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Carrier frequency tuning of few-cycle light pulses by a broadband attenuating mirror

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We demonstrate the performance of a novel multilayer dielectric reflective thin-film attenuator capable of reshaping the super-octave spectrum of near-single-cycle visible laser pulses without deteriorating the phase properties of the reflected light. These novel broadband attenuating mirrors reshape in a virtually dispersion-free manner the incident spectrum such that the carrier wavelength of the reflected pulses shifts from ∼700 nm ($E_\gamma = 1.77$ eV) to ∼540 nm ($E_\gamma = 2.25$ eV) or beyond while maintaining their initial near-single-cycle pulse duration. This constitutes a viable approach to convert a number of established few-cycle ultrafast laser systems into sources with a selectable excitation wavelength to meet the requirements of single-color/multicolor high temporal resolution spectroscopic experiments.

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1. INTRODUCTION

With the advent of few-cycle laser pulses, ultrafast pump-probe spectroscopy became a powerful tool that allows the real-time observation of electron dynamics in atomic, molecular, and condensed matter systems [1–4]. This prospect propelled the development of a variety of systems synthesizing laser pulses comprising less than two oscillation cycles of the electric field of light and corresponding pulse durations in the few-fs range.

To date, most of these systems are either Ti:sapphire based with subsequent spectral broadening [5–7], relying on optical parametric amplification [8–10] or fiber laser schemes [11]. While Ti:sapphire lasers emit at a carrier wavelength of ∼800 nm, the latter systems typically generate few-cycle pulses in the visible and infrared spectral region beyond 1 μm.

Such laser sources offer an abundance of pulse energy and spectral width, and are readily employed in ultrafast pump-probe schemes (e.g., in attosecond time-resolved experiments [4]); however, the choice of laser system predetermines the central photon energy/carrier wavelength of the excitation pulse in the experiment. As a result, pump-probe spectroscopy with subfemtosecond temporal resolution is typically carried out in the near-infrared and infrared wavelength region.

Most ultrashort pump-probe spectroscopy experiments in the visible and near infrared integrate observables over multiple oscillation periods of the pump laser field due to its rapidity. Attosecond spectroscopy, in contrast, resolves subcycle dynamics in a sample and proves that the time-dependent light-matter interaction is fundamentally depending on the oscillation frequency of the impinging electric field [12]. For a Fourier-limited (compressed) ultrashort laser pulse, possessing a constant instantaneous frequency, this oscillation period naturally defines the carrier frequency and vice versa. Furthermore, for many physical systems, especially those of chemical and biological interest and band-structure materials (e.g., for solar cell applications), the electronic response to visible and ultraviolet light is of central interest. Even though the discussed laser systems generate such spectral components, their interactions are easily obscured or the samples destroyed by the much stronger near-infrared components. Merging subfemtosecond temporal resolution with availability of the excitation wavelengths in the visible and ultraviolet spectral ranges would substantially expand the toolbox of ultrafast spectroscopy.

Here we present a viable approach to tune the excitation wavelength of an established near-infrared few-cycle source already used for the purposes of the attosecond spectroscopy from the traditional near-infrared spectral range to the visible range and beyond. The tuning is achieved via an implementation of a single piece of a tailored reflective optics.

2. THIN-FILM ATTENUATOR

To alter the carrier frequency of a compressed ultrashort pulse one needs to shift the "center of mass" of its spectrum. One possibility is to reshape its spectrum using wavelength selective optics. For this purpose we developed a thin-film multilayer dielectric broadband attenuating mirror (BAM) coating. While the coating is reflective in a broad spectral range spanning upon specification to more than an optical octave, its reflectance is not constant over the entire spectrum. The coating has a >90% reflectance for a relatively narrow part of the spectrum around the desired wavelength, while only ~10% of the rest of the spectral range is reflected. Therefore, the reflected spectrum is inhomogeneously attenuated. Meanwhile, the spectral phases of the incident and reflected pulses remain identical. This is achieved by careful control over the dispersion that is introduced by the attenuator itself. The combination of both allows for the desired reshaping of the pulse spectrum and shift of the carrier wavelength while the initial pulse duration is preserved. For a compressed pulse, the resultant carrier frequency is determined as the first moment of the product of the incident spectrum and the reflectance profile of the attenuator.

Despite the BAM appearing to be similar to well-known dichroic beam splitters (DBSs) and edge filters, its functionality is substantially different. First, unlike in the case of DBSs, the spectrum outside the desired range is not intended to be fully transmitted; it is in fact partially reflected, though with a high attenuation factor. Second, special care is taken of the dispersion properties of the component. While exhibiting negligible group-delay dispersion (GDD) within the reflectance band, DBSs introduce strong GDD ripples at the edges of the band, thus severely impairing the spectral phase and, consequently, the temporal profile of the reflected pulse [13]. The multilayer stack of the BAM, on the contrary, is optimized to maintain smooth, nearly zero GDD over all of the reflected and transmitted spectrum, not only inside but also outside the high reflectance band. Therefore, the reflected spectrum maintains a spectral phase nearly identical to the phase of the incident spectrum, while the transmitted spectrum will only accumulate the dispersion introduced by the substrate of the BAM. By proper choice of the substrate, significant lengthening of the transmitted pulse can be avoided enabling an arrangement producing a spectrally separated few-cycle pulse pair.

For demonstration we have developed a BAM that allows the selection of a central wavelength in the visible range; its reflectance and GDD are presented in Fig. 1.

The basis of the multilayer design is an assembly of several alternating quarter-wave-thick layers corresponding to the preferred wavelength. The layer thicknesses are then numerically tuned to maintain control over the ripples in the GDD. For illustration, the layer sequence of an attenuator selecting 550 nm is presented in the inset in Fig. 1.

The coatings are produced from suitable dielectric materials, defined by the specified working spectral range. To select ultraviolet light, a combination of HfO_2/SiO_2 is most appropriate. For visible light, Ta_2O_5/SiO_2 and Nb_2O_5/SiO_2 are material pairs of choice. In our case the coatings were deposited by magnetron sputtering ensuring high precision of the deposited layers, needed to fulfill the requirement for controlled dispersion, and providing coatings with sufficiently high damage

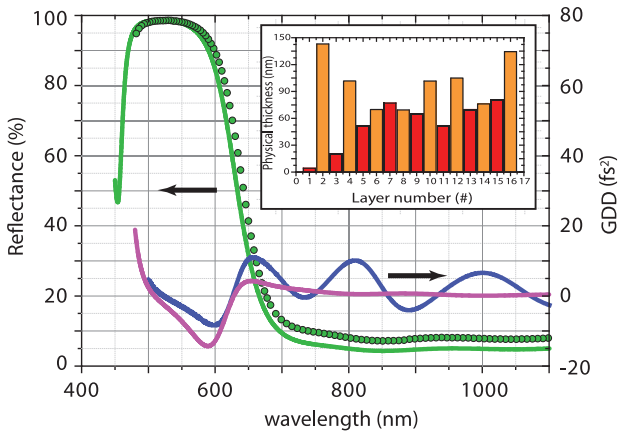

Fig. 1. Broadband attenuator mirror (BAM). Solid lines: designed reflectance at 8° angle of incidence (green) and GDD on reflection (blue) and transmission (magenta). Dotted green line: measured reflectance, 8° angle of incidence. Inset: layer sequence of the BAM. Red bands: high-; orange: low-index layers.

thresholds. The reflectance of the produced BAM, Fig. 1, was measured in a spectrophotometer (Perkin Elmer, Lambda-950) at 8° angle of incidence. Dispersive properties were evaluated in the time domain using the attosecond streak camera technique.

3. EXPERIMENT AND RESULTS

As the course of light-matter interaction on the subcycle time scales is fundamentally depending on the oscillation frequency of the impinging electric field [12], this oscillation frequency is a central quantity in attosecond spectroscopy.

To unambiguously determine whether the implementation of a BAM permits the tuning of the carrier wavelength, we have to resort to a measurement technique that provides direct access to the temporal evolution of the electric field under the pulse envelope. Recording of a time-integrated spectrum of the reflected pulses will allow only the identification of the spectral centroid, not providing direct access to the oscillation period around the pulse maximum. Well-established nonlinear techniques for the measurement of femtosecond pulses such as FROG [14], SPIDER [15], or the rather recent D-scan [16] require reconstruction algorithms and do not directly determine the time-dependent light field oscillation. Moreover, the techniques suffer from a range of limitations when dealing with near single-cycle pulses [17–19], and extreme care needs to be taken to correctly perform and interpret the measurement.

The attosecond streak camera [20,21] is proven to be suitable for the measurement of few-cycle, one-cycle, and even subcycle pulses [22] without phase-matching constraints of the abovementioned techniques. Moreover, it offers direct access to the electric field evolution and therefore the carrier frequency. Hence, we used it for the temporal characterization of both the incident, nonmodified and the spectrally shifted pulses (see Fig. 2).

The laser source is built on the basis of a commercial Ti:sapphire amplifier, which is delivering ~25 fs, 1 mJ pulses at 4 kHz repetition rate at a central wavelength of ~800 nm (femtolasers). The amplified pulses are then spectrally broadened in a noble gas filled hollow-core fiber and postcompressed

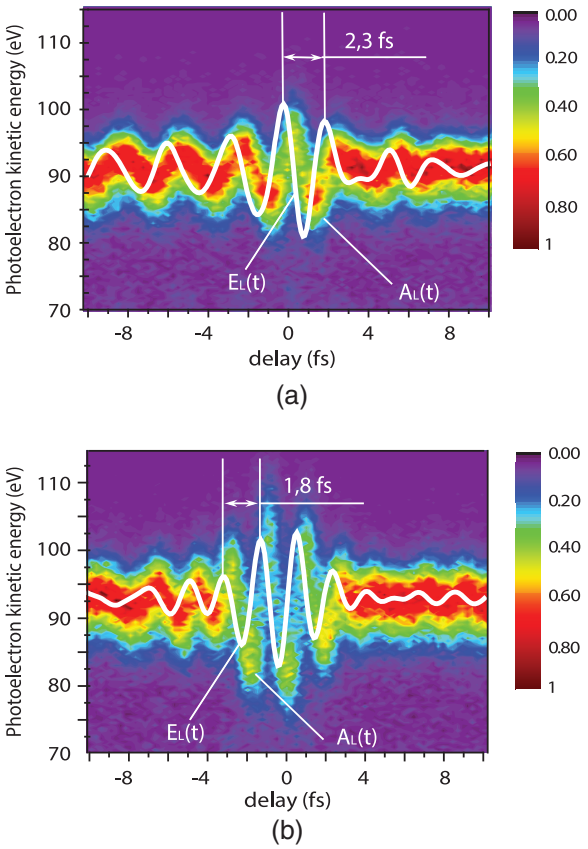

Fig. 2. Attosecond streak camera spectrograms. (a) Incident nonattenuated pulse; (b) reshaped pulse. The solid curve $E_L(t)$ (white solid line) is the temporal evolution of the instantaneous laser electric field extracted from the vector potential $A_L(t)$ (color map) recorded by streaking spectroscopy. The amplitude setting was chosen arbitrarily in the experiments and does not reflect the pulse energy attenuation due to the BAM.

with a set of chirped mirrors to sub-4-fs duration. The spectrum covers the superoctave range, spanning from 450 nm to 1000 nm [7].

Recorded spectrograms before and after reflection from the BAM together with the reconstructed electric fields are presented in Fig. 2. From the laser electric field (white solid line) it is seen that the pulse reflected from the BAM, Fig. 2(b), has an oscillation period of ~1.8 fs—substantially shorter than the oscillation period of ~2.3 fs of the nonattenuated pulse, Fig. 2(a). The oscillation period of 1.8 fs corresponds to a carrier wavelength of ~540 (2.25 eV) nm, while the 2.3 fs period translates to ~700 nm (1.77 eV).

The reconstructed intensity envelopes of the incident and reflected pulses, Fig. 3(a), show that the pulse duration before and after the BAM stays virtually unchanged (~3 fs). Reconstructed group delays of the incident and reflected pulses, Fig. 3(c), demonstrate preservation of the spectral phase across the whole working range of the BAM, confirming its dispersion free operation. Comparison of the recorded spectra of the incident nonmodified (blue) and reflected (red), Fig. 3(b), pulses illustrates the spectral reshaping performed by the BAM. The recorded time-integrated spectra are well reproduced by those

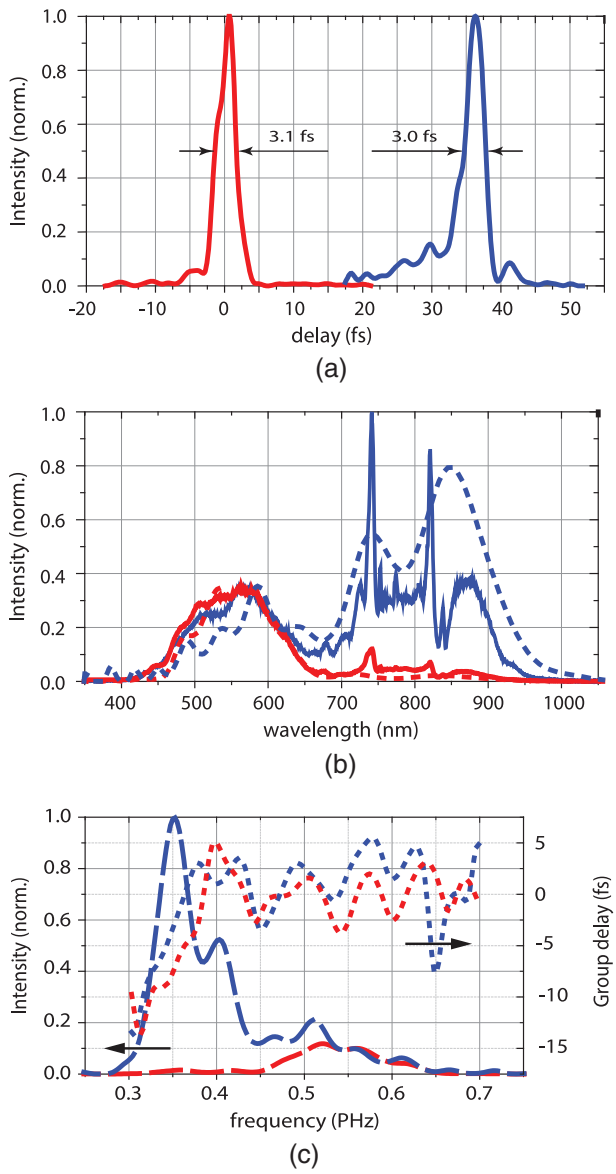

Fig. 3. Temporal and spectral characterization. (a) Intensity envelopes of incident, nonmodified (blue) and reflected by the BAM (red) pulses. The temporal delay between the pulses is artificial and introduced for illustration purposes. (b) Measured (blue) and reconstructed (blue-dashed) spectra of the incident, nonmodified pulse compared to measured (red) and reconstructed (red-dashed) spectra of the spectrally tuned pulse. (c) Reconstructed spectra of the incident, nonmodified (blue-dashed) and reflected, spectrally tuned (red-dashed) pulses together with reconstructed group delays for the incident, nonmodified (blue-dotted) and reflected, spectrally tuned (red-dotted) pulses in the frequency domain.

reconstructed from the attosecond streak camera spectrograms (red- and blue-dashed), confirming the presence of all spectral components in the streaking window.

As a BAM itself introduces only a negligible phase, the quality of the spectrally shifted pulse is closely related to the properties of the incident pulse. The temporal profile of a compressed pulse is preserved besides spectral narrowing at full width half-maximum. However, the substructure in the incident pulse will

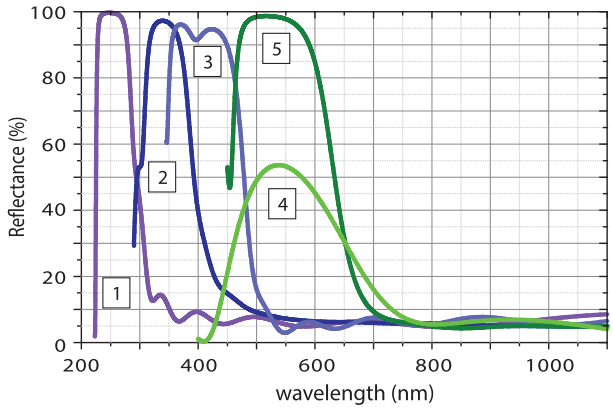

Fig. 4. Family of BAMs designed for (1) 260 nm ± 5 nm, (2) 350 nm ± 5 nm, (3) 400 nm ± 20 nm, (4) and (5) 530 nm ± 20 nm carrier wavelengths. Solid lines: designed reflectance.

be mitigated or magnified depending on the fraction of the spectral components it is rooted in to the final pulse.

Implementation of the attenuator naturally decreases the pulse energy of the reshaped pulse as the spectrally integrated reflection of the BAM is rather modest, in our particular case 27.5% for a balanced spectrum. Nevertheless, the residual pulse energy is still abundant for the purposes of attosecond spectroscopy.

Based on our findings, it is possible to design a family of BAMs, thus enabling tunability over a broad selection of carrier wavelengths. Curves 1, 2, and 3 in Fig. 4 demonstrate the capability to select carrier wavelengths in the ultraviolet for ultrabroadband sources that have components in the relevant spectral range, while curve 4 additionally performs controlled attenuation, e.g., to avoid sample damage. Curve 5 displays the design discussed above. Implementation of several properly designed BAMs (on thin substrates) in a row enables extension to multicolor ultrafast sources.

The presented approach is intrinsically power-/energy-scalable limited only by the damage threshold of the BAM itself. Therefore, it is possible to extend its application well beyond hollow-core fiber based systems to future high-power broadband sources.

4. CONCLUSIONS AND OUTLOOK

In conclusion, we have demonstrated a broadband wavelength-selective thin-film attenuating mirror, BAM. By applying linear spectral filtering, the BAM effectively tunes the carrier wavelength of few-fs sub-mJ light pulses from ~700 nm (1.77 eV) to ~540 nm (2.25 eV) while preserving their initial pulse duration.

Overall, the suggested arrangement is a viable approach to immediately convert a number of well-established broadband few-cycle near-infrared and infrared driving lasers commonly used in the attosecond spectroscopy into few-cycle sources with selectable excitation wavelengths by integration of only one additional cost-efficient component. Moving the carrier wavelength of the excitation pulses away from the well-explored near-infrared and infrared ranges into the visible and ultraviolet allows us to unambiguously study a broad class of materials and to significantly simplify the interpretation of experimental results. In addition, the possibility to select the excitation wavelength empowers studying the wavelength dependence of crucial physical observables in the subcycle regime, thus unfolding another dimension of attosecond spectroscopy.

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