Characterization of attosecond pulse trains from high-harmonic generation

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It was demonstrated that high harmonics, generated in a gas jet by a 40-fs laser pulse, have a well-defined phase relation with respect to each other, and that the relative phases result in strong amplitude beating between the various harmonics. Due to this beating, the radiation emerging from the gas jet consists of a train of attosecond pulses (1 as $= 10^{-18}$s), each 250 as long, and repeating every 1.35 fs.

It is described how mixed-color two-photon ionization can be used to determine the relative phases needed for detailed reconstruction of the temporal profile.

1 Introduction

At high electric field strength, the atomic polarization reacts non-linearly to the driving field. If this driving field is the electric component of an electromagnetic wave with frequency ω, focused to an intensity around 10^{14}W/cm^2, the nonlinear behavior causes the production of harmonics with frequency $N\omega$. In principle, even the polarization of a single atom would be the source of dipole radiation. In practice, the emitted radiation is only observable when millions of atoms, all driven in phase by the laser, enhance each others fields by constructive interference. This constructive interference only occurs in the direction of propagation of the driving beam, where the path length traveled by the light is to a good approximation independent of the position of the polarized atom. The harmonic radiation is thus confined to a narrow cone around the symmetry axis of the incoming beam, and the energy of the produced harmonics grows quadratically with the number of atoms in the focus.

The emission of the Nth harmonic is caused by the Nth-order perturbation of the atomic charge distribution. One might expect consecutive orders of the perturbation to decrease in magnitude as I/I_o, where I_o represents some characteristic intensity where the atom succumbs to the onslaught of the driving field, resulting in total divergence of the perturbation series. At intensities below this catastrophic intensity, the successive terms of perturbation series then decrease exponentially with their order. For the lower harmonics this behavior was well known.

It therefore came as a surprise when it was discovered that the spectrum of emitted harmonic radiation does show a completely different behavior [1]. Many harmonics, sometimes more than 100, can have comparable intensities, and this region of the spectrum is known as the plateau. At the high-energy side the plateau does end in a rather abrupt cutoff, and the decrease of harmonic strength with order

beyond this cutoff becomes indeed exponential. Due to the high order, the harmonic spectrum extends to far in the vacuum ultraviolet (VUV), even if the driving laser was in the infrared (IR).

The peculiar shape of the harmonics spectrum can be understood by realizing that the atomic continuum already leads to divergence of the perturbation series at very low intensities, especially if the fundamental frequency of the driving laser is low. Such continuum states represent an ion and a free electron, and the latter basically offers no resistance at all to acceleration by an electric field. Inertia of the electron, however, limits the excursion (in position as well as momentum) of its motion, but at low ω the quiver amplitude of a free electron can easily exceed a hundred bohr radii, at intensities where the ground state is hardly perturbed.

The process of harmonic generation can thus be thought of as consisting of three steps [2], a (rate-limiting) field ionization of the atom at times near the electric field maxima, followed by acceleration of the free electron, which, due to the ac character of the driving light, eventually makes the electron recollide at high energy with its parent ion. Radiative recapture of this fast electron into the ground state, strongly favored due to the remaining coherence between these two parts of (initially) the same wave function, completes the harmonic-generation process without any change to the atom.

2 Harmonic beating

The fact that multiple harmonics are generated simultaneously, necessarily means that the total field must have a complicated time structure. Symmetry requirements dictate that only odd harmonics are emitted by centro-symmetric media like atomic gases. The beating between such a set of frequency components, spaced by 2ω, repeats after a time T/2, (where $T=2\pi/\omega$ is the cycle of the fundamental), albeit with opposite sign [3].

The simplest example of harmonic interference consists of two harmonics beating together. In that case the beating causes a sinusoidal modulation of the total field amplitude, with two maxima and two minima per fundamental cycle T. The relative phase of the two harmonics merely determines the timing of the beats, not their shape, and thus has little physical significance. This changes with the presence of additional harmonics: depending on its phasing a third harmonic can enhance and sharpen the maxima in the beat wave of the other two, and deepen and broaden the minima between them. Or, conversely, it can weaken the maxima and beef up the minima to the extent that intensity becomes nearly constant and only a frequency-modulated wave results. So although the spectral content of the harmonic radiation guarantees an intensity and frequency pattern that repeats with periodicity T/2, the exact nature of this pattern can not be determined without knowledge of the relative phases of the various harmonics.

3 Phase measurements

To measure the relative phase of the various harmonics, a nonlinear process is required. Linear responses merely cause each frequency component to evoke a response at its own frequency. The beating between the responses will be just as fast as that in the driving radiation, impossible to measure. Non-linear media, on the other hand, can convert frequencies by sum or difference frequency mixing, and originally different frequencies converted to the same value cause a static interference that can be easily measured without the need to time-resolve anything. If the conversion process somehow preserves the phase of the converted radiation, the latter is easily deduced from this static interference.

To find a suitable non-linear process to drive with the harmonic radiation is far from trivial, since the harmonic conversion efficiency is quite small. Even with laser pulses of tens of mJ, the total energy produced in the VUV is much less than a μJ, and individual harmonics are often well below 10 nJ. Even with tight focusing, which is difficult for the scarcity of good reflective surfaces and the total absence of transparent refractive materials, such low energies hardly produce any measurable non-linear response.

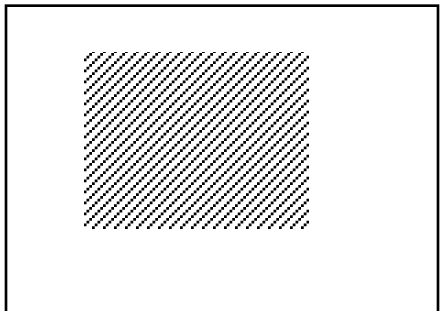
VUV radiation, however, can easily ionize atoms by single-photon processes. A photo-electron ejected with high kinetic energy is very susceptible to low-frequency laser radiation. Like mentioned in the introduction, a laser of frequency ω modulates the electron velocity by E/ω, and thus the energy [4].

A mixed-color two-photon ionization process is thus driven easily at comparatively low laser intensity of 10^{11}W/cm^2, and with laser pulse energies in the mJ range such intensities can be realized in large volumes. In the perturbative limit, the two-photon ionization amplitude is proportional to the product of the two driving fields, and thus preserves the phase of each of them.

Using ionization as a non-linear process might seem quite unusual. In non-linear optics it is much more common to utilize non-linear responses that result in the production of photons at the sum or difference frequencies of a number of driving fields. Ionization does have some advantages, though. For example, phase matching is not an issue: electrons produced from different atoms will never interfere but rather will add incoherently, since the quantum states they represent are distinguishable from the ion that remains in the gas sample. The optical methods leave the medium unaltered, and thus do represent identical quantum states no matter in which atom of the medium the new photon was generated.

4 RABBITT

To measure the relative phases of neighboring harmonics, both harmonics thus have to be converted to photo-electrons of the same energy. The processes that do this are the sum and difference two-photon processes that have absorption of a VUV photon go accompanied by absorption and mission, respectively, of an IR laser



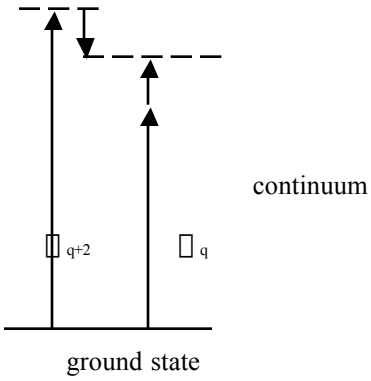


Figure 1: Two distinct two-photon pathways to the same final continuum state interfere in a way depending on the relative phases of the involved light fields.

photon. Where absorption of a single photon of harmonic q would produce electrons with an energy $E_o+q\omega$, the presence of sufficiently intense IR fundamental causes appearance of 'sidebands' $E_o+(q\pm1)\omega$. The VUV+IR process of harmonic q produces the same quantum state as the VUV-IR utilizing harmonic q+2. The resulting yield is given by the square of the sum of the amplitudes of each quantum path contributing to the state.

In the perturbative limit, when the IR intensity is very weak (a requirement that makes the experiment easier!), this can be expressed quantitatively [5]. The amplitude of the VUV+IR pathway is given as

$$A = M_q^+ F_q F_{IR} e^{-i\varphi_q} e^{-i\varphi_{IR}},$$

and that of the VUV-IR pathway to the same state as

$$B = M_{q+2}^- F_{q+2} F_{IR} e^{-i\varphi_{q+2}} e^{+i\varphi_{IR}},$$

where the fields are explicitly factorized in a real amplitude F and a phase factor. (Note the opposite sign of φ_{IR} due to emission!) The total yield is then given by

$$|A+B|^2 = \text{constant} + C\cos(\varphi_q-\varphi_{q+2}+2\varphi_{IR}+\alpha),$$

where the newly appearing phase α is a consequence of the fact that $M^{\pm}$ are complex numbers.

Recording the sideband yield as a function of the IR phase thus should produce an oscillating behavior, from which $\varphi_q-\varphi_{q+2}+\alpha$ can be immediately deduced. The small atomic phase contribution α can be reliably obtained from calculations [6],

and when the observed phase of the sideband oscillations is corrected for it we obtain the sought phase difference of the harmonics. This way of measuring relative phases has been given the name RABBITT, for Reconstruction of Attosecond Beating By Interference of Two-photon Transitions, and can be viewed as a VUV adaptation of techniques like SPIDER (Spectral Interferometry for Direct Electric-field Reconstruction), which are popular in the visible part of the spectrum [7].

The way the phase determination works can also be easily understood from classical arguments. The two involved harmonics cause beats in the amplitude of their combined field, and the production of photoelectrons follows the instantaneous amplitude of this resultant field. The presence of the IR has little effect on the ionization itself, but changes the time-averaged energy of the escaping photo-electrons. Those electrons produced near the IR field maxima are affected in a different way than those produced near IR field zero crossings. So it matters if the beat maxima occur during such field maxima, or during zero crossings. In fact we are measuring the timing of the beats between the harmonics against a time 'ruler' set up by the IR wave.

By repeating this measurement for every pair of neighboring harmonics (something that is done automatically if we take the entire electron spectrum as a function of IR phase), we thus measure the (temporal) position of the beat maxima between all such pairs along the same ruler. This allows us to reconstruct every detail of the temporal shape of the harmonics radiation with respect to this ruler.

5 Experimental setup

To perform the RABBITT measurement on the VUV pulses emerging from high-harmonics generation, only a few modifications to the experimental setup were needed [5]. We used the (by now standard) annular-beam method to separate the generated harmonics from the IR driver pulse. With this method a beam stop block the center of the laser beam. The lens used to focus the resulting beam of annular cross section into the gas jet casts an image of this beam stop a safe distance behind the focus. In this plane, an aperture blocks the annular IR beam, but passes the harmonics (which are generated on axis) together with the shadow of the beam stop. A tungsten-coated mirror refocuses the light passing the aperture into a second gas jet in a magnetic-bottle electron spectrometer. Photo-electrons produced by ionization events in this second gas jet follow the magnetic field lines on their way to the channel-plate detector, and are apertured in such a way that only electrons emerging from a collimated part of the beam, 2 mm behind the focus, are detected.

To admit IR light into the ionization region, a 2-mm hole was made in the beam stop. The beamlet passing through the hole was focused together with the annular beam into the harmonic-generation gas jet, but due to the strong diffraction of such a narrow beam most of the light passes around the region where the actual harmonic

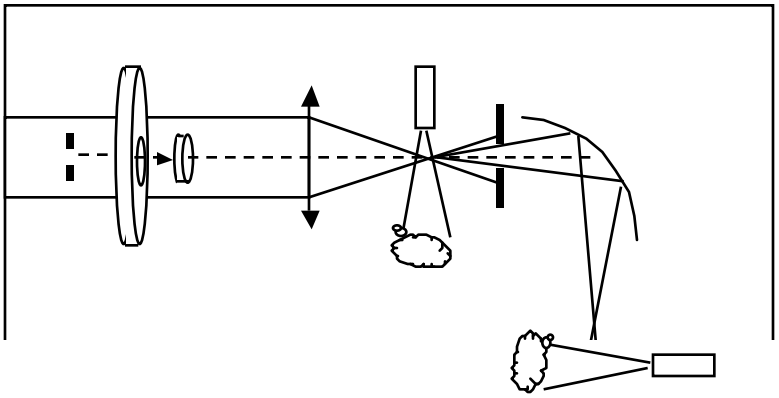


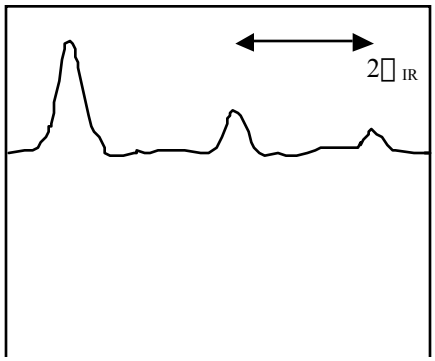
Figure 2: Setup for RABBITT. Inner and outer part of the same beam pass through independently rotatable glass plates for adjusting their phases. The outer part focus tightly in a first gas jet for harmonic generation, the harmonics and the inner part (dashed) reach a second jet where two-color two-photon ionization takes place.

generation takes place. This IR beamlet automatically overlaps the generated harmonics in time and space. To make it possible to tune its phase, it was passed through a piece of glass cut from the center of a 6-mm thick laser window; the annular beam was passing through the remaining (annular) piece of the same window. By tilting the two window pieces with respect to each other around the direction normal to the beam, very precise (sub-cycle) tuning of the delay (of either sign) between the two beams was possible without affecting the beam pointing.

6 Results

The sideband strength is indeed highly dependent on the phase of the IR light at the ionization point, all other factors (such as light intensities) remaining equal. The contrast between constructive and destructive interference is more than a factor 2 for all sidebands, showing that the relative phase of the harmonics is quite stable over the observed volume of the beam, during the laser pulse and between laser shots.

In a scan over IR phase (figure 2, left), the sideband strength shows a sinusoidal modulation with two cycles for each full wave the IR is delayed. This modulation at double the IR phase is exactly what is expected from the interference between the sum and difference quantum paths. Most conceivable artifacts, such as interference between different IR beams (e.g. the intentional one and some scattered IR from the harmonics-generation focus) would result in modulation at the single IR phase, with one fringe per 800-nm wave length. The fact that the modulation is sinusoidal is



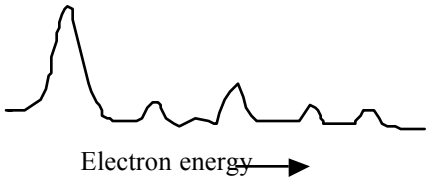


Figure 3: Photoelectron spectra taken in the second gas jet. The upper trace is produced by the harmonics only , the lower trace shows the appearance of sidebands when IR is admitted. The area of these sideband peaks is the quantity of interest.

evidence for the fact that processes involving more than a single IR photon (higher-order sidebands) do not significantly contribute to the signal; interference due to those quantum paths would result in 'overtones' of the modulation (i.e. depending on the IR phase as 4 or 6 φ_{IR}.)

The interference fringes in the various side bands seem to be in register, showing that the relative phases of the various pairs of neighboring harmonics is the same (ignoring the small atomic phase η). This implies an almost linear dependence of the phase of the individual harmonics on their frequency, with almost no quadratic or higher order dependence. The absence of these higher order phase dependences shows that the beat pulses are band-width limited, and the linear dependence is related to the timing of the beats with respect to the IR light. Since the amplitudes of the various harmonics can be easily deduced from the strength of the various ionization signals these individual harmonics cause in the absence of the IR, a complete reconstruction of the beats is possible and shown in figure 5.

7 Outlook

The nice thing about the RABBITT method is that it can in principle be applied to any harmonics, even quite high ones. The higher the energy of the photo-electrons, the lower the IR intensity required to produce sidebands of a given magnitude. This is a consequence of the fact that a small modulation (imposed by the laser) on a high velocity produces a large modulation on the energy. And as soon as the energy modulation approaches the IR photon energy, the sidebands grow strong. If the electron energies grow so high that our electron spectrometer has difficulty separating the sidebands from the single-photon signals, we can switch to a target gas with a much higher ionization potential (e.g. by employing ionization from an inner shell).

The RABBITT method also holds the promise for resolving temporal structure in non-periodic pulses, such as a single attosecond pulse rather than a train, or a short train of non-identical attosecond pulses. Such XUV pulses do not have a

spectrum that consists of narrow lines. In stead the lines around the harmonic positions broaden and acquire internal phase and amplitude structure. In the limit of a single attosecond pulse the completely merge into a broad feature. How many independent frequency components have to be measured (their amplitude and phase!), depends on the total duration of the XUV burst; short bursts have a much smoother spectrum than long ones.

A spectrum that is no loger line-like makes it impossible to separate the single- and two-photon signals based on their energy. Recording the angular distribution of the photo-electrons offers an alternative, and in general allows unambiguous separation of the sidebands from the ordinary ionization (which has opposite parity). If, for instance, the target atom has its electrons in an s-state (such as He), the single-photon events produce p-continuum states with a nodal plane perpendicular to the VUV polarization. The only signal in the direction of this plane is then due to the sidebands, allowing background-free measurement of those even without recording the full angular distribution.

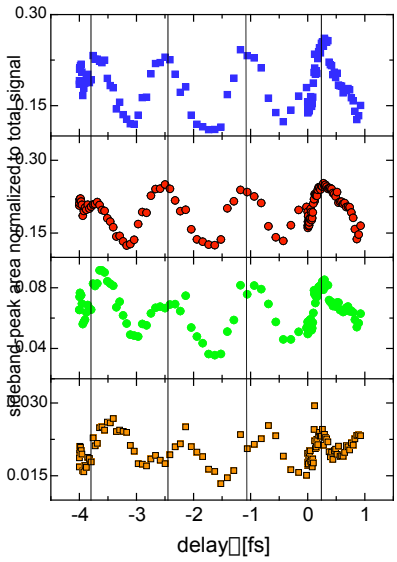


Figure 4: Strength of the various sideband signals as a function of the IR phase (given as a time delay; one full cycle equals 2.7 fs). From top to bottom we see the signal 12, 14, 16 and 18 infrared (800-nm) photon energies above the atomic ground state.

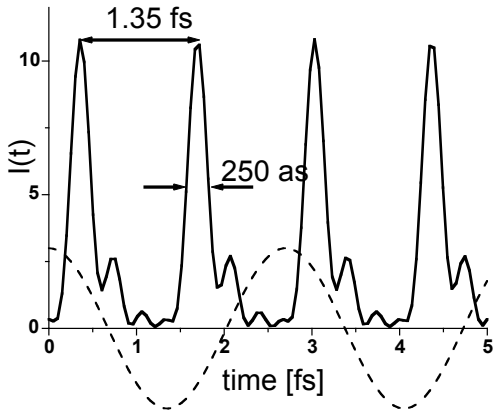


Figure 5: The reconstructed VUV intensity as a function of time. The dashed line indicates the infrared field that was used for the phase determination.

The concept of 'fundamental' of course becomes meaningless in the case of non-periodic XUV pulses. One should merely consider the IR as an auxiliary field causing (simultaneous) up and down conversion of the photo-electrons, and thus causing interference between the processes employing XUV frequencies spaced by 2ω. If a 2ω grid does not offer sufficiently fine sampling to fully characterize the XUV pulse, (because it lasted longer than T/2 and thus had spectral structure within 2ω), a finer sampling can be affected by using IR of a longer wavelength. An alternative is to repeat the measurement at two different IR frequencies ω and ω' close to the frequency of the driving laser (e.g. from the opposite wings of a chirped-out version of that pulse). This is equivalent to sampling on a frequency grid of the greatest common divisor of ω and ω', and can handle quite high-density sampling of the spectrum while avoiding the use of mid or far infrared radiation. In fact the use of two rather large frequencies with a small difference is superior to using a single small frequency, since it directly compares frequencies far apart, the relative phase of which should otherwise be derived from summing phase differences over many small frequency steps (with a corresponding pile-up of the experimental error). The worst limitation in this case seems to be the resolution of the electron spectrometer.

The icing on the cake of course would be the actual application of attosecond pulses or pulse trains to other problems, to study processes on the attosecond time scale. Prime candidates in the field of atomic physics are processes similar to the non-linear process used in the phase measurement. An attosecond pulse train can be used as means to effect ionization in attosecond bursts. This allows the possibility

to inject electrons during specific phases of the fundamental IR field, and several processes driven by that IR field can then be studied [8].

In particular, the theory of harmonic generation by ionization, acceleration, recollision and recapture predicts that electrons leaving the atom at different times recollide at different energies, and thus produce harmonics in different energy ranges. This can be directly tested by seeding a harmonic-generation setup that is driven below the intensity where the gas is field ionized by the IR (e.g. because it has a high ionization potential, like He or Ne) with an attosecond train from another harmonics source. The relative phasing of the attosecond train with respect to the IR should then allow control of the generated harmonic spectrum (which might include harmonics of order very much larger than the original train).

Another good candidate would be the study of non-sequential double ionization [9]: the attosecond pulses would create photo-electrons (by single-photon single-ionization events) only during a particular phase of a strong IR field, and thus select the trajectories along which the IR field could drive these electrons. This way it could be experimentally determined which trajectories are effective in recolliding with the parent ion and producing a second ionization (e,2e) event.

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