Extreme Nonlinear Optics (high harmonic generation and attosecond generation)

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(Dated: May 1, 2007)

This paper wants to cover the most important aspects of the generation of attosecond pulses and higher harmonics and introduce an interesting application. Phase-matching is critical to achieve a high output intensity, since efficiencies are currently only about 10^{-5} for high harmonic generation.

I. FOURIER SYNTHESIZING PULSES BY STIMULATED RAMAN SCATTERING

This method relies on the possibility of Fourier-Synthesis of discrete lines over a broad spectrum (similar to mode-locking). A Raman medium is characterized by its Raman frequency ω_0, which can be the difference between two rotational or vibrational lines. When we apply a laser excitation $\omega_L - \omega_s$ close to ω_0, we can obtain gain at the stokes $(\omega_L - n\omega_0)$ and anti-stokes $(\omega_L + n\omega_0)$ frequencies, the Raman effect gets resonance-enhanced. The distance between these lines determines the repetition frequency $\tau_{rep} = \frac{2\pi}{\omega_0}$ (as in ML) and therefore we wish to have smaller distances that help us isolate single pulses. For a $100as$ pulse, we would need a locked bandwidth of about $5 \times 10^3 THz$. The generated **equidistant sidebands** (fig. 1) are already phase-locked. In a medium like Hydrogen, about 40 rotational Raman-lines have been obtained, covering all the spectrum from $200nm$ upto $800nm$ (fig. 2) with $\nu_0 = 507cm^{-1}$, equivalent to $17.6THz$. The repetition rate of $\tau_{rep,vib} = 57fs$ is much more suitable for single pulses than the repetition rate for rotational lines $\tau_{rep,rot} = 8fs$. In this example the medium was pumped with approx. $1ps$ pulses. For the two different wavelengths Nd:YAG (SHG) and Ti:Sapphire ($805nm$) lasers were used. As the molecular oscillation modulates the index of refraction (optical Kerr effect), the bandwith gets bigger and a white light continuum is generated. That decreases the Raman gain but increases the spectrum of possible lines. Therefore this parameter has to be optimized. The technique is much more efficient than High Harmonic Generation, because a significant part of the energy is pumped in the Raman lines, whereas HHG only has an efficiency of 10^{-5}. Pulse width measurement with autocorrelation methods is possible with these field strenghts.

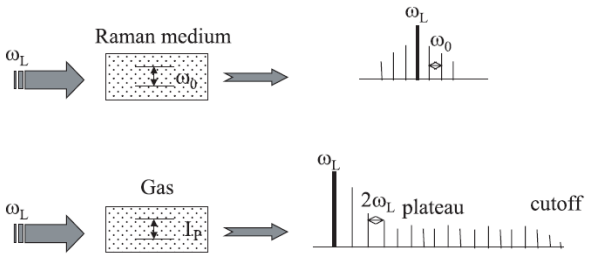

FIG. 1: principles of attosecond generation

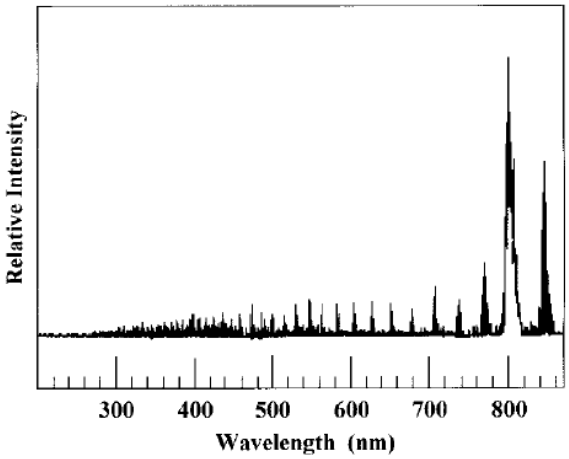

FIG. 2: rotational Raman lines of Hydrogen

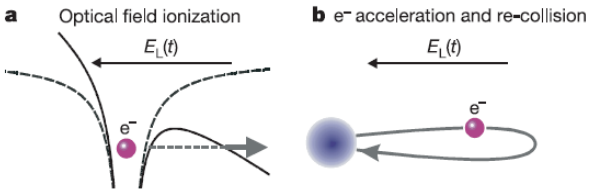

FIG. 3: ionization and re-collision

II. HIGH HARMONIC GENERATION (HHG)

As well-known from NLO, an isotropic material can only produce harmonics of uneven orders. It is the same with the gas atoms used when generating higher harmonics through ionization processes. The electrical field of the fs-pulse (typically few-cycle pulses) suppresses the effective Coulomb potential of the valence electrons, which can then tunnel through or escape above the potential barrier (fig. 3a) . As the probability for this ionization depends heavily on the field intensity the process is confined to within $\frac{T_0}{10}$ with pulsewidths of about $T_0 = 2.5fs$. This is why the Carrier-Envelope-Phase (phase between the visible carrier frequency and the fs-period envelope) gets especially important. The freed electrons get accelerated by the light field and can recollide with the atoms (fig. 3b). This leads to several effects like secondary electron-emission, excitation of electrons and emission of soft x-rays. As the electrons will

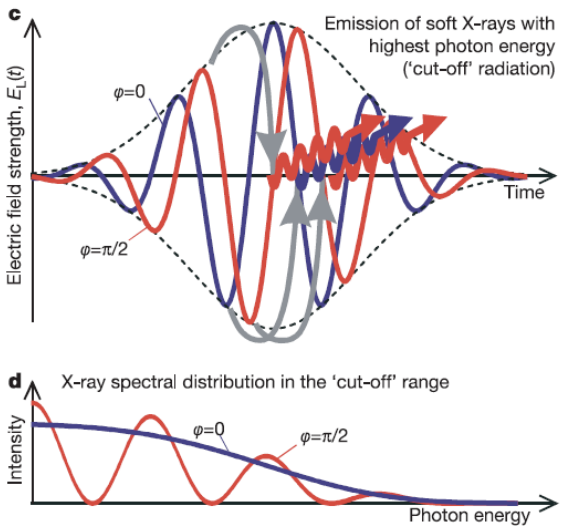

FIG. 4: Carrier-Envelope-phase

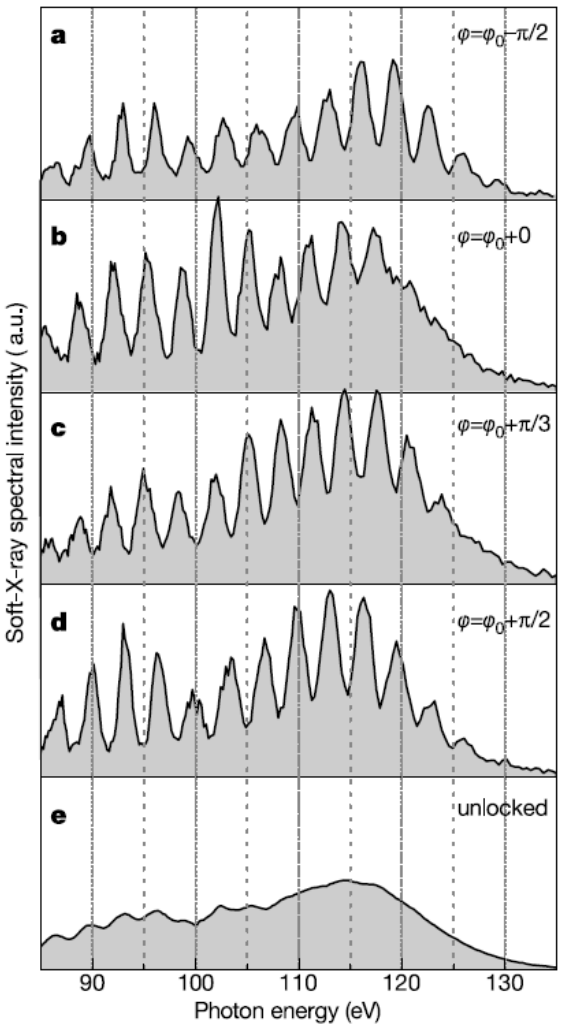

FIG. 5: influence of the CE-phase on the cut-off region

only re-collide with a linearly polarized laser field, there is the opportunity to isolate single pulses by polarization gating (very fast gating with two different laser frequencies at orthogonal polarization). This is because the electrons take different trajectories at for the external field that changes with time, therefore creating different harmonics. The time delay between the harmonics leads to chirp in the produced pulses, that can be used to compress those pulses further but is also what limits the pulsewidth. For each harmonic, there always exist two different trajectories (except for the cut-off harmonic, the one with the shortest wavelength, that is controlled by the CE-phase). The short trajectory is produced before, the long one after the cut-off harmonic. Through phase-matching, the experimentator can favor one of these. The cut-off energy is given by:

$$\omega_c = I_p + 3.17U_p \tag{1}$$

where $U_p = \frac{e^2E^2}{4m\omega_0^2}$ is the pondermotive and I_p the ionization potential. The efficiency of HHG is heavily determined by phase-matching (see that section). With an uncontrolled CE-phase, we observe different trajectories, because of different initial (field) conditions. When we chose the pulse peak of the field to hardly reach the ionization threshold, we can get single isolated electronic wave packets. With a CE-phase $\phi = 0$ (fig. 4c) only one pulse will appear, whereas $\phi = \pi/2$ lead to two x-ray burst with the same intensity which leads to a maximum modulation of the spectral intensity in the cut-off range (fig. 4d). To make this observable (those modulations would average to 0 for an unlocked phase), we need **phase-stabilized** light pulses. The experiment (using $5fs$ pulses in a neon gas sample) demonstrated the expected modulation and moreover showed that the top 40% of the harmonic peaks shift with the phase (fig. 5). Thats the mechanism that makes them smear out for unlocked phases (fig. 4e). Therefore these peaks are not referred to as genuine harmonics. They can be utilized to determine the CE-phase to within $\pi/10$.

III. PHASE-MATCHING HIGHER HARMONICS

Phase-matching is what currently limits the efficiency of HHG, therefore making the obtained pulses difficult to analyze. Like in regular NLO, the propagation effects determine the intensity we can achieve. These effects are **Absorption** (other gas atoms get excited),

dephasing (there is a phase velocity missmatch between fundamental and higher harmonic, a phasemismatch of π is refered to as one coherence length L_c). The different wavelengths experience different Gouy phase-shifts and ionization and recollision leads to a phase shift that can be evaluated quantum-mechanically. And finally there also occurs **defocusing**(As the pulse creates

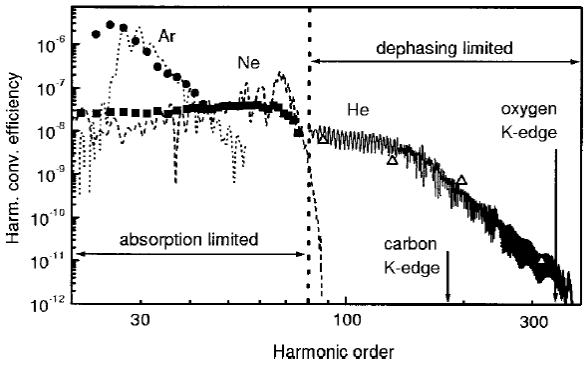

FIG. 6: limitations for higher harmonics

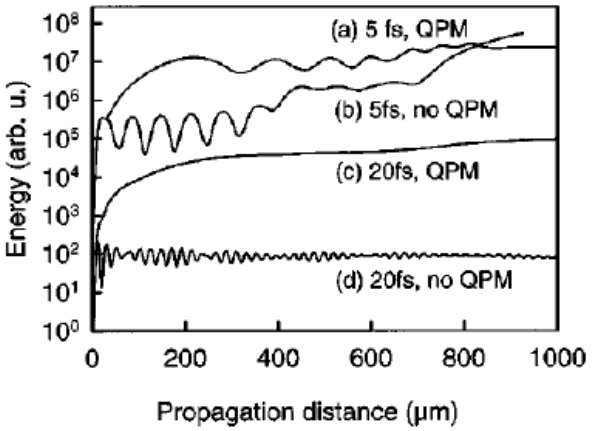

FIG. 7: Phase-matching of different pulse widths

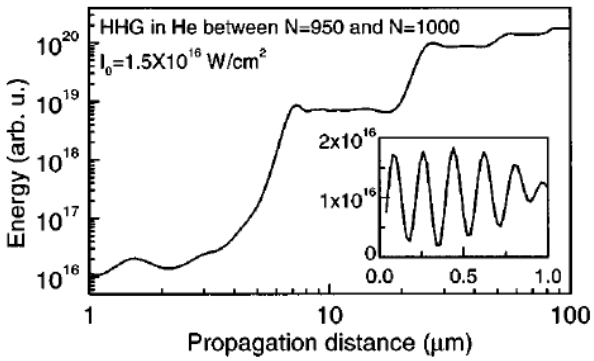

FIG. 8: efficiency over propagation distance for a very high intensity

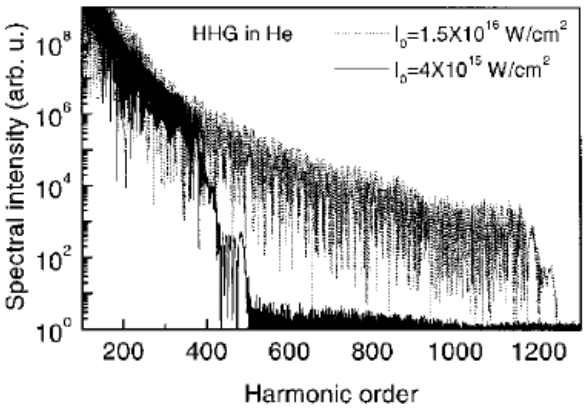

FIG. 9: higher harmonics up to 1200 get self-phase-matched by higher intensities

a free electron density profile, the effective index of refraction changes with $n_2 < 0$, which leads to defocusing. This is similar to semiconductor materials with frequencies over the bandgap.) The geometric limitations (Gouy, defocusing) can be suppresed by higher radius and pulse energy, so that intensity remains constant. In soft x-ray regime ($\lambda \leq 10nm$) dephasing becomes the dominant limitation, whereas absorption limits in the xuv regime ($\lambda \geq 40nm$) (fig. 6). **Adiabatic** phasematching means techniques, that compensate a phase that grows linearly with distance, we can compensate this phase with Quasi-Phase-matching, putting single gas cells at distances L_c. **Non-adabatic** phasematching takes care of the fact that the change of the free-electrons during one cycle is not neglicable any more and leads to dephasing. This is particularly important for high ionizations with few-cycle pulses. Fortunately these effects compensate for each other for very short driving pulses, leading to what is called **self-phase-matching**. As can be seen from the figures, with self-phase-matching, a lot more harmonics can be obtained (1200 !) (fig. 9), leaving no need to start with Quasi-PM for small driving pulses (fig. 7). With high intensities we can increase the efficiency by several orders. In (fig. 8) we can see osciallations in the efficiency upto $1\mu m$. Those are the same we can observe in SHG. At higher distances, the oscillations disappear and the efficiency grows even step-like. On future TW-scale sub-$10fs$ lasers this promises very high ouput intensities and could lead to Nonlinear x-ray optics.

IV. APPLICATIONS

Until now the simultaneous requirements of high photon energy and sub-fs pulse duration have prevented the time resolved pump-probe spectroscopy from examining ultrafast electronic processes in the inner shell of atoms. The recent availability of as pulses in the XUV-range allows now experiments which resolve those transitions. Emission of secondary Auger electrons can be triggered with as-pulses in the 100 eV range by using the streak-camera-concept. Here the pulse excites a inner shell electron. The hereby created vacancy is filled after its typical lifetime by an electron from the outer shell. The released energy leaves the atom either as a XUV/X-Ray photon or by a secondary Auger electron. The photoelectrons get emitted instantaneously when the as-pulse probes the sample. Whereas the Auger process is spontaneous. Therefore it is delayed with the emission time of the Auger electron. This effect is used by applying a

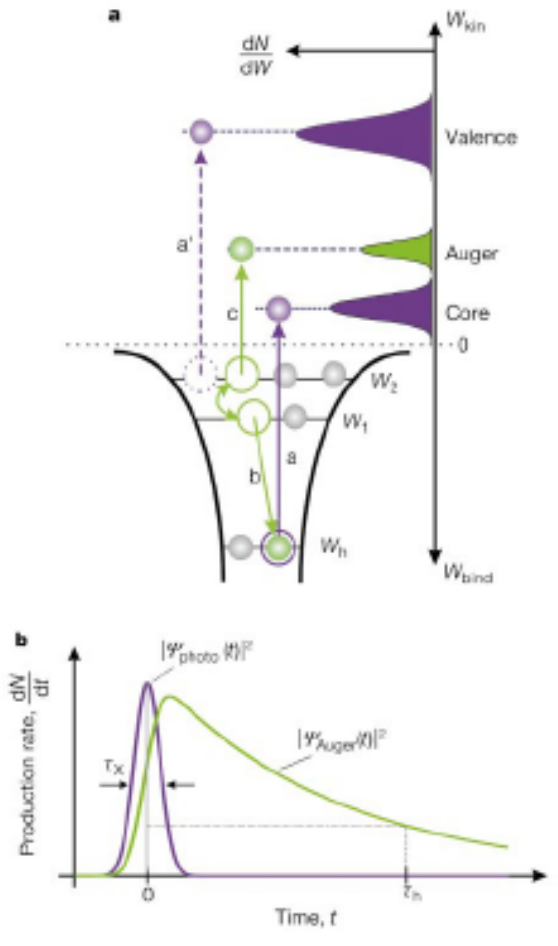

FIG. 10: Schematic illustration of atomic excitation and relaxiation process a) atomic response to an ultrashort X-Ray pulse emission of an Auger electron: process a b c, of an photoelectron: process a' b) plot of temporal behaviour of those processes

delayed light pulse, which imprints it's electric field on the trajectorie of the Auger- but not the photoelectrons. Using a photoelectron spectrometer one can now distinguish between photoelectrons (from the valenz band) and the Auger electrons. Fig (10) shows the two ways of decays (fig(10) a) and the timal spectrum (fig(10) b). A 900 as pulse with 97 eV energy from a HHG neon source ionizes a 3d electron in Krypton. The vacancy in the M-shell undergoes the described Auger decay. A 5 fs Ti-Sapphir pulse This experiment yields a Auger life time of 7.9 ± 1.0 fs. Which is in good accordance to recent energy domain measurements.

V. CONCLUSION

Variations of the experimental techniques are being investigated to get a better spatial resolution and to make experiments on shorter time scales possible. Furthermore more intense short pulse sources are highly regarded. This would allow to resolve ionization processes in atoms in temporal domain. Reactions of matter to high laser fields as the dynamics of strong field ionization and the depletion of atomic ground states by laser fields could be explored. But also many body relaxation proceses, dizzipation and viscosity of atomic nuclei might be able to observed.

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