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Picosecond All-Optical Wavelength Conversion using Hot Carrier Intraband Absorption in Colloidal PbS Nanocrystals

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Owing to their widely tuneable optical properties and strong light-matter interaction, colloidal quantum dots (QDs) are considered for next-generation photonic devices such as all-optical wavelength conversion. Using QDs for this application is limited by either slow interband dynamics (106-109 s-1) or energy consuming multi-exciton dynamics (109-1011 s-1). Here we show, using white light pump-probe spectroscopy, that the interplay between two intrinsic material properties of PbS QDs, intraband absorption and interband bleach can lead to a very strong modulation of near-infrared light on an ultrafast, picosecond, timescale. After a few picoseconds upon non-resonant photo-excitation with a femtosecond pulse, hot carriers have thermalized to the band edges and a strong reduction of the QD absorption A ($\Delta A<0$) is observed around the band gap due to well-known occupation of the band-edge states by electrons and holes. Adversely, at energies below the band gap, a photo-induced absorption ($\Delta A>0$) is observed showing the same decay dynamics as the band gap bleach, attributed before to intraband transitions involving the cooled electrons and holes. As both bleach and intraband show a distinct spectral dependence and scale with the exciton density, a wavelength range exists where they both cancel out independent of pump fluence, leaving the excited dots with a similar cross section as the unexcited dots, i.e. $\Delta A = 0$. In the first picoseconds after photo-excitation however, no bleach is observed (as the carriers are not occupying the band edge) and the strong photo-induced absorption of the hot exciton cooling down is dominating the transient response ΔA, leading to a net positive ΔA at the matching wavelength. As such, a peculiar dynamic arises at the matching wavelength: a strong burst of absorption induced by the pump lasts for roughly 1 picosecond and vanishes, leaving the dots as they were before the pump pulse arrived. This provides an excellent platform to convert wavelengths without residual absorption, high conversion efficiency and picosecond speed. To characterize the conversion, the normalized absorption change ($\Delta A/A0$) is chosen as a figure-of-merit (FOM): reaching up to 23 for a single exciton population, both in colloidal solution and film, we deduce the strength of the absorption burst as 5200 cm-1. To shown the potential for high speed conversion, a pump pulse sequence of up to 4 femtosecond pulses ,separated by 2.2 and/or 4.4 ps, is converted to a probe wavelength while preserving the intrinsic strength, speed and zero background of the single pulse case, showing the ability for handling 450 and 225 Gb/s datastreams. Combining the QDs with on-chip or fiber based devices is shown viable with conversion energies as low as a few femtojoule per bit. Moreover, due to the strong light-matter interaction of direct gap colloidal semiconductor QDs, the integrated device footprints of a few hundred micron are much smaller than existing approaches.

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----- PHOTOS ARE HERE -----

All technical sessions and non-technical events were held at Lille Grand Palais.

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PLENARY SESSION

Main session (Wednesday May 13, 16:00 - 19:00)

MATERIALS FOR ENERGY AND ENVIRONMENT

- **A** Materials, mechanism and devices in nano energy
  INCLUDING ONE DAY on Carbon dioxide recovery and circular economy of carbon
- **B** Materials for applications in water treatment and water splitting
- **C** Advanced inorganic materials and structures for photovoltaics
- **D** Earth abundant and emerging solar energy conversion materials
- **E** Materials design and processing concepts for efficient and stable organic, hybrid, perovskite and dye solar cells
- **F** Scientific basis of the nuclear fuel cycle – III