dominated by the changes in the temperature of the Co/Pt film. On SiO₂/Si substrates we find that a Pt thickness of 50 nm (corresponding to approximately five times the optical penetration depth) is required to guarantee that TDTR and TR-MOKE ratios are identical after equilibration of the transducer.

B. Thermal conductance of Pt/a-SiO₂ interfaces



FIG. 6: (a) TR-MOKE ratio data versus delay time t measured using Co/Pt transducers with thicknesses of 4.2 nm (red), 5.5 nm (green), and 8.2 nm (blue) on 440 nm (thermally thick) SiO₂. Black lines are fits of thermal models to the data obtained treating $G_{\rm Pt}$ as only free parameter and considering an adsorbed layer with a heat capacity of $C_{\rm ad} = 2.8 \text{ mJ m}^{-2} \text{ K}^{-1}$. Black numbers are conductances in MW m⁻² K⁻¹. (b) Results for conductances from this work and data by Liu et al.¹⁶. All error bars are calculated assuming same relative uncertainty in experimental parameters.

Thermal conductance G_{Pt} of Pt/a-SiO₂ interfaces was measured on four different samples with thin Co/Pt transducers. TR-MOKE data and fit curves are presented in Fig. 6(a) for 4.2 nm, 5.5 nm, and 8.2 nm Co/Pt on 440 nm SiO₂, as well as in Fig. 8 for 4.2 nm Co/Pt on 26 nm SiO₂. Considering the sensitivity each sample provides with respect to G_{Pt} we find an average value of $G_{\rm Pt} = 310 \pm 50$ MW m⁻² K⁻¹. Here, the error originates from the scattering of best fit results for the individual samples and does not represent an experimental uncertainty based on model parameters which is larger, compare Fig. 6(b).

Figure 6(b) summarizes our results and compares them to the work of Liu et al. which provided the highest sensitivity to a comparable interface prior to this work. Experimental advancement through using transducers of greatly reduced thermal mass is reflected in the width of error bars. By using 4.2 nm thin transducers, the uncertainty can be reduced down to approximately 20 %, compared to 60 % uncertainty when using 20 nm thick Co/Pt films. Note that the advancement is masked by the fact that values for S with respect the $G_{\rm Pt}$ as well as to other experimental parameters (most critically $\Lambda_{\rm SiO_2}$) depend on the absolute conductance values measured. The higher the interface conductance, the more difficult it is to separate its contribution from the conductivity of the SiO₂.

The values we measure for the interface conductance $G_{\rm Pt}$ between metal and an amorphous material are higher than the ones reported for comparable interfaces to date^{12,14,16}. As shown both experimentally and in molecular dynamics simulations, conductance can be significantly increased by enhancing interfacial bonding^{14,29,30}. Bonding at the interface between a metal and a-SiO₂ can be weakened by layers of contamination adsorbed on the oxide surface. Based on the results of earlier studies^{18,19}, we believe that the Pt/a-SiO₂ interface is relatively free of such contamination layers because we use the electron-cyclotron resonance (ECR) sputtering technique to deposit the first part of the Pt seed layer on SiO₂, followed by an additional 1 nm Pt by magnetron sputtering. During ECR sputter deposition high energy Ar⁺ ions (≤ 1.2 keV) reflected at the target bombard the surface of the SiO₂ substrate and remove contamination. Pt atoms impinge the surface with kinetic energies of $\approx 30 \text{ eV}^{19}$. This enhances bonding and potentially drives some degree of intermixing of atoms at the interface¹⁸.

Since in a metal heat is mainly carried by electrons and in amorphous SiO₂ it is carried only by vibrational modes, the effective conductance can be considered as the series of two thermal resistances. The conductance value $G_{\rm Pt}$ we measure is thus given by $G_{\rm Pt} = 1/(G_{\rm el-ph}^{-1} + G_{\rm vib}^{-1})$. Assuming a coupling constant of $g = 5 \cdot 10^{17} {\rm W m^{-3} K^{-1}}$, $G_{\rm el-ph} = g h$ is 2 GW m⁻² K⁻¹ and higher for our samples. The vibrational energy transfer across the interface is thus the main source of thermal resistance.

For a crystalline material, the corresponding conductance $G_{\rm vib}$ is given by $G_{\rm vib}$ =

 $\frac{1}{4}\sum_{j}\int t_{\omega}v_{\omega}c_{\omega}d\omega_{j}^{5,31}$ where v_{ω} are the group velocities and c_{ω} are the heat capacities of phonons with frequency ω_{j} and polarization j. $c_{\omega} = \hbar\omega D_{\omega}\partial n_{\omega}/\partial T$, where D_{ω} is the density of states, n is the Bose-Einstein distribution function. We define a maximum conductance value G_{max} for a material by setting the transmission coefficient t = 1. G_{max} for an interface is determined by the material that has the lower intrinsic G_{max} value. Following the calculation described in Ref.³¹, we obtain a theoretical value of $G_{\text{max}} = 0.9 \text{ GW m}^{-2} \text{ K}^{-1}$ for Pt. For interfaces between metals and dielectric crystals, the experimental values for G typically lie around $G_{\text{max}}/3$, and up to $G_{\text{max}}/2$ for strongly bonded interfaces. The values we measure fall into this range.

C. Thermal model and the role of adsorbates



FIG. 7: TR-MOKE ratios normalized by models for $C_{ad} = 0 \text{ mJ m}^{-2} \text{ K}^{-1}$, no Pt/SiO₂ interface, and no adjusted parameters to highlight deviations between data and model curves. Data points and black curves are same as in Fig. 6(a). Dashed lines are models representing experimental uncertainties (black numbers are conductance values in MW m⁻² K⁻¹). Red lines are best fits obtained not considering the additional heat capacity of adsorbates (C_{ad} values in mJ m⁻² K⁻¹). Green model line in (a) includes an interface with conductance $G_{ad} = 400 \text{ MW m}^{-2} \text{ K}^{-1}$ between Co/Pt transducer and adsorbed layer.

To better see deviations between model curves and experimental data, we plot the data for 4.2 nm and 8.2 nm Co/Pt shown in Fig. 6(a) again in Figs. 7(a) and (b), normalizing all curves to models with the thermal resistance of the Pt/SiO₂ interface and the additional heat capacity $C_{\rm ad}$ due to adsorbates set to zero. All model parameters are listed in Tab. I. As discussed in Sec. II C we use contour plots (compare Fig. 3) to estimate the heat capacity of adsorbed hydrocarbons and water and add the averaged value of $C_{\rm ad} = 2.8 \pm 0.6 \,\mathrm{mJ}\,\mathrm{m}^{-2}\,\mathrm{K}^{-1}$ to the thermal model for all samples. Black lines in Fig. 7 are the same fit curves as in Fig. 6(a) obtained treating $G_{\rm Pt}$ as the only remaining free parameter. Dashed black lines are model curves representing the error bars based on uncertainties in the model parameters.

For comparison, we also plot the best fits obtained neglecting the additional heat capacity due to an adsorbed layer and treating $G_{\rm Pt}$ as only free parameter, see red lines in Fig. 7. Not considering $C_{\rm ad}$ reduces the thermal masses of the transducers and thus remarkably increases the sensitivity to $G_{\rm Pt}$, especially for the 4.2 nm sample. Consequently, we see a notable deviation of the normalized curves from one (representing the model for infinite conductance), even though the fitted conductance values are higher than 600 MW m⁻² K⁻¹. The delay time of highest sensitivity to $G_{\rm Pt}$ is at approximately 20 ps, compared to 80 – 100 ps for the models including an adsorbed layer. Comparison to the curvature in our data clearly underlines the validity of our thermal model considering the additional heat capacity.

The green line in Fig. 7(a) is a model considering the interface between Co/Pt transducer and adsorbed layer as additional free parameter. Adjusting $G_{\rm Pt}$ and the conductance $G_{\rm ad}$ of this interface at delay times between 10 and 500 ps yields $G_{\rm Pt} = 250$ MW m⁻² K⁻¹ and $G_{\rm ad} = 400$ MW m⁻² K⁻¹. Introducing this interface does not significantly change the result of our thermal analysis. However, the fit between model curve and data clearly improves at short delay times. This demonstrates that ultra-thin Co/Pt transducers provide a platform for a more profound study of the thermal properties of adsorbed molecular layers and facilitate access to heat diffusion processes occurring at picosecond time scales.

D. Thermal conductance of $a-SiO_2/Si$ interfaces

Our TR-MOKE method also gives access to the conductance $G_{\rm Si}$ between a-SiO₂ and Si for a carefully chosen thickness of the oxide layer that provides a separation of the delay times at which maximum sensitivities to $G_{\rm Pt}$ and $G_{\rm Si}$ occur.



FIG. 8: TR-MOKE ratio data measured with 4.2 nm thin Co/Pt transducers on 26 nm SiO₂. Thermal models include an adsorbed layer with a heat capacity of $C_{\rm ad} = 2.8 \text{ mJ m}^{-2} \text{ K}^{-1}$ added on top of the Co/Pt. Conductance $G_{\rm Pt} = 320 \pm 110 \text{ MW m}^{-2} \text{ K}^{-1}$ of the Pt/SiO₂ interface was fitted between 30 ps and 500 ps. Black numbers are conductances $G_{\rm Si}$ between SiO₂ and Si in MW m⁻² K⁻¹ fitted between 800 ps and 3600 ps. All curves are normalized by models for $C_{\rm ad} = 0 \text{ mJ m}^{-2} \text{ K}^{-1}$ and no Pt/SiO₂ and SiO₂/Si interfaces.

We characterized a sample with 4.2 nm Co/Pt on 26 nm SiO₂/Si by TR-MOKE thermometry. Figure 8 depicts ratio data and model curves normalized to models with the thermal resistances of the Pt/SiO_2 and SiO_2/Si interfaces and the additional heat capacity due to adsorbed hydrocarbons and water set to zero. We find $G_{\rm Si} = 1.4 \ {\rm GW} \ {\rm m}^{-2} \ {\rm K}^{-1}$. Based on the small sensitivity to such a high conductance value, the error bars are large, $\pm 1.6 \text{ GW m}^{-2} \text{ K}^{-1}$. Since sensitivity coefficients scale with G_{Si} , symmetrical error bars do not reflect the physical reality. The dashed lines in Fig. 8 representing the uncertainties are thus drawn for $G_{\rm Si} = 3 \,\rm GW \,m^{-2} \,\rm K^{-1}$ and $G_{\rm Si} = 0.6 \,\rm GW \,m^{-2} \,\rm K^{-1}$. Our lower limit for $G_{\rm Si}$ considers error propagation from all uncertainties by considering the 'worst-case' combination of model parameters. This 'worst-case' combination of those parameters was selected as follows: For each model parameter α an uncertainty is assumed (compare Table I) that sets the minimum and maximum values possible for α . Depending on the sign of the sensitivity coefficient S_{α} compared to the sign of $S_{G_{Si}}$ either the minimum or maximum value of α was used. Fitting a thermal model with those parameters to the experimental data results in the lowest value possible for $G_{\rm Si}$ within our uncertainties. Our result agrees well with the lower limit we calculate from data reported by Costescu et al.²².

Costescu et al. measured the thermal conductivity of thin SiO_2 layers on Si by TDTR using 60 nm thick TiN transducers²². In their experiments oxide layers were thin enough for

the TiN/SiO₂ and SiO₂/Si interfaces to notably reduce the effective thermal conductivity. From their room-temperature data on 6.5 and 11 nm thin SiO₂ we determine a lower limit for the SiO₂/Si interface of 0.5 GW m⁻² K⁻¹ by assuming an infinite conductance for the first interface. Based on the data by Costescu et al. it is unclear how the thermal resistance splits between the TiN/SiO₂ and SiO₂/Si interfaces. If we assign a value of 600 GW m⁻² K⁻¹ to the TiN/SiO₂ interface, we calculate $G_{\rm Si} = 4$ GW m⁻² K⁻¹. The agreement with our result supports the assumption that the conductance of SiO₂/Si interfaces is very high.

Molecular dynamics simulations also predict high conductances on the order of $1 \,\mathrm{GW}\,\mathrm{m}^{-2}\mathrm{K}^{-132}$. According to Ref.³¹, the theoretical value of G_{\max} for Si is $0.8 \,\mathrm{GW}\,\mathrm{m}^{-2}\mathrm{K}^{-1}$. Our experimental result for G_{Si} suggests that the transmission coefficient for vibrational heat flow at a Si/SiO₂ interface is close to unity.

IV. CONCLUSION

Selectively probing the temperature of magnetic thin films via TR-MOKE facilitates the use of transducers with significantly reduced thermal mass. Using this thermometry method, sensitivity to the conductance of interfaces with low thermal conductivity materials can be increased by approximately one order of magnitude compared to TDTR. The 4.2 nm to 8.2 nm thin Co/Pt transducers used in our study provide sufficient sensitivity to quantify the interface thermal conductance between Pt and amorphous SiO₂. We find an average value of $G_{\rm Pt} \approx 0.3$ GW m⁻² K⁻¹. For SiO₂/Si interfaces we find a lower limit of $G_{\rm Si} = 0.6$ GW m⁻² K⁻¹.

Perpendicularly magnetized Co/Pt thin films can be grown on almost any sample^{19,33}, do not require annealing, and are comparably inert. They are thus ideal transducers for the TR-MOKE thermometry method demonstrated here. A critical aspect to consider which will vary depending on the transducer material selected, is the additional heat capacity due to adsorbed water and hydrocarbons, as well as due to possible oxide formation. In our samples adsorbates increase the heat capacity by up to 20 %.

As discussed in detail in the Appendix Sec. V B, the optical absorption behavior and the Kerr rotation in our samples are a function of the magneto-optic constants and thicknesses of the transducer and the underlying sample structure. Ferromagnetic transducers have thus to be optimized depending on the sample requirements with respect to magneto-optic prop-

erties, sensitivities, and time resolution. In our case, a transducer thickness of approximately 4 nm minimizes the thermal mass while maintaining a large Kerr signal.

We anticipate that magnetic thin film transducers will provide a platform for measurements of small heat capacities, for example of adsorbed molecular layers, and make it possible to study thermal processes on the picosecond time scale. The time resolution in optical pump-probe experiments is typically limited by the time scale $\tau_{\rm D} = \frac{h^2 C}{\Lambda}$ of heat diffusion through a transducer with thickness h. In TDTR experiments with opaque Al transducers $\tau_{\rm D}$ is on the order of 100 ps, while it is only approximately 4 ps for TR-MOKE thermometry with a 5 nm thin Co/Pt transducer. Access to such short time scales might allow for the observation of non-equilibrium effects of heat carriers directly in the time domain³⁴.

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- * Electronic address: jkimling@illinois.edu
- ¹ A. Shakouri, P. IEEE **94**, 1613 (2006).
- ² D. Weller and A. Moser, IEEE Trans. Magn. **35**, 4423 (1999).
- ³ D. G. Cahill, P. V. Braun, G. Chen, D. R. Clarke, S. Fan, K. E. Goodson, P. Keblinski, W. P. King, G. D. Mahan, A. Majumdar, et al., Appl. Phys. Rev. 1, 011305 (2014).
- ⁴ M. D. Losego and D. G. Cahill, Nat. Mater. **12**, 382 (2013).
- ⁵ R. J. Stoner and H. J. Maris, Phys. Rev. B 48, 16373 (1993).
- ⁶ D. G. Cahill, W. K. Ford, K. E. Goodson, G. D. Mahan, A. Majumdar, H. J. Maris, R. Merlin, and S. R. Phillpot, J. Appl. Phys. **93**, 793 (2003).
- ⁷ R. Cheaito, J. T. Gaskins, M. E. Caplan, B. F. Donovan, B. M. Foley, A. Giri, J. C. Duda, C. J. Szwejkowski, C. Constantin, H. J. Brown-Shaklee, et al., Phys. Rev. B **91**, 035432 (2015).
- ⁸ D. G. Cahill, S. K. Watson, and R. O. Pohl, Phys. Rev. B 46, 6131 (1992).

- ⁹ W. Lv and A. Henry, New Journal of Physics **18**, 013028 (2016).
- ¹⁰ P. E. Hopkins, ISRN Mechanical Engineering **2013** (2013).
- ¹¹ A. J. Schmidt, R. Cheaito, and M. Chiesa, Review of Scientific Instruments 80, 094901 (2009).
- ¹² J. Zhu, D. Tang, W. Wang, J. Liu, K. W. Holub, and R. Yang, J. Appl. Phys. **108**, 094315 (2010).
- ¹³ W. S. Capinski, H. J. Maris, T. Ruf, M. Cardona, K. Ploog, and D. S. Katzer, Phys. Rev. B 59, 8105 (1999).
- ¹⁴ P. J. Orien, S. Shenogin, J. Liu, P. K. Chow, D. P. Laurencin, P. Mutin, M. Yamaguchi, P. Keblinski, and G. Ramanath, Nat. Mater. p. 118 (2013).
- ¹⁵ P. Hopkins, J. Serrano, L. Phinney, S. Kearney, T. Grasser, and C. Harris, ASME. J. Heat Transfer **132**, 081302 (2010).
- ¹⁶ J. Liu, G.-M. Choi, and D. G. Cahill, J. Appl. Phys. **116**, 233107 (2014).
- ¹⁷ D. H. Hurley, M. Khafizov, and S. L. Shinde, J. Appl. Phys. **109**, 083504 (2011).
- ¹⁸ H. Stillrich, C. Menk, R. Frömter, and H. P. Oepen, J. Appl. Phys. **105**, 07C308 (2009).
- ¹⁹ G. Winkler, A. Kobs, A. Chuvilin, D. Lott, A. Schreyer, and H. P. Oepen, J. Appl. Phys. **117**, 105306 (2015).
- ²⁰ K. Kang, Y. K. Koh, C. Chiritescu, X. Zheng, and D. G. Cahill, Rev. Sci. Instrum. **79**, 114901 (2008).
- ²¹ D. G. Cahill, Rev. Sci. Instrum. **75**, 5119 (2004).
- ²² R. M. Costescu, M. A. Wall, and D. G. Cahill, Phys. Rev. B 67, 054302 (2003).
- $^{23}\,$ M. Kochsiek, Metrologia ${\bf 18},\,153$ (1982).
- ²⁴ F. E. Jones and R. M. Schoonover, *Handbook of Mass Measurement* (Chapman and Hall, 2002).
- ²⁵ P. J. Cumpson and M. P. Seah, Metrologia **33**, 507 (1996).
- ²⁶ G. T. Hohensee, R. Wilson, and D. Cahill, Nat. Commun. **6**, 6578 (2015).
- ²⁷ Y. K. Koh, S. L. Singer, W. Kim, J. M. O. Zide, H. Lu, D. G. Cahill, A. Majumdar, and A. C. Gossard, J. Appl. Phys. **105**, 054303 (2009).
- ²⁸ J. Kimling, J. Kimling, R. B. Wilson, B. Hebler, M. Albrecht, and D. G. Cahill, Phys. Rev. B 90, 224408 (2014).
- ²⁹ M. Hu, P. Keblinski, and P. K. Schelling, Phys. Rev. B **79**, 104305 (2009).
- ³⁰ M. D. Losego, M. E. Grady, N. R. Sottos, D. G. Cahill, and P. V. Braun, Nat. Mater. **11**, 502

(2012).

- ³¹ R. B. Wilson, B. A. Apgar, W.-P. Hsieh, L. W. Martin, and D. G. Cahill, Phys. Rev. B 91, 115414 (2015).
- ³² J. Chen, G. Zhang, and B. Li, J. Appl. Phys. **112**, 064319 (2012).
- ³³ M. Albrecht, G. Hu, I. L. Guhr, T. C. Ulbrich, J. Boneberg, P. Leiderer, and G. Schatz, Nat. Mater. 4 (2005).
- ³⁴ R. B. Wilson and D. G. Cahill, Nat. Commun. 5, 5075 (2014).
- ³⁵ Z. Q. Qiu and S. D. Bader, Rev. Sci. Instrum. **71**, 1243 (2000).
- ³⁶ S. Fiedler, H. Stillrich, and H. P. Oepen, J. Appl. Phys. **102**, 083906 (2007).

V. APPENDIX

A. Sample details

TABLE II: Magnetic properties of Co/Pt films on 440 nm SiO₂. h: total thickness of Co/Pt transducer, K_1 : effective magnetic anisotropy constant, $\mu_0 H_{\rm C}$: coercive field.

h	$h_{ m Pt}/h_{ m Co}/h_{ m Pt}$	K_1	$\mu_0 H_{ m C}$	
(nm)	(nm)	$(kJ m^{-3})$	(mT)	
4.2	1.8/0.7/1.7	230 ± 20	6	
5.5	2.8/0.7/2	380 ± 40	14	
8.2	4.5/0.7/3	400 ± 20	19	

Table I lists the sample parameters relevant to the thermal modeling of our TR-MOKE experiment. In Table II we provide information about the magnetic properties of the Co/Pt transducer films measured by static MOKE at a wavelength of 635 nm. Anisotropy constants were determined from hard-axis hysteresis loops. Coercive fields were measured with the external magnetic field applied along the easy-axis, perpendicularly to the sample plane. Table III summarizes additional information about the samples relevant to the optimization of sample design.

B. Optimization of sample design

TABLE III: Sample parameters. h: total thickness of Co/Pt transducer, C: volumetric heat capacity, A: absorption, R: reflectance (calculated), R_{ex} : measured reflectance, θ_{K} : static Kerr rotation at room temperature and wavelength of 785 nm, $d\theta_{\text{K}}/dT$: temperature dependence of θ_{K} from TR-MOKE data.

Substrate	h	$h_{\mathrm{Pt}}/h_{\mathrm{Co}}/h_{\mathrm{Pt}}$	C	A	R	$R_{\rm ex}$	$ heta_{ m K}$	$d heta_{ m K}/dT$
	(nm)	(nm)	$(10^6~{\rm J}~{\rm m}^{-3}~{\rm K}^{-1})$				(10^{-3} rad)	$(10^{-5} \text{ rad } \text{K}^{-1})$
Al_2O_3	43	43/-/-	2.82	0.22	0.77	0.72		
Al_2O_3	4.2	1.8/0.7/1.7	2.97	0.29	0.26	-	$2.5{\pm}0.3$	-0.5 ± 0.1
$26 \text{ nm SiO}_2/\text{Si}$	4.2	1.8/0.7/1.7	2.97	0.18	0.45	0.43	$1.4{\pm}0.2$	-0.3 ± 0.1
440 nm $\rm SiO_2/Si$	4.2	1.8/0.7/1.7	2.97	0.58	0.08	0.09	$5.6{\pm}0.3$	-1.2 ± 0.1
$440 \text{ nm SiO}_2/\text{Si}$	5.5	2.8/0.7/2	2.94	0.59	0.15	0.16	$5.2 {\pm} 0.4$	-1.1 ± 0.1
$440 \text{ nm SiO}_2/\text{Si}$	8.2	4.5/0.7/3	2.9	0.56	0.27	0.29	$2.6{\pm}0.2$	-0.5 ± 0.1

In TDTR and TR-MOKE measurements the ratio signal $R = -V_{\rm in}/V_{\rm out}$ at short delay times is approximately proportional to $\frac{\sqrt{\Lambda_{\rm S}C_{\rm S}}}{hC}$. $\sqrt{\Lambda_{\rm S}C_{\rm S}}$ is the effusivity of the sample within the penetration depth of the thermal wave and hC is the thermal mass of the transducer. As the latter is drastically reduced in our TR-MOKE experiments, the ratio signals are larger than the ratio signals measured in typical TDTR experiments. Noise in our experimental ratio data mainly originates from the out-of-phase signal. $V_{\rm out}$ is inversely proportional to the effusivity and is on the order of $1 \cdot 10^{-6}$ V for our fluence setting. The noise level in our signal is on the order of $5 \cdot 10^{-8}$ V.

To minimize the time of data acquisition it is thus of interest to maximize the amplitude of the measurement signal. In TR-MOKE thermometry, the signal strength is proportional to the temperature rise ΔT in the transducer (given by the laser fluence as well as the absorption and thermal mass of the transducer), the reflectance R of the sample, and the temperature dependence of the Kerr rotation $d\theta_{\rm K}/dT$ (the equivalent to the thermoreflectance coefficient in TDTR experiments). To guarantee linear response the temperature rise has to be limited, so the signal cannot be increased significantly by increasing the laser fluence.

Absorption behavior and magneto-optic properties of our samples strongly depend on the thicknesses of the Co/Pt and SiO₂ layers. In what follows, we present model calculations to illustrate how R and $\theta_{\rm K}$ can be optimized by adjusting sample parameters. Assuming constant base temperature and stable magnetic properties, $d\theta_{\rm K}/dT$ is proportional to the static Kerr rotation $\theta_{\rm K}$.

We use a transfer-matrix model as described in Ref.³⁵ to calculate absorption A, reflectance R and Kerr rotation $\theta_{\rm K}$ for our Co/Pt samples. Results are summarized in Table III and Fig. 9.

For simplicity we neglect the sandwich structure here and model the Pt/Co/Pt stack as one layer. Optical constants were measured by ellipsometry. As the dependence of the optical constants on layer thickness is weak (below 3 percent), we use the following complex refractive indices for all samples: $n_{\rm Co/Pt} = 3.0 + 5.45i$; $n_{\rm SiO_2} = 1.45$; $n_{\rm Si} = 3.7 +$ 0.007i; $n_{\rm Al_2O_3} = 1.76$. As discussed by Fiedler et al., magneto-optic constants Q for thin Pt/Co/Pt sandwiches depend on the individual layer thicknesses and are fingerprints of the complex structure of the magneto-optic active part of the film, probably determined also by intermixing at the Co/Pt interfaces³⁶. They find $Q_{\rm m} = 0.041 - 0.040i$ for a magnetronsputtered Co layer. For our optical modeling we determine the effective magneto-optic constants for the entire layer by scaling $Q_{\rm m}$ to roughly adjust the model curves for $\theta_{\rm K}$ to our experimental data, compare Fig. 9(d). We find good agreement using Q = 0.0063 - 0.062ifor the 4.2 and 5.5 nm thin films, and Q = 0.0046 - 0.044i for the 8.2 nm thick film.

 $\theta_{\rm K}$ and thus $\theta_{\rm K}/dT$ can critically depend on the thickness of the transducer film, see Fig. 1(c), and has a surprisingly strong dependence on the thickness of the underlying oxide due to optical cavity effects, compare Fig. 1(d). On the quest for the ideal transducer material with high $d\theta_{\rm K}/dT$, one should thus compare the magneto-optic constants rather than the actual Kerr rotation angles measured.

Optical cavity effects also lead to a strong wavelength dependence of A and $\theta_{\rm K}$, so static Kerr rotations presented in Fig. 1 (measured with laser wavelength $\lambda = 635$ nm) and Table III ($\lambda = 785$ nm) differ notably.

The change in Kerr rotation due to the temperature rise in the SiO₂ is more than an order of magnitude smaller than $d\theta_{\rm K}/dT$ based on the temperature change in the metal transducer itself for all samples studied, and thus negligible in our experiment.



FIG. 9: Absorptions A (dotted) and reflectances R (dashed lines) calculated with a transfer-matrix model³⁵ for (a) Co/Pt transducers with varying thickness $h_{\text{Co/Pt}}$ on 440 nm SiO₂. (b) Models and measured reflectance values for $h_{\text{Co/Pt}} = 8.2$ nm (blue), 5.5 nm (green), and 4.2 nm (red) for varying SiO₂ thicknesses. (c) Kerr rotation angles θ_{K} versus $h_{\text{Co/Pt}}$ and (d) versus h_{SiO2} including experimental values. If not stated differently Q = 0.0063 - 0.062i was used as magneto-optic constant.