Mixing Waves with Free Electron Lasers

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The invention of lasers in 1960 [1] was the beginning of the *lasers* revolution that kept on continuing till today. This revolution made accessible amazing tools for researchers as testified by the number of Nobel prizes awarded starting from 1964 to A. M. Prokhorov, C. H. Townes and N. G. Basov for the creation of the maser and laser. Since then novel spectroscopies were developed ranging from holography [D. Gabor was Nobel Laureate in 1971] to non-linear optics [N. Bloembergen and A. L. Schawlow were awarded with the Nobel Prize in 1981], until more lately femtosecond spectroscopy was proposed by H. Zweil [Nobel Prize in 1999] to *take pictures of* chemical reactions as they occur.
The research on nonlinear optics started immediately after the construction of the first laser with the detection of the second harmonic generation process [2] and now includes investigations of any interactions among two or more fields with matter. Although the nonlinear response can, in principle, be appreciated despite the nature of the sample and the input radiation, its observation requires rather high field intensities and, most importantly, phase coherence of the fields. The availability of optical lasers satisfying these necessities has permitted the development of a large variety of non-linear methods providing central information on diverse dynamical processes in a range of systems and timescales ranging, e.g., from sub-picosecond reconstruction of wavefunctions in reacting molecules [3] to microsecond dynamics in complex bio-systems [4]. However, the relatively long photon wavelength of table-top lasers imposes constraints for selectively probing specific atomic species. The recent advent of Free Electron Laser sources makes the shorter wavelength range accessible.
In this paper, we will discuss how recentadvances in the performance of the FELs allowed non-linear experiments at sub-optical wavelengths. In particular Second Harmonic Generation (SHG) [5] and Transient Grating (TG) [6] experiments have finally demonstrated the high potential of VUV/soft X-ray wave mixing techniques. SHG is one of the second order non-linear responses of systems that are non-zero only for non-centrosymmetric materials as surfaces or interfaces. The advantages of using electronic resonance would imply a significantly higher surface specificity than existing soft X-ray methods[5].TG experiments at sub-optical wavelength are relevant for the study of nanoscale dynamics in disordered systems as well as in semiconductors. Exciting phonon modes with nanometer wavelength would allow shedding light on a plethora of scientific open problems ranging from the thermal anomalies in glasses to understanding nanoscale thermal transport. Indeed the study of thermal transport approaching the nanometer is extremely important and motivated by relevant technological needs such as thermal management of electronic devices[7]. Wave mixing in the soft X-ray can be used as well to investigate drug/target intermolecular vibrational dynamics, possessing the potential to understand the marked differences in biological activities ofenantiomer or easily follow the dynamic of metal complexes[8].

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