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An inexpensive nonlinear medium for intense ultrabroadband pulse characterization

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Abstract The ability of pellets made up of compressed iron iodate nanocrystals to frequency-double the whole visible spectrum is demonstrated. We suggest their use for complete characterization of intense ultrabroadband laser pulses.

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1 Introduction

The interest in few- or single-cycle pulses (FCP) has increased in the last years particularly because of their implementation for attosecond pulse generation [1, 2]. FCP pulses, which can be prepared for example by spectral broadening via (co-)filamentation in rare gases [3, 4], exhibit very broad (octave spanning) bandwidths, which makes their characterization highly challenging. Standard pulse diagnostic techniques [5, 6], in fact, are trimmed down by limited spectral acceptance, as they are based on nonlinear frequency mixing in bulk crystals [7]. Ideally, one should posses a nonlinear medium capable of producing an intense nonlinear signal, fulfilling the phase-matching condition over a very broad spectral domain, and inducing none or negligible temporal dispersion on the pulses to be measured. Recently, we successfully used individual $Fe(IO_3)_3$ nanocrystals as nonlinear medium for complete pulse characterization within a microscope focus [8]. In that case, we showed that, as the average crystal size is much smaller than the coherence length, no phase-matching constraints apply to the frequency-mixing process, allowing for complete spectral doubling of broadband femtosecond pulses. Due to the large nonlinear coefficients of $Fe(IO_3)_3$ and the cost-effective synthesis of the nanocrystals [9], it seemed tempting to employ them also for the routine characterization of intense ultrashort and ultrabroad laser pulses. In the following, we demonstrate that pellets of compressed $Fe(IO_3)_3$ nanocrystals can efficiently frequency-convert broadband infrared and ultraviolet femtosecond laser pulses through various nonlinear processes, enabling their spectral and temporal characterization.

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2 Experimental

2.1 Sample preparation

Two aqueous solutions of iron nitrate and iodic acid salts with molar concentrations are mixed at room temperature under vigorous stirring. Nanosized powders are obtained after filtering and drying the yellow-green precipitate. The grain size, estimated by the broadening of X-ray diffraction peaks, is around 30–40 nm which is consistent with High Resolution Scanning Electron Microscope (SEM) images (Fig. 1). Pellets of compressed nanocrystals (3 mm thickness and 8 mm diameter) were obtained by exerting a 250 MPa pressure on dried powder for 15 minutes. The surface roughness is found to be in the micrometer range.

2.2 Optical set-up

The direct output of a Ti:Sapphire amplified laser system (from hereon labeled as **R** pulses: central wavelength $\lambda_{\mathbf{R}} =$

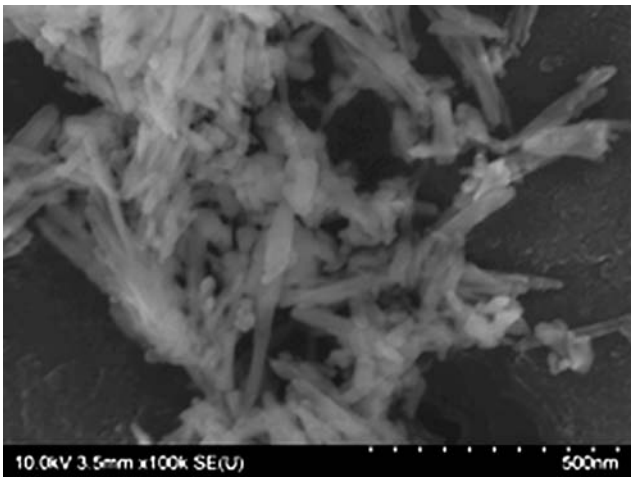

Fig. 1 High resolution SEM image of iron iodate nanosized powders

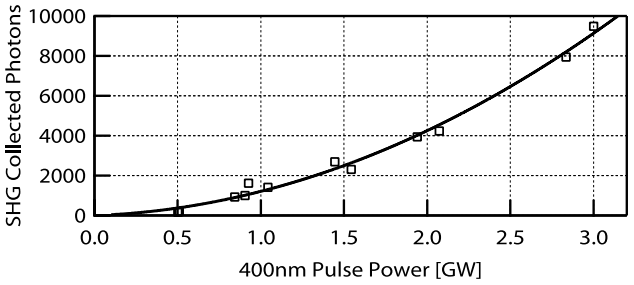

Fig. 2 Power dependance of the 200 nm signal as a function of the 400 nm input pulse power (experimental data points (*squares*) and its quadratic fit (*line*)). The collection solid angle corresponds to 5×10^{-2} srd

792 nm, bandwidth 30 nm, energy 2.5 mJ) are frequency-doubled in a 150 μm thick BBO crystal, providing a typical output energy of 150–300 μJ. In a first experiment, the second harmonic pulses (**B**) are directly focused onto a nanoparticle pellet by an aluminium-coated spherical mirror ($f = 50$ cm). The scattered nonlinear UV signal (**UV**) (see the power dependence in Fig. 2) is collected by a fused-silica lens-doublet and coupled into a monochromator with 0.4 nm spectral resolution. The spectrally-resolved signal is finally detected by a solar blind photomultiplier (Hamamatsu, R166UH). Alternatively, the **B** pulses are temporally characterized using a collinear interferometric setup: the pulse train is separated by a 2 μm thick 50/50 pellicle beamsplitter and the relative delay between the two arms adjusted with a motorized delay stage (temporal resolution ∼0.1 fs). In a third experiment, the **B** pulses are slightly focused by a $f = 1$ m lens and injected collinearly with **R** pulses in an Ar-filled cell, via co-filamentation the spectral bandwidth broadens up from the initial 6.3 to 30 nm FWHM. The **B** pulses are finally focused on the pellet for characterization using the generated **UV**-200 nm signal. In a last experiment, both **R** and **B** are focused with adjustable

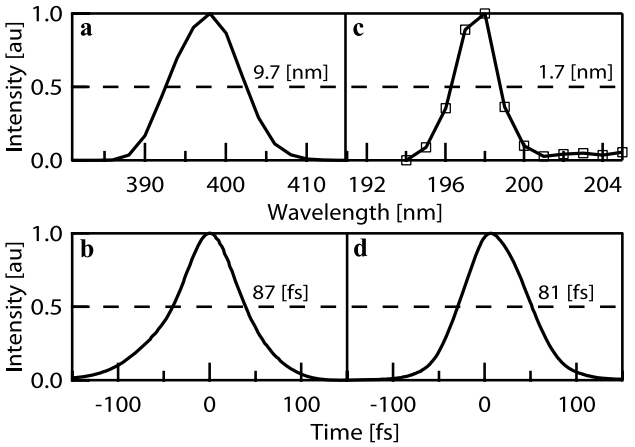

Fig. 3 (**a**) SHG spectrum of a **R** pulse performed on a single nanocrystal and (**b**) its autocorrelation [8]. (**c**) SHG spectrum of the **B** pulse generated on the pellet and (**d**) its autocorrelation

delay and polarization onto the nonlinear sample to measure their sum-frequency signal at 266 nm. All the measurements around 200 nm are performed in an argon saturated environment to prevent oxygen absorption below 190 nm.

3 Results

Since $Fe(IO_3)_3$ does not exist in bulk form, in a previous work we characterized its nonlinear optical response by studying the properties of single nanocrystals [10], determining a value of the order of several pm/V for its $\chi^{(2)}_{ijk}$ tensor elements. Thanks to their small size ($l = 20$–150 nm) and large second order susceptibility, we succeeded in frequency-doubling 800 nm radiation on individual nanocrystals (see Fig. 3(a)). As reported in Fig. 3(b), the nonlinear response of the nanoparticle was used to acquire a pulse autocorrelation trace at the focal plane of a high numerical aperture objective [8]. Due to the geometry imposed by the microscopic setup, the interference fringes typical of collinear measurements were removed by an ad hoc cut-off filter in the Fourier space [11].

Since $Fe(IO_3)_3$ nanocrystals are smaller than the coherence length, the material can simply be considered as an ensemble of nonlinear radiating dipoles, providing complete doubling of an incident spectrum centered at any wavelength independently from any phase-matching consideration. This property is exemplified by the plot in Fig. 3(b), showing the result of the spectral doubling of an input 6.3 nm broad **B** pulse. As reported in Fig. 3(d), the corresponding autocorrelation trace indicates a **B** pulse duration of 81 fs.

The above results demonstrate the ability of $Fe(IO_3)_3$ to generate an easily detectable scattered SHG signal from a UV–Vis pulse focused onto the pellet. As seen in Fig. 2, even

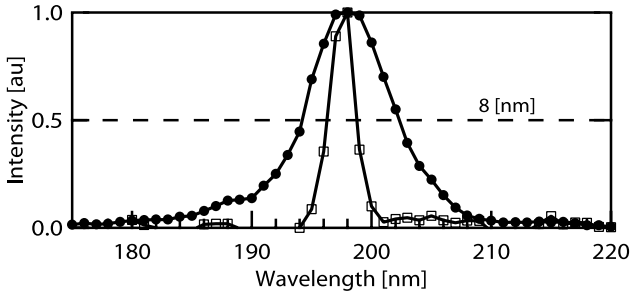

Fig. 4 UV spectrum (at 200 nm) of a **B** broad source obtained by co-filamentation (*dots*). Original spectrum corresponding traces for a 400 nm pulse without broadening (*squares*) (data reported from (Fig. 3(c)))

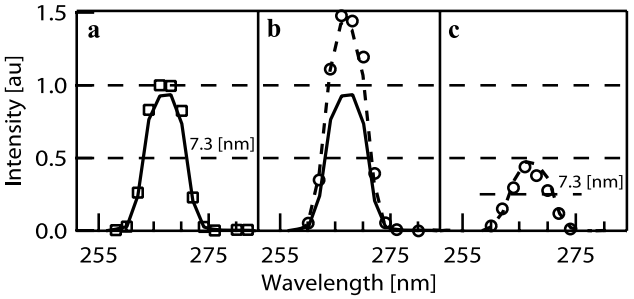

Fig. 5 *Solid line*: signal generated only by the IR beam. (**a**) Signal (*squares*) for UV and IR laser pulses temporally separated by 750 fs, (**b**) simultaneous and parallel (*dotted line*) (respectively perpendicular (*circles*)) **B** and **R** laser pulses. (**c**) Signature of the SFG signal

though the detection efficiencies was limited by the low numerical aperture of the collection lens, a clear background-free quadratic dependence is observed on the scattered 200 nm signal. Within the scope of FCP characterization, we show now that the bandwidth increase does not limit this capability, by repeating the measurement using as input filamentation-broadened **B** pulses with spectral bandwidth spanning from 350 to 440 nm. As illustrated in Fig. 4, their doubled spectrum extends from 175 to 220 nm fully matching the spectral range of the input pulse. The measured width is 8 nm FWHM, which could theoretically be recompressed down to 7 fs pulse duration.

In order to accurately perform a FCP autocorrelation, the nonlinear material has to efficiently support a nonlinear polarization $\mathcal{P} \propto E^2(t)$, where $E(t)$ is the *real* electric field. In the Fourier reciprocal space, this nonlinear source term corresponds to all the frequency-mixing pairs within the spectrum. We show that $Fe(IO_3)_3$ pellets support sum-frequency generation (SFG). In particular, when launching simultaneously **R** and **B** pulses onto the nanoparticles agglomerate, a delay-dependent signal centered at 264 nm was recorded both for orthogonal and parallel polarization of the two input pulses (Fig. 5(b)). In Fig. 5 we summarize the results of these measurements: in (a) one can observe that, also in the absence of the **B** pulse on the sample, a weak 264 nm signal is present, which is a signature of **R** pulse frequency tripling. By subtracting from the zero-delayed curve (Fig. 5(b)) this third harmonic component, we obtain the signature of SFG spectrum at 264 nm (about 7.3 nm FWHM) (Fig. 5(c)).

4 Conclusion

In conclusion, we have successfully demonstrated that $Fe(IO_3)_3$ pellets can support both ultrabroadband SHG and broadband SFG. Because the nonlinear material is used in a reflective configuration, no dispersion occurs, allowing its use for FCP measurement. Moreover, since the phase-matching condition does not apply because of the nanometric dimensions of the doubling particles, this approach opens the way to the utilization of this material with ultrabroadband spectra in its whole transparency region spanning up to 13 μm. Unlike nonlinear crystals which need a careful polishing and are quite expensive, this material is inexpensive enough to be used in adverse conditions such as within a pulse core where the intensity can exceed the damage threshold.

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