

VILNIUS UNIVERSITY
FACULTY OF PHYSICS
LASER RESEARCH CENTER

Gytis Zaremba

**Transient absorption investigation of photonic microstructures with
dielectric coatings**
MASTER'S THESIS

Laser Physics and Optical Technology program

Student	Gytis Zaremba
Supervisor	Dr. Mikas Vengris
Director of Laser Research Center	Dr. Dalia Kaškelytė

Vilnius 2026

Contents

1	Introduction	3
2	Theory and literature	5
2.1	Transient absorption measurement	5
2.2	Photonic crystals. Woodpile structures	8
2.3	Epsilon-near-zero (ENZ) materials	12
2.4	Pump-probe investigation of photonic crystals	14
2.5	Tantalum pentoxide (Ta_2O_5)	15
2.6	Pump-probe investigation of grating resonances	16
3	Experiment methodology	19
3.1	Samples	19
3.2	Experimental scheme	21
3.3	White light continuum generation and beam sizes	22
4	Experimental results	26
4.1	Transmittance of the samples	26
4.1.1	AZO coated photonic crystals	26
4.1.2	Ta_2O_5 coated SiO_2 grating	28
4.2	Pump-probe investigation of the AZO coated PCs	30
4.3	Pump-probe investigation of Ta_2O_5 coated SiO_2 grating	37
5	Main results and conclusions	45
6	References	46
7	Santrauka	50
8	Summary	51

1 Introduction

Photonic structures are widely studied because they allow light propagation to be controlled through geometry rather than only through the intrinsic properties of the material. Periodic structures such as photonic crystals and dielectric gratings can modify the optical density of states, form photonic band gaps, support localized optical modes, and produce narrow resonant features. These effects make them attractive for applications in optical filtering, sensing, waveguiding, resonant enhancement, and wavelength-selective light manipulation.^{1,2}

Although photonic crystals and dielectric gratings differ in geometry, their optical response is governed by the same general principle: periodic modulation of the refractive index modifies the propagation of light and can create resonant optical modes.^{3,4} These resonances arise when incident light couples to modes supported by the structure, leading to strong spectral features in transmission or reflection.^{4,5} Because the resonance condition depends on the effective refractive index, absorption, and geometry of the structure, even small pump-induced changes in the optical properties of the material can shift the resonance wavelength or modify its amplitude.^{5,6} Transient absorption spectroscopy is therefore a useful tool for studying these effects, as it can monitor pump-induced changes in transmission or reflection as a function of both wavelength and delay time.⁶

In this work, two types of photonic structures are investigated: aluminum-doped zinc oxide (AZO) coated photonic crystals and a tantalum pentoxide (Ta_2O_5) coated silicon dioxide (SiO_2) grating. Photonic crystals provide spectral selectivity through periodic modulation of the refractive index and the formation of photonic modes, while dielectric gratings can support guided-mode or Fano-type resonances through diffraction and phase-matching effects. These resonances are highly sensitive to changes in the local refractive index and absorption, making such structures promising platforms for studying light-matter interaction in thin-film-coated systems.^{1,2}

The functionality of photonic structures can be further expanded by integrating them with optically active coatings. Structures made only from passive dielectric materials usually exhibit limited ultrafast response under optical excitation. In contrast, transparent conducting oxides (TCOs), such as AZO, have high free-carrier densities and strongly dispersive optical properties. Optical excitation can modify the carrier density and scattering rate in these materials, leading to wavelength-dependent changes in the complex dielectric function, especially near the plasma frequency or epsilon-near-zero region.^{7,8,9} Therefore, combining TCO coatings with photonic structures offers a way to couple ultrafast material dynamics with resonant photonic behavior. Similar principles can also be applied to dielectric-coated gratings, where pump-induced refractive index or absorption changes may shift or modulate narrow optical resonances.

Such hybrid structures are interesting not only in terms of their steady-state optical properties, but also because of their dynamic response. Pump-induced changes in refractive index, absorption, or carrier concentration can modify the spectral position, amplitude, and relaxation dynamics of optical resonances. This makes coated photonic structures promising candidates for ultrafast optical modulators, wavelength-selective switches, and dynamically tunable photonic components. In resonant systems, even relatively small material changes can produce pronounced optical modulation if they occur in the spectral region of a photonic mode or resonance.^{2,10}

The short-wave infrared region is particularly relevant for this type of study. Many microscale photonic structures fabricated by optical or lithographic methods have periods and feature sizes that naturally support resonances in the near-infrared or short-wave infrared range. In addition, the optical response of TCOs is often strongest in the near-infrared region close to their plasma or ENZ wavelength.^{7,8} The spectral range from approximately 1 to 2 μm is therefore well suited for investigating the interaction between photonic resonances and pump-induced changes in active thin films.¹¹

Transient absorption spectroscopy provides a direct way to investigate such pump-induced dynamics. The technique was originally developed in the form of flash photolysis experiments in the 1940s, while modern femtosecond laser systems now allow photophysical and photochemical processes to be studied on ultrafast time scales.^{12,14} In a pump-probe experiment, an excitation pulse induces a transient change in the sample, while a delayed broadband probe pulse monitors the resulting changes in transmission or reflection. This makes the method suitable for studying carrier relaxation, optical nonlinearities, resonance shifts, and transient modulation in coated photonic structures.

However, conventional pump-probe setups typically require relatively large and homogeneous sample areas, which limits their applicability to microscale patterned structures. To overcome this limitation, a micro pump-probe setup was used in this work. Reflective microscope objectives were employed to focus the probe beam to a spot size of approximately 9 μm , enabling measurements on photonic crystal structures as small as $50 \times 50 \mu\text{m}$. In order to study the response of the samples in the short-wave infrared range, white light continuum generation was optimized to obtain a broadband probe spectrum extending from 1 μm to 2.08 μm , and its stability and generation conditions were characterized.

The goal of this work is to investigate the ultrafast optical dynamics of coated microscale photonic structures in the short-wave infrared spectral range. Specifically, AZO coated photonic crystals and a Ta_2O_5 coated SiO_2 grating are studied under ultraviolet excitation using transient transmission and transient reflection measurements. By comparing these two platforms, the work

evaluates how photonic resonances influence the transient optical response of coated structures and assesses their potential for ultrafast optical modulation and wavelength-selective switching.

2 Theory and literature

2.1 Transient absorption measurement

A transient absorption (TA) experiment, also known as a pump-probe experiment, aims to detect how a sample's absorbance changes when it is optically excited compared to when it is not. This involves using two light pulses: the pump pulse, which excites the sample, and the probe pulse, which monitors how the sample's transmission changes because of that excitation. 0.1 % to tens of percents of absorbing molecular states in solution, or a corresponding photoexcited carrier population in solid-state materials, is typically excited during a pump-probe experiment. Probe pulse should be sufficiently weak not to trigger unwanted multiphoton or nonlinear absorption effects, whereas the pump pulse should be intense enough to effectively excite the sample.¹⁵

A low intensity probe pulse is directed through the sample after a controlled delay time τ relative to the pump pulse. The result is a difference absorption spectrum, which is calculated by subtracting the absorption spectrum of the sample in the ground-state from the excited sample, as shown in equation 1:

$$\Delta A(\lambda) = \log_{10} \left(\frac{I(\lambda)_{unpumped}}{I(\lambda)_{pumped}} \right) \quad (1)$$

By systematically varying the delay time τ and recording a ΔA spectrum at each value, a dataset $\Delta A(\lambda, \tau)$ is generated, showing how the absorption changes over both time and wavelength. This dataset reveals dynamic processes occurring within the investigated system, such as energy transfer in excited electronic states, electron or hole relaxation, trapping and recombination in solids, as well as molecular processes including isomerization, intersystem crossing, or proton transfer. To interpret this information, global analysis techniques are often employed. A key advantage of time-resolved absorption spectroscopy over time-resolved fluorescence is its ability to monitor the evolution of non-emissive states, which is particularly important in solid-state and amorphous materials where radiative emission may be weak or absent.^{15,16}

In general, a difference absorption (ΔA) spectrum reflects contributions from several distinct processes:

1. The main contribution is caused by ground-state bleaching. When the pump pulse excites a portion of the absorbing species, the population of the ground state is reduced. In molecular

systems, this corresponds to depletion of molecular ground states, while in solid-state materials it is often associated with band filling or occupation of electronic states near the band edges. As a result, the sample's absorption in these spectral regions decreases, leading to a negative signal in the ΔA spectrum.^{15,16}

2. Stimulated emission is another important process contributing to the ΔA spectrum. In a two-level system, the probability of absorption and stimulated emission is described by identical A_{12} and A_{21} Einstein coefficients. Once the pump pulse populates the excited state, the probe pulse can induce stimulated emission, resulting in amplification of the probe and a negative ΔA signal. In solid-state systems, stimulated emission or optical gain may occur between populated electronic states and lower-energy states, though it does not necessarily correspond to detectable fluorescence, but at high excitation densities can manifest as a very fast decrease of the signal. Spectrally, this contribution often appears near or slightly redshifted (Stokes-shifted) from the absorption band.^{15,16}
3. A third contribution to the ΔA spectrum arises from excited-state absorption, which occurs when excited species absorb additional probe photons and transition to higher electronic states. In molecular systems, these are discrete electronic transitions, while in solids they may involve interband, intraband, or defect-related transitions. As a result, a positive signal appears in the ΔA spectrum in the spectral regions where these transitions take place.^{15,16}
4. A fourth potential contribution arises from absorption by photoproducts or long-lived excited states. In molecular systems, these may include triplet states, charge-separated states, or isomerized species. In solid-state and amorphous materials, analogous contributions can originate from trapped carriers, defect states, or metastable electronic configurations, which often exhibit distinct absorption features and longer lifetimes. In transmission-based pump-probe measurements, the detected signal may also contain contributions from pump-induced changes in sample reflectivity, since variations in reflectance directly affect the amount of transmitted probe light reaching the detector. Such contribution results in a positive signal in the ΔA spectrum.^{15,16}

On ultrafast (femtosecond) timescales, transient absorption signals may include additional contributions beyond those arising from population dynamics. These non-resonant effects are often collectively referred to as coherent artifacts or cross-phase modulation. Since transient absorption is governed by third-order nonlinear optical processes, described by the third-order susceptibility $\chi^{(3)}$, the signal can include light-matter interactions that are not directly related to electronic state transitions.

These undesired contributions result from coherent interactions rather than sequential absorption and emission events. Typically, they can be minimized or excluded from analysis by ignoring the earliest time points in the kinetic trace, where such artifacts dominate. Alternatively, if these effects are to be accounted for, their physical basis must be modeled explicitly.

One prominent non-resonant effect is cross-phase modulation (CPM), which arises from transient changes in the refractive index of the solvent and the optical cuvette, or the sample itself, induced by the intense pump pulse. This phenomenon can generate oscillatory features near time-zero delay, obscuring real population dynamics.¹⁷ In practice, such artifacts can be identified and corrected by performing reference measurements using a solvent-only sample in the same optical cell, if the sample is in liquid form.

The resolution of an ultrafast transient absorption measurement primarily depends on the laser pulse duration. Laser pulses as short as 5 femtoseconds are now available for transient absorption spectroscopy, enabling the investigation of extremely fast processes. However, such short pulse durations come with an inherent limitation: a shorter pulse necessarily has a broader spectral bandwidth. For instance, a 10-fs pulse centered at 800 nm exhibits a spectral bandwidth of approximately 100 nm at full width at half maximum (FWHM).

This broad spectral coverage can lead to a significant drawback in experiments where spectral selectivity is important. In complex systems where multiple chromophores absorb in distinct but closely spaced wavelength regions, a broad-band excitation pulse may simultaneously excite several components. As a result, it becomes challenging to isolate and interpret the excited-state dynamics of a specific chromophore or pigment. Therefore, researchers must carefully balance temporal precision and spectral specificity, depending on the goals and complexity of the system under study.

The advancement of fast and sensitive pump-probe imaging methods has opened up applications across various fields, including materials science¹⁸, biology¹⁹, and art²⁰.

In material characterization, certain nanostructured materials, such as graphene, exhibit exceptionally high optical susceptibilities but very weak fluorescence, making them ideal targets for pump-probe spectroscopy. Femtosecond transient absorption spectroscopy has been extensively used to investigate the ground and excited state dynamics of nanomaterials like single-walled carbon nanotubes, offering deep insights into their photophysical behavior. These include the role of triplet states, the influence of environmental factors such as solvent dielectric properties, and the dynamics of exciton generation, diffusion, and recombination. However, traditional bulk spectroscopy lacks spatial resolution and averages out inherent sample heterogeneities. For instance, graphene often shows variations in thickness, structure, and substrate interaction, while individual nanoparticles can differ in size, aggregation, and local environment - factors that significantly impact their transient absorption signatures. To achieve reliable data, especially for carbon nanotubes, it is crucial to study samples of uniform type and size, which can be ensured through techniques like gradient ultracentrifugation.¹³ One of the more exciting studies were done by Gabriel et al.²¹, who designed a pump-probe microscopy system that allows for photoexcitation of a single nanostructure at one point and probing it at a different location. Using this setup on silicon nanowires, they were able to directly

observe how a charge cloud, generated by localized photoexcitation, spreads along the nanowire axis. The time-resolved imaging revealed distinct phases of behavior, including fast diffusion and recombination of free carriers, followed by slower migration of trapped carriers.

In biology, pump-probe techniques have found valuable applications, particularly for investigating ultrafast processes such as energy transfer, charge separation, and conformational changes in biomolecules. Femtosecond transient absorption has been used to study light-harvesting pigment-protein systems in both extracted spinach leaves and bacterial membranes, revealing energy migration pathways and kinetic rates of charge separation.²² Similarly, sub-20 fs pump-probe systems have probed carotenoid-to-chlorophyll energy transfer in bacterial light-harvesting complexes.²³ In live-cell imaging, pump-probe optical microscopy provides label-free contrast for non-fluorescent biomolecules, such as melanin and hemoglobin, by exploiting mechanisms like excited state absorption and ground-state depletion, enabling high-resolution mapping of biochemical composition in tissues - including early detection of melanoma.²⁴

In the field of art, pump-probe spectroscopy offers a powerful, non-invasive method to explore subsurface layers of paintings. By analyzing the transient optical responses of different materials, this technique can differentiate pigments and structural features beneath the surface. It allows for the creation of virtual cross-sections, aiding in authentication, preservation, and the detailed study of artistic methods without causing any harm to the original work.²⁵

In this work, the region of interest in the sample's transient absorption spectrum lies in the range of 1-2.5 μm . Consequently, achieving sufficient supercontinuum redshift with conventional femtosecond solid-state lasers presents a significant challenge. From the overview of Dubietis et al.²⁶ it can be seen that only a few works have reached 2 μm wavelength mark while using fundamental femtosecond laser radiation.

2.2 Photonic crystals. Woodpile structures

In wave optics, layered structures - such as interference coatings - with alternating high and low refractive indices spaced at quarter-wavelength intervals have become essential components. This periodic modulation in space creates a photonic band gap in the electromagnetic dispersion relation, at least for light traveling perpendicular to the layers. Within this gap, wave propagation is prohibited, making these dielectric multilayer structures highly effective as reflective mirrors, for example, in Fabry-Perot resonators. Extending this concept, a fully three-dimensional refractive index modulation with periodicity of $\lambda/2$ can produce a complete photonic band gap near wavelength λ , regardless of the direction of light propagation - similar to how electronic band gaps form in crystals. If this photonic band gap overlaps with the electronic band edge by more than a few kT in energy, it can strongly

suppress radiative electron-hole recombination. This suppression can be even more effective than that achieved in metallic waveguides, as dielectric structures can, in principle, be entirely lossless.²⁷

A one-dimensional potential well structure - where each layer has an optical thickness of one-quarter wavelength ($\lambda/4$) provides a useful conceptual foundation for understanding the operation of photonic crystal, shown in Fig. 1. The spatially varying dielectric constant, $\varepsilon(x)$, can be expressed as:

$$\varepsilon(x) = \varepsilon_0 + \varepsilon(x) \quad (2)$$

where ε_0 represents the average dielectric constant, and $\varepsilon(x)$ accounts for its spatial modulation. In the absence of absorption, the refractive index n is related to the dielectric constant by the relation $n = \sqrt{\varepsilon}$. It is assumed that the total dielectric constant remains real and positive throughout the structure. If the periodic structure has a lattice constant a , with L_{low} and L_{high} denoting the physical thicknesses of the low- and high-index layers respectively, then the period is $a = L_{low} + L_{high}$. The optical thickness of each layer is defined as the product of its physical thickness and refractive index.²⁸

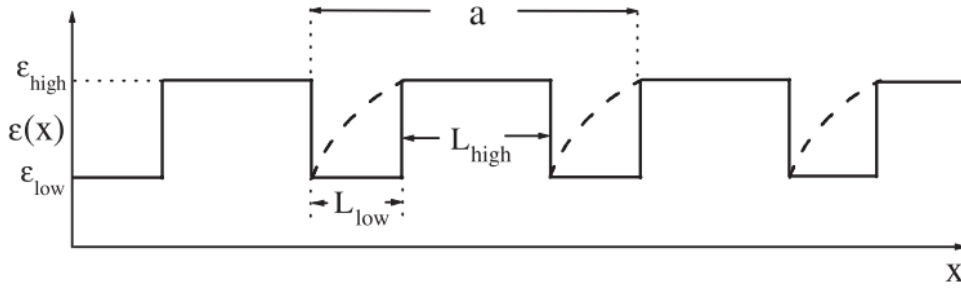


Figure 1. The structure consists of a one-dimensional square-well potential with a periodicity a , where each unit cell is made up of low- and high-index layers with thicknesses L_{low} and L_{high} , respectively. This type of periodic system exhibits Bragg scattering. The dashed line within the low-index region represents a length equal to one-quarter of the optical wavelength - this is the thickness required to achieve maximum reflectivity through constructive interference. Taken from Lipson et al.²⁸

In such a periodic structure, two complementary optical scattering mechanisms can occur. The first is Bragg scattering, which arises when the total geometric period of the structure, a , is equal to an integer multiple of half the optical wavelength, i.e., $m\lambda/2 = a$, where m is an integer ($\pm 1, \pm 2, \pm 3, \dots$). The second mechanism is internal reflection within each layer, which is maximized when the optical thicknesses of the high- and low-index segments are each equal to a quarter of the wavelength, that is, $n_{high} \cdot L_{high} = n_{low} \cdot L_{low} = \lambda/4$. A photonic band gap (or stop band) is formed when both conditions are satisfied at the same wavelength, resulting in constructive interference of the reflected light and enhanced overall reflection from the structure. Typically, such periodic arrangements of dielectric materials create a one-dimensional photonic band gap. The width of this band gap increases with the

refractive index contrast between the high- and low-index materials. For normal incidence, the edges of the stop band centered at frequency ω_0 are located at $\omega_0 \pm \Delta\omega$, where

$$\frac{\Delta\omega}{\omega_0} = \frac{2}{\pi} \arcsin \left(\frac{n_{high} - n_{low}}{n_{high} + n_{low}} \right) \quad (3)$$

This relationship is independent of the polarization of light. However, when light enters at an angle, the reflectivity and band structure become dependent on polarization.

The first experimental demonstration of a three-dimensional photonic crystal (3D PC) was carried out by Yablonovitch et al.²⁹, who constructed a face-centered cubic (fcc) structure, similar to an opal lattice, within a low-loss dielectric material. This structure exhibited a photonic band gap in the microwave range. It was created by mechanically drilling tens of thousands of holes with radii slightly larger than those required for close packing, allowing the voids to connect and form a continuous network. This design is commonly referred to as the three-cylinder structure, or a “Yablonovite.” The drilling angles along the three equivalent (110) directions used to generate the three-cylinder structure are shown in Fig. 2. Both the Yablonovite and its inverse structures are found to possess a complete band gap in three dimensions.²⁸

Studies have shown that the highest $\omega_0/\Delta\omega$ ratio (0.46) can be obtained in an inverse diamond lattice composed of overlapping air holes within a dielectric material, with a filling fraction of 0.81. In practical terms, this structure consists primarily of air voids supported by an extremely thin dielectric framework. Given that achieving reflection in the visible range requires submicron-scale periodicity, it is not surprising that fabricating such a lattice for high-frequency operation remains a significant challenge.²⁸

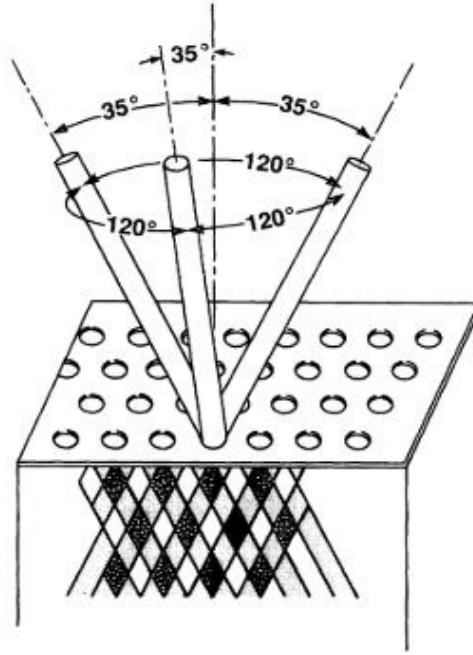


Figure 2. Drilling method used to make a fcc structure, drilling holes in the specified angles. Taken from Yablonovitch et al.²⁹

A relatively new emerging promising 3D photonic crystal structure is a so called “woodpile structure”. The woodpile structure, illustrated in Fig. 3, was the first three-dimensional photonic crystal with a complete bandgap to be fabricated at the micrometer scale for infrared light. Originally proposed by Ho et al.³⁰, it is composed of stacked layers of parallel dielectric rods, with each successive layer oriented perpendicular to the one below.

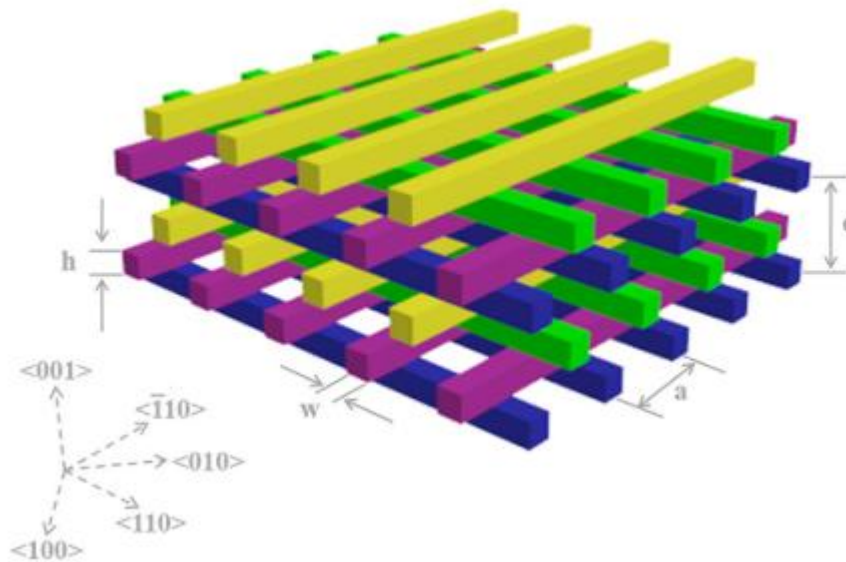


Figure 3. Structural layout of the woodpile crystal and its corresponding lattice parameters.

Woodpile photonic crystals are typically fabricated using layer-by-layer lithography, where patterned dielectric rods are stacked in alternating directions with high precision. This method is

effective for infrared applications, especially in silicon. For visible wavelengths, two-photon polymerization enables true 3D printing of the structure with sub-micron resolution. Other methods, like interference lithography and colloidal self-assembly, offer simpler fabrication but with less design flexibility.³¹

Some of the most compelling applications of photonic crystals emerge when defects are intentionally introduced to locally disrupt their spatial periodicity. These defects create localized states within the photonic band gap, enabling light to propagate specifically through the regions defined by the defect configuration.²⁸ They have potential in development of optical filters, waveguides, and high-efficiency mirrors. One of their most promising uses lies in creating photonic band gap materials that can inhibit spontaneous emission, making them valuable for enhancing the performance of lasers, LEDs, and quantum optical devices. Additionally, when engineered with defects, 3D photonic crystals can guide light with high precision, enabling compact, low-loss optical circuits for photonic computing and telecommunication systems.³²

The optical response of photonic crystals is highly sensitive to changes in refractive index and absorption within the structure. Therefore, integrating active materials such as aluminum-doped zinc oxide (AZO) into photonic crystal architectures enables dynamic tuning of their optical properties. Under optical excitation, the free carrier concentration in AZO changes, modifying both its absorption and refractive index. These changes can alter the effective refractive index of the photonic crystal and shift or modify resonant modes within the structure. As a result, transient transmission and reflection measurements of AZO coated photonic crystals can reveal pump-induced spectral shifts and changes in resonance intensity, providing insight into ultrafast light-matter interactions in photonic structures.

2.3 Epsilon-near-zero (ENZ) materials

The refractive index n is a fundamental optical property that is both simple to understand and interesting to manipulate. While typical dielectric materials have refractive indices greater than one, the concept of near-zero-index (NZI) materials challenges this norm by allowing n to be tuned below one and even close to zero. Since $n = \sqrt{\mu\epsilon}$, where μ is permeability and ϵ is permittivity, NZI behavior can be achieved through three routes: μ -near-zero (MNZ), ϵ -near-zero (ENZ), or ϵ - μ -near-zero (EMNZ). MNZ and EMNZ involve adjusting magnetic permeability and are studied separately. ENZ materials, however, have gained more attention - largely due to their relatively easier experimental realization. The specific wavelength or frequency at which the real part of the permittivity approaches zero is referred to as the ENZ point, and the surrounding spectral region is called the ENZ region or ENZ zone, although its exact width isn't strictly defined. A common interpretation defines the ENZ region as the spectral range where $-1 \leq \epsilon \leq 1$ or $0 \leq n \leq 1$. In practical ENZ materials, such as transparent conducting oxides (TCOs), intrinsic material losses are often present within the ENZ

region, leading to a complex-valued permittivity and refractive index.³³ The most usual TCO used is indium tin oxide (ITO), but aluminum-doped zinc oxide (AZO) exhibits enhanced ENZ behavior in the spectral range around 1500 nm, aligning well with the wavelengths commonly used in telecommunication technologies.³⁴

From a classical optics perspective, when the refractive index n approaches zero, it enables phenomena that do not occur in conventional dielectric materials. Take Snell's law for example: $n_1 \sin \theta_1 = n_2 \sin \theta_2$. If $n_1 \rightarrow 0$ while n_2 remains finite, the transmitted angle θ_2 will always approach zero regardless of the incident angle, resulting in light emerging normal to the surface. This implies that in ideal or near-ideal ENZ materials, the emission direction can be precisely controlled and tailored by shaping the medium itself, enabling highly directional radiation, much like a custom-designed optical antenna.^{33,35}

The relative permittivity is usually described by the Drude model:

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\Gamma\omega} \quad (4)$$

where ε_∞ represents the high-frequency permittivity contribution, ω_p denotes the plasma frequency, and Γ is the damping constant. The plasma frequency depends on the free-carrier concentration N_c , the elementary charge q , and the electron effective mass m^* , and is given by:

$$\omega_p^2 = \frac{N_c q^2}{\varepsilon_0 m^*} \quad (5)$$

The ENZ region is observed in the spectral range surrounding the plasma frequency.⁷

A defining property of ENZ materials is the strong local electric field enhancement imposed by electromagnetic boundary conditions at material interfaces. While the tangential electric field remains continuous, continuity of the normal component of the electric displacement field requires the $\varepsilon_1 E_{\perp 1} = \varepsilon_2 E_{\perp 2}$ relation to be satisfied to keep the electric flux density constant. As the permittivity of ENZ material approaches zero, the electric field E inside the material needs to increase. As a result, the electric field strength increases within the ENZ layer. This enhancement originates at the interface and persists even for ENZ layers much thinner than the electromagnetic skin depth, enabling deep subwavelength field confinement.⁷

The ENZ condition leads to a dramatic increase in the phase velocity $v_p = \omega/k = c/n$ as the refractive index n approaches zero, resulting in an expanded effective wavelength $\lambda_{ENZ} = \lambda_0/n$. In the case of negligible optical loss in the ENZ material, such that $\varepsilon'' \rightarrow 0$, the wavevector k also approaches zero. Consequently, phase accumulation across the ENZ medium is strongly suppressed, effectively converting propagating optical waves into spatially quasi-static (“DC”) fields. This reduction in phase accumulation significantly relaxes the phase-matching requirements for nonlinear

optical processes, including second-harmonic generation and four-wave mixing, for which phase matching is normally essential to avoid destructive interference of the generated nonlinear fields during propagation. In ENZ media, the near absence of phase accumulation over the interaction length substantially alleviates this constraint, largely independent of the incident photon direction.⁷

2.4 Pump-probe investigation of photonic crystals

There have been efforts to investigate 3D photonic crystals using the pump probe technique. Based on photonic band structure calculations for photonic band-gap materials exhibiting Kerr nonlinearity, under intense optical pumping, the photonic band gap is anticipated to shift on a femtosecond timescale, driven by changes in the refractive index of the photonic crystal's backbone material caused by the Kerr nonlinear effect.³⁶

Euser et al.¹ has reported a significant and ultrafast blueshift of the photonic band gap occurring within 300 fs, and then relaxation within 18 ps, in a Si woodpile photonic crystal. The mechanism which caused the appearance of the photonic band gap was thought to be the generation of free carriers. The 18 ps timescale is related to the recombination of carriers in the polysilicon structure. According to their modelling and experimental results, they observe both a reflectivity increase of 19 % and a decrease of 10 % at the blue edge of the photonic band gap.

Becker et al.³⁷ observed a large nonlinear optical transmission changes of 134% and stop band shifts reaching approximately 150 nm towards the blue, in a Si inverse opal photonic crystal structure. Under these excitation conditions, two-photon absorption, associated with the imaginary component of the third-order nonlinear susceptibility, becomes the dominant mechanism. This behavior contrasts with earlier theoretical models of photonic crystal nonlinearities, which primarily focused on changes in the real part of the third-order susceptibility, linked to the nonlinear refractive index n_2 .

Sakellari et al.³⁶ also observed a photonic band gap shift when exciting a CdS doped woodpile photonic crystal with a femtosecond laser, but a surprising result was observed: at low pump power, the transmittance rises, while at high pump power, it decreases, indicating significant alterations in the system's response caused by the pump. These effects may result from a pump-induced shift of the photonic band gap.

There have also been studies which have shown the ability to control the absorption and tuning of the photonic bandgap in the timescale of 1 ps, indicating a possibility of using these structures as ultrafast all-optical switches.^{37,9}

Saha et al.⁸ manufactured a bi-material coating of two ENZ materials - titanium nitride (TiN) and AZO. They have combined a faster and slower materials to achieve ultrafast switching when the

coating is pumped, which also has different speeds for different wavelengths. According to them, the interband pump creates free carriers in AZO, increasing absorption and lowering the refractive index via changes to the Drude response. They attributed the fast AZO decay times to defect assisted Shockley-Read-Hall recombination, a recombination mechanism commonly observed in heavily doped TCOs. They have also noted that the reflection resonance is blueshifted when AZO is pumped as compared to unpumped.

According to Li et al.³⁸, the decay function of a TCO material pumped with UV pulses should be fitted with a bi-exponential function since both the carrier concentration and the Fermi level are increased, giving rise to two distinct recovery mechanisms associated with carrier recombination (slow) and electron-phonon interactions (fast).

To date, to the best of my knowledge, only a limited number of studies have explored ultrafast dynamics in photonic structures incorporating transparent conductive oxides. The only one that I could find was done by Moscardi et al.⁹, they carried out pump-probe experiments in one-dimensional photonic crystals incorporating transparent conductive oxides, notably multilayer structures composed of alternating ITO and dielectric TiO₂ layers. In these crystals, optical pumping induces transient carrier dynamics in the ITO layers, leading to rapid changes in their permittivity and a corresponding ultrafast ~1 ps modulation of the photonic bandgap. This work illustrates that embedding TCOs into photonic crystal architectures enables dynamic control of photonic band structures via carrier-driven nonlinearities.

To sum up, studies using pump-probe techniques on photonic crystals have shown ultrafast shifts of the photonic band gap driven by Kerr effect induced refractive index changes and photoinduced free carrier effects. Kerr-induced refractive index changes lead to nearly instantaneous spectral shifts occurring on femtosecond timescales, whereas free carrier generation via one- or two-photon absorption produces additional band gap shifts with characteristic buildup times of a few hundred femtoseconds and relaxation dynamics spanning several picoseconds to tens of picoseconds. Significant femtosecond blueshifts and picosecond relaxations were observed in silicon structures, with two-photon absorption playing a key role at high intense excitation. Complex transmission changes depending on pump power suggest intricate dynamics, requesting further, more detailed investigation in this area. Furthermore, measurements have not previously been reported for 3D photonic crystals coated with TCOs, such as AZO.

2.5 Tantalum pentoxide (Ta₂O₅)

Tantalum pentoxide (Ta₂O₅) high refractive index dielectric material that has attracted considerable interest in optical coating technology, integrated photonics, and nonlinear optical

applications due to its favorable combination of optical, structural, and chemical properties. Ta₂O₅ thin films have been extensively employed in dielectric mirrors, antireflection coatings, optical filters, waveguides, and resonant photonic structures, where low optical losses and high environmental stability are required. One of the main advantages of Ta₂O₅ is its relatively high refractive index, typically in the range of $n \approx 2$ -2.2 in the visible and near-infrared spectral regions. This high refractive index enables strong optical confinement and pronounced modulation of guided or resonant optical modes when integrated into photonic devices. In addition, Ta₂O₅ exhibits a wide optical transparency window extending from the near-ultraviolet to the mid-infrared, together with comparatively low propagation losses, making it highly suitable for both passive and active photonic applications.³⁹

The optical and electronic properties of Ta₂O₅ are strongly influenced by its microstructure and fabrication conditions. Thin films deposited by electron-beam evaporation, sputtering, ion-beam deposition, or atomic layer deposition are often amorphous in the as-deposited state, although thermal treatment can induce crystallization and modify the optical constants. Reported optical bandgap values typically lie between approximately 4.0 and 4.5 eV, which places the absorption edge in the ultraviolet region. As a result, excitation with ultraviolet photons can generate real electronic excitations within the material, either through direct interband absorption or through defect-mediated transitions involving oxygen vacancies or localized trap states.^{40,41}

The relaxation dynamics of photoexcited carriers in Ta₂O₅ are expected to depend strongly on defect density, film morphology, and interface quality. Oxygen vacancy related trap states in amorphous Ta₂O₅ can introduce localized electronic states within the bandgap, potentially enabling carrier trapping and delayed recombination on picosecond to longer timescales, particularly in thin-film or interfacial geometries.^{42,43}

Beyond its linear optical properties, Ta₂O₅ has also emerged as a promising material for nonlinear photonics. Recent studies have demonstrated that Ta₂O₅ exhibits a measurable third-order nonlinear susceptibility and Kerr nonlinear refractive response, while maintaining relatively low propagation losses compared with many other integrated photonic materials. Combined with its compatibility with silicon-based fabrication processes, these characteristics make Ta₂O₅ attractive for nonlinear waveguides, micro resonators, and all-optical modulation platforms.⁴⁴

2.6 Pump-probe investigation of grating resonances

An optical diffraction grating is a periodically structured optical element designed to manipulate the propagation of light through diffraction and interference. Optical gratings are periodic structures in which the refractive index or surface profile is modulated on the scale of the optical wavelength. This periodicity enables diffraction of incident light into multiple propagation orders,

with the optical response determined by the grating period, groove depth, angle of incidence, and the refractive indices of the surrounding materials. When light interacts with such a periodic structure, each grating period acts as a secondary source of scattered electromagnetic waves. Constructive and destructive interference between these scattered waves gives rise to discrete diffraction orders, whose propagation directions are determined by the grating equation and depend on the wavelength of light, grating period, angle of incidence, and refractive indices of the surrounding media. Due to their ability to spectrally separate or redirect light with high precision, diffraction gratings are widely used in spectrometers, monochromators, laser cavities, pulse compression systems, and wavelength-selective optical components.⁴⁵

When the grating is fabricated from dielectric materials such as silicon dioxide (SiO_2), and the grating period becomes comparable to or smaller than the optical wavelength, the optical response can extend beyond conventional diffraction behavior. Under appropriate conditions, incident light can couple into waveguide-like modes supported by the periodic structure. Since these modes are not fully confined and can radiate back into free space, they are commonly referred to as guided mode resonances (GMRs) or leaky-mode resonances. The excitation of such resonances leads to rapid spectral variations in transmission or reflection, often appearing as narrow transmission minima or reflection maxima. Ideal reflection and transmission spectra are shown in Figure 4. The spectral position, linewidth, and polarization dependence of these resonances are determined by the grating period, groove depth, fill factor, refractive-index contrast, and angle of incidence.

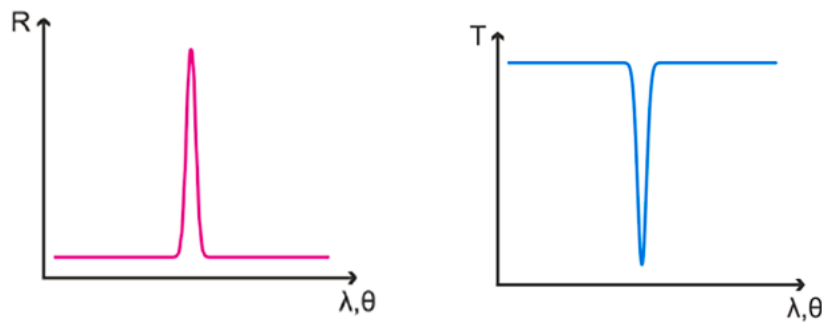


Figure 4. Ideal reflection and transmission spectra of guided mode resonance gratings. Taken from Gambino et al.⁴⁶

One of the most important characteristics of guided-mode resonances is the strong localization of the electromagnetic field within or near the grating structure. This enhanced field confinement increases the interaction between the optical mode and surrounding materials, making resonant dielectric gratings highly sensitive to changes in the local refractive index or optical absorption. As a result, dielectric resonant gratings have found widespread applications in optical filtering, biosensing, beam shaping, narrowband reflection, and integrated photonics.⁴⁶

S. S. Wang et al.⁴ defined the effective propagation constant of the waveguide grating:

$$\beta \rightarrow \beta_i = k(\sqrt{\varepsilon_g} \sin \theta - \frac{i\lambda}{\Lambda}) \quad (6)$$

where β_i is the propagation constant of the i -th diffracted order coupled into the guided mode, $k = \frac{2\pi}{\lambda}$ is a free space vector, ε_g is the dielectric constant of the incident medium, θ is the angle of incidence, i is the diffraction order, Λ is the grating period and λ is the free space wavelength. Effective refractive index is also give as $N_i = \frac{\beta_i}{k}$.

By taking a normal angle, the (6) equation can be simplified to $\beta_i = -i \frac{2\pi}{\Lambda}$. From the effective refractive index definition, we know that $\beta = \frac{2\pi}{\lambda} N$. At the resonance condition $\beta = |\beta_i|$. Then the equation can be solved to express the resonant wavelength condition:

$$\frac{2\pi}{\lambda} N = \frac{2\pi|i|}{\Lambda} \quad (7)$$

$$\lambda_{res} = \frac{N\Lambda}{|i|} \quad (8)$$

Fano resonances can be understood using a simple model of two coupled oscillators: one driven oscillator with a broad spectral response interacting with a second oscillator that has a discrete resonance. When the driving frequency is tuned near the resonance of the second oscillator, the response of the driven oscillator changes sharply. In particular, the driven oscillator can exhibit a near-zero response close to the resonance condition of the second oscillator, effectively corresponding to an infinite system impedance and resulting in complete reflection of the incoming energy. This mechanism can lead to very high sensitivity, as absorption and scattering can be precisely tailored within extremely narrow frequency ranges.⁴⁷

The phenomenon was originally introduced by U. Fano⁴⁸ in the context of atomic spectroscopy, where quantum interference between localized and continuum states produced characteristic asymmetric absorption profiles. In photonic structures, analogous effect occurs when incident light couples to a narrow optical resonance, such as a guided or leaky mode of a periodic dielectric structure, while simultaneously interacting with directly transmitted or reflected non-resonant radiation. The interference between these two optical pathways produces the characteristic Fano line shape, which differs from a conventional symmetric Lorentzian resonance. In dielectric gratings and photonic crystal slabs, these resonances are often associated with guided-mode excitation and can exhibit extremely steep spectral slopes, high quality factors, and strong electromagnetic field localization. Because of this high spectral sensitivity, even small pump-induced changes in the optical constants of the surrounding materials can produce pronounced modulation of the resonance position, amplitude, or linewidth, making Fano-resonant structures attractive for optical filtering, sensing, and all-optical switching applications.²

Several recent studies have demonstrated the use of pump-probe or transient optical spectroscopy to dynamically control guided-mode and related photonic resonances.

A recent study particularly relevant to the present work was carried out by S. Bej et al.⁴⁹ In this work, the authors investigated ultrafast optical modulation in a dielectric guided-mode resonance grating fabricated from silicon nitride on a fused silica substrate. The structure consisted of square-shaped pillars in a square lattice, with an overall thickness below 300 nm, was designed to support narrow resonances in the near-infrared spectral region. Under excitation with intense ultrashort laser pulses of ~ 744 nm, the optical Kerr effect induced a transient refractive index change in the silicon nitride structure, resulting in a pump power dependent spectral shift of the transmission resonance. The authors observed more than 50% modulation of the resonance magnitude and reported an effective enhancement of the Kerr nonlinearity by more than three orders of magnitude compared to an unstructured film of the same material. This enhancement was attributed to strong electromagnetic field confinement associated with the guided-mode resonance. Although the study did not employ conventional time-resolved pump-probe detection, it demonstrated that even small ultrafast refractive index changes can be converted into pronounced spectral modulation in resonant dielectric gratings, highlighting their potential for all-optical switching and dynamic photonic applications.

A related example of ultrafast resonance modulation was demonstrated by M. Shcherbakov et al.⁵⁰ In this work, femtosecond pump-probe spectroscopy was used to investigate resonant gallium arsenide (GaAs) meta surfaces under above-bandgap optical excitation. Photoexcited free carriers transiently modified the dielectric function of the semiconductor, resulting in resonance shifts of several tens of nanometers on picosecond timescales. The study demonstrated that carrier-induced refractive index changes in resonant photonic structures can enable efficient all-optical modulation of spectrally selective optical modes.

3 Experiment methodology

3.1 Samples

The first sample is a “woodpile” photonic crystal made by Dimitra Ladika, described in her PhD thesis⁵¹. It was manufactured using a SZ2080TM organic-inorganic silicon hybrid composite photoresist. The samples were created by drop-casting onto 100 μm thick glass substrates that had undergone chemical treatment via a silanisation process. The deposited photoresist droplets were left to dry at room temperature inside a vacuum chamber for several days prior to photopolymerization. After exposure, the photopolymerized droplets were developed using a specific mixture of solvents to remove the unpolymerized resin and reveal the final 3D structure. The fabrication of the 3D structures

was obtained via the Multiphoton Lithography (MPL) technique. The deposition of AZO thin films onto both glass substrates and 3D structures was carried out using pulsed laser deposition (PLD) technique. The AZO layers were formed under high vacuum conditions with a conventional PLD setup. A scanning electron microscope (SEM) pictures of the 3D photonic crystals are shown in Fig. 5.

An array of samples has been investigated. There were three inlayer periodicities of 1.2 μm , 1.3 μm and 1.4 μm , and three thicknesses of the deposited AZO coating of 150 nm, 200 nm and 300 nm.

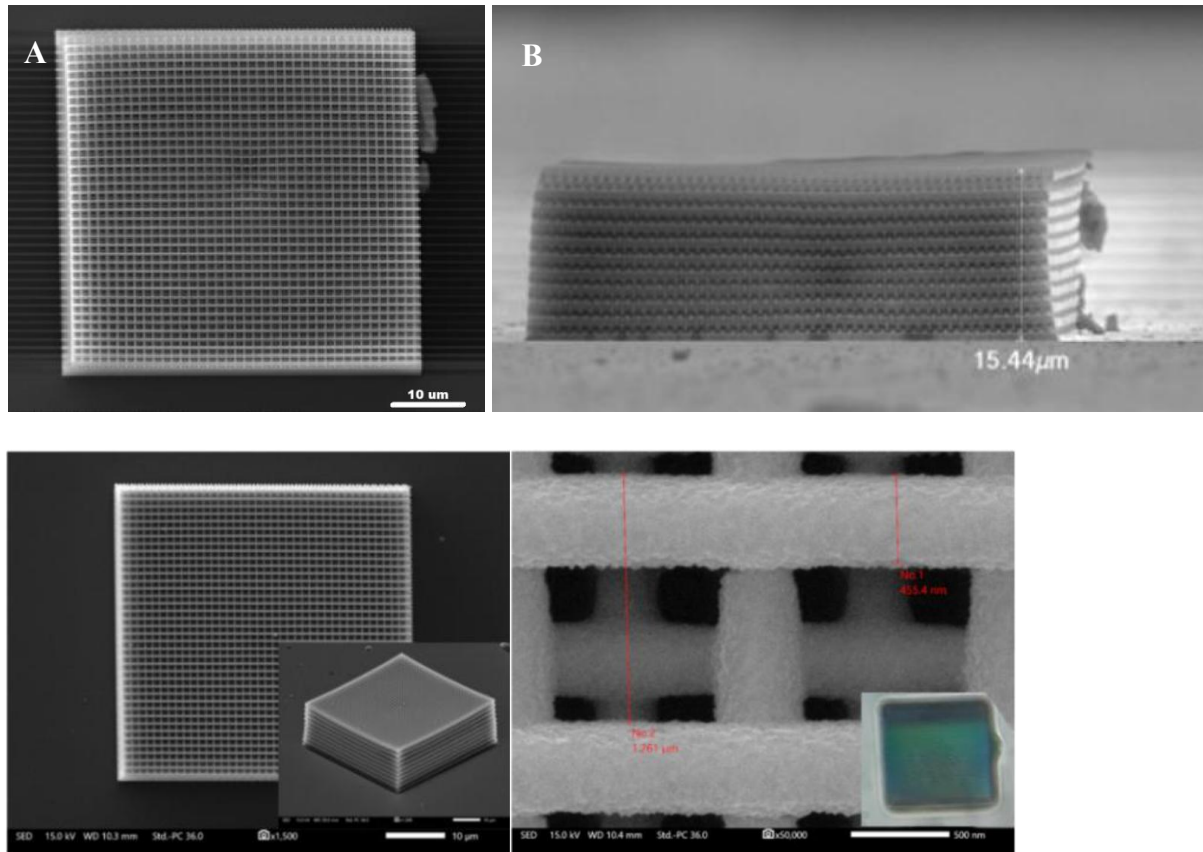


Figure 5. (A) Top-view SEM image displaying the 3D woodpile photonic crystal (PC) with inlayer periodicity of 1.3 μm . (B) Side-view of the same structure. (C) Top- and side-view of the 1.3 μm inlayer periodicity PC, coated with 150 nm of AZO. (D) Zoomed in top view of the same structure as in picture C.

The other sample was a tantalum pentoxide (Ta_2O_5) coated fused silica (SiO_2) grating. The grating had a period of 833 nm and a groove depth of around 300 nm. The Ta_2O_5 coating was deposited using the ion beam sputtering (IBS) technology, with a system developed by Cutting Edge Coatings GmbH (Hannover, Germany). The thickness of the deposited Ta_2O_5 coating was 390 nm and the refractive index was 2.09.

3.2 Experimental scheme

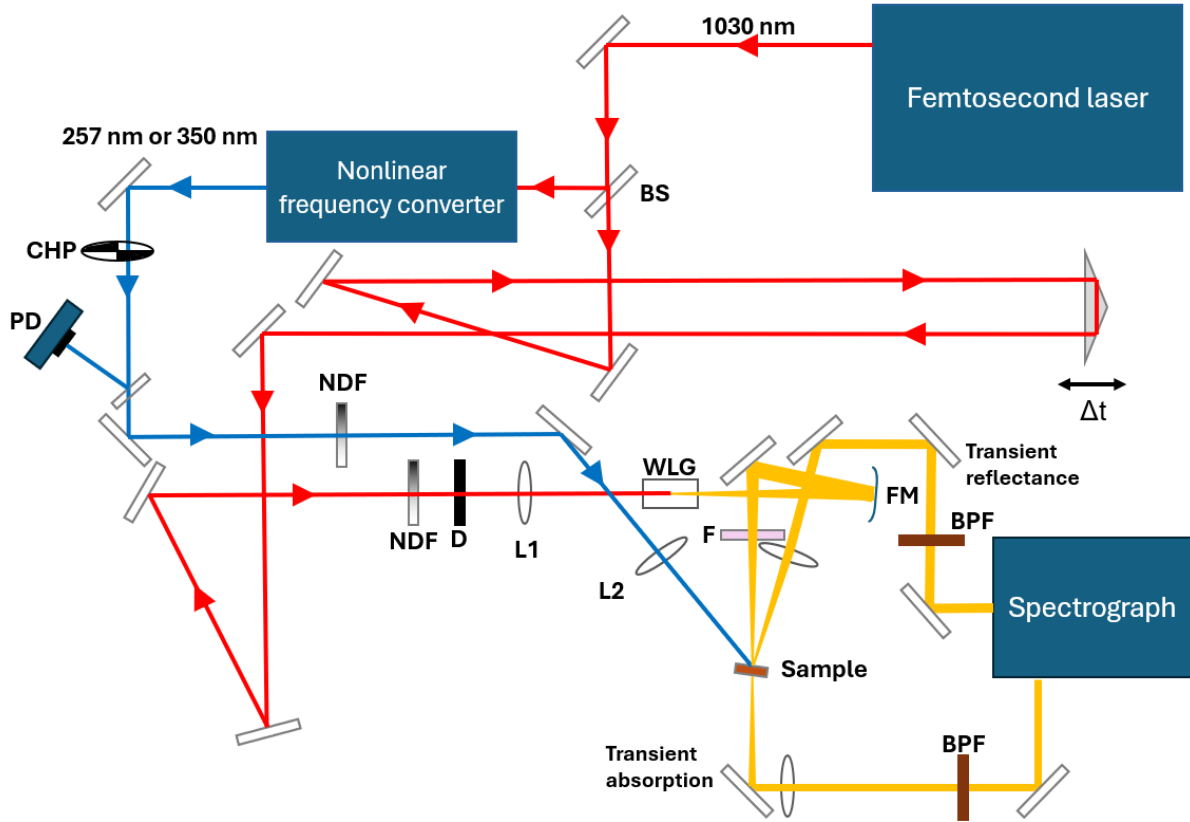


Figure 6. Principal transient absorption experimental scheme. OPA - optical parametric amplifier, BS - beam splitter, CHP - chopper, PD - photodiode, NDF - neutral density filter, D - diaphragm, L1 & L2 - lenses, WLG - white light generation, F - fundamental frequency filter, BPF - bandpass filter, FM - focusing mirror. Transmitted probe was used for transient absorption measurements, while the reflected probe was used for transient reflectance. During the investigation of AZO coated photonic crystals, two reflective objectives were used to focus the probe light.

Yb:KGW femtosecond laser (Pharos, Light Conversion) was used at a repetition rate of 26.7 kHz, 190 fs pulse duration at 1030 nm center wavelength, and pulse energy of 23 μ J. Reflected part of the radiation from the beamsplitter was used to pump the OPA (Orpheus, Light Conversion), which has a wavelength range of 310-2600 nm. 350 nm was used in the experiments as the pump source for photonic crystal samples. A neutral density (ND) filter was used to control the power used for the samples. A +150 mm lens were used to focus the pump beam.

The transmitted part of the 1030 nm radiation was guided through a 150 mm delay line, towards the white light generation part, which was used as a probe in the experiments. An iris diaphragm was used to control the NA for white light generation (WLG) and a ND filter was used to change pumping energy of the white light. L1 is a +150 mm lens. White light is generated in a 10 mm YAG crystal. Then the light was collimated with a 200 mm radius focusing mirror and directed towards the

objective. During the investigation of AZO coated photonic crystals, probe light was focused with aluminum coated 15X reflective objectives (LMM15X-UVV, Thorlabs) with a NA=0.3. After the second objective, the white light was guided towards the spectrograph (77480, Newport) through a bandpass filter of >1050 nm, which is used to block the visible part of the continuum. The spectra were registered using a 256-pixel InGaAs linear image sensor (G9208-256WB, Hamamatsu). Each recorded spectrum was obtained by integrating 31 consecutive shots on the sensor.

For the investigation of the Ta₂O₅ coated SiO₂ grating, the fundamental 1030 nm laser output was first frequency-doubled and subsequently converted to its fourth harmonic, resulting in a final wavelength of 257 nm used to pump the sample. Sample dimensions no longer required to focus the white light probe with the objectives, hence the 200 mm radius focusing mirror was used to focus the white light on the sample. For the transient absorption measurements, the transmitted probe beam was directed to the spectrograph. For the transient reflectance measurements, the probe beam reflected from the sample was guided to the spectrograph.

3.3 White light continuum generation and beam sizes

For the probing of the differences in sample absorbance white light continuum was used. The generation conditions were adjusted by controlling the NA of the input beam using an iris diaphragm, thus altering the focusing condition, and fine tuning the pump pulse energy using a ND filter.

Since the goal is to investigate how the AZO coated photonic crystals respond to excitation in the IR range, (ideally in the 1-2.5 μm) generation conditions were optimized to achieve the most redshifted spectrum as possible. It was done by iterating between the pulse energy and the input beam diameter.

Fig. 7 shows how the shape of the continuum spectrum varies with the input pulse energy. At 0.6 μJ , the spectrum intensity in the IR range is low, then with increasing energy the spectrum expands further into the IR range with increasing spectral intensity, until at 1.04 μJ it becomes significantly modulated in the 1200-1350 nm range. The ideal pump pulse energy is around 0.92 μJ , which yields a relatively flat spectrum with strong IR intensity.

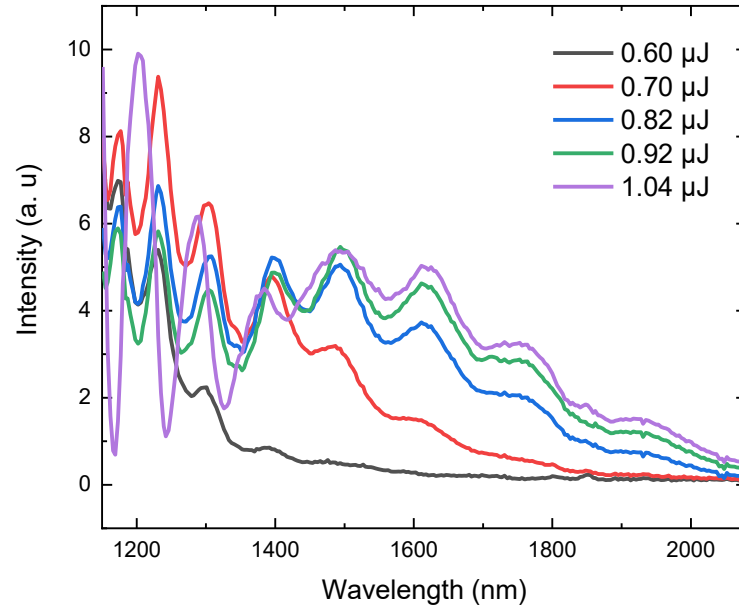


Figure 7. White light spectrum dependence on input pulse energy.

The best continuum shape is achieved just as the 2nd nonlinear self-focus in the WLG YAG crystal forms at 0.92 μJ . However, a similarly good spectral shape can also be observed with only a single self-focusing event, just before the threshold of the 2nd self-focus. The picture of the YAG crystal during WLG with two foci is shown in Fig. 8, with the second self-focus forming at the very end of the crystal. The power before the iris diaphragm was 41.7 mW, after the iris - 27.3 mW, after the lens - 24.5 mW.

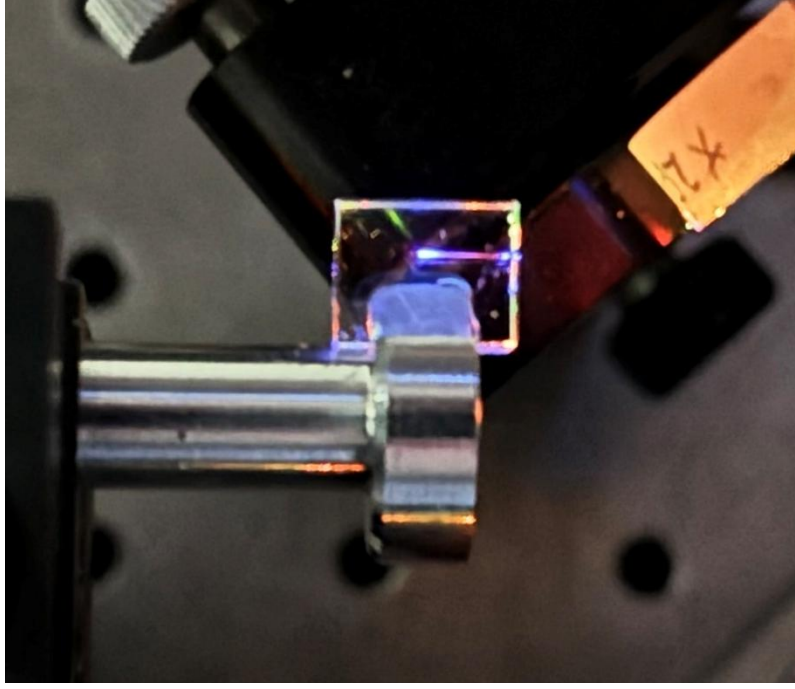


Figure 8. 2 filament formation in YAG crystal during WLГ.

The stability of the white light was also evaluated. It was done by taking a normalized root mean square deviation (NMRSD) normalized by the mean of the dataset:

$$NMRSD = \frac{\sqrt{\frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^2}}{\bar{x}} \quad (5)$$

The stability was measured at 1400, 1600, 1800 and 2000 nm wavelengths over 250 spectra taken to calculate each value. As mentioned before, each spectrum was integrated over 31 shots. The results are shown in Fig. 9. The stability is highest towards the blue end of the spectrum and continuously deteriorates towards the red part. But overall, the stability falls under 1% across the spectrum.

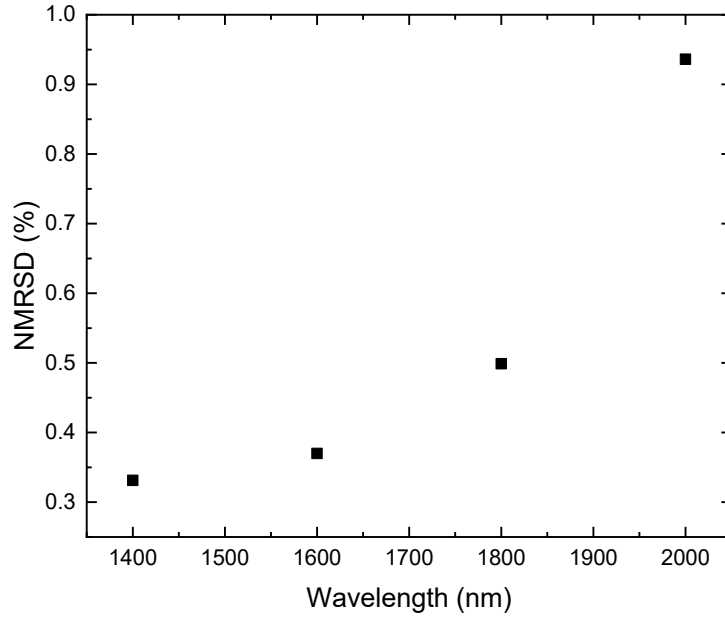


Fig 9. NMRSD values calculated over 250 white light spectra over different wavelengths.

Using a wavelength centered around $\sim 1 \mu\text{m}$, this is one of the furthest continuum extensions towards the red part of the spectrum reported in literature. To my knowledge, only Galinis et al.⁵² has reached a similar result, where a Nd:glass based 1.3 ps laser was used to generate a continuum in a YAG crystal. Their continuum also spanned up to $2 \mu\text{m}$, but they did not have the means to measure beyond this point.

The micro pump-probe setup allows for a probe spot size as small as $9 \pm 1.5 \mu\text{m}$ (at e^{-2}) as evaluated via the knife-edge method shown in Fig. 10 (a), providing sufficient spatial resolution to investigate photonic crystal structures with approximately $50 \times 50 \mu\text{m}$ dimensions. The diameter of the 350 nm pump beam at the sample position was determined to be $80 \pm 1.5 \mu\text{m}$ (at e^{-2}), based on measurements obtained using both the knife-edge technique (shown in Fig. 10 b)) and imaging with a camera.

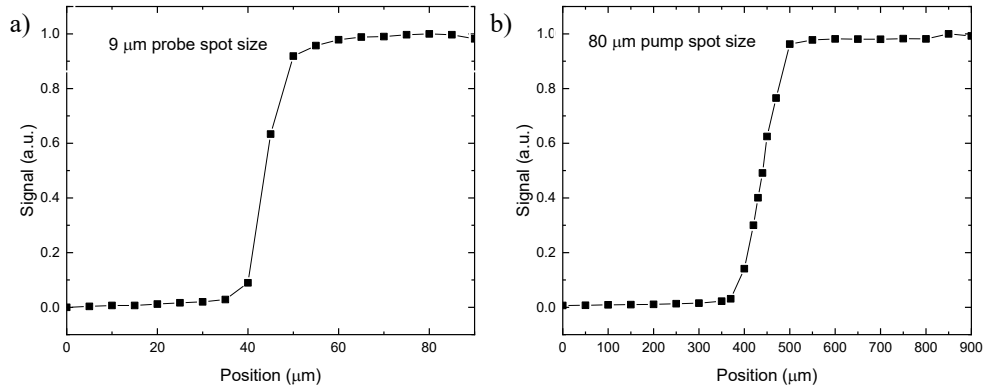


Figure 10. Knife-edge measurements used for estimating the probe beam spot size (a) and the pump beam spot size (b).

During the measurements of transient response of the Ta₂O₅ coated SiO₂ grating, white light was focused with a curved mirror. Probe spot size on the sample was evaluated to be 103 μm on the sample via the knife-edge method, and the 257 nm pump spot size was determined to be 630 μm using the same method.

4 Experimental results

4.1 Transmittance of the samples

4.1.1 AZO coated photonic crystals

As was discussed in the samples section, numerous photonic crystals manufactured with varying inlayer periodicities and different coating thicknesses of AZO. Crystals coated with thicknesses of 200 nm and 300 nm did not show traces of a photonic bandgap in transmittance and pump-probe dynamics spectra, evidenced with a spectral dip, so only the crystals coated with 150 nm thickness of AZO coating will be discussed. Inlayer periodicities were 1.2 μm, 1.3 μm and 1.4 μm, but the 1.2 μm samples did not have any narrow spectral features in the many measured pump probe spectra, so they will be excluded from further analysis. As a result, only 1.3 μm and 1.4 μm inlayer periodicity samples coated with a 150 nm AZO coating will be investigated in this work. There were around 7 PCs of each combination (periodicity plus coating thickness), but only a couple out of this subset exhibit interesting optical properties, and the observed peculiarities also differ between the crystals intended to be identical.

Transmittance of just the 150 nm AZO coating was evaluated using the probe white light source focused with the objectives in Fig. 12. White light transmitted through the substrate and

through the coating deposited on the substrate near the structures was compared. The values measured match the values found in literature and also with D. Ladika's work¹, but the spectrum appears spiky, even though the expected transmittance of a TCO material should be broadband and smooth. This behavior could be attributed to the high numerical aperture objectives used. Slight changes in thickness of the substrate as compared to the substrate and the coating may result in different angular transmission, causing interference effects to become more pronounced. Transmittance of the 1.4 μm inlayer periodicity and 150 nm thickness AZO coating is also shown in Fig. 12 for the two samples chosen. Transmittance spectra look vastly different than just the AZO coating transmittance. Shapes become intricate and vary sample by sample. In the spectrum of the 1st sample, there is a very pronounced dip at ~ 1990 nm. In the 2nd sample, there is a dip at ~ 1830 nm, though not as pronounced as in the 1st sample. Both spectral regions also exhibit a distinct ultrafast response in the pump-probe response.

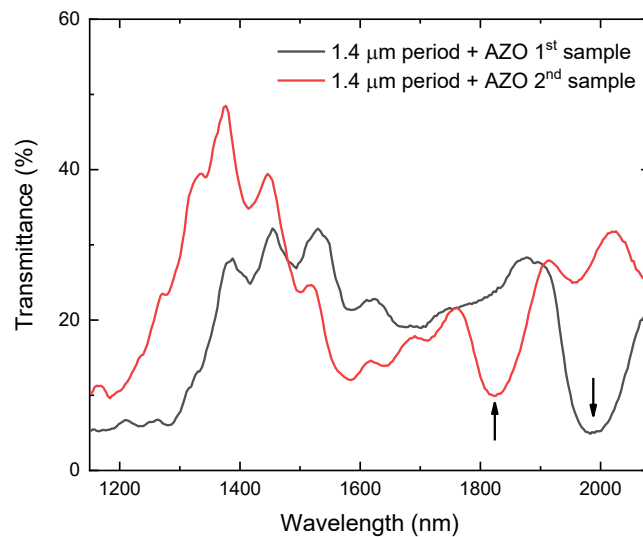


Figure 12. Transmittance spectra of 150 nm thickness AZO coating and of two samples chosen of a 1.4 μm inlayer periodicity photonic crystals coated with 150 nm thickness AZO coating. Spectral dips are marked with arrows.

Transmittance of the 1.3 μm inlayer periodicity photonic crystal coated with a 150 nm thickness AZO coating is shown in Fig. 13 for the two samples chosen. In the 1st sample, the most pronounced dip is centered at ~ 1830 nm. In the 2nd sample, there are 2 pronounced dips, but only the ~ 1750 nm is visible in the pump-probe response, while the ~ 1470 nm is absent.

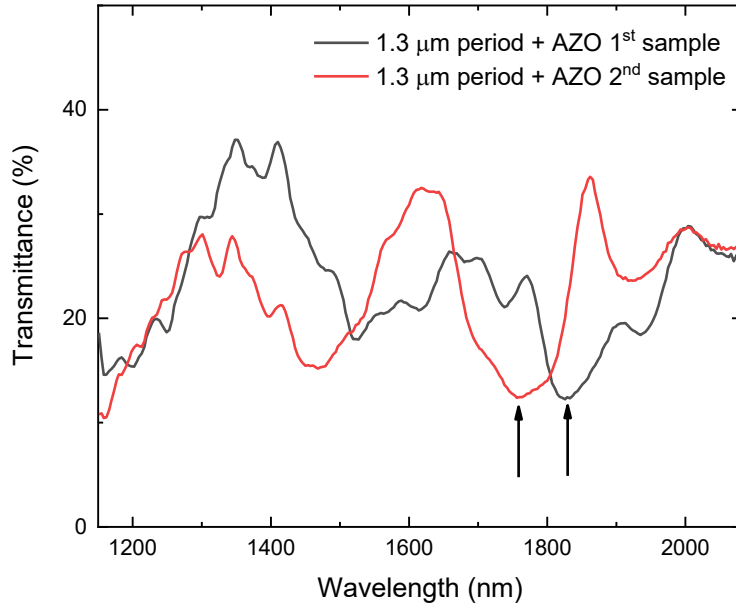


Figure 13. Transmittance spectra of 150 nm thickness AZO coating and of two samples chosen of a 1.3 μm inlayer periodicity photonic crystals coated with 150 nm thickness AZO coating. Spectral dips are marked with arrows.

4.1.2 Ta₂O₅ coated SiO₂ grating

Transmittance of the Ta₂O₅ coated SiO₂ grating sample was evaluated using the probe light, by comparing the spectra with and without the sample in the beam path. The transmittance spectra of the Ta₂O₅ coating deposited on a SiO₂ grating for both s- and p-polarized incident probe light are shown in Fig. 14. For s-polarization, the electric field is oriented parallel to the grooves, while for p-polarization, it is oriented perpendicular to the grating lines.

Reflectance was evaluated by taking a reflected probe of the sample's surface and comparing it with the aluminium mirror reflection mounted in the same holder. The resulting reflectance spectra for s- and p-polarized incident probe light is shown in Fig. 15. As expected, the transmittance and reflectance spectra are roughly inverted, and the shape of the spectra for p-polarized light shows a guided mode resonance.

The grating was rotated by 6° relative to the normal incidence direction of the probe beam, allowing the reflected signal to be collected and directed to the spectrograph. Transmission map of the sample obtained by spectrophotometric measurements is shown in Fig. 16. At 0° angle the transmission minimum is centered at ~1367 nm and then, by changing the angle of incidence the resonance is moved towards the longer wavelengths, allowing tunability of the minimum point.

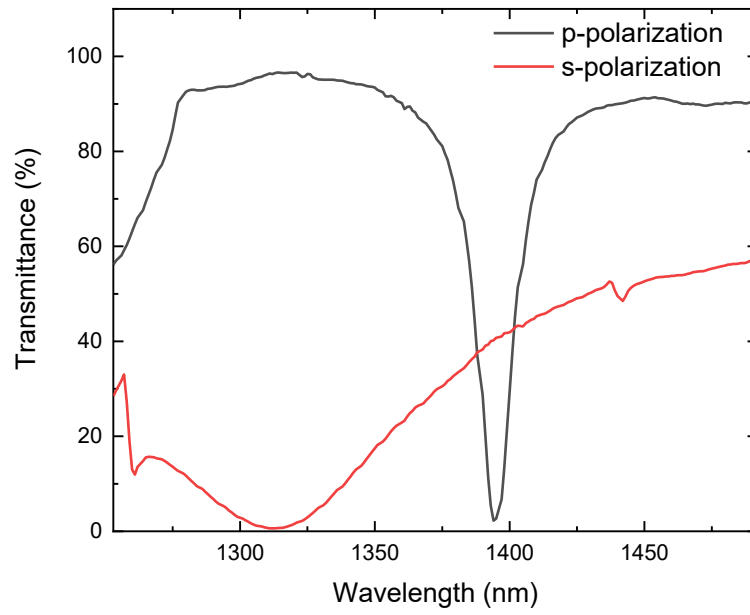


Figure 14. Transmittance spectra of Ta₂O₅ coated SiO₂ grating for s- and p-polarized incident probe light.

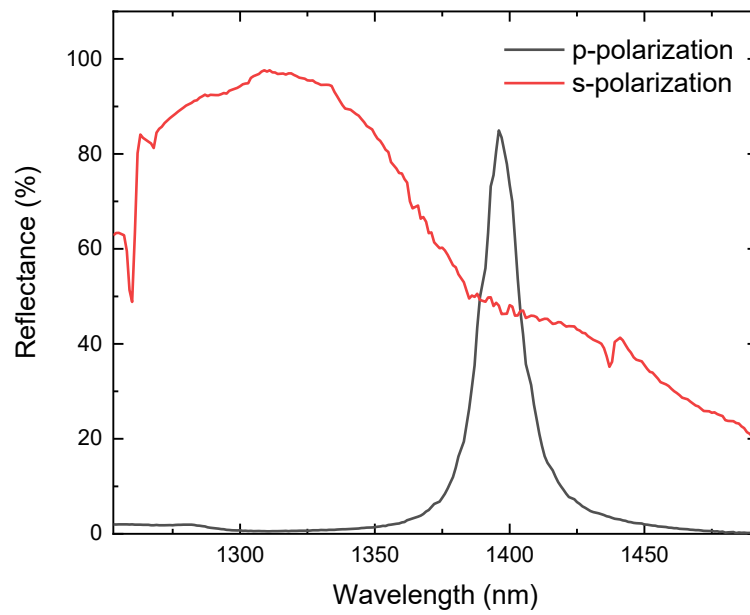


Figure 15. Reflectance spectra of Ta₂O₅ coated SiO₂ grating for s- and p-polarized incident probe light.

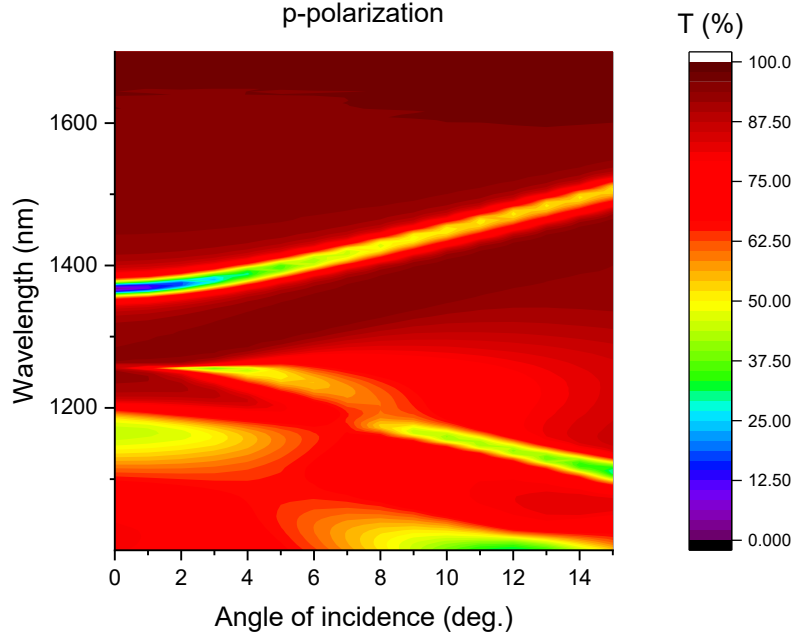


Figure 16. Angular dependence of the transmittance for p-polarized incident light on the Ta₂O₅ coated SiO₂ grating.

4.2 Pump-probe investigation of the AZO coated PCs

Firstly, only the 150 nm AZO coating was investigated with the pump-probe method. The pump wavelength was the same as used for all the samples, i.e. 350 nm and the peak intensity of the pump was $\sim 0.3 \text{ GW/cm}^2$. Transient absorption dataset of probe delay time versus wavelength is shown in Fig. 17. From the trace dynamics on the right, it can be seen from the trace of 1800 nm and 2000 nm that the shape is bi- or multi- exponential, as has been shown in literature³⁸. The slower picosecond delay shows little spectral dependence, likely indicating intrinsic carrier cooling and recombination processes. The initial high difference absorption signal is increasing gradually with increasing wavelengths. This behavior is consistent with an instantaneous Kerr-type optical response associated with Drude-like free carrier dynamics in AZO. At lower photon energies, where the optical response is dominated by intraband carriers, pump-induced changes in carrier density and scattering rate lead to an enhanced modulation of the dielectric function. Decay times have been fitted with a bi-exponential function, and the two constants were weighed to get a single decay constant τ . Decay constant times varied depending on probe wavelength: $\tau_{1400 \text{ nm}} = 4.6 \pm 0.4 \text{ ps}$, $\tau_{1600 \text{ nm}} = 8.9 \pm 1.6 \text{ ps}$, $\tau_{1800 \text{ nm}} = 14.9 \pm 2.9 \text{ ps}$, $\tau_{2000 \text{ nm}} = 2.9 \pm 1.5 \text{ ps}$. Up to 1800 nm the decay constants were increasing, but at 2000 nm the initial fast response begins to dominate and the difference in absorption decays faster.

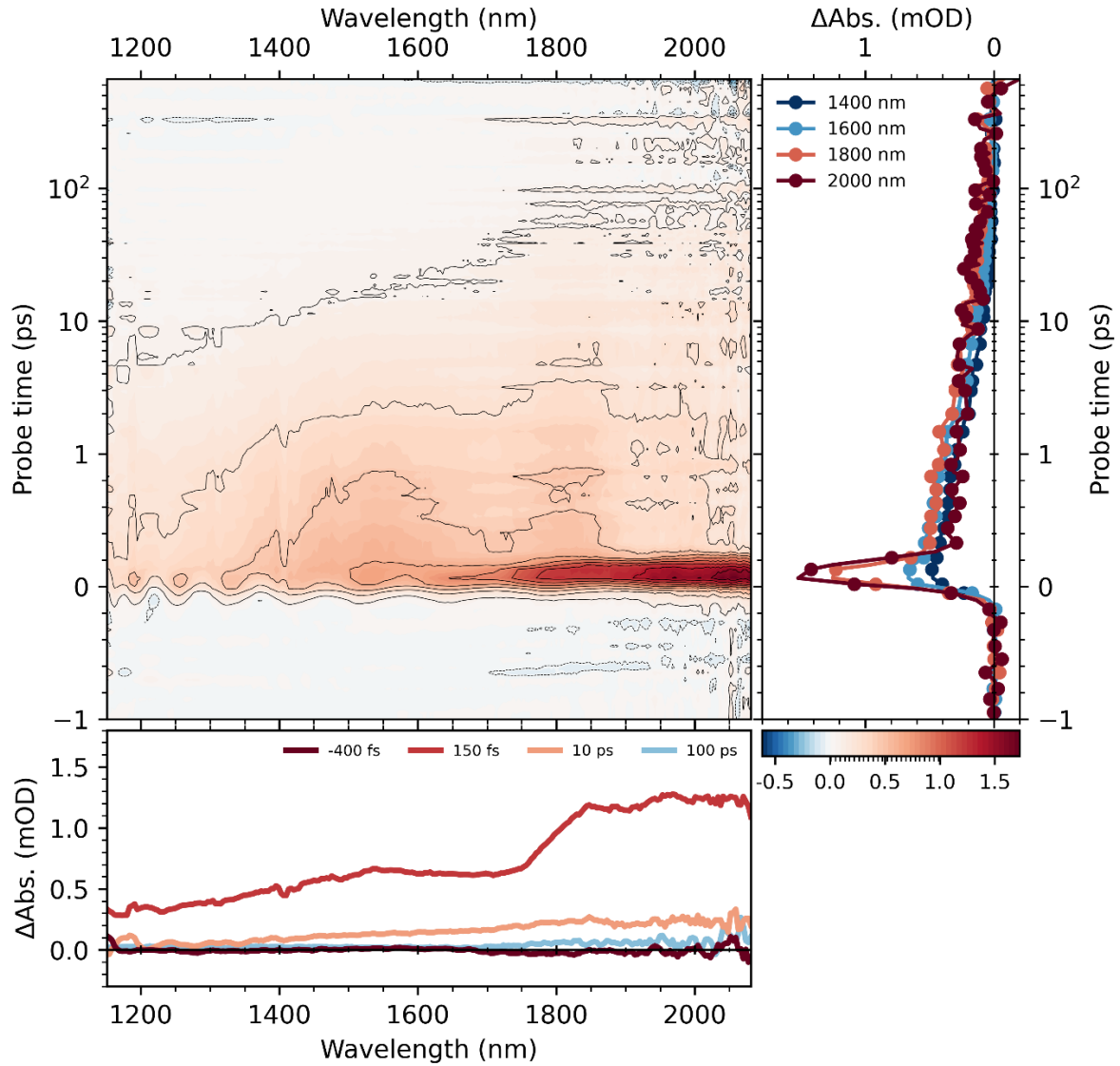


Figure 17. Spectral transient absorption color-map modulation over time of 150 nm AZO coating. Below - spectra at different delay times are shown, on the right - traces of different wavelengths are shown.

A crystal of 1.4 μm inlayer periodicity without an AZO coating was also investigated. To observe a difference absorption signal, the peak intensity had to be increased by 25 times to $\sim 7.4 \text{ GW}/\text{cm}^2$. The measured transient absorption spectra at different delay times are shown in Fig. 18. The difference absorption does increase a little bit near time 0, but the observed signal is very weak, and it clearly accumulates, since the signal does not fully decay. The signal could be mostly attributed to thermal effects and coherent artifacts, with the observable peak in signal vanishing under 1 ps. This means that the photonic crystal itself absorbs the 350 nm pump very weakly and has a small contribution to overall pump probe signal observed when the photonic crystal is coated with AZO (shown later).

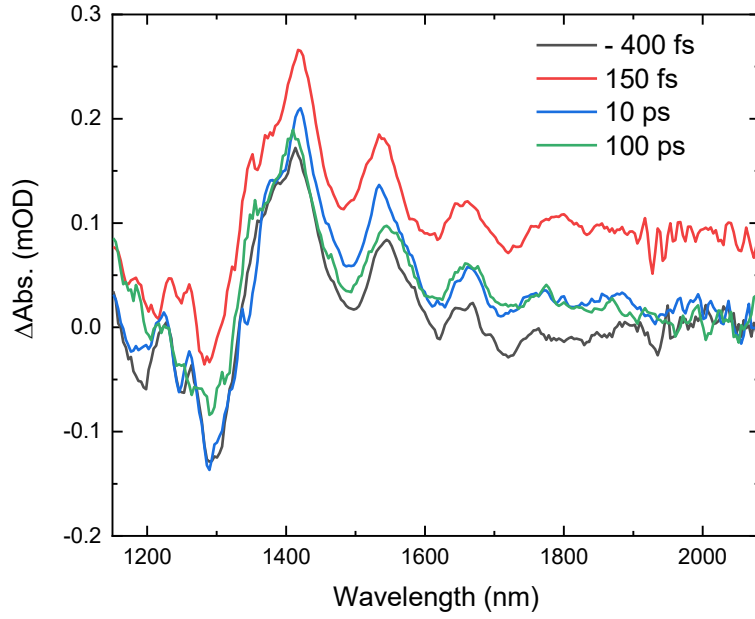


Figure 18. Transient absorption spectra at different delay times of a 1.4 μm inlayer periodicity photonic crystal.

Transient absorption spectrum over time of 1.4 μm inlayer periodicity sample and 150 nm AZO coating 1st sample (also noted the same in the transmission discussion) is shown in Fig. 19, the intensity of the 350 nm pump was $\sim 0.3 \text{ GW}/\text{cm}^2$, and it was kept the same for all photonic crystal with AZO coating samples. The spectral shape resembles pure AZO response, except for the sharp spectral dip around 2025 nm. The dip is approximately $\sim 35 \text{ nm}$ redshifted as compared to the dip in the transmission spectrum. Decay time constants were also evaluated the same way as for just the AZO coating. The same base wavelengths of 1400, 1600, 1800 and 2000 nm were evaluated, with the addition of spectral dip location. In the case of this sample the constants were: $\tau_{1400 \text{ nm}} = 9.0 \pm 1.3 \text{ ps}$, $\tau_{1600 \text{ nm}} = 13.2 \pm 1.0 \text{ ps}$, $\tau_{1800 \text{ nm}} = 14.8 \pm 1.1 \text{ ps}$, $\tau_{2000 \text{ nm}} = 20.5 \pm 4.3 \text{ ps}$, $\tau_{2023 \text{ nm}} = 28.8 \pm 4.6 \text{ ps}$. Contrary to decay constants of just the AZO coating, the τ constants kept increasing with increasing probe wavelength. The fast component was present and was also increasing with higher wavelengths, but it was not as pronounced. This suggests different internal decay dynamics. Decay constant $\tau_{2023 \text{ nm}}$ at the spectral dip seems to be shorter than the $\tau_{2000 \text{ nm}}$, but they are within error.

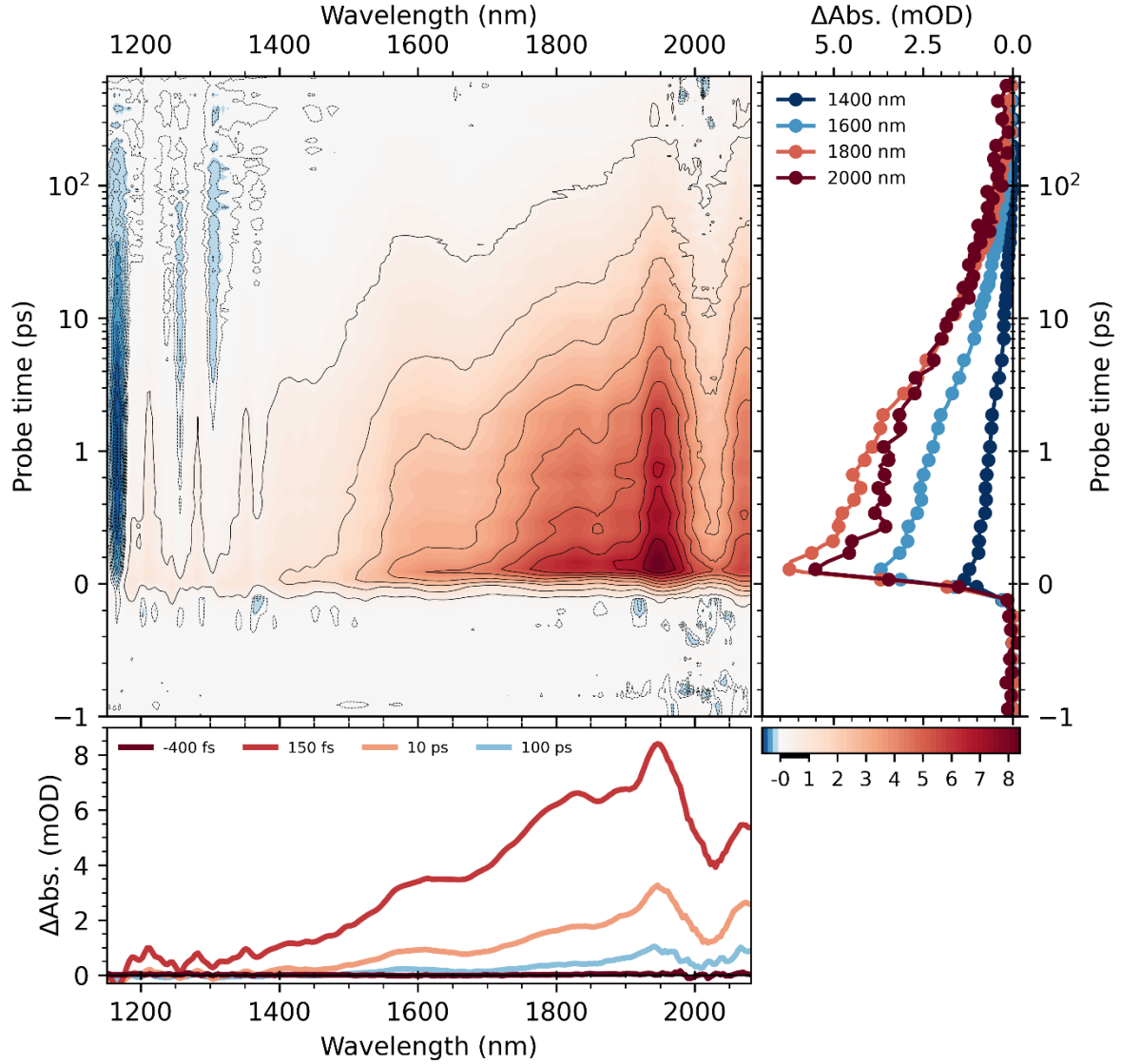


Figure 19. Spectral transient absorption color-map modulation over time of a 1.4 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (1st sample). Below - spectra at different delay times are shown, on the right - traces of different wavelengths are shown.

Transient absorption spectra at different delay times and traces at different wavelengths of 1.4 μm inlayer periodicity crystal and 150 nm AZO coating 2nd sample are shown in Fig. 20. Even though the samples were meant to be the same, the transient absorption spectra are quite different. It also has a spectral dip, but at a different wavelength, same as in the transmission graphs. The dip is roughly centered at ~ 1855 nm, is also redshifted as compared to the transmission dip, in this case by ~ 25 nm. Decay time constants kept increasing with increasing wavelength as in the 1st sample: $\tau_{1400 \text{ nm}} = 6.0 \pm 0.5$ ps, $\tau_{1600 \text{ nm}} = 9.0 \pm 0.9$ ps, $\tau_{1800 \text{ nm}} = 15.1 \pm 1.1$ ps, $\tau_{2000 \text{ nm}} = 19.1 \pm 1.9$ ps, $\tau_{1855 \text{ nm}} = 7.3 \pm 0.6$ ps. Decay times were quite similar, except for the decay constant at the dip $\tau_{1855 \text{ nm}}$, in this case the dip decayed faster than the surrounding wavelengths, but it was the only sample which showed such behavior.

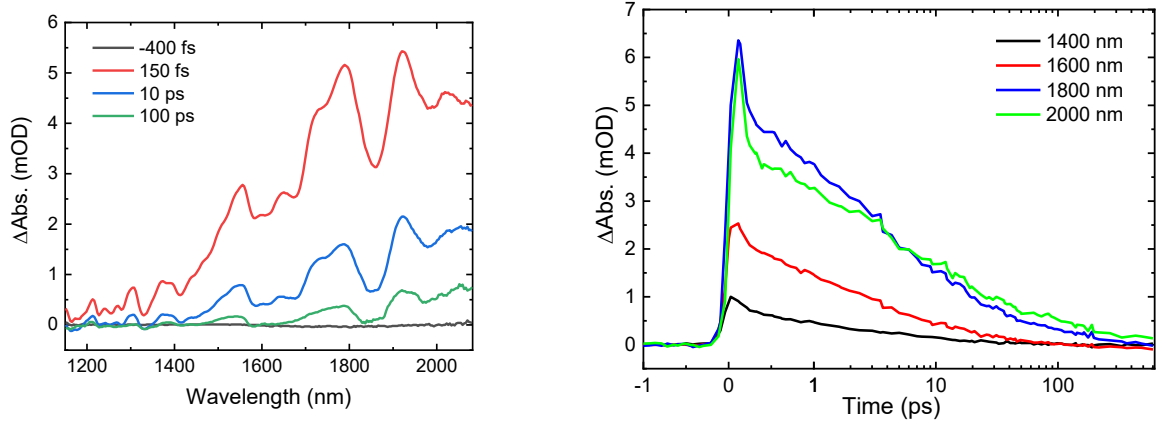


Figure 20. Transient absorption spectra at different delay times (left) and traces at different wavelengths (right) of a 1.4 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (2nd sample).

Moving on to 1.3 μm inlayer periodicity crystals coated with 150 nm of AZO, transient absorption spectra at different delay times and traces at different wavelengths of the 1st sample are shown in Fig. 21. Once again, the spectral dip in the transient absorption spectrum is redshifted as compared to the transmission spectrum dip by ~ 20 nm in this case. As in the samples before, decay time constants kept increasing with the increasing probe wavelength: $\tau_{1400 \text{ nm}} = 4.3 \pm 0.5$ ps, $\tau_{1600 \text{ nm}} = 11.7 \pm 0.7$ ps, $\tau_{1800 \text{ nm}} = 16.8 \pm 1.2$ ps, $\tau_{2000 \text{ nm}} = 25.0 \pm 1.6$ ps, $\tau_{1851 \text{ nm}} = 15.33 \pm 1.7$ ps. Decay time constants roughly match the values in the samples before, in this case the decay time $\tau_{1851 \text{ nm}}$ was only slightly smaller as compared to wavelengths not in the dip.

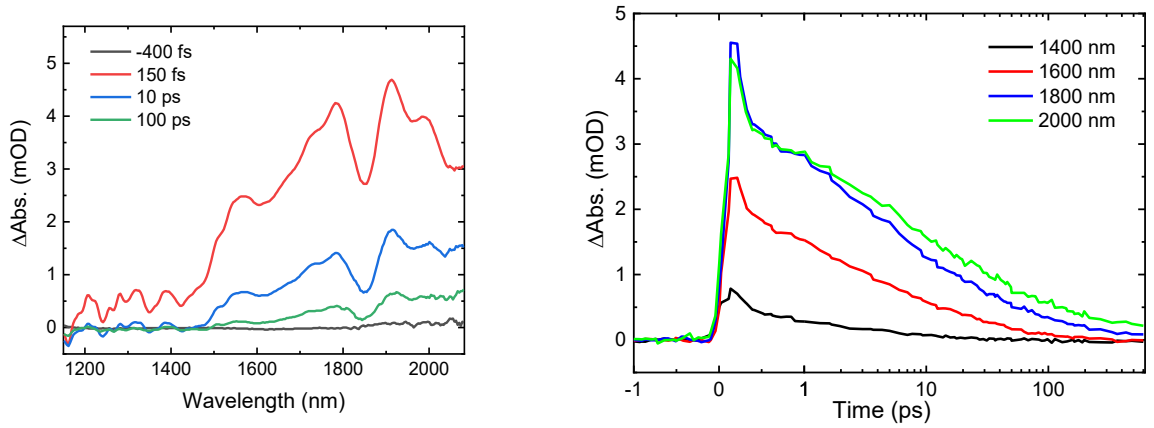


Figure 21. Transient absorption spectra at different delay times (left) and traces at different wavelengths (right) of a 1.3 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (1st sample).

The last sample in this analysis is a 2nd sample of 1.3 μm inlayer periodicity and a 150 nm AZO coating. Transient absorption spectra at different delay times and traces at different wavelengths are shown in Fig. 22. The observed spectral dip in the transient absorption spectrum is the smallest of all samples investigated, and in this case, it is blueshifted by ~ 15 nm as compared to the dip in the

transmission spectrum. This contradicts with the structures investigated before, but this sample exhibits the shortest wavelength spectral dip and the most strongly blueshifted broadband absorption.

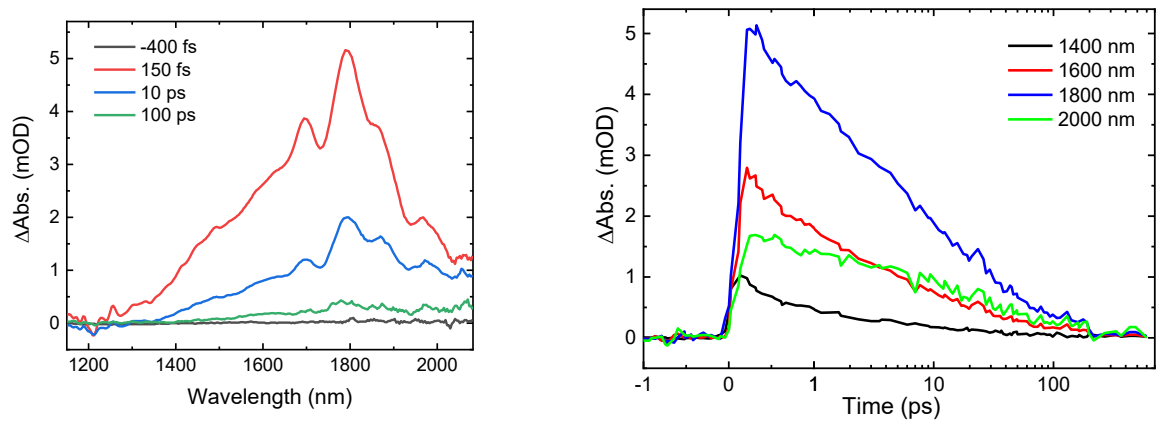


Figure 22. Transient absorption spectra at different delay times (left) and traces at different wavelengths (right) of a 1.3 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (2nd sample).

Linear transmission measurements of the AZO coated photonic crystals reveal well defined spectral dips corresponding to photonic resonances whose positions vary across different photonic crystal geometries. Crystals which are meant to be identical vary vastly in the transmission spectral shape they produce. Not only is the shape different, but the location of the photonic resonance is too. Transmittance of AZO coating is gradually declining towards the longer wavelengths and there are no enhanced spectral dips.

Pump probe measurements of only AZO coating exhibit a broadband transient absorption response across the probed near-infrared range, governed by intrinsic AZO carrier relaxation, with no spectral selectivity. The measurements on the bare photonic crystal show only weak, thermally induced signals, which were only achieved by increasing the pumping intensity by 25 times. These pump probe experiments demonstrate that the ultrafast optical response originates from the AZO coating, whereas the photonic crystal itself acts more as a passive optical structure.

When the AZO coating is integrated with the photonic crystal, the transient response retains the broadband character, similar to just the AZO coating response, but acquires a pronounced spectral dip, or a modulation, correlated with the photonic resonances observed in linear transmission. The linear transmittance spectra overlayed with the transient absorption spectra at a 150 fs delay are shown in Fig. 23.

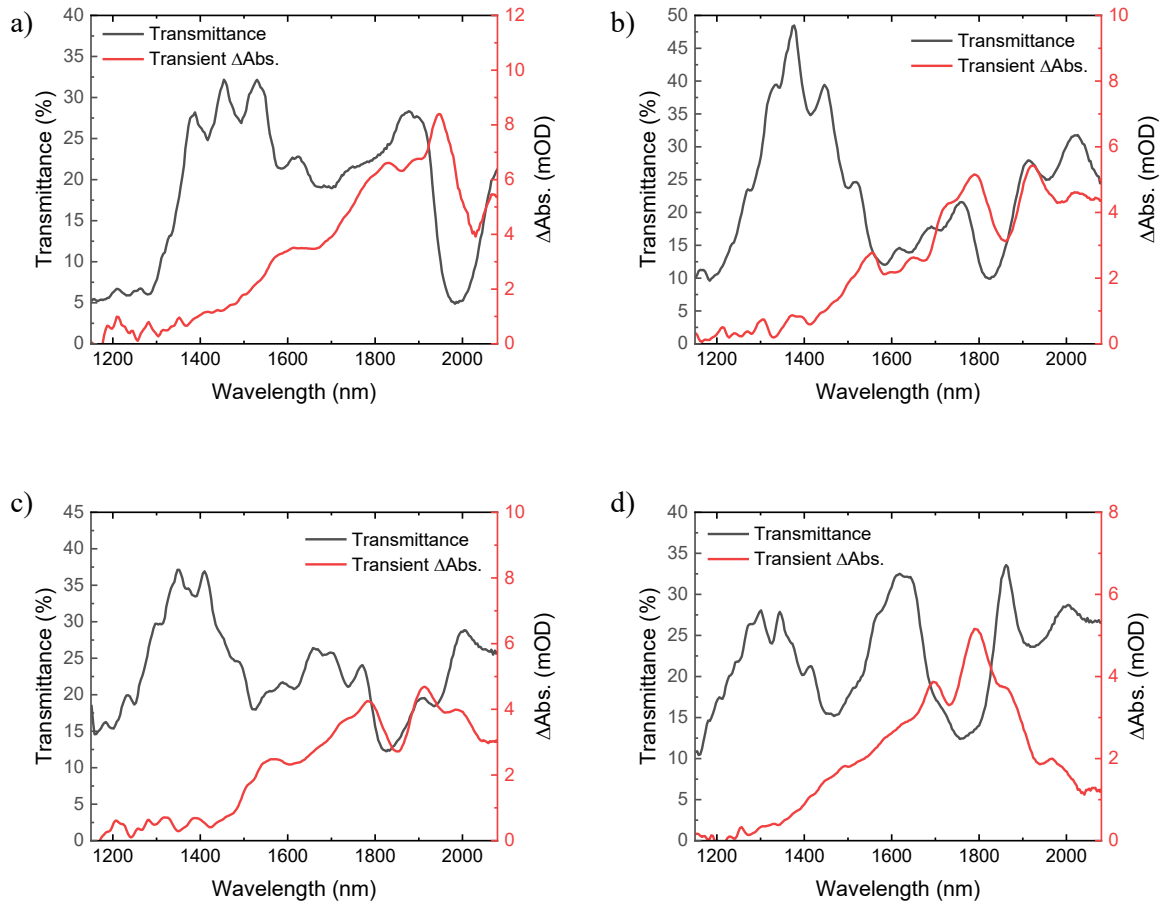


Figure 23. Transmittance spectra overlayed with transient absorption spectra at 150 fs for all AZO coated photonic crystals investigated. a) 1.4 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (1st sample), b) 1.4 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (2nd sample), c) 1.3 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (1st sample), d) 1.3 μm inlayer periodicity photonic crystal coated with 150 nm of AZO (2nd sample).

Whether a transmission spectral dip also appears in the pump probe signal probably is thought to depend on how well the field is overlapped within the AZO coating, whether a significant fraction of its electromagnetic energy density resides within the AZO layer. In that case, pump-induced changes in the AZO dielectric function effectively perturb the resonance condition of the mode. The degree of overlap depends on the spatial field distribution of the mode, which is determined by the photonic crystal geometry, as well as on the AZO thickness, refractive index contrast. To sum up, the photonic crystal geometry determines where the optical field is localized, while the AZO coating determines how that field responds in ultrafast time scale.

When the AZO coated photonic crystal is pumped, the free carrier density in AZO increases, increasing absorption while reducing the refractive index. This causes the modes overlapping with AZO to appear in the transient absorption measurements, at wavelengths very similar to unpumped transmission dips, just offset by 15-35 nm. Since the transient spectra did not exhibit a clear bipolar

spectral profile characteristic of a distinct resonance shift, the exact origin of the spectral offset remains uncertain. Three samples had a redshifted resonance, while one had a blueshifted resonance, so it is difficult to draw any conclusions.

Analysis of the transient decay dynamics reveals a gradual increase of relaxation times toward longer wavelengths in all AZO coated photonic crystals. At the photonic resonance wavelengths, the decay times are only slightly shorter (or not at all) than those at neighboring wavelengths, indicating that the photonic modes primarily influence the spectral amplitude of the response rather than the underlying carrier relaxation mechanisms.

The hybrid system of an ENZ material AZO, coated on a photonic crystal, exhibits spectrally selective ultrafast modulation arising from the interplay between photonic resonances, created by the photonic crystal, and broadband AZO carrier dynamics. These properties make hybrid photonic crystal and AZO coated structures promising platforms for ultrafast, wavelength selective optical modulators, or switches, and dynamically tunable photonic filters.

4.3 Pump-probe investigation of Ta₂O₅ coated SiO₂ grating

First, the ultrafast transient absorption response of just the Ta₂O₅ coating deposited on a fused silica substrate was investigated. Pump peak intensity on the sample was 150 MW/cm². Transient absorption spectra at different delay times and traces at different wavelengths are shown in Fig. 24. The spectra show broadband increase in difference absorption and from the traces it can be seen that there is almost no difference between wavelengths chosen. By fitting 1400 nm trace with a bi-exponential decay function, the weighed decay constant of the positive TA signal was determined to be $\tau = 95 \pm 15$ ps.

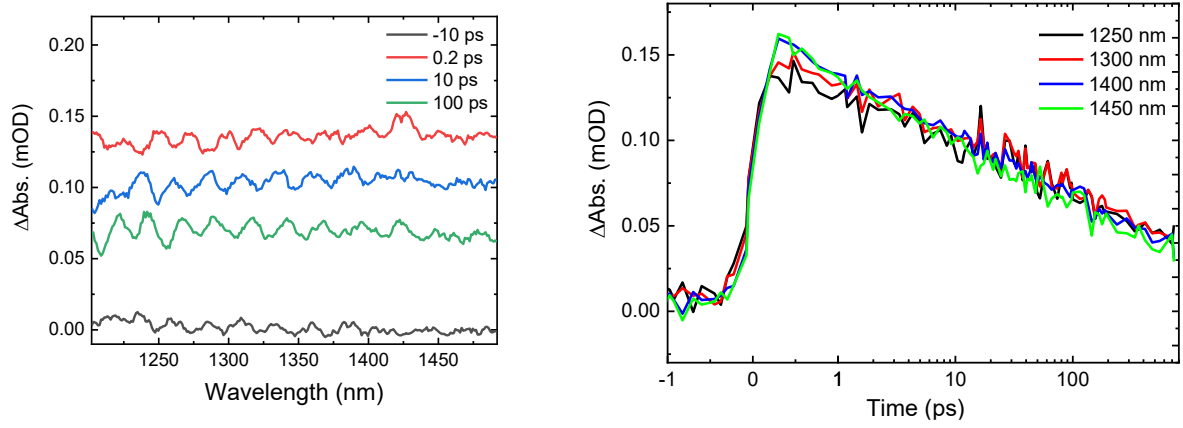


Figure 24. Transient absorption spectra at different delay times (left) and traces at different wavelengths (right) of a Ta_2O_5 coating on a glass substrate.

For the investigation of the grating, the intensity was decreased to 74 MW/cm^2 . The repetition rate of the laser was also decreased to 4.8 kHz, since the transient absorption signal did not fully decay between successive pulses at 26.7 kHz. The transient absorption spectra at different delay times for the s-polarized incident probe light is shown in Fig. 25. There are two distinct peaks of increased absorption at $\sim 1260 \text{ nm}$ and $\sim 1430 \text{ nm}$. The signal at 1310 nm is mostly noise, since the transmission at that wavelength is near-zero. The weighed decay time constant was within error the same as just the coating, $\tau = 95 \pm 7 \text{ ps}$.

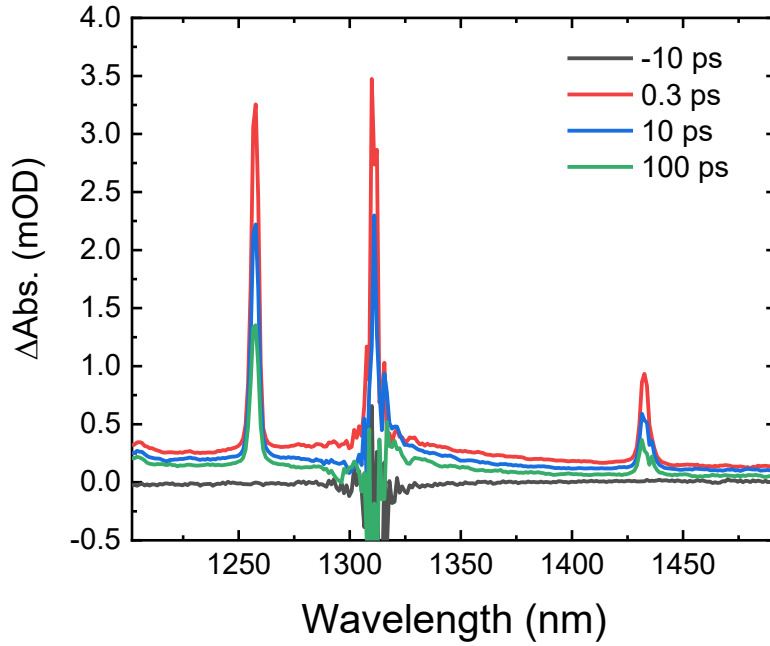


Figure 25. Transient absorption spectra at different delay times of a Ta₂O₅ coated SiO₂ grating, for s-polarized incident probe light.

For the investigation of the transient absorption for the p-polarized incident probe light, the grating inside the spectrograph was changed to have better resolution, since in both linear transmittance and ultrafast transient absorption spectra there is only one peak of interest around 1400 nm. Transient absorption spectra at different delay times and traces at different wavelengths for the p-polarized light are shown in Fig. 26. After 100 fs delay of 257 nm pump excitation, the coated grating initially exhibits an increase in absorption near the peak of the guided-mode resonance, observed as a positive transient absorption signal. This could be attributed to Ta₂O₅ UV absorption and free carrier generation. After another 400 fs, a dip in transient absorption spectrum appears and within 1 ps becomes negative. Such a spectral shape indicates that the transmission dip of the sample becomes redshifted. According to the resonance condition of dielectric gratings noted in equation 9, such a shift corresponds to an increase in the effective refractive index of the optical mode.

By fitting the 1398 nm peak with an exponential decay function, decay constant of the positive TA signal was determined to be $\tau = 150 \pm 9$ ps, but the full decay could not be observed due to physical limitations of the delay line. The decay constant of the 1396 nm negative TA signal peak could not be evaluated, as the signal remained close to its maximum even at a delay of 700 ps. The rise of the signal was also slow, with the negative peak reaching its maximum only after approximately 80 ps. The fact that delay times differ substantially suggests that multiple pump-induced processes contribute simultaneously. Longer lived refractive index modulation could be

potentially associated with trapped carriers, defect-state occupation, or slow dielectric relaxation in the Ta₂O₅ coating.

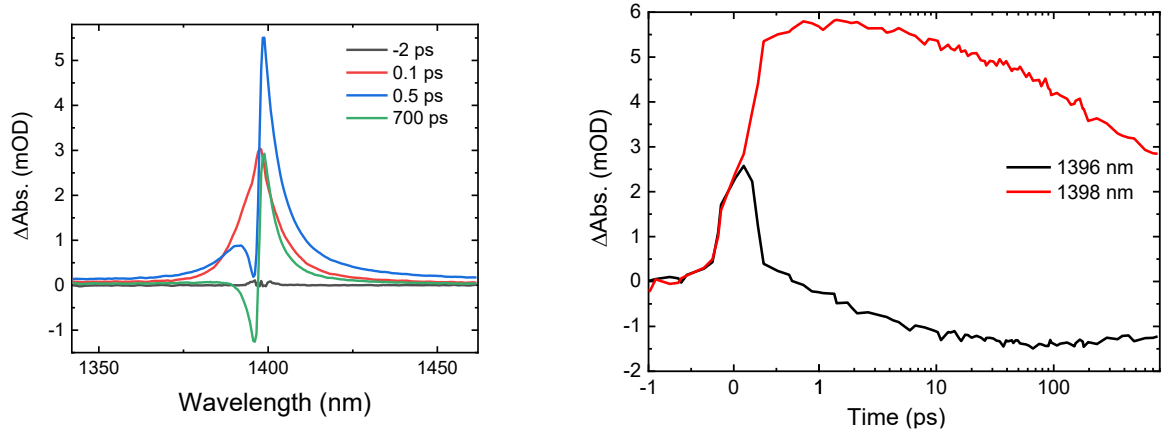


Figure 26. Transient absorption spectra at different delay times (left) and traces at different wavelengths (right) of a Ta₂O₅ coated SiO₂ grating, for p-polarized incident probe light.

By increasing the pump peak intensity by 2 times to 150 MW/cm², the transient absorption signal also increased by ~ 2 times, in both negative and positive contributions, showing potential to improve the contrast under more intense pumping conditions. The spectra are compared in Fig. 27. Transient absorption dataset of probe delay time versus wavelength with 150 MW/cm² pump intensity is shown in Fig 28.

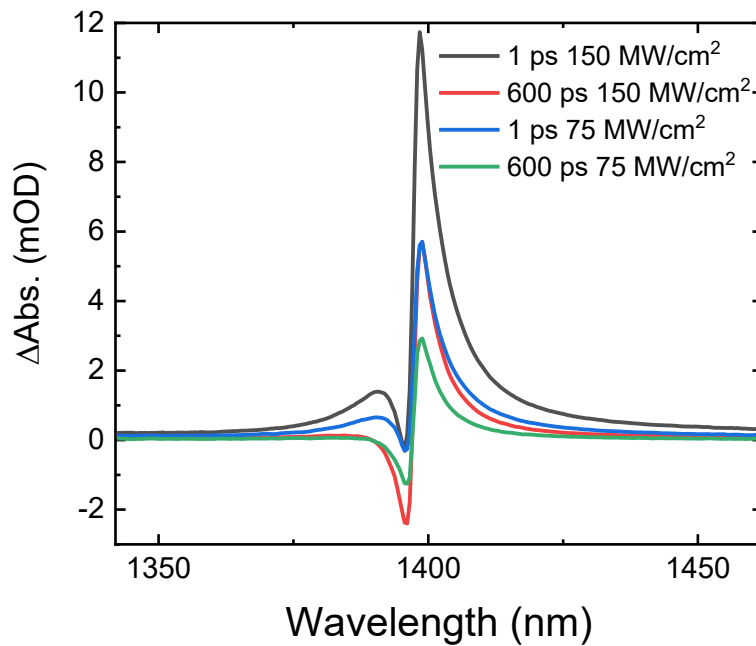


Figure 27. Transient absorption spectra comparison between peak intensities of the pump at 1 and 600 ps delay times.

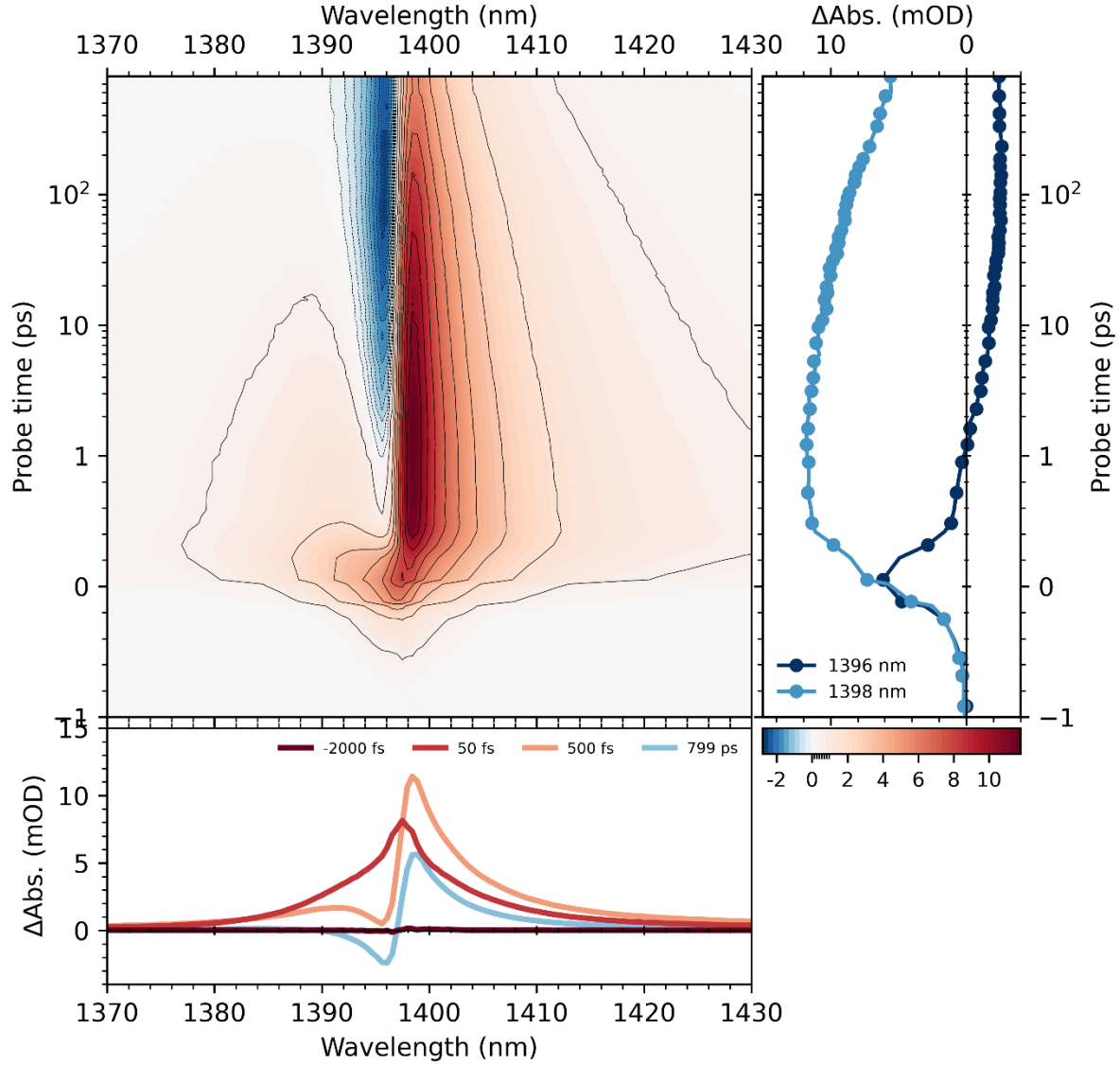


Figure 28. Spectral transient absorption color-map modulation over time of a Ta₂O₅ coated SiO₂ grating, for p-polarized incident probe light. Below - spectra at different delay times are shown, on the right - traces of different wavelengths are shown.

Transient reflectance of the Ta₂O₅ grating was also investigated. Transient reflectance spectra at different delay times for the s-polarized incident probe light is shown in Fig. 29. Transient reflectance is expressed in $\Delta R/R$ (10^{-3}) units. The same spectral features as in transient absorption measurement appear at approximately 1260 nm and 1430 nm in transient reflectance measurement, indicating a decrease in reflectance at these wavelengths. The decay constant τ at 1260 nm is 99 ± 5 ps, which is within error of the decay constant of the transient absorption signal at the same wavelength. In contrast to the transient absorption measurements, no spectral feature is observed around 1310 nm, as sufficient transmitted probe light was present. This supports the hypothesis that the spectral features near 1310 nm in Fig. 25 originated from the lack of transmitted probe signal

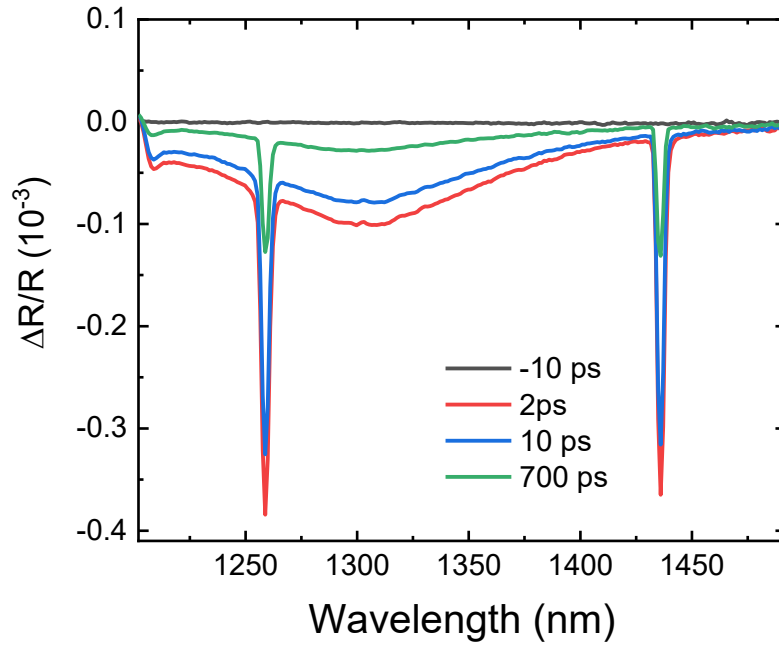


Figure 29. Transient reflectance spectra at different delay times of a Ta₂O₅ coated SiO₂ grating, for s-polarized incident probe light.

Transient reflectance spectra at different delay times for the p-polarized incident probe light is shown in Fig. 30. Transient reflectance measurements performed under the same excitation conditions revealed a spectrally similar feature near the guided-mode resonance. A decrease in reflectivity was observed on the blue side of the resonance, while the reflectivity increased on the red side. This sign reversal is consistent with the pump-induced redshift observed in transmission and provides independent evidence that ultraviolet excitation modifies the resonance condition of the grating rather than solely inducing broadband absorption changes in the coating. Compared to transmission, the transient reflectance feature appeared broader and less pronounced, which may reflect the different spectral line shape and lower resonance contrast in reflection, as well as additional background contributions. Decay constant at 1390 nm is $\tau = 95 \pm 5$ ps, while for 1414 nm it cannot be evaluated as was the case with the transient absorption measurement.

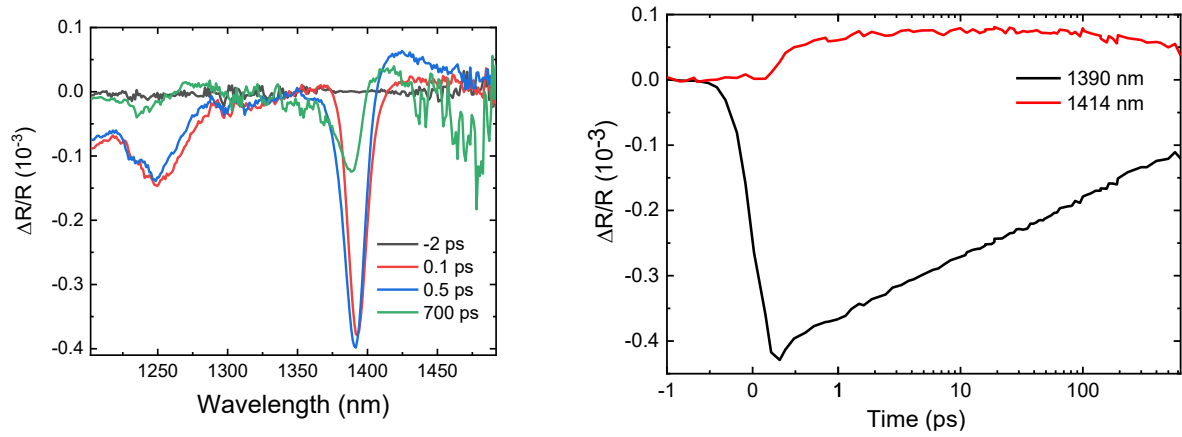


Figure 30. Transient reflectance spectra at different delay times (left) and traces at different wavelengths (right) of a Ta₂O₅ coated SiO₂ grating, for p-polarized incident probe light.

The combined transient transmission and reflectance measurements demonstrate that ultraviolet excitation induces a pronounced and spectrally selective optical response in the Ta₂O₅ coated SiO₂ grating for p-polarized light near the guided-mode resonance at approximately 1400 nm. While the transient response initially appears as a broadband increase in optical loss, it subsequently evolves into a dispersive spectral feature, characterized by adjacent positive and negative signal components in both transmission and reflection. Such a spectral response is consistent with a pump-induced redshift of the resonance, indicating a transient modification of the effective refractive index of the optical mode, likely caused by photoinduced changes in the Ta₂O₅ coating. The different decay dynamics observed between the spectral lobes, as well as between transmission and reflection, suggest that the transient response cannot be fully described by a single relaxation process and may involve overlapping contributions from short-lived absorptive changes and longer-lived refractive index modulation. Although the exact origin requires further investigation, the results clearly demonstrate that combining dielectric resonant structures with photoactive oxide coatings enables dynamic tuning of narrow spectral resonances. These findings highlight the potential of such hybrid structures for wavelength-selective all-optical modulation and ultrafast photonic switching applications.

A comparison between the two investigated hybrid photonic platforms reveals notable differences in both the spectral response and the temporal dynamics. The AZO coated photonic crystal structures exhibited clear pump-induced modulation near selected photonic resonances, confirming that the interaction between the photonic modes and the photoactive AZO layer can locally enhance or spectrally shape the transient optical response. However, the exact spectral position, modulation amplitude, and line shape varied significantly between nominally similar photonic crystal samples, indicating a strong sensitivity to fabrication tolerances, structural geometry, and modal overlap with the AZO coating. As a result, while resonance-related effects were observed, the reproducibility of the transient response across the sample series remained limited. In contrast, the Ta₂O₅ coated SiO₂

grating exhibited a highly spectrally defined and easily identifiable resonant response, with clear pump-induced modulation observed in both transmission and reflection near the Fano-like resonance. Although only a single grating sample was investigated in the present work, the sharp resonance lineshape and the mature fabrication technology of dielectric gratings and IBS deposited coatings suggest a potentially more controlled and reproducible optical platform. From a dynamical perspective, the AZO based structures exhibited faster relaxation on picosecond timescales, which may be advantageous for high-speed optical switching, whereas the grating-based structure showed slower but spectrally more selective modulation, potentially offering greater precision for wavelength-selective photonic applications.

5 Main results and conclusions

- Linear transmission measurements of AZO coated photonic crystals revealed distinct near-infrared resonances, while pump-probe measurements of the individual components showed broadband carrier-induced absorption in AZO and only weak transient response from the bare photonic crystal.
- In the hybrid AZO-photonic crystal structures, spectrally localized transient features were observed at wavelengths corresponding to the linear optical resonances, confirming that photonic modes can selectively shape the broadband ultrafast response of AZO. The relaxation dynamics remained comparable to standalone AZO films, indicating that the temporal response is primarily governed by intrinsic carrier relaxation in AZO.
- Linear optical characterization of the Ta₂O₅ coated SiO₂ grating revealed a sharp resonance near 1400 nm for p-polarized incident light, consistent with a Fano-type guided-mode resonance and indicating strong spectral selectivity of the dielectric grating platform.
- Ultraviolet pump-probe measurements of the grating revealed a pronounced transient modulation in both transmission and reflection, evolving into a bipolar spectral response consistent with a pump-induced redshift of the optical resonance. Compared to the AZO-based structures, the grating exhibited a more spectrally defined and experimentally reproducible response, although with slower relaxation time.
- Both investigated structures showed promising but distinct transient optical behavior. The AZO coated photonic crystals were characterized by faster dynamics, whereas the dielectric grating produced a more clearly resolved resonant response.

6 References

1. Euser, T. G. *et al.* All-optical octave-broad ultrafast switching of Si woodpile photonic band gap crystals. *Phys. Rev. B - Condens. Matter Mater. Phys.* **77**, 1–6 (2008).
2. Sturmberg, B. C. P., Dossou, K. B., Botten, L. C., McPhedran, R. C. & De Sterke, C. M. Fano resonances of dielectric gratings: symmetries and broadband filtering. *Opt. Express* **23**, A1672–A1686 (2015).
3. Joannopoulos, J. D., Villeneuve, P. R. & Fan, S. Photonic crystals: putting a new twist on light. *Nature* **386**, 143–149 (1997).
4. Wang, S. S. & Magnusson, R. Theory and applications of guided-mode resonance filters. *Appl. Opt.* **32**, 2606–2613 (1993).
5. Zhou, W. *et al.* Progress in 2D photonic crystal Fano resonance photonics. *Prog. Quantum Electron.* **38**, 1–74 (2014).
6. Pasanen, H. P., Vivo, P., Canil, L., Abate, A. & Tkachenko, N. Refractive index change dominates the transient absorption response of metal halide perovskite thin films in the near infrared. *Phys. Chem. Chem. Phys.* **21**, 14663–14670 (2019).
7. Fomra, D. *et al.* Nonlinear optics at epsilon near zero: From origins to new materials. *Appl. Phys. Rev.* **11**, (2024).
8. Saha, S. *et al.* Engineering the temporal dynamics of all-optical switching with fast and slow materials. *Nat. Commun.* **14**, 1–9 (2023).
9. Moscardi, L., Varas, S., Chiasera, A., Scotognella, F. & Guizzardi, M. Ultrafast broadband optical modulation in indium tin oxide/titanium dioxide 1D photonic crystal. *J. Eur. Opt. Soc. Publ.* **18**, 1–5 (2022).
10. Kinsey, N. *et al.* Epsilon-near-zero Al-doped ZnO for ultrafast switching at telecom wavelengths. *Optica* **2**, 616–622 (2015).
11. Roelkens, G. *et al.* III–V/silicon photonics for short-wave infrared spectroscopy. *IEEE J. Quantum Electron.* **48**, 292–298 (2011).
12. Porter, G. Flash photolysis and spectroscopy: A new method for the study of free radical reactions. *Life Sci. Leg. Georg. Porter* 41–60 (2006).
13. Fischer, M. C., Wilson, J. W., Robles, F. E. & Warren, W. S. Invited Review Article: Pump-probe microscopy. *Rev. Sci. Instrum.* **87**, 1–21 (2016).
14. Maiuri, M., Garavelli, M. & Cerullo, G. Ultrafast spectroscopy: State of the art and open challenges. *J. Am. Chem. Soc.* **142**, 3–15 (2019).
15. Berera, R., van Grondelle, R. & Kennis, J. T. M. Ultrafast transient absorption spectroscopy: Principles

- and application to photosynthetic systems. *Photosynth. Res.* **101**, 105–118 (2009).
16. Hilton, D. J., Prasankumar, R. P. & Taylor, A. J. Ultrafast pump-probe spectroscopy. *Optical Techniques for Solid-State Materials Characterization* 42 at (2011).
 17. Kovalenko, S. A., Dobryakov, A. L., Ruthmann, J. & Ernsting, N. P. Femtosecond spectroscopy of condensed phases with chirped supercontinuum probing. *Phys. Rev. A* **59**, 2369 (1999).
 18. Grinberg, D. & Paz, Y. Recent progress in transient spectroscopy studies of solid state photocatalysts. *Curr. Opin. Green Sustain. Chem.* **41**, 100768 (2023).
 19. Berera, R., van Grondelle, R. & Kennis, J. T. M. Ultrafast transient absorption spectroscopy: principles and application to photosynthetic systems. *Photosynth. Res.* **101**, 105–118 (2009).
 20. Samineni, P., deCruz, A., Villafañá, T. E., Warren, W. S. & Fischer, M. C. Pump-probe imaging of historical pigments used in paintings. *Opt. Lett.* **37**, 1310–1312 (2012).
 21. Gabriel, M. M. *et al.* Direct imaging of free carrier and trap carrier motion in silicon nanowires by spatially-separated femtosecond pump-probe microscopy. *Nano Lett.* **13**, 1336–1340 (2013).
 22. Ombinda-Lemboumba, S. *et al.* Femtosecond pump probe spectroscopy for the study of energy transfer of light-harvesting complexes from extractions of spinach leaves. *S. Afr. J. Sci.* **105**, 376–386 (2009).
 23. Cerullo, G., Manzoni, C., Lürer, L. & Polli, D. Time-resolved methods in biophysics. 4. Broadband pump—probe spectroscopy system with sub-20 fs temporal resolution for the study of energy transfer processes in photosynthesis. *Photochem. Photobiol. Sci.* **6**, 135–144 (2007).
 24. Wei, L. & Min, W. Pump-probe optical microscopy for imaging nonfluorescent chromophores. *Anal. Bioanal. Chem.* **403**, 2197–2202 (2012).
 25. Villafana, T. E. *et al.* Femtosecond pump-probe microscopy generates virtual cross-sections in historic artwork. *Proc. Natl. Acad. Sci. U. S. A.* **111**, 1708–1713 (2014).
 26. Dubietis, A., Tamošauskas, G., Šuminas, R., Jukna, V. & Couaïron, A. Ultrafast supercontinuum generation in bulk condensed media. *Lith. J. Phys.* **57**, 1–34 (2017).
 27. Yablonovitch, E. Physical review letters. **58**, 2059–2062 (1987).
 28. Lipson, R. H. & Lu, C. Photonic crystals: A unique partnership between light and matter. *Eur. J. Phys.* **30**, (2009).
 29. Case, F. Letters 21. *Phys. Rev. Lett.* **67**, 2295 (1991).
 30. Ho, K. M., Chan, C. T., Soukoulis, C. M., Biswas, R. & Sigalas, M. Photonic band gaps in three dimensions: New layer-by-layer periodic structures. *Solid State Commun.* **89**, 413–416 (1994).
 31. Pudis, D., Goraus, M. & Urbancova, P. Optical properties of woodpile structures for application on the surface of photonic devices. *Appl. Surf. Sci.* **461**, 227–232 (2018).

32. Ruckebusch, C., Sliwa, M., Pernot, P., de Juan, A. & Tauler, R. Comprehensive data analysis of femtosecond transient absorption spectra: A review. *J. Photochem. Photobiol. C Photochem. Rev.* **13**, 1–27 (2012).
33. Wu, J., Xie, Z. T., Sha, Y., Fu, H. Y. & Li, Q. Epsilon-near-zero photonics: infinite potentials. *Photonics Res.* **9**, 1616 (2021).
34. Tian, W. *et al.* Highly efficient super-continuum generation on an epsilon-near-zero surface. *ACS omega* **5**, 2458–2464 (2020).
35. Enoch, S., Tayeb, G., Sabouroux, P., Guérin, N. & Vincent, P. A metamaterial for directive emission. *Phys. Rev. Lett.* **89**, 213902 (2002).
36. Sakellari, I. *et al.* Quantum dot based 3D printed woodpile photonic crystals tuned for the visible. *Nanoscale Adv.* **1**, 3413–3423 (2019).
37. Becker, C. *et al.* Two-color pump-probe experiments on silicon inverse opals. *Appl. Phys. Lett.* **87**, 1–4 (2005).
38. Li, C. *et al.* Invertible optical nonlinearity in epsilon-near-zero materials. *Phys. Rev. Res.* **5**, 1–8 (2023).
39. Chaneliere, C., Autran, J. L., Devine, R. A. B. & Balland, B. Tantalum pentoxide (Ta₂O₅) thin films for advanced dielectric applications. *Mater. Sci. Eng. R Reports* **22**, 269–322 (1998).
40. Chen, X., Bai, R. & Huang, M. Optical properties of amorphous Ta₂O₅ thin films deposited by RF magnetron sputtering. *Opt. Mater. (Amst)*. **97**, 109404 (2019).
41. Ren, W. *et al.* Annealing effects on the optical and electrochemical properties of tantalum pentoxide films. **10**, 704–713 (2021).
42. Online, V. A. *et al.* Charge carrier dynamics in tantalum oxide overlayered and tantalum doped hematite. 3206–3215 (2019).
43. Lee, J., Lu, W. D. & Kioupakis, E. Electronic and optical properties of oxygen vacancies in amorphous Ta₂O₅ from first principles. *Nanoscale* **9**, 1120–1127 (2017).
44. Tantara Kerr nonlinear integrated photonics. **8**, (2021).
45. Palmer, C. H. *Diffraction Grating Handbook. Journal of the Optical Society of America* vol. 46 (1970).
46. Gambino, F., Giaquinto, M., Ricciardi, A. & Cusano, A. (INVITED)A review on dielectric resonant gratings: Mitigation of finite size and Gaussian beam size effects. *Results Opt.* **6**, 100210 (2022).
47. Grineviciute, L., Nikitina, J., Babayigit, C. & Staliunas, K. Fano-like resonances in nanostructured thin films for spatial filtering. *Appl. Phys. Lett.* **118**, (2021).
48. Fano, U. Effects of configuration interaction on intensities and phase shifts. *Phys. Rev.* **124**, 1866–1878 (1961).

49. Bej, S., Tkachenko, N., Fickler, R. & Niemi, T. Ultrafast Modulation of Guided-Mode Resonance in a Nonlinear Silicon Nitride Grating. **2402632**, 1–10 (2025).
50. Shcherbakov, M. R. *et al.* semiconductor metasurfaces. *Nat. Commun.* **8**, 1–5 (2017).
51. Ladika, D. Linear and Nonlinear optical properties of materials for the development of 3D photonic nanostructures at Telecommunication wavelengths. (2023).
52. Galinis, J. *et al.* Filamentation and supercontinuum generation in solid-state dielectric media with picosecond laser pulses. *Phys. Rev. A - At. Mol. Opt. Phys.* **92**, 1–5 (2015).

7 Santrauka

Gytis Zaremba

Fotoninių struktūrų su dielektrinėmis dangomis dinamikos tyrimas žadinimo-zondavimo spektroskopijos metodais

Fotoninės struktūros, tokios kaip fotoniniai kristalai ir dielektrinės gardelės, leidžia valdyti šviesos sklaidimą per geometrines formas, sudarydamos fotoninius draustinius tarpus, nukreiptąsias modas ir siaurus rezonansinius spektrinius požymius. Jas sujungus su optiškai aktyviomis plonasluoksniėmis dangomis, žadinimo impulso sukelti lūžio rodiklio, sugerties ar krūvininkų koncentracijos pokyčiai gali modifikuoti šiuos rezonansus. Todėl tokios hibridinės sistemos yra perspektyvios ultra greitai optinei moduliacijai ir bangos ilgio atžvilgiu selektyviam perjungimui artimojoje bei trumpabangėje infraraudonojoje srityje.

Šio darbo tikslas buvo ištirti padengtų mikrometrinių fotoninių struktūrų ultra greitąją optinę dinamiką taikant žadinimo zondavimo sugerties spektroskopiją. Buvo tirtos dvi sistemos: aliuminiu legiruoto cinko oksido (AZO) danga padengti fotoniniai kristalai ir tantalio pentoksido (Ta_2O_5) danga padengta silicio dioksido (SiO_2) gardelė. Mažiems fotoninių kristalų bandiniams matuoti naudota mikroskopinė žadinimo-zondavimo sistema, kurioje zondavimo pluoštas sufokusuotas iki maždaug $9\ \mu m$. Baltos šviesos kontinuumo generacija YAG kristale buvo optimizuota siekiant gauti plačiajuostį zondo spektrą nuo maždaug $1,2\ \mu m$ iki $2,08\ \mu m$, leidžiantį atlikti laike kintančio pralaidumo ir atspindžio matavimus mažesniu nei mOD jautrumu.

AZO danga padengtų fotoninių kristalų tiesinio pralaidumo spektruose stebėti aiškūs artimosios infraraudonosios srities spektriniai požymiai, siejami su fotoniniais rezonansais arba fotoninio draustinio tarpo pobūdžio elgsena. Matavimai parodė, kad atskiros AZO dangos pasižymi plačiajuosčiu trumpalaikiu sugerties atsaku, o nepadengti fotoniniai kristalai sukuria tik silpną sugerties pakitimo signalą. Hibridinėse struktūrose trumpalaikis atsakas apjungė plačiajuostį AZO pobūdį su papildoma spektrine moduliacija ties fotoniniais rezonansais. Tai rodo, kad fotoninis kristalas veikia žadinimo impulsu sukulto AZO atsako spektrinę išraišką, o relaksacijos dinamika daugiausia išlieka nulemta vidinės krūvininkų relaksacijos AZO dangoje.

Ta_2O_5 danga padengta SiO_2 gardelė pasižymėjo ryškesniu ir spektriškai aiškiau apibrėžtu atsaku. Fano tipo rezonansas ties maždaug $1400\ nm$, esant p-poliarizacijos krintančiai šviesai, buvo stipriai moduluojamas žadinant $257\ nm$ spinduliuote. Trumpalaikio pralaidumo signalas nuo pradinio teigiamo sugerties požymio perėjo į bipolinį spektrinį atsaką, atitinkantį žadinimo impulsu sukeltą rezonanso raudonąjį poslinkį. Trumpalaikio atspindžio matavimai parodė atitinkamą ženklą keičiantį atsaką, palaikantį šią interpretaciją. Gardelės atsakas buvo lėtesnis, relaksacija vyko šimtų pikosekundžių skalėje, o tai leidžia manyti, kad prie jo prisideda ilgesnės gyvavimo trukmės fotoindukuotos būsenos Ta_2O_5 .

Apibendrinant, AZO danga padengti fotoniniai kristalai pasižymėjo greitesne krūvininkų nulemta dinamika, tačiau mažesniu atsikartojamumu tarp bandinių, o Ta_2O_5 danga padengta SiO_2 gardelė užtikrino tikslesnę ir geriau atkuriamą rezonansinę moduliaciją. Šie rezultatai rodo, kad padengtos mikrometrinės fotoninės struktūros gali formuoti ultragreituosius optinius atsakus trumpabangėje infraraudonojoje srityje ir būti naudingos bangos ilgio atžvilgiu selektyviai fotoninei moduliacijai.

8 Summary

Gytis Zaremba

Transient absorption investigation of photonic microstructures with dielectric coatings

Photonic structures such as photonic crystals and dielectric gratings control light propagation through geometry, enabling photonic band gaps, guided modes, and narrow resonant spectral features. When combined with optically active thin-film coatings, pump-induced changes in refractive index, absorption, or carrier concentration can modify these resonances. Such hybrid systems are therefore promising for ultrafast optical modulation and wavelength-selective switching in the near- and short-wave infrared range.

The aim of this work was to investigate the ultrafast optical dynamics of coated microscale photonic structures using transient absorption spectroscopy. Two systems were studied: aluminum-doped zinc oxide (AZO) coated photonic crystals and a tantalum pentoxide (Ta_2O_5) coated silicon dioxide (SiO_2) grating. A microscopic pump-probe setup was used to measure small photonic crystal samples, with the probe focused to approximately 9 μm . White-light continuum generation in YAG was optimized to provide a broadband probe from approximately 1.2 μm to 2.08 μm , enabling transient transmission and reflection measurements with sub-mOD sensitivity.

For AZO coated photonic crystals, linear transmission spectra showed distinct near-infrared spectral features associated with photonic resonances or band-gap-like behavior. Measurements confirmed that standalone AZO films exhibit a broadband transient absorption response, while bare photonic crystals produce only a weak pump-probe signal. In the hybrid structures, the transient response combined the broadband character of AZO with additional spectral modulation near the photonic resonances. This indicates that the photonic crystal influences how the pump-induced AZO response appears spectrally, while the relaxation dynamics remain mainly governed by intrinsic carrier relaxation in the AZO coating.

The Ta_2O_5 coated SiO_2 grating showed a sharper and more spectrally defined response. Fano-type resonance near 1400 nm for p-polarized incident light was strongly modulated under 257 nm excitation. The transient transmission signal evolved from an initial positive absorption feature into a bipolar spectral response, consistent with a pump-induced redshift of the resonance. Transient reflectance measurements showed a corresponding sign-changing response, supporting this interpretation. The grating response was slower, with relaxation on hundreds of picoseconds, suggesting contributions from longer-lived photoinduced states in Ta_2O_5 .

Overall, AZO coated photonic crystals showed faster carrier-driven dynamics but greater sample-to-sample variability, while the Ta_2O_5 coated SiO_2 grating provided more precise and reproducible resonant modulation. These results demonstrate that coated microscale photonic structures can shape ultrafast optical responses in the short-wave infrared and may be useful for wavelength-selective photonic modulation.