Role of Magnetic Circular Dichroism in All-Optical Magnetic Recording

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Using magneto-optical microscopy in combination with ellipsometry measurements, we show that all-optical switching with polarized femtosecond laser pulses in ferrimagnetic GdFeCo is subjected to a threshold fluence absorbed in the magnetic layer, independent of either the excitation wavelength or the polarization of the laser pulse. Furthermore, we present a quantitative explanation of the intensity window in which all-optical helicity-dependent switching (AO-HDS) occurs, based on magnetic circular dichroism. This explanation is consistent with all the experimental findings on AO-HDS so far, varying from single- to multiple-shot experiments. The presented results give a solid understanding of the origin of AO-HDS, and give novel insights into the physics of ultrafast, laser controlled magnetism.

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Controlling the magnetic state of a medium with the help of femtosecond laser pulses is a recently emerging and rapidly developing research direction in modern magnetism. The interest to the problem of the ultrafast optical control of magnetism was triggered by pioneering work by Beaurepaire *et al.* [1], who found that excitation of magnetic Ni with a femtosecond laser pulse resulted in the quenching of the magnetization within a few picoseconds. This is much faster than one might expect supposing that the demagnetization is defined by the characteristic time of the spin-lattice interaction in the ground state. Therefore, it was suggested that the ultrafast laser-induced demagnetization was due to ultrafast angular momentum transfer from light to the medium. Soon it was argued however, that such a mechanism is negligible since the total angular momentum of photons in realistic experiments is not large enough to contribute significantly to the observed changes of the magnetization of the media [2].

The discussion about the mechanisms of light-matter interaction in optical control of magnetism was revived after the discovery of all-optical magnetization reversal with the help of circularly polarized (*CP*) light [3]. Namely, it was shown that in a certain range of pulse energies **M** is not only quenched, but is fully reversed when the medium is exposed to one light-helicity while it is not affected if the helicity is opposite. This *All Optical Helicity Dependent Switching* (AO-HDS) was observed in ferrimagnetic GdFeCo in the absence of an external magnetic field. The role of light, light helicity, and angular momentum in AO-HDS are still open fundamental issues and subject of intense discussions in modern magnetism [4].

It was hypothesized that *CP* light acts as a strong effective magnetic field pulse B_{eff} on the spins of the medium through the inverse Faraday effect (IFE). The direction of B_{eff} is then defined by the helicity of the light. Atomistic spin simulations performed under the assumption that light acted as $B_{\text{eff}} = 20$ T were able to describe some key features of AO-HDS, such as the existence of an intensity window at which this phenomenon occurs [5].

Although the existence of such effective magnetic field was demonstrated by the experiments on dielectrics [6], so far this effective field has been described only phenomenologically. The equation describing it was derived in a nondissipative approximation and thus the microscopical origin of the IFE-field in metallic GdFeCo is still a subject of research. Furthermore, these simulations have shown only a qualitative agreement with AO-HDS.

Hence, the microscopic mechanism of the light-matter interaction responsible for AO-HDS has not been revealed yet. In general, the effects of light-matter interaction can be divided into two groups: those that result in an absorption of light and those that lead to a change of the phase of the radiation. Consequently, helicity-dependent effects in optics of magnetic media can be seen as magnetic circular dichroism (MCD) or magnetic circular birefringence (MCB), respectively. It is the aim of this Letter to reveal which one of these two groups is responsible for AO-HDS.

To achieve this goal we performed spectral studies of all-optical switching as a function of light polarization, and intensity. We compared this with the spectral dependencies of both MCD and MCB. We find that AO-HDS can be explained quantitatively with MCD. This explanation is consistent with all the experimental findings on AO-HDS so far, varying from single- to multiple-shot experiments. Furthermore, we show that all-optical switching is

subjected to a threshold fluence of 2.6 ± 0.2 mJ/cm^2, independent of the wavelength and polarization of the excitation pulse.

The structure of the material we studied is SiO_2/AlTi(10)/SiN(5)/$Gd_{26}Fe_{65}Co_9$(20)/SiN(60). The values in parentheses represent the layer thicknesses in nm. The GdFeCo layer is a ferrimagnetic film with perpendicular magnetic anisotropy. The switching was studied by exciting the material with a single laser pulse ($\tau \approx 60$ fs) at different wavelengths using magneto-optical microscopy to determine the final magnetic state of the exposed area. Subsequently, an external magnetic field pulse was applied to bring the ferrimagnet back to its initial state. The initial magnetization was always in the "up" direction, while the polarization was varied from left- (LC), via linearly (LP) to right-handed circularly polarized (RC). It should be noted that it is symmetrically equivalent to fix the helicity and vary the magnetic state from "M up" to "M down" (see Table I). This procedure was repeated about 20 times at every pulse energy and allowed us to measure the switching probability P_σ as a function of fluence F and light-polarization σ. We define the fluence as the pulse energy divided by the beam area (defined as the standard deviation of a Gaussian distribution). The beam area at the sample surface was measured accurately at each wavelength [8]. All measurements were performed at room temperature. Further details of the experimental setup are described elsewhere [3,5].

Figure 1 shows the results for $\lambda = 700$ nm, demonstrating that for "M up" $P_{LC} \leq P_L \leq P_{RC}$, and thus that AO-HDS occurs in a certain intensity window. In the ideal case of no pulse-to-pulse fluctuations of the laser (e.g., fluctuations in the pulse energy and/or the spatial beam profile), P_σ is given by a step function centered at the threshold fluence F_σ, as illustrated in the inset of Fig. 1. More generally, in the realistic case of finite pulse-to-pulse fluctuations (dashed lines in the inset of Fig. 1), P_σ is given by an error function

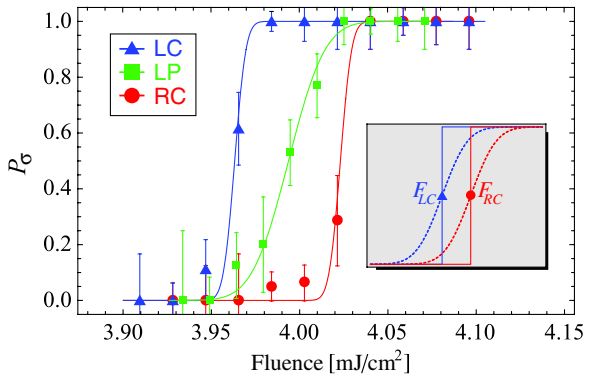

FIG. 1 (color online). Switching probability P_σ as a function of the fluence at $\lambda = 700$ nm for three different polarizations. The measurements with RC and LC pulses were performed at a different time than the ones with LP pulses, and therefore the laser stability ϵ was different. Inset: Illustration of the switching probabilities in case of zero (solid) and nonzero (dashed) laser fluctuations using Eq. (1). F_{LC} and F_{RC} denote the switching threshold of GdFeCo for LC and RC excitation pulses, respectively.

$$P_\sigma(F) = \frac{1}{\sqrt{\pi}\epsilon F_\sigma} \int_0^F dF' e^{-(F'-F_\sigma)^2/(\epsilon F_\sigma)^2}, \qquad (1)$$

where ϵ is a unitless quantity representing the relative magnitude of the pulse-to-pulse laser fluctuation, which varies typically between 0.5%–2%.

We define the window (Δ) of AO-HDS, as

$$\Delta = \frac{F_{RC} - F_{LC}}{\frac{1}{2}(F_{RC} + F_{LC})}. \qquad (2)$$

Note that, while pulse-to-pulse fluctuations do affect the shape of P_σ, the value Δ is not affected by these fluctuations (see inset Fig. 1).

From Fig. 1 it can be seen that at $\lambda = 700$ nm, F_{RC} and F_{LC} are separated by a window of $\Delta \approx 1.5\%$. Furthermore, it shows that switching occurs even when the system is excited with LP light. The threshold fluence at which this

TABLE I. Results from single-shot magneto-optical microscopy measurements, ellipsometry measurements and from calculations at four different excitation wavelengths. F_{LP} denotes the switching threshold fluence using LP light. The errors originate from the uncertainties in the spot sizes, which were accurately measured with the Liu method [7]. The window Δ is defined in Eq. (2). From ellipsometry measurements the refractive index of GdFeCo for LP light n_{LP}, and Δn[a] were obtained. A_{LP} denotes the total absorption of LP light in the GdFeCo layer, and MCD the calculated magnetic circular dichroism in GdFeCo.

	Single-shot magneto-optical microscopy		Ellipsometry measurements		Calculations	
λ (nm)	F_{LP} (mJ/cm^2)	Δ (%)	n_{LP}	$\Delta n(+M)$	A_{LP}(%)	MCD (%)
500	5.04 ± 0.75	1.50 ± 0.21	$1.76 + 2.514i$	$0.024 + 0.052i$	53.0	1.46 ± 0.26
600	4.84 ± 0.96	1.65 ± 0.08	$1.86 + 2.939i$	$0.040 + 0.044i$	59.5	1.51 ± 0.21
700	3.99 ± 0.38	1.49 ± 0.10	$2.41 + 3.454i$	$0.082 + 0.056i$	62.0	1.45 ± 0.15
800	4.90 ± 1.05	1.54 ± 0.10	$2.66 + 3.604i$	$0.088 + 0.050i$	51.8	1.33 ± 0.13

[a]The refractive index for LC and RC light is calculated from $n_{LC}(M) = n_{LP} + \frac{1}{2}\Delta n(M)$ and $n_{RC}(M) = n_{LP} - \frac{1}{2}\Delta n(M)$, respectively, where $\Delta n(-M) = -\Delta n(M)$. Note that the reversal of light-helicity is symmetrically equivalent to the reversal of the magnetization.

occurs, F_{LP}, lies exactly in between F_{RC} and F_{LC}. The experimental results at the different wavelengths are summarized in Table I, showing that Δ is almost independent of the excitation wavelength.

In order to understand the origin of the window it was hypothesized that the latter is due to an effective magnetic field generated by *CP* light via the IFE [3,5]. Pershan *et al.* [9] showed using arguments of thermodynamical equilibrium that *CP* light may act on a medium as an effective magnetic field

$$\mathbf{B}_{\text{eff}} = \epsilon_0 \alpha \mathbf{E}(\omega) \times \mathbf{E}(\omega)^*, \tag{3}$$

where α denotes the magneto-optical susceptibility, and $\mathbf{E}$ the electric field of the incoming pulse. Note that the effective magnetic field is maximum for *CP* light, while it vanishes for *LP* light. In a nondissipative approximation, the strength of this field for *CP* light is given by [4]

$$B_{\text{eff}} = \frac{\theta_F \lambda \tilde{n} F}{\pi M_0 c \tau d}, \tag{4}$$

where θ_F is the Faraday rotation at saturated magnetization, $\tilde{n}$ and d the real part of the refractive index and layer thickness of GdFeCo, respectively, $F \approx 5$ mJ/cm^2 the fluence of the excitation pulse, $M_0 \approx 10^5$ A/m [3] the static magnetization, c the speed of light and $\tau \approx 60$ fs the temporal pulse length.

We would like to stress that although the existence of B_{eff} has been demonstrated experimentally [6], there is no experimental evidence that Eq. (4) is an adequate representation of the effective magnetic field generated by *CP* light in GdFeCo. This equation intrinsically neglects effects of absorption, taking into account only those effects of light-matter interaction that are related to a phase change of the light waves. Consequently, this approach accounts for MCB and neglects MCD in a medium. Note that these two effects have different spectral dependencies.

From spectral measurements of θ_F (inset Fig. 2) and n (Table I), we calculated B_{eff} as a function of the wavelength. This is plotted in Fig. 2(a) (open dots) together with the switching window Δ (closed dots). It can be seen that the calculated B_{eff} has a strong wavelength dependence, similar as in Ref. [10], while the switching window is almost constant in this spectral range.

Alternatively, we will consider MCD as the possible origin of AO-HDS. GdFeCo is well known for its strong magneto-optical effect. Therefore, *RC* and *LC* pulses experience different refractive indices in this material and consequently a difference in absorption of *RC* and *LC* pulses may be present. The refractive indices of the GdFeCo layer were measured using ellipsometry. The thus obtained values for n_{LP}, n_{LC}, and n_{RC} of GdFeCo are shown in Table I. We calculated [8] the total absorption in the GdFeCo layer A_σ for each polarization σ, with a model based on linear absorption in a multilayer stack [11]. For the refractive indices we used for GdFeCo the measured values n_σ given in Table I, and for the other layers the values from Palik [12]. Subsequently, we can calculate the MCD from

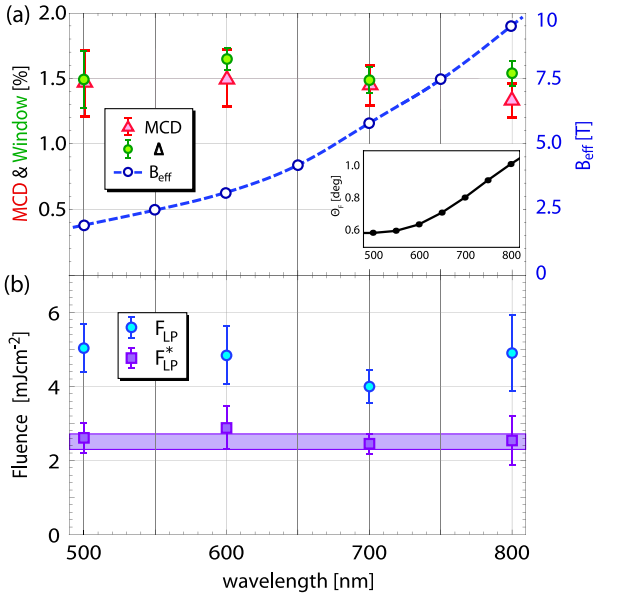

FIG. 2 (color online). (a) The spectral dependence of the window Δ, IFE field B_{eff}, and MCD in GdFeCo. In the inset the spectral dependence of the Faraday rotation θ_F is shown. (b) The switching threshold fluence F_{LP} and the effective threshold fluence F^* plotted against λ. The solid line gives the intersection of the different values of F^*_{LP} and is equal to 2.6 ± 0.2 mJ/cm^2.

$$\text{MCD} = \frac{A_{LC} - A_{RC}}{A_{LP}}. \tag{5}$$

The absorption of *LP* light is equal to $A_{LP} = \frac{1}{2}(A_{RC} + A_{LC})$. The thus obtained values for MCD in GdFeCo [8] are given in Table I. These values show that the switching window in GdFeCo, expressed in terms of relative fluence Δ, exactly corresponds to the relative absorption of *RC* and *LC* light, given by the MCD values. This correspondence can also be seen clearly from Fig. 2(a), where the spectral dependence of MCD and the window Δ are plotted together.

Note that this correlation shows that the switching thresholds for *LC* and *RC* light are different by exactly the same amount as the difference between the total absorption in the GdFeCo layer for *LC* and *RC* light, respectively. The effective switching threshold $F^*_\sigma(\lambda) = F_\sigma(\lambda) A_\sigma(\lambda)$, i.e., the actual absorbed energy density in the GdFeCo layer at which switching occurs, is therefore independent of the polarization: $F^*_{RC}(\lambda) = F^*_{LP}(\lambda) = F^*_{LC}(\lambda)$. In fact, the effective switching threshold is also independent of the wavelength and is equal to $F^* = 2.6 \pm 0.2$ mJ/cm^2, as can be seen in Fig. 2(b).

These results demonstrate that given a 60 fs excitation pulse, all-optical switching depends only on the amount of energy absorbed by the magnetic system, independent of

the wavelength or helicity of the laser pulse. A natural consequence is that switching can be achieved with any polarization as long as the absorbed intensity in the GdFeCo layer is sufficiently strong. Because of different absorption coefficients for *RC* and *LC* light in GdFeCo, the switching threshold is helicity dependent. We can conclude unambiguously that AO-HDS originates from MCD. Considering the errors in our measurements, possible contributions of other mechanisms to AO-HDS are at least 10 times smaller than MCD.

The observation that all-optical switching relies on absorption only, supports the recent simulations showing that a heat pulse can be sufficient to switch the magnetic state of GdFeCo [13]. It was shown that strongly nonequilibrium conditions in multisublattice magnetic systems such as GdFeCo, due to an ultrashort heating pulse, can lead to sublattices that evolve temporarily against their intersublattice exchange interaction. This results in a transient ferromagneticlike state within a picosecond after excitation [14]. On a longer time scale, the sublattices will align antiparallel again due to their intersublattice exchange interaction. This results in an overall reversal of the net magnetization.

It is interesting that with multiple-shot exposures it is possible to obtain helicity-dependent switching of large areas, even for intensities much higher than the regime where AO-HDS occurs with a single pulse [3]. This observation is illustrated in Fig. 3. *CP* pulses are swept over the surface (from left to right in Fig. 3) with a 1 kHz repetition rate. For the given helicity, more energy is absorbed in the black (''*M* down'') than the white domain (''*M* up'') due to MCD. In the center of the excited area, the intensity is high enough to switch both magnetic states. Near the edge of the excited area however, there is a small ring-shaped region where only black domains switch to white. The left side of the white ring will not switch back to black when sweeping the beam to the right. Hence, the beam will leave a trail of white domain. This trail is visible when the underlying **M** is ''down'' (black) but invisible when it is up (white), in exact agreement with the observations in Ref. [3]. In the same article another multiple-shot experiment on GdFeCo is shown. The beam was fixed at a position near a domain wall with a repetition rate of 1 kHz. In the Supplemental Material we illustrate how MCD can also explain very accurately the observations in this experiment [8].

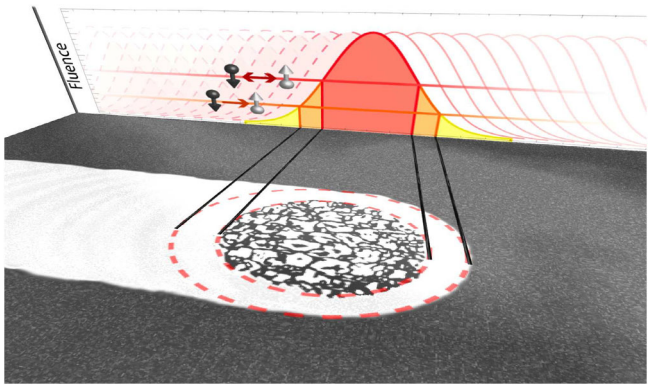

FIG. 3 (color online). Illustration of multiple-shot switching with *CP* excitation pulses. In the center of the excited area, both magnetic states can switch because of the high fluence, while at the edges (i.e., between the dashed borders) helicity-dependent switching occurs and therefore a single domain state is formed. Hence, by sweeping multiple *CP* excitation pulses over the surface, a large area can be switched.

The effective switching threshold $F^* \approx 3\ \mathrm{mJ/cm^2}$, compares favorably with the $2 \times 10^4\ \mathrm{mJ/cm^2}$ of writing fluence per bit in current hard drives and FLASH solid state memories, and even with the $10\ \mathrm{mJ/cm^2}$ in magnetic random access memory [15]. The present switching area, however, is on the order of micrometers compared to tens of nanometers of bit size in the afore-mentioned technologies. Reducing the spot size of the excitation pulse, for example, with optical near-field microscopy [16] or plasmonic antennas [17], could lead easily to a smaller switched area, as was recently demonstrated [18]. This would make all-optical magnetic recording a promising alternative to conventional recording technologies with respect to both writing speed [5] and energy per bit.

The new understanding of AO-HDS may have far reaching consequences for the realization of all-optical magnetic recording. The intensity regime at which this phenomenon occurs is the most relevant parameter for possible applications of all-optical magnetic recording, as in this regime the final magnetic state is completely determined by the helicity of the excitation pulse. For such applications, however, it is necessary to overcome pulse-to-pulse fluctuations of the laser intensity to have fully controllable switching. The ultimate solution would be the enhancement of the switching window, or equivalently the MCD value in the GdFeCo layer. The latter could be easily enhanced by optimizing the design of the multilayer structure, even without changing the properties of the magnetic layer. As an example, we calculated the MCD in GdFeCo for different structures of the capping layer, see Fig. 4. In the calculations, the 60 nm thick SiN capping layer was replaced by SiN(y)/amorphous-Si(x)/SiN(5), where the values between parentheses represent the layer-thickness in nm. For $y = 5$ and $x = 40$ the window appears to be more than doubled, i.e., from 1.5% to roughly 3.5%. Adjusting the material and/or thickness of other layers, including the magnetic layer, could enhance the window even further.

Summarizing, we have shown that all-optical switching in GdFeCo is subjected to a threshold fluence of $2.6 \pm 0.2\ \mathrm{mJ/cm^2}$ absorbed by the magnetic layer, independent of either the wavelength or the polarization of the excitation pulse. With MCD we can quantitatively explain AO-HDS. Furthermore, this explanation is consistent with all the

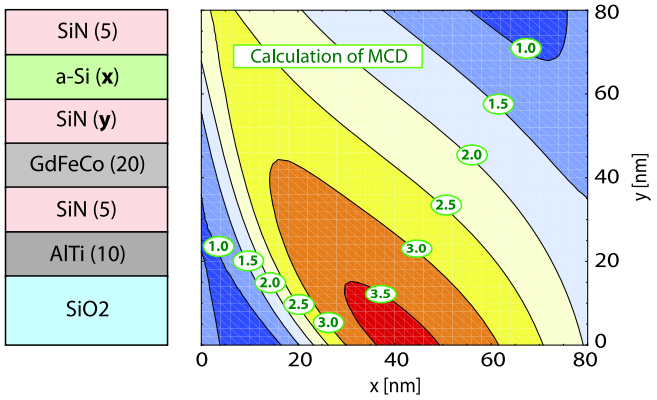

FIG. 4 (color online). Calculation of the MCD in the GdFeCo layer (in percentages) at 700 nm for different thicknesses of top layers. The multilayer structure is shown left, where the values in parentheses indicate the layer thickness in nm.

experimental findings on AO-HDS so far, varying from single- to multiple-shot experiments. The new understanding of this phenomenon enables the possibility to design multilayer structures with an enhanced intensity window in which AO-HDS occurs, which is important for its potential application for all-optical magnetic recording.

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[1] E. Beaurepaire, J. C. Merle, A. Daunois, and J. Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996).
[2] B. Koopmans, M. van Kampen, J. T. Kohlhepp, and W. J. M. de Jonge, Phys. Rev. Lett. **85**, 844 (2000).
[3] C. D. Stanciu *et al.*, Phys. Rev. Lett. **99**, 047601 (2007).
[4] A. Kirilyuk, A. V. Kimel, and T. Rasing, Rev. Mod. Phys. **82**, 2731 (2010).
[5] K. Vahaplar *et al.*, Phys. Rev. Lett. **103**, 117201 (2009).
[6] A. Kimel *et al.*, Nature (London) **435**, 655 (2005); F. Hansteen, A. Kimel, A. Kirilyuk, and T. Rasing, Phys. Rev. Lett. **95**, 047402 (2005); T. Satoh *et al.*, *ibid.* **105**, 077402 (2010).
[7] J. M. Liu, Opt. Lett. **7**, 196 (1982); J. Chalupsky *et al.*, Opt. Express **15**, 6036 (2007); A. R. Khorsand *et al.*, *ibid.* **18**, 700 (2010).
[8] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.108.127205 for (i) a detailed explanation of how we obtained the MCD value of GdFeCo layer with ellipsometry measurements and absorption profile calculations, (ii) the demonstration of how switching at a domain wall using multiple CP pulses, as observed in Ref. [13], can be explained with MCD, and (iii) a brief explanation of the method we used for accurate spot size characterization.
[9] P. S. Pershan, J. P. Vanderzi, and L. D. Malmstro, Phys. Rev. **143**, 574 (1966).
[10] D. Steil, S. Alebrand, A. Hassdenteufel, M. Cinchetti, and M. Aeschlimann, Phys. Rev. B **84**, 224408 (2011).
[11] D. L. Windt, Comput. Phys. **12**, 360 (1998).
[12] *Handbook of Optical Constants of Solids II*, edited by E. D. Palik (Academic Press, San Diego, 1998).
[13] J. H. Mentink *et al.*, Phys. Rev. Lett. **108**, 057202 (2012); T. Ostler *et al.*, Nature Commun. (to be published).
[14] I. Radu *et al.*, Nature (London) **472**, 205 (2011).
[15] J. P. Liu, E. Fullerton, O. Guteisch, and D. Sellmyer, *Nanoscale Magnetic Materials and Applications* (Springer-Verlag, New York, 2009).
[16] M. Savoini *et al.*, Opt. Lett. **34**, 3571 (2009).
[17] P. Biagioni, J. S. Huang, L. Duo, M. Finazzi, and B. Hecht, Phys. Rev. Lett. **102** (2009).
[18] M. Savoini *et al.*, Moris 2011, Book of Abstracts.