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Executive Summary of the Thesis

Ultrafast spectroscopy for semiconductor metasurfaces

Laurea Magistrale in Engineering Physics - Ingegneria Fisica

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1. Introduction

Active polarization control is an essential aspect of telecommunications and is been extensively used for information encoding. However, conventional polarization optics are quickly reaching their limits in terms of miniaturization and speed. In this context, metasurfaces have been proposed as platforms for polarization control due to their incredible ability to shape light on a sub-wavelength scale. Furthermore, the response of these devices is characterized by ultrafast processes taking place in a few picoseconds after excitation, enabling Terahertz polarization switching. The ability to access such short time scales has been made possible only by advances in ultrafast spectroscopy, an ensemble of experimental techniques aimed at studying ultrafast electron dynamics. The simplest of these techniques is called pump-probe spectroscopy and consists of using a strong pulse (pump) to induce variations in the optical properties of the sample which are then monitored by a second weaker pulse (probe) at different time delays with respect to the pump. Here we present a reflective anisotropic nonlocal metasurface based on AlGaAs nanowires for active polarization control and reconfigurable by all-optical means. A customized pump-probe setup was used to perform two different experiments aimed at characterizing the sample transient dichroism and birefringence.

2. Introduction to metasurfaces

2.1. Optics of periodic media

Since the discovery of diffraction gratings in the 18^{th} century, periodic media have been at the center of intensive research in the development of technologies for the control of optical beams, offering the capability to tailor selective behavior in frequency, direction of propagation and polarization. In the last decades, the increasing demand for more complex structuring and localization of light on a sub-wavelength scale has led to the development of artificial materials with unusual optical properties that are not observed in nature. Initially, the interest focused on Photonic crystals (PCs) consisting of a periodic arrangement of low and high dielectric constant unit cells which lead to diffraction effects resulting in frequency ranges for which light cannot propagate. A decade later, Metamaterials (MMs) were proposed as platforms capable of shaping light with extreme efficiency via the interaction with intense near fields produced by a sub-wavelength periodic array of highly polarizable resonant nanoparticles. These devices pro-

vide additional degrees of freedom in tailoring the optical response compared to PCs, together with a more convenient modeling approach since the sub-wavelength nature of the array allows to define effective parameters for its characterization. In the last decade, attention has shifted towards the 2D counterpart of MMs, known as Metasurfaces (MSs), due to easier and cheaper fabrication processes, smaller sizes and lower losses.

2.2. Resonant nanoparticles

Particles of sizes between 1 and 100 nm, known as nanoparticles (NPs), exhibit unique scattering properties suitable for the control of light on the nanoscale. The optical response of these structures shows a strong resonantly enhanced polarization dependent on their size, shape and surrounding medium, making them ideal constitutive blocks in the development of artificial materials with desired optical properties. The first NPs exploited plasmonic resonances in noble metals. However, the metallic nature of these structures presents several drawbacks due to ohmic losses and thermal dissipation. Moreover, their optical response is dominated by resonances of electric dipolar type, limiting the ability to engineer the directionality of the scattered light. To overcome these issues, high-index dielectric NPs, which exploit Mie-resonances instead of plasmonics, have been proposed. These structures can support different orders of electric and magnetic modes, enabled by the excitation of displacement currents, providing additional features such as unidirectional scattering and high-field confinement with very low losses.

2.3. Metasurfaces

MSs are two-dimensional optically thin planar arrays of densely packed scatterers having subwavelength size and periodicity. The incident field is strongly coupled to the near fields produced by the inclusions, which usually consist of resonant plasmonic or dielectric NPs, resulting in abrupt phase changes that ultimately define the MS optical response in the far field. Therefore, a proper arrangement of meta-atoms enables control over the phase, amplitude and polarization of the scattered fields. MSs were initially proposed as platforms to shape light into a desired wavefront by engineering a phase gradient acting on the inclusion geometric parameters [1]. These devices implemented a local response, i.e. resonances are strongly confined in the single unit cell with no interaction between the near fields of adjacent meta-atoms, since it allows straightforward designs of arrays where each scatterer is independently tailored to apply a specific transformation without affecting the behavior of the others. However, spatial localization results in a broadband spectral response precluding the ability to achieve frequency selectivity. Additional control over the spectral features of light can be obtained in structures having extended states that are coupled to the incident field only for those frequencies matching their spatial dispersion. Further extensions in MSs functionalities are provided by implementing tuning mechanisms that dynamically modify the static features of the array. A particularly important reconfiguration approach consists of exploiting an all-optical scheme where an external optical stimulus is used to induce a transient variation in the MS properties. Nowadays, MSs are used in a wide variety of applications, as compact alternatives to conventional optics or for completely new functionalities. In particular, they provide the ability to realize active polarization-sensitive devices via reconfigurable arrays of anisotropic meta-atoms.

3. Introduction to ultrafast spectroscopy

3.1. Pulse propagation in nonlinear media

Waves propagating in nonlinear media induce a polarization $P(z,t) = P^{(l)}(z,t) + P^{(nl)}(z,t)$, comprising a linear part and a nonlinear term. Starting from Maxwell's equations in scalar approximation, the evolution of the field can be described by [2]:

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 D(z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P^{(nl)}(z,t)}{\partial t^2} \tag{1}$$

where $D(z,t) = \varepsilon_0 E(z,t) + P^{(l)(z,t)}$. The nonlinear polarization acts as a source term and is responsible for a huge variety of effects due to its nonlinear dependence on $E(z,t)$, which is usually expressed as a power series of the electric

field:

$$P = \epsilon_0 \left(\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right) \quad (2)$$

where $\chi^{(n)}$ are the nth order susceptibilities of the medium. As the intensity of the field increases, the nonlinear terms start becoming increasingly important so that the polarization drifts from its linear behavior and nonlinear effects arise.

3.2. Second order nonlinear effects

Second order nonlinear effects are mediated by a nonlinear polarization $P^{(2)} = \varepsilon_0 \chi^{(2)} E^2$ which is responsible for the following processes:

- **Second Harmonic generation (SHG):** waves interact with themselves generating new components with twice the frequency of the initial one.
- **Sum Frequency Generation (SFG):** the interaction between waves at frequencies ω_1 and ω_2 results into a new component oscillating at $\omega_3 = \omega_1 + \omega_2$.
- **Difference Frequency Generation (DFG):** waves at frequencies ω_3 and ω_1 interact to produce a new wave at frequency $\omega_2 = \omega_3 - \omega_1$.
- **Optical Rectification (OR):** waves interact with themselves to produce a constant signal.

A particularly important DFG process, called **Optical Parametric Amplification**, relies on the interaction between a weak and an intense pulse oscillating at different frequencies. In this process an intense beam at ω_3 (pump) transfers energy to the beam at ω_1 (signal) amplifying it, and generating light at $\omega_2 = \omega_3 - \omega_1$ (idler) as a consequence of energy conservation. Maximizing the conversion efficiency of these processes requires the induced polarization to be in phase with the driving field, a condition called phase matching.

3.3. Third order nonlinear effects

Intense laser pulses propagating in a centrosymmetric medium, for which $\chi^{(2)} = 0$, experience a nonlinear polarization $P^{(3)} = \varepsilon_0 \chi^{(3)} E^3$. This term introduces a time-dependent nonlinear modulation of the refractive index resulting in a spectral broadening of the pulse through a time-dependent phase modulation. This effect is known as Self Phase Modulation and can be exploited to generate ultra-broadband pulses.

3.4. Pump-probe spectroscopy

Pump-probe spectroscopy is an experimental technique used to study electron dynamics with femtosecond resolution. It consists of employing an intense laser pulse (pump) to induce changes in the optical properties of the system under examination, which are then monitored by a weaker pulse (probe) at different time delays with respect to the pump arrival, providing information on the relaxation dynamics of the electrons in the system. In our case, the measured quantity of interest is the relative differential reflection of the sample $\Delta R/R = \frac{R'-R}{R}$, where R and R' are the unperturbed and perturbed reflectivity, respectively. This quantity is directly connected with variations in the electronic populations via the differential absorption of the sample $\Delta\alpha$. In particular, three different processes can be discriminated by the sign of $\Delta\alpha$:

- **Ground State Bleaching (GSB)**: ground state population is decreased by pump-induced promotion of carriers in the excited state resulting in lower absorption ($\Delta\alpha < 0$).
- **Excited State Absorption (ESA)**: probe absorption can further promote carriers from the excited state ($\Delta\alpha > 0$).
- **Stimulated Emission (SE)**: the probe can induce stimulated emission from the level populated by the pump ($\Delta\alpha < 0$). In this case we have the same sign of GSB but SE occurs at lower energies.

Moreover, it can be shown that the measured signal is proportional to the convolution between the system response and the cross-correlation between pump and probe pulses. As a consequence, the temporal resolution of this technique is only limited by the duration of the pulses.

4. Experimental setup

4.1. Sample description and fabrication

The fabricated AlGaAs nanowires MS (see fig.1) consists of two replicas of a 4 x 7 matrix of 70 μm x 70 μm $Al_{0.2}Ga_{0.8}As$ nanowires supported by a composite AlOx-GaAs substrate, each im-

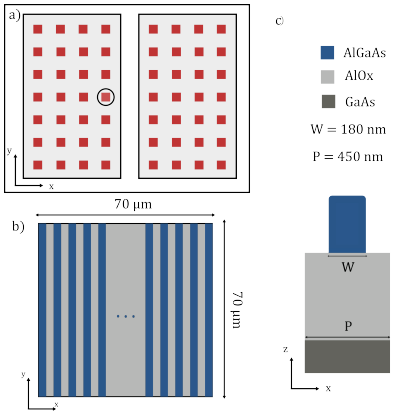

Figure 1: (a) sketch of the sample. (b) top view of one MS. (c) vertical cross-section.

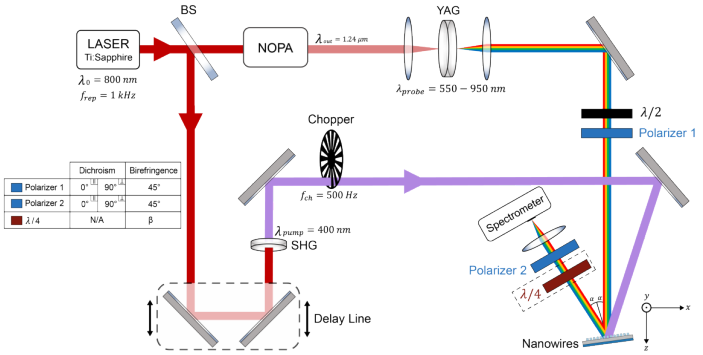

Figure 2: Schematic of the experimental setup. The box on the left specifies waveplate and polarizer configurations for both experiments.

plementing different values of periodicity P and thickness W. The MS investigated, circled in figure1a, corresponds to the geometrical parameters $P = 450$ nm and $W = 180$ nm. The fabrication procedure relies on the epitaxial growth of AlGaAs, through a combination of electron-beam lithography and reactive-ion-etching, on an AlAs substrate which is then oxidized into Al-Ox.

4.2. Dynamical polarization reconstruction

Here we discuss the working principle of a detection line used in one of the experiments for polarization reconstruction, comprising a quarter-wave plate rotated at a certain angle β followed by a polarizer. It can be shown that the intensity of light transmitted by this line can be expressed as a linear combination of the Stokes parameters, where the coefficients only depend on the angle β [3]. This way, by recording the light intensity for 4 different β values, we can write an algebraic system whose solution provides the Stokes parameters. This, combined with a pump-probe setup, allows the reconstruction, at each time and wavelength, of the transient variation in the polarization state of light upon reflection from the MS.

4.3. Experimental setup

The experimental setup (fig. 2) is based on a Ti:Sapphire laser providing 100 fs pulses at 800 nm with 1 kHz repetition frequency. The laser is used to pump a Beta-Barium Borate (BBO) crystal to produce second harmonic pulses centered at 400 nm used as control pulse in the experiments. The probe pulse is produced via supercontinuum generation by pumping a YAG crystal with 1240 nm pulses, obtained by a non-collinear optical parametric amplifier (NOPA). To record the signal $\Delta R/R$ as a function of the delay between pulses, an optical delay line is inserted along the pump path and the pump beam is modulated at 500 Hz with a mechanical chopper. This setup, combined with waveplates and polarizers, allows to perform two types of polarization-resolved pump-probe measurements. In particular, for the dichroism experiment the input polarization is controlled by inserting a half-wave plate followed by a linear polarizer oriented along either the x or y-axis. An additional polarizer, having the same angle as the first one, is placed after the sample. On the other hand, in the birefringent experiment, polarizer 1 and polarizer 2 are both set at an angle of 45°, and an additional quarter-wave plate rotated at an angle β is inserted between the sample and polarizer 2 for polarization reconstruction. In both experiments, the signal is recorded via a spectrometer coupled to a linear photodetector array.

5. Experimental results

5.1. Ultrafast transient linear dichroism

The results of the dichroic experiment are reported in the upper panel of figure 3., showing polarization-resolved pump-probe maps of the relative differential reflection of the sample for TM and TE modes. We can notice huge modulations of the dichroic response for TM-polarized light, up to 470% at 2 ps in a narrow

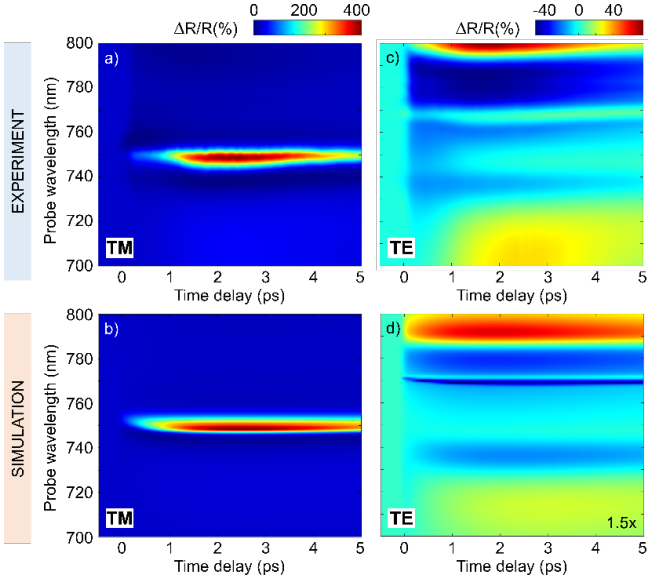

Figure 3: (a-b) Experimental and simulated temporal cross-sections, taken at wavelengths specified in the legend, for both TE and TM modes.

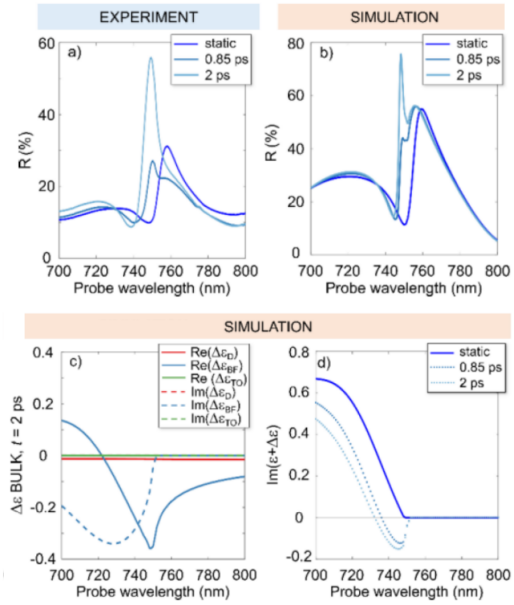

Figure 4: (a) TM reflection measured in static conditions, at 0.85 ps and at 2ps after pump arrival. (b) Simulated values for the same three cases. (c) Contributions to the real (solid lines) and imaginary (dashed lines) parts of the permittivity variation for Drude (red), band filling (blue) and thermo-optics effects (green). (d) Imaginary part of the total permittivity before and after pump arrival.

spectral range centered at 750 nm. In contrast, the TE reflectivity is modulated only up to a maximum of 70% and has a relatively low value (10%) at 750 nm. Our results are in excellent agreement with the simulations of the optical response reported in figure 3 (b-d). These were performed following a semiclassical modeling approach which takes into account a series of ultrafast processes taking place after excitation (photoinduced carrier generation, diffusion followed by nonradiative recombination, and consequent lattice heating) each of which contributes to a variation of the sample permittivity. Once the permittivity is computed, the optical response is retrieved via full-wave electromagnetic simulations. This model can be also used to investigate the origin of this particular TM response. Specifically, a comparison between the different contributions to the permittivity variation (fig. 4c) shows that, in the spectral region of interest, the dominant mechanism is represented by the so-called band-filling effect arising from the saturation of absorption channels. This corresponds to a negative variation of the real permittivity of the sample, coherent with the blueshift of the static TM resonance observed in the experiments and simulations (fig. 4 (a-b)). Interestingly, plots of the total imaginary permittivity at different time delays (fig. 4d) reveal the opening of a gain window (738-750 nm) within the first picosecond which gets larger at 2ps. The efficient increase in reflection can then be understood as an interplay between stimulated emission, triggered by the probe and caused by a pump-induced population inversion, and the shift in the TM resonance towards this spectral region.

5.2. Ultrafast transient birefringence

In the birefringence experiment, we recorded $\Delta R/R$ maps for 15 different angles of the quarter-wave plate, which were then used for polarization reconstruction as explained above. In particular, the results reported here were obtained as an average of reconstructed values from 3 different sets of 4 angles. Figure 5 shows both experimental and simulated values in terms of the relative phase φ as a function of the probe wavelength. In figure 5 (a-b), static (φ) and perturbed response at 2 ps (φ') are compared, while figure 5 (c-d) displays the transient variation $\Delta\varphi = \varphi' - \varphi$. At 749 nm, 2 ps after pho-

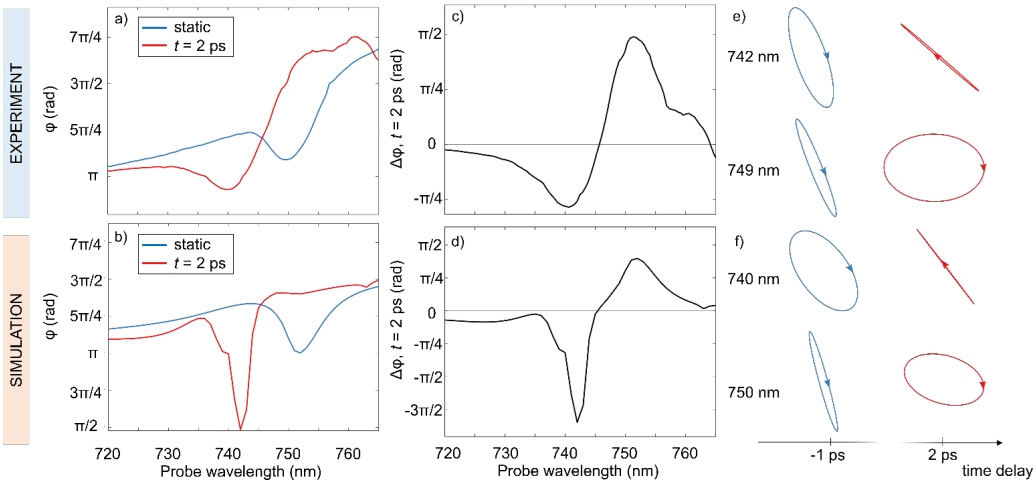

Figure 5: (a-b) Comparison between phases in static (φ) and perturbed (φ') conditions at 2 ps, for both experimental and simulated results. (c-d) Experimental and simulated transient relative phase shift between components at 2 ps. (e-f) Reconstructed polarization ellipse in static (-1 ps) and perturbed (2ps) conditions from experimental and simulated values.

toexcitation, the sample generates an additional $\pi/2$ relative phase shift, acting like a transient quarter-wave plate. In other words, if the static response is linearly polarized, perturbation results in ultrafast linear-to-circular polarization conversion, as can be seen from figure 5 (e-f). Note that also in this case the relevant features of the response are located in the spectral region where the band-filling dominates, suggesting that this is the mechanism responsible for the transient birefringent features of the sample.

6. Conclusions

We have presented an active AlGaAs nanowire MS working in reflection, together with a customized pump-probe setup for ultrafast polarization analysis. This device is capable of huge modulations of dichroism, up to 470% for TM polarized light, and waveplate-like functionalities with relative phase modulations up to $\pi/2$. The origin of this response is explained as the interplay between the shift of the static extended state resonance near the bandgap of the semiconductor and the photoinduced band-filling effect.

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