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Direct measurement of group delay dispersion in metamagnetics for ultrafast pulse shaping

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Abstract: In this paper, we explore the use of magnetic resonant metamaterials, so called metamagnetics, as dispersive elements for optical pulse shaping. We measure both positive and negative group delay dispersion (GDD) values in a metamagnetic material using the multiphoton interference phase scan (MIIPS) technique and show pulse temporal profiles numerically. The results are compared with finite element models. These GDD properties of metamagnetics, along with previously shown tunability and loss control with gain media, enable their use in ultrashort pulse optical applications.

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References and links
1. Introduction

Metamaterials offer the potential to engineer optical responses for technological and scientific needs. Media with negative permittivity and permeability have renewed discussions on the fundamental aspects of group velocity and its dispersion [1–5]. Metamaterials with custom reflectivity and group delay dispersion (GDD) are essential for many applications in ultrafast optics. Existing metamaterials enable new possibilities for GDD management, including tunable resonances of metamagnetics in the visible range [6] and loss compensated negative index materials [7]. Synthetic dispersion in metamaterials can be designed to either maximize or compensate GDD within a given band. Here, we test a metamagnetic with engineered dispersion using a multiphoton interference technique and demonstrate that a desired phase step for shaping 10-15 fs pulses (spectrally centered around the magnetic resonance at 800 nm) can be achieved with a relatively thin 136-nm sample. Hence, by proving effective dispersion engineering at a nanoscale, our study brings realism to integrated ultrafast nanophotonics and coherent control.

The ability to synthesize broadband, ultrafast optical pulses has increased over the last few decades to enable optical waveform generation [8], i.e. the design of nearly arbitrary temporal pulse profiles at optical frequencies. This has led to a rapid development of pulse characterization [9–11] and nonlinear optical spectroscopy [12,13] techniques employing pulse shaping. Essential to the control of optical pulse profiles is the introduction of tailored dispersion into the optical train, typically accomplished by spatially separating and addressing individual frequency channels or collinearly done with acousto-optic modulators. Nanostructured systems offer the possibility of compact, monolithic devices for complicated phase engineering [4,14]. In addition, controlling the phase modulation of pulses can enable the spatial concentration of ultrafast energy [15–17].

For an ultrafast pulse with at least a few optical cycles (the bandwidth $\Delta \omega$ is much smaller than the central frequency $\omega_c$), a slowly varying envelope formalism can be used where a complex representation of the field in the frequency domain can be written as [18]:

$$\tilde{E}(\omega) = E(\omega - \omega_c)e^{-i\varphi(\omega - \omega_c)}$$

where $E(\omega - \omega_c)$ is the frequency spectrum of the pulse centered at the carrier frequency, $\omega_c$, and $\varphi(\omega - \omega_c)$ is the frequency-dependent spectral phase of the pulse. Dispersive effects can be formulated in a general way that is also applicable to the case of sub-wavelength thick materials. Specifically, $\varphi(\omega)$ is the spectral phase imparted on a pulse transmitted through a material, and it can be found directly from the complex transmission:

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where $t_{12}, t_{23}$ and $r_{12}, r_{23}$ are the complex Fresnel transmission and reflection coefficients at the air-material and material-substrate boundaries and $\beta(\omega)$ is the optical thickness of the material given at normal incidence by $\beta(\omega) = \omega n(\omega) L / c$ where $\omega$ is the frequency of incident light, $n$ is the effective refractive index of the material, and $c$ is the speed of light. Because $\tilde{E}_{\text{out}}(\omega) = \tilde{E}_{\text{in}}(\omega) \tilde{T}(\omega), \varphi_{\text{out}}(\omega) = \varphi_{\text{in}}(\omega) + \varphi(\omega)$, so the spectral phase imparted by some material is the difference between the phase on the pulse before and after that material.

In a band-limited pulse, one can expand the phase on a pulse in a Taylor series about the carrier frequency and the leading two terms do not affect the shape of the temporal envelope [18]. The first term in the series that does affect the shape is the GDD, and it is defined as

$$GDD \equiv \frac{d^2 \varphi(\omega)}{d\omega^2} \bigg|_{\omega = \omega_0}$$

Consequently, the GDD is the most important property of a material in ultrafast optics. Another commonly used description of the dispersion of a homogenous material is the group velocity dispersion (GVD), which is GDD per physical length of the material.

2. Experimental setup

In general, measurement of the change in the pulse’s electric field (amplitude and phase) versus time or frequency also provides sufficient knowledge to reconstruct its complex transmission. There are a number of techniques developed for making such a measurement, including FROG, SPIDER, etc [19]. The multiphoton intrapulse interference phase scan (MIIPS) technique, which is implemented with a calibrated pulse-shaping device capable of adding suitable reference phases to the pulse under test [20], is insensitive to the constant and first derivative terms, but directly measures the second derivative term (GDD).

In this paper, we explore the use of magnetic resonant metamaterials, so called metamagnetics, as dispersive elements for optical pulse shaping. To demonstrate a large range of both positive and negative GDD values, we measure the GDD in a metamagnetic material directly and show pulse temporal profiles numerically. These new properties of metamagnetics, along with previously shown tunability and loss control with gain media, enable their use in ultrashort pulse optical applications.

Our metamagnetic samples are composed of subwavelength-periodic metal strip pairs with dielectric spacers [21]. The schematic, FE-SEM, and AFM images of a fabricated sample are shown in Fig. 1. When using normally incident transverse magnetic (TM) polarized light, the magnetic field of the incident plane wave is aligned along the metal strips separated vertically by a distance that results in a magnetic resonance near 800 nm. The electric field couples into the periodic silver lines with a resonance near 525 nm, which is slightly less than twice the period of the grating structure. These resonances can be clearly seen in Fig. 1(b) that compares the experimental transmission measurement for the structure of Fig. 1(a) to the simulated data from the finite element method (FEM) and spatial harmonic analysis (SHA) [22].
The spectral phase and the GDD of the transmitted light for the structure dimensions given in Fig. 1(a) was calculated using FEM, and these are shown in Fig. 2. The materials used were air \((n = 1, k = 0)\), glass \((n = 1.52, k = 0)\), \(\text{Al}_2\text{O}_3\) (dispersion from [23]), and Ag (Drude-Lorentz model with a loss factor of 3 from [24], which is typical for nanomaterials with internal grained structure [25]). An electric resonance near 500 nm and a magnetic resonance near 800 nm are evident from the transmission spectra (Fig. 1(b)) and in the phase and GDD curves (Figs. 2(a), 2(b)). Near these resonances the spectral dispersion of the material changes quickly and this results in large GDD values.

To experimentally measure the GDD of a metamagnetic material, we used the MIIPS technique [20] with an ultrafast oscillator, the Griffin-5, from KMLabs that had a 100 nm root-mean-squared (RMS) spectral bandwidth capable of transform limited pulses below 15 fs. The RMS bandwidth is defined as the full width at half of the average maximum across the spectrum, and the shape of the spectral bandwidth was rounded on the sides but roughly flat on top. As a control experiment the dispersive glass SF5 of 3.2 mm in thickness was characterized. Good agreement was obtained between MIIPS and spectroscopic ellipsometry followed by calculation of GDD from the refractive index dispersion. At 800 nm, MIIPS retrieved a GDD of \(442 \pm 52\) fs\(^2\), and the derived GDD from ellipsometry was \(429 \pm 2\) fs\(^2\). This agrees well with a Cauchy model using data from Schott that gives a GDD of 429 fs\(^2\) at 800 nm. The GVD using the ellipsometry and Schott value is 134 fs\(^2\)/mm.

3. Results and discussion

The measured GDD for our metamagnetic material is shown in Fig. 2(c) (solid green line) and is compared with the FEM and SHA simulations (orange line and red circles, respectively).
The FEM and SHA simulations trace together, and reasonable agreement can be seen between the measurements and simulations. One can observe that the experimental dispersion lies between about ±2000 fs² and crosses zero near 801 nm. The level of agreement between simulation and experiment reflects a possible difference between the simulation design and the real geometry. It includes inhomogeneous size distribution over the sample, roughness, and asymmetry in the trapezoidal shape, which are taken into account only in average. Each point of the representative measured spectrum is a moving-average over 12 consequent experimental points corresponding to 4 nm of spectral interval. The error bars in Fig. 2 (c) show an estimate of the root-mean-square deviation from the averaged values.

The effect of this dispersion on the temporal shape of a 10 fs pulse (95.3-nm Gaussian FWHM spectrum), with the inflection point of the phase step placed at the central frequency, can be seen in Fig. 3 and Fig. 4. For simplicity, a Gaussian profile was used such that a simple analytical pulse shape could be inspected for changes due to material GDD. The effect was calculated with the Fourier analysis of the dispersion using FEM (Fig. 3) and through a finite-difference time-domain (FDTD) simulations (Fig. 4). For the Fourier analysis, the structure was as given above and the center wavelength of laser spectrum was 801 nm. For the FDTD calculation, a scaled version of the structure was used with a period of 240 nm and a bottom-up thickness-material sequence being 10-nm-Al₂O₃|30-nm-Ag|40-nm-Al₂O₃|30-nm-Ag|10-nm-Al₂O₃ layers (here Ag permittivity is modeled with a loss factor of 1), and the center wavelength of a 10-fs pulse was 774 nm.

Fig. 3. Simulated effect of the 136-nm thick metamagnetic material in the time domain using Fourier analysis. Simulation of the effect that a magnetic grating with 136-nm physical thickness of material in the grating lines has on 10-fs and 30-fs ultrafast pulses. The envelopes of the input and output field amplitudes are shown in the left and right panels, respectively. A strong dip with a small broadening in the output pulse can be seen clearly in Panels (b) and (d) for 10-fs and 30-fs input pulses, respectively.

Fig. 4. FDTD simulation of an input pulse traveling through a metamagnetic material. (a) The 10-fs incident pulse field amplitude. (b) Reflected pulse. (c) The transmitted "pulse" shows a strong dip indicative of a π-phase step near the center of the bandwidth of the pulse. Inset in (c) shows the narrow-band spectrum of the incident pulse (orange) along with the broadband reflectance (blue) and transmittance (red) spectra (colors match those in panels (a)-(c)).

In both types of simulations some broadening, related to the sharpness of the phase step transition, is observed due to non-zero dispersion over a large bandwidth. More interestingly,
there is a strong dip in the center of the temporal field intensity profile of the pulse. This is
due to the fact that the dispersion is anti-symmetric across the full laser bandwidth. The
resulting transmitted pulse has approximately half of the pulse’s spectral bandwidth chirped
with positive GDD and the rest chirped with negative GDD, resulting in a field intensity dip
seen in the time domain. The reflected pulse, however, maintains its temporal profile
(Fig. 4(b)), which indicates most of the reflection occurs due to the impedance mismatch
going into the material. The inset of Fig. 4(c) indicates that the pulse probes only the magnetic
resonance without engaging a blue-shifted electric resonance.

Note that the applicability of any material for short pulse shaping depends on the
dispersion in transmission. An example of 10 fs pulses presented in Figs. 4(a)-4(c) shows
reasonably high amplitudes of the transmitted and reflected pulses despite near-zero
transmission at 800 nm. Moreover, the MIIPS technique we used in the experiments requires
pulses of sufficient intensity be transmitted to interact with a nonlinear material. This
demonstrates directly and conclusively that the transmission of the material is not a barrier for
its use.

Potential control over the dispersion properties is also important. Here we present direct
measurements of the GDD in a metamagnetic grating, which is wavelength dependent. By
changing the resonance position (by changing the liquid crystal temperature or applied
voltage or through angle of incidence changes) one can control the GDD at certain
wavelengths. The temperature control over the magnetic resonance position was previously
demonstrated for a metamagnetic grating combined with liquid crystal [6]. Another way to
control the resonance properties was demonstrated in the pump-probe 120 fs experiments on
the hybrid of a cross-gratings and gain medium (rhodamine 800 in epoxy) [7].

4. Conclusions

In conclusion, GDD values of metamaterials with strong resonances, including magnetic
resonances, can create changes in the spectral phase that are several times larger than a typical
dispersive glass like SF5 that is four orders of magnitude thicker; and if one considers a
metamagnetic structure to act like bulk material, the GVD for a metamagnetic material is
about four orders of magnitude larger than SF5. Possible applications of dispersive
metamaterials include pulse shaping and compression with less than a micron of material. In
the future, more carefully designed metamaterials should yield thin and precise temporal pulse
shape control. Closely spaced, narrow, and interacting resonances can be positioned within
the bandwidth of a pulse providing complex dispersion features [14]. Such highly tailored
profiles could potentially be useful in ultrafast systems, integrated nanophotonics, and
coherent control. By controlling the GDD in a metamaterial, it could be possible in the future
to compress or stretch ultrafast laser pulses with only nanometers to microns of material
thickness rather than the millimeters to centimeters of glass and path length used in a typical
ultrafast laser compressor. Future efforts will explore more advanced designs of
metamaterials and perform GDD measurements over a broader bandwidth of laser
wavelengths to look at the effects of dispersion on even shorter pulses.

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