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D32.2
Report on ultrafast spectroscopy, and strong field physics applications in the mid-IR

Lead Beneficiary: 13 (LENS)

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Deliverable Nature	
R = Report, P = Prototype, D = Demonstrator, O = Other	R
Dissemination Level	
PU = Public PP = Restricted to other programme participants (incl. the Commission Services) RE = Restricted to a group specified by the consortium (incl. the Commission Services) CO = Confidential, only for members of the consortium (incl. the Commission Services)	PU

A. Abstract / Executive Summary

Novel mid-IR sources have been developed and applied to the analysis of different aspects of laser/matter interactions, from the dynamics of laser/cluster interactions, to linear and nonlinear spectroscopy with high-order harmonics generated from mid-IR pump sources, to the mid-IR spectroscopy of molecules with high precision and sensitivity.

B. Deliverable Report

1 Introduction

The development of novel sources in the mid-IR part of the spectrum is opening new perspectives in the study of laser/matter interactions. Ultrafast and ultraintense mid-IR sources can either be used directly for investigating matter, like in the generation of coherent electron wavepackets and in the explosion of molecular clusters, or, more indirectly, as a driver for the production of high-order harmonics. In this case, either the harmonic generation process itself is used to reveal the spectroscopic properties of the medium, or the produced XUV radiation becomes the source for spectroscopy.

Another indirect way of using ultrafast and accurately phase-stabilized laser sources is to extend the concept of frequency-comb measurements to the mid-IR spectral region. In this case, mid-IR combs can be used as ultra-precise frequency references for locking more powerful narrowband lasers. In combination with the development of advanced detection techniques, this allows one to achieve ultra-high sensitivity in absolute frequency spectroscopic measurements.

2 Objectives

Demonstration of novel spectroscopic tools and strong-physics applications based on ultrafast and/or ultraintense mid-IR sources.

3 Work performed / results / description

POLIMI explored the possibility to steer, control or switch electron wave packets with light [1]. Sharp metallic nanotapers irradiated with few-cycle laser pulses are a source of highly confined coherent electron wave packets with attosecond duration and strong directivity. Such electron pulses were generated by strong-field-induced tunnelling and acceleration of electrons in the near-field of a sharp gold tip, illuminated by an infrared laser field. The few-cycle infrared pulses with stable and controllable CEP were generated by difference frequency generation between light from two NOPA stages, delivering broadband pulses in the 540-650 nm range, and 870-890 nm, respectively. The difference frequency resulted in passively CEP-stabilized pulses as short as 14 fs, tunable between 1.3 and 1.9 microns. We showed the effect of the carrier-envelope phase of the laser field on the generation and motion of electrons emitted from the tips. We observed clear variations in the width of plateau-like photoelectron spectra characteristic of the subcycle regime.

POLIMI also extended recent studies involving the interaction of near-IR pulses with atomic clusters to the interaction of intense mid-IR pulses with molecular clusters [2]. Clusters excited by intense laser pulses are a unique source of warm dense matter that has been the subject of intensive experimental studies. **POLIMI** studied CO2 cluster explosion dynamics driven by a few-cycle 1.45-μm laser pulse and probed by a delayed 800-nm pulse; the dynamics was traced by recording XUV fluorescence lines emitted by different ionic species as a function of the pump-probe delay. Fluorescence is a powerful probe of the cluster environment, since it is sensitive to the electron temperature as well as to collisional excitation and ionization mechanisms at work. Its potential can be fully accomplished by time-resolved techniques, as in a pump-probe measurement, providing access to dynamical processes occurring in the cluster. The main finding of the investigation is that lower ionization stages emit earlier than higher ones along the pump-probe delay scan. This points to intriguing opportunities for probing phenomena occurring inside clusters excited by intense laser pulses. In spite of the relatively high peak intensity, the interaction of a mid-IR few cycle driving pulse with a CO2 cluster leads to the generation of a cold nanoplasma. This result shows that intense mid-IR pulses, which are usually considered as ideal drivers for accelerating electrons to very high ponderomotive energies, can be effectively exploited as ultrafast drivers of nanoscale warm matter with solid-state density and electron temperatures in the range of a few to a few tens of electron volts, allowing to reproduce warm and dense states of matter in a laser laboratory.

ICFO carried out several investigations with the uniquely developed few-cycle and CEP stable 160 kHz OPCPA. The wavelength scaling of strong field interaction of mid-IR pulses for high harmonic generation was investigated [3] together with the first measurement of strong field ionization momentum distributions in simple noble gas atoms and molecules in the deep tunneling regime [4]. Unexpected features were found which point at trapping at high lying states.

ICFO also successfully completed the development of a Ti:Sapphire pumped OPA source which delivers 1.8-cycle pulses with <90 mrad CEP stability over 72 hour, 600 microjoule pulse energy at 1850 nm central wavelength and 1 kHz repetition rate (see D32.1). This source exhibits a power stability of better than 1% and demonstrated HHG at the oxygen K shell edge at 530 eV. It allowed the first demonstration of near edge x-ray absorption fine structure (NEXAFS) measurement in condensed matter at the carbon K-shell edge at 294 eV with a HHG based source [5]. The resulting CEP stable soft X-ray radiation was also characterized and corresponds to the first isolated 355 as pulse at 300 eV [6].

SLIC performed nonlinear harmonic spectroscopy in molecules, driven by elliptically polarized mid-IR light produced with an OPA-based mid-IR source at 1.1-2 µm [7]. The polarization of harmonic light was measured by means of an optical polarizer. Variations of the harmonic polarization with ellipticity/intensity of the mid-IR field partially reveals multiple orbital dynamics in tunnel ionization in, e.g., SF6 molecule in the gas phase [8].

SLIC also studied the resonant photoionisation dynamics in atoms (He, Ar) using phase spectroscopy (RABBIT). By combining the OPA-based tunable mid-IR source and a high resolution electron spectrometer, SLIC showed that the spectral phase inside a Fano resonance could be directly accessed. This allows in turn the reconstruction of the time-dependent electron wavepacket released in the continuum at the resonance, with attosecond resolution.

LENS actively phase-locked a quantum cascade laser to a difference-frequency-generated (DFG) non-linear source referenced to a near-IR optical frequency comb, achieving a subkilohertz linewidth and a Cs-traceable mid-IR source delivering tens of mW. This novel source has been used for sub-Doppler spectroscopy of CO2 transitions at 4.3 um, performing absolute frequency measurements of several lines with few kHz uncertainties [9,10].

LENS developed a new spectroscopic technique, named intracavity quartz-enhanced photoacoustic spectroscopy (I-QEPAS), and employed it for sensitive trace-gas detection in the mid-IR [11]. It relies on a distributed-feedback QCL emitting at 4.33 µm and on a combination of photoacoustic spectroscopy with a build-up optical cavity. The possibility of realizing comb-referenced Mid-IR sources with high power and narrow linewidths, combined with novel spectroscopic techniques [12] for high sensitivity, allowed us to perform optical radiocarbon dioxide concentration measurements at unprecedented levels [13].

LENS also demonstrated a Mid-IR frequency comb with more than 400 GHz span, 1 GHz rep. rate and sub-kilohertz-linewidth teeth, based on intra-cavity DFG in a Ti:sapphire laser [14]. This multi-frequency highly coherent source was used for frequency referencing a DFB quantum cascade laser and to perform ambient air direct frequency comb spectroscopy with the Vernier technique, by de-multiplexing it with a high-finesse Fabry-Perot cavity [15].

4 Conclusions

The activities carried out in this deliverable D32.2 demonstrated the successful use of advanced mid-IR sources, mostly developed within the JRA framework, for spectroscopic analysis and strong field applications. The development of these advanced new sources and tools in the JRA context has strong potential to grant improved access opportunities to the involved partner laboratories.

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