Laser and Particle Beams (2004), *22*, 199–202. Printed in the USA.

DOI: 10.1017/S0263034604223011

High resolution spectroscopy in the XUV with pairs of mutually coherent and time-delayed laser harmonics

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(Received 1 November 2003; Accepted 17 February 2004)

Abstract

We present a Ramsey technique using high-order harmonics for high-resolution atomic spectroscopy in the extreme ultraviolet. Pairs of time-delayed and phase-coherent harmonic pulses generated by the interaction of ultrashort and intense laser pulses with a noble gas are used to study excited states of krypton.

Keywords: High-order harmonic generation; Ramsey spectroscopy; Ultrashort laser pulses; XUV radiation

1. INTRODUCTION

High-order harmonics generated by the interaction of amplified femtosecond laser pulses in noble gases are an efficient source of coherent radiation in the XUV and soft X rays. Thanks to their unique properties, harmonic sources have the potential for a wide range of applications, from atomic and molecular spectroscopy to plasma and surface studies (Salières *et al.*, 1999*a*). Unfortunately, the ultrashort duration of the pulses corresponds to a broad spectral width that limits the instrumental linewidth to the inverse of the pulse duration and is thus not compatible with high-resolution spectroscopic investigations. However, if pairs of time-delayed and phase-locked pulses in a Ramsey-type configuration are used, the resulting field, sum of the two temporally separated pulses is given by

$$E_{tot}(t) = E(t) + E(t+\tau) \tag{1}$$

and a simple Fourier transformation shows that the corresponding spectrum

$$|\tilde{E}_{tot}(\omega)|^2 = 2|\tilde{E}(\omega)|^2(1+\cos(\omega\tau)) \tag{2}$$

maintains the broad envelope but also acquires a sinusoidal modulation with a spectral period given by the inverse of the temporal separation between the two pulses τ. It is the time delay between the two pulses and thus, practically, the length and the stability of the delay line that now sets the instrumental resolution and allows one, in principle, to investigate very fine spectral features if long time delays are available (Fourkas *et al.*, 1989; Haberle *et al.*, 1995; Bellini *et al.*, 1997; Cavalieri & Eramo, 1998). Moreover, combining this technique with the short wavelength radiation of harmonics, one can study high-lying bound atomic states or auto-ionizing levels by means of one-photon transitions.

The use of this technique in the extreme ultraviolet is, however, not straightforward because the Michelson interferometer normally used to create time-delayed pulses cannot be built to work in the XUV due to the lack of good mirrors and beamsplitters. A possible solution to the problem is to invert the two steps of harmonic generation and pulse splitting, placing the interferometer in the path of the pump laser pulses and relying on the fact that the phase coherence among them is transferred to the harmonic pulses. A simple test of the mutual phase coherence between time-delayed harmonic pulses was performed by generating them in two spatially separated regions obtained by focusing two phase-coherent laser pulses obtained from a Michelson interferometer. The existence of clear and stable interference patterns of high visibility in the harmonic far field showed that two secondary sources could preserve a memory of the phases of their parent pulses, demonstrating that two phase-locked pump pulses can indeed produce two phase-locked harmonic pulses (Bellini *et al.*, 1998).

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However, to obtain the modulated spectrum predicted by Eq. (2), the pulses need to be collinear, and they thus have to focus into the same spot of the gas jet and interact with the same atoms to produce harmonics. In this case one cannot simply extend the previous results, because harmonic generation always implies a certain degree of medium ionization and the second pulse of the pair may generate harmonics in a less efficient way or with significant phase disturbances due to the presence of free electrons. A second experiment was then performed to observe the spectra corresponding to sequences of collinear harmonic pulses with different time delays: in Figure 1 we show modulated two-pulse spectra corresponding to the fifth (a and b) and to the seventh harmonics (c) at different time delays between the pump pulses. According to expectations based on Eq. (2), the two-pulse spectra present the broad envelope of the single pulses with a superposed sinusoidal modulation, leading us to conclude that also in the collinear case and for harmonics of medium order and peak intensities of the order of 10^{14} W/cm^2 there is no degradation of the fringe contrast due to the generation process itself. Thus, the generation of collinear phase-locked harmonic pulses is possible and such pulses can be used to achieve high-resolution spectroscopy in the short-wavelength region (Salières *et al.*, 1999*b*; Bellini *et al.*, 2001).

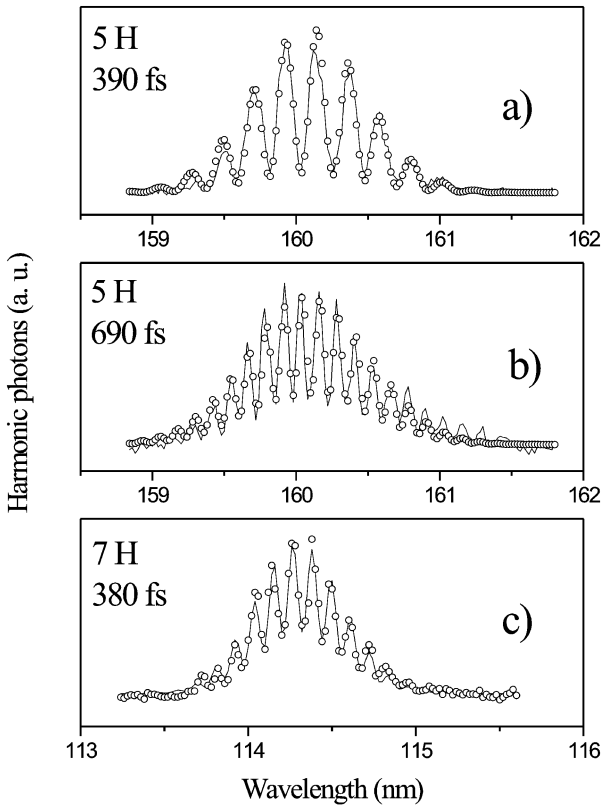

Fig. 1. Two-pulse spectra showing interference fringes with a period $\delta\lambda = \lambda^2/c\tau$ are shown for the fifth harmonic at $\tau \approx 390$ fs (a) and $\tau \approx 690$ fs (b). The spectrum of the seventh harmonic at $\tau \approx 380$ fs is shown in c.

2. EXPERIMENTAL RESULTS AND DISCUSSION

From a temporal point of view, two short, time-delayed harmonic pulses that would no longer show optical interference due to their temporal separation may be made to overlap again thanks to the spreading of their temporal profiles induced by a spectral filtering, given in the above case by the exit slit in the focal plane of the monochromator. If the two light pulses are phase locked, interference effects show up while scanning the delay and last as long as the time separation is compensated by the pulse spreading. The width of the spectral filter sets the maximum time delay for the existence of interference fringes, whereas its shape determines the type of decay. It is then possible to replace the instrumental spectral filter of the monochromator with the spectral profile of an atomic transition, and look for interference fringes in some of the atomic excitation-related observables, thus realizing the first example of Ramsey spectroscopy in the XUV (Cavalieri *et al.*, 2002).

Using the ninth harmonic (at 88 nm) of our Ti:sapphire amplified laser (providing 100-fs-duration pulses, with a wavelength centered at 792 nm and 1 kHz repetition rate) we excited the $4p^5(^2P_{1/2})6d'$ and $4p^5(^2P_{1/2})8s'$ autoionizing states of krypton. These two states have an energy separation of about 29 meV, well below the 90 meV of the single-pulse spectral width, so that both of them are simultaneously excited by the ninth-harmonic pulses. The first pulse induces a coherence in the system, creating a dynamical polarization of the medium that oscillates at the transition frequency with a decaying amplitude during the dephasing time. The second pulse, depending on its phase with respect to the polarization oscillation, can enhance or destroy the residual system excitation. The quantum interference on the states is expected to manifest as a fringe pattern in the ionization signal versus the delay between the pulses, with a fringe spacing given by the atomic transition period (Cavalieri & Eram, 1998), about 0.29 fs in this case.

The experimental setup, sketched in Figure 2, is composed of a Michelson interferometer, used to provide the laser pulse pairs and to control their relative delay by means of a stepping motor and a piezoelectric crystal, a vacuum chamber, where the nonlinear generation of radiation is achieved in a pulsed jet of Xenon atoms (generation chamber), a monochromator to select the harmonics, and an interaction chamber, where the harmonic field interacts with a jet of krypton atoms. The interaction chamber is provided with an electron-energy and ion-mass time-of-flight spectrometer (TOF). A 200-mm focal length plane-convex lens was used to focus the laser pulses to peak intensities up to 1.5×10^{14} W/cm^2 in the gas jet, near the exit nozzle of the pulsed valve in the generation chamber. The electrons produced by the photoionization of krypton atoms leave the interaction region and are analyzed by the TOF spectrometer, and the spectrally unresolved field of the selected harmonic is observed downstream from the interaction region

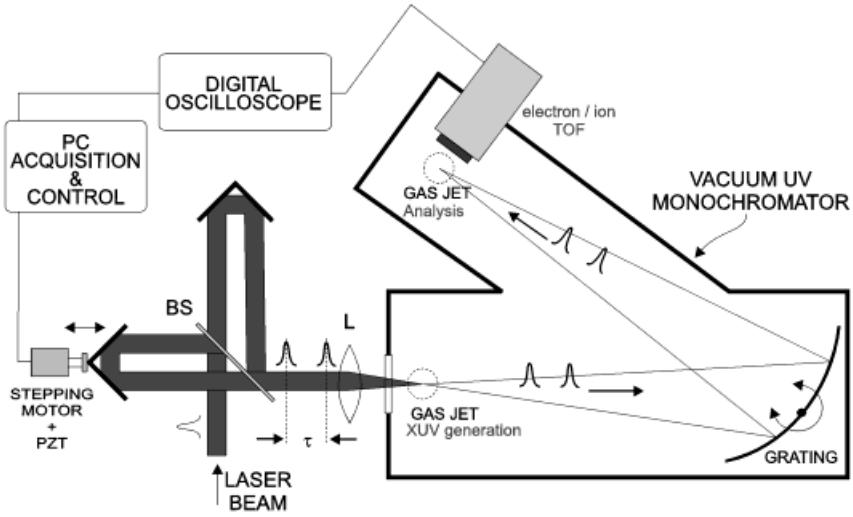

Fig. 2. Experimental setup: The ninth harmonic, generated by focusing the phase-locked pulse pair in the xenon jet is selected by the monochromator grating and crosses the the krypton jet in the interaction chamber. The electrons ejected in the one-photon ionization process are energy analyzed by a TOF spectrometer as a function of the delay between the incoming pulses.

by a phosphor/photomultiplier pair, whose signal, recorded by a digital oscilloscope, gives a pulse-by-pulse monitor of the harmonic signal. The interference fringes in the electron signal when the delay is smoothly varied around 110 fs are shown in Figure 3. As the $8s'$ and $6d'$ states have an energy separation that is not resolved in the TOF electron-energy spectrum, both continua contribute to the observed ionization signal. The modulation is clearly visible above the statistical fluctuations, and has the expected period. The observation of the modulation, a signature of the quantum interference effect, is indeed a stringent demand for the Ramsey-type approach with high-order harmonic radiation.

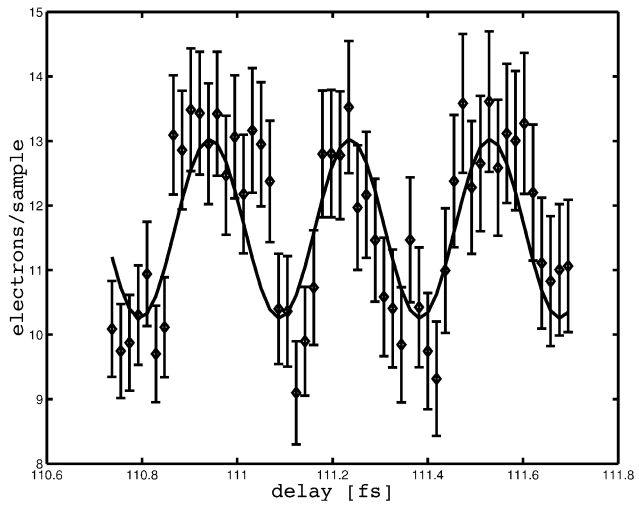

Fig. 3. Detail of the fringe pattern around the 110-fs delay. The fringe spacing corresponds to the 0.29-fs period of the atomic transition. Experimental points result from summing over 10 laser pulses and smoothing on a small fraction ($\approx$ 10%) of the fringe period. The data bars measure the statistical errors.

The simultaneous detection of the transmitted harmonic intensity did not show any modulation, thus verifying the absence of a residual optical interference between the harmonic pulses. The fringe contrast as a function of the pulse delay can be compared with the theoretical curves obtained by assuming an essential-state model for the interaction (Cavalieri & Eramo, 1998). The model parameters, that is, the states' energies, energy widths, ionization constants, and the Fano parameters, are the ones reported in the synchrotron measurements of Wu *et al.* (1990). The energy difference between the two states is expected to give rise to a beating with a period of 143 fs; our experimental data for the fringe contrast as a function of the delay shows such modulation in a fairly good agreement with the calculated curve.

3. CONCLUSIONS

We have presented the first demonstration of a Ramsey-type spectroscopic technique involving high-order harmonic pulses in the XUV. Preliminary experiments have first proved the preservation of phase coherence in the harmonic generation process, in the case of both spatially separated and collinear sources. Finally, an experimental setup for Ramsey spectroscopy has been realized and used to study a pair of autoionizing states of krypton. This novel spectroscopic tool, based on relatively simple table-top equipment, has the potential to become an attractive alternative to synchrotron radiation sources for a range of selected applications in the XUV region.

ACKNOWLEDGMENT

This research has been performed at LENS, under EEC contract HPRI-CT1999-00111.

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