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Phase-Locked High-Order Harmonic Sources

Raoul Zerne,1 Carlo Altucci,1 Marco Bellini,2 Mette B. Gaarde,1,3 T. W. Hänsch,2,4 Anne L’Huillier,1 Claire Lyngå,1 and C.-G. Wahlström1

1Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden
2L.E.N.S, Largo E. Fermi, 2, I-50125 Florence, Italy
3Niels Bohr Institute, Ørsted Laboratory, 2100 Copenhagen, Denmark
4Max-Planck-Institut für Quantenoptik, P.O. Box 1513, D-85740 Garching, Germany

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We demonstrate that two harmonic sources generated independently in a xenon gas jet using the same picosecond Nd:YAG laser are locked in phase. The experiment is performed by separating a laser beam into two parallel beams focused at different locations under the nozzle of a gas jet, and therefore producing two independent sources of harmonic radiation, and studying the pattern obtained in the far field when the radiations from these sources interfere. A high and robust fringe visibility is obtained. [S0031-9007(97)03735-6]

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High-order harmonic generation (HG) in gases has attracted considerable attention in the last few years, motivated by the potential of the harmonic radiation to become a useful coherent source in the extreme ultraviolet (XUV) range. Although fundamental aspects of HG have been extensively studied [1], many issues regarding the use of harmonics as a practical source of high-frequency radiation remain to be explored. Efforts are being made in improving and measuring the conversion efficiency and to study its sensitivity with respect to differences in the intensities of the driving fields for the two sources.

Our experimental setup presents some analogies with Young’s double slit experiment. We create harmonics in two different foci separated by a small distance and observe the interference pattern in the far field. Note the fundamental difference with Young’s double slit experiment: The radiations from the two sources are not directly emerging from the same plane wave (and therefore locked in phase), but are generated through the process of high-order harmonic generation. The experiments are performed using a picosecond laser system focused in a jet of xenon atoms. We find that the generated harmonics, from the 7th to the 17th, lead to a good and robust fringe visibility. We have performed detailed calculations simulating the experimental conditions and find that intensity-dependent phase shifts can indeed remain negligible, well into the plateau region of high-order harmonic generation, i.e., into a very interesting wavelength range in the extreme ultraviolet.

We begin by introducing some notations, which are similar to those used to describe Young’s double slit experiment. We consider two harmonic sources of radiation separated by a distance Δ and measure the interference pattern in an observation plane placed at a distance L from the sources. In the ideal case, where the sources are perfectly phase locked, quasimonochromatic, with no intensity-dependent phase and no chirp, the intensity of the total field at a point with ordinate Y in the observation plane reads as

\[ I = I_1 + I_2 + 2\sqrt{I_1I_2}\cos(2\pi \frac{\Delta Y}{\lambda L}), \tag{1} \]

where \(I_1, I_2\) are the intensities of the two harmonic beams at the same point and \(\Lambda\) the harmonic wavelength. The visibility, or contrast of the fringes, defined by \(V = (I_{\text{max}} - I_{\text{min}})/(I_{\text{max}} + I_{\text{min}})\) is, in this simple case, equal to \(2\sqrt{I_1I_2}/(1 + I_1/I_2)\). V is equal to 1 when the
harmonic pulses have the same intensity and decreases with the harmonic intensity ratio. How rapidly $V$ decreases with the intensity ratio of the two fundamental pulses reflects, in this ideal case, how rapidly the harmonic strength varies with the laser intensity.

For a generation process inducing a frequency chirp, i.e., a temporal variation of the instantaneous frequency during the laser pulse, however, the decrease of the visibility with the intensity ratio depends also on how rapidly the harmonic phase varies with the laser intensity. The instantaneous fringe pattern, at any given time $(t)$ during the laser pulse, reads as

$$I(t) = I_1(t) + I_2(t) + 2\sqrt{I_1(t)I_2(t)} \times \cos \left[ 2\pi \frac{\delta Y}{\lambda L} + \phi_1(t) - \phi_2(t) \right],$$

where $\phi_1(t), \phi_2(t)$ are the phases of the generated fields at the observation point at time $t$. The observed interference pattern is obtained by integrating $I(t)$ over the pulse duration. If $\phi_1(t) - \phi_2(t)$ varies significantly during the laser pulse (i.e., by more than $\approx \pi$ over the time during which harmonics are generated), the fringe pattern will be washed out upon time integration.

The experimental setup is shown in Fig. 1. The output from a mode-locked Nd:YAG laser (1064 nm, 35 ps) [13] is focused by a 255 mm focal length lens ($f\# = 30$) and then split into two spatially displaced parallel beams of equal intensities but orthogonal polarizations in a beta barium borate (BBO) crystal. The axes of the crystal are oriented at 45° with respect to the laser polarization. A polarizer placed after the crystal transmits a common polarization component in the two beams. By rotating the polarizer, the intensity ratio between the two beams can be continuously varied. The distance between the two beams, and consequently between the two foci, is $\delta \approx 150 \mu m$, while the beam waist diameter in each focus is less than 50 $\mu m$. The two sources of harmonic radiation are hence well separated in space and independent of each other. The maximum intensity in each of the two foci is estimated to be slightly above $10^{13}$ W/cm². Harmonic radiation of a particular order (ranging between the 7th and the 17th) is selected by a normal-incidence spherical grating and imaged onto a micro-channel-plate detector (MCP), coupled to a charged-coupled device camera. The distance ($L$) from the focal plane of the spectrometer and the observation plane is about 75 cm.

An example of a recorded far-field pattern for the 13th harmonic is shown in Fig. 2(a). The intensities in the two beams are approximately equal. Interference fringes are clearly visible. Their position is very stable on a

![Experimental setup](image)

**FIG. 1.** Experimental setup.

![Interference pattern](image)

**FIG. 2.** (a) Interference pattern for the 13th harmonic for equal intensities of the two laser pulses, (b) integration of the fringe pattern shown in (a), and (c) theoretical simulation in the same conditions. The solid line has been obtained for equal intensities, the dashed line for an intensity ratio of 0.75.
shot-to-shot basis. The recording has been obtained by integrating over 30 laser shots. However, similar results are obtained in single-shot recordings as well. Figure 2(b) shows the integration of the fringe pattern in the vertical direction. The contrast of the fringes is 30%. The fringe interval is measured to be 0.44 mm, which agrees with the expected interval $i = \lambda L / \delta = 0.41$ mm. We find that the interference pattern is rather robust, and that it can be observed, with varying degrees of contrast, even when the peak intensities in the two foci differ by almost a factor of 2. It is also observed to be independent of the position of the gas jet relative to the laser focus, over the length over which harmonics can be observed (a few mm). In Fig. 3, we show two fringe patterns obtained with the 9th harmonic. The fringe interval is larger than for the 13th harmonic, 0.6 mm, directly reflecting the longer wavelength. Figure 3(a) is obtained with approximately equal intensities for the two laser pulses, while in Fig. 3(b) the laser intensity ratio is about 0.6. The contrast decreases in this case from 30% to 6%. The contrast is never observed to be better than $-30\%$ for single-shot recordings and does not improve significantly with averaging. Some of the laser shots, apparently in a random fashion, do not lead to any fringe pattern: we believe this might reveal some instability of the driving laser itself. Unfortunately, this prevents us from performing systematic quantitative measurements, e.g., of the contrast as a function of the laser intensity ratio.

To interpret the experimental results, we have performed numerical simulations for conditions close to the experimental ones. The method is described in a previous Letter [11]. It uses the strong field approximation for the single-atom response and the slowly varying envelope approximation for the propagation of the generated field. The fringe pattern is obtained by numerically superposing, in the far field, the two harmonic fields generated in each focus. The result obtained for the 13th harmonic is shown in Fig. 2(c). The solid line shows the result for equal intensities of the two laser pulses ($1.5 \times 10^{13}$ W/cm$^2$); the curve in the dashed line is obtained for an intensity ratio of 0.75. The theoretical results compare well with the experimental one. The fringe interval and the total number of fringes observed (which depends on the harmonic angular distribution) are very similar. The contrast is equal to 1 for equal intensities, as expected, and decreases with the intensity ratio (it is 30% for an intensity ratio of 0.75).

In Fig. 4, we examine the variation of the total phase for the 13th harmonic field in xenon as a function of time for two different peak intensities: $0.75 \times 10^{13}$ (dot-dashed line) and $1.5 \times 10^{13}$ W/cm$^2$ (solid line). We consider a point in the observation plane on axis. The phase variation of the propagated field is found to be approximately quadratic. The total phase variation $\phi_1(t) - \phi_2(t)$ over 30 ps is about 0.8 rad. This does not induce a significant displacement of the fringe pattern. In Fig. 5, we plot the contrast as a function of the laser intensity ratio, using a maximum intensity of $1.5 \times 10^{13}$ W/cm$^2$ (full squares). As in the experiment, the contrast decreases rather slowly with the intensity ratio. The influence of the
A direct application of the phase locking is the generation of coherent XUV radiation whose degree of polarization can be continuously controlled. This can simply be achieved in the present setup by removing the polarizer after the BBO crystal (see Fig. 1), so that the two generated harmonic beams are cross polarized and the total field has a degree of ellipticity varying from circular to linear in space. To check this idea, in addition to removing the polarizer, we place an XUV polarizer (a gold-coated mirror at a 45° angle of incidence) before the MCP detector. We indeed obtain interference patterns similar to the ones shown in Figs. 2 and 3. By placing a slit perpendicular to the direction defined by the two foci, it is possible to obtain XUV radiation with a given degree of polarization.

In conclusion, we have demonstrated that harmonics of “moderately” high orders, such as those produced by picosecond laser systems at relatively modest intensities, are coherent, in the sense that they lead to stable interference patterns. This makes it possible to split the laser beam into two and to generate two independent but phase-locked beams at the new frequency, which can be directly used, for example, in an interferometry experiment. This result opens perspectives for applications such as polarimetry, interferometry, holography, or even high resolution Ramsey spectroscopy in this short wavelength region.

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