# Ramsey-Type Spectroscopy with High-Order Harmonics

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We present an experiment where Ramsey-type spectroscopy is applied to autoionizing states of krypton by using the ninth harmonic (88 nm) of a femtosecond Ti:sapphire laser. The ionization process, detected by an electron-energy spectrometer, shows the characteristic quantum interference pattern. The behavior of the fringe contrast compares favorably with a simple essential-state model, whose parameters have been taken from the literature. The experiment shows the feasibility of high-resolution spectroscopy in the extreme ultraviolet by using high-order laser harmonics.

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Rapid progress in the development of high-intensity short-pulse-laser systems permits efficient generation of coherent radiation in the vacuum ultraviolet, extreme ultraviolet (XUV), and soft x-ray regions by means of highly nonlinear optical mixing in polarizable media. High-order harmonic generation has now the potential to find a wide range of applications, from atomic and molecular spectroscopy to plasma and surface studies [1]. One of the main limitations in its use is the large bandwidth associated with the short duration necessary for an efficient generation. A technique to overcome this limitation is the Ramsey-type spectroscopy, whose resolution is limited only by the length and the stability of the delay line [2–5]. The technique, based on the use of a pair of collinear phase-locked pulses, has been applied to bound state spectroscopy with optical sources in single-photon as well as in multiphoton and multistep transitions [6]. In its simplest scheme, a short pulse induces a coherence in a two-level system, creating a dynamical polarization of the medium. The induced polarization 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. As a result, any polarization-related observable exhibits interference fringes when varying the delay between the two pulses. In more complex excitation schemes, involving more than two interacting states, the modulation of the fringe pattern gives information on the energy separation of nearby states and on their lifetimes. Recently, the use of Ramsey-type excitation has been suggested also as a spectroscopic tool for states embedded in the continuum, i.e., autoionizing states, even when the continuum part of these states plays a relevant role in the dynamic of the system [7]. This is the typical case for high-lying excited states, where high-order harmonics are good candidates for one-photon induced transitions. However, it is not straightforward to obtain a pair of phase-locked harmonic pulses; in fact, due to the lack of suitable optics in the XUV region, a Michelson interferometer is not available to create the pair of delayed pulses. Experiments have been recently done to check the possibility of producing the phase-locked pulse pair from two phase-locked collinear laser pulses: The results of the optical tests suggest that this is indeed possible, at least in a delimited intensity range [8,9].

In this paper, we present an experiment where Ramsey-type spectroscopy is applied to autoionizing states of krypton by using the ninth harmonic of a Ti:sapphire laser. To our knowledge, this is the first time that Ramsey-type spectroscopy is employed in conjunction with high-order harmonics and, in perspective, this could be a resource in the field of high-resolution laser spectroscopy at short wavelengths.

As schematically shown in Fig. 1, two phase-locked harmonic pulses excite the $4p^5(^2P_{1/2})6d'$ and $4p^5(^2P_{1/2})8s'$ autoionizing states of Kr. These two states have an energy separation of about 29 meV, well below the 90 meV of the single-pulse spectral width. Both of them are thus simultaneously excited by the ninth-harmonic pulse. The presence of the two autoionizing states within the bandwidth of the light pulse does not affect the Ramsey fringes visibility, but results in a nonexponential decay due to the beating of the two states. In fact, the quantum interference on the states is expected to manifest as a fringe pattern in the electron signal versus the delay between the pulses, with a fringe spacing given by the atomic transition period [7], in this case about 0.29 fs. The modulation of the fringe contrast on the scale of twice the state lifetime, which amounts to tens or hundreds of fs, will reflect the decay and the possible beating of the autoionizing states.

  

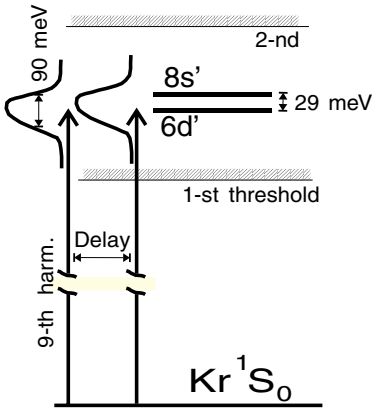


FIG. 1 (color online). Scheme of the interaction, showing the involved autoionizing levels of the krypton atom.

The experimental setup, sketched in Fig. 2, is composed basically of four units plus the laser system not included in the figure: a Michelson interferometer, used to provide the pulse pair and to control the relative delay, 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). The Ti:sapphire amplified laser provides 100 fs-duration pulses, with a wavelength centered at 792 nm (ninth harmonic at 88 nm on resonance with the Kr states), and with 1 kHz repetition rate. Large scans of the time delay are performed with a stepper motor controlled translation stage, whereas shorter and smoother scans are obtained by applying a triangular wave voltage to a piezoelectric crystal, in both cases translating the mirror in one arm of the interferometer. Piezoelectric transducers are also used for the fine alignment of the Michelson interferometer, which is enclosed in a Plexiglas box and is placed on an antivibration plastic sheet. We used a 200 mm-focal length plane-convex lens to focus the laser pulses to peak intensity up to $1.5 \times 10^{14}$ W/cm$^2$ in the gas jet, near the exit nozzle of the pulsed valve in the generation chamber.

After being separated by the normal incidence grating, the harmonics are either directed into the interaction chamber, or can be deviated by a mirror towards the exit slit of the monochromator, where they are detected by a phosphor screen and a photomultiplier. The high-resolution measurement of the two-pulse spectrum at this position is important not only for the knowledge of the harmonic wavelength: This, indeed, allows us to test the phase-lock condition, as shown in Ref. [9], and also to check the alignment of the spectrometer on the scale of fraction of the harmonic wavelength. It is important to point out that the requirement on the stability of the alignment is thus very challenging, and acquisition times cannot exceed a few minutes. The fine optimization of this alignment must be routinely carried out by means of the piezoelectric controls of the Michelson interferometer.

The electrons produced by the photoionization of krypton atoms leave the interaction region and are analyzed by the TOF spectrometer, described in more detail in Ref. [10]. The krypton jet valve works at 10 Hz repetition rate, and this is the limiting rate of our acquisition. A microchannel plate assembly detects the electrons at the end of the flight tube; the time-resolved electron signal,

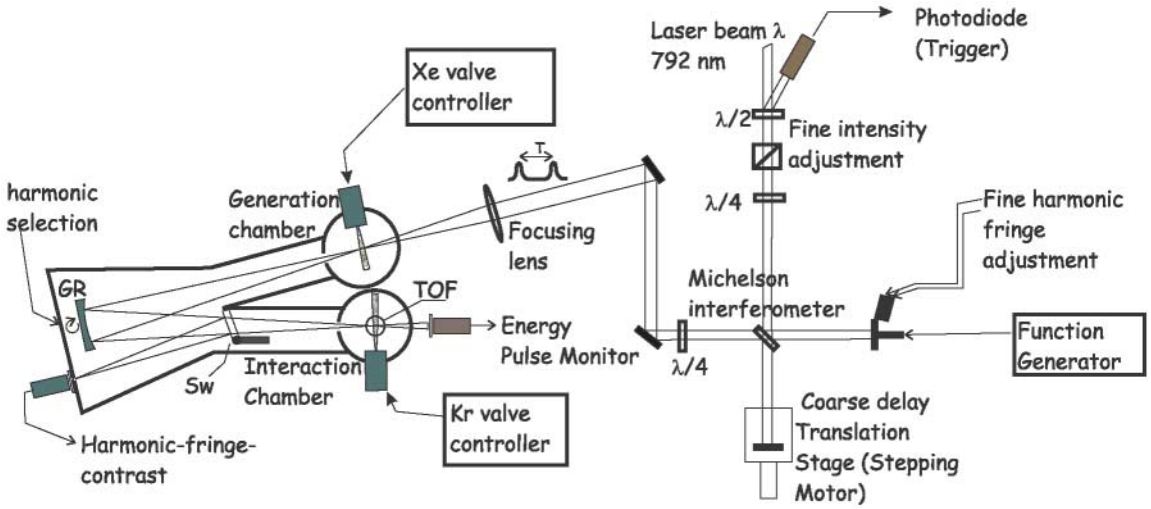


FIG. 2 (color online). Experimental setup. The ninth harmonic, generated by focusing the phase-locked pulse pair in the xenon jet and selected by the XUV monochromator, is focused in the krypton jet in the interaction chamber. The electrons ejected in the one-photon ionization process are energy analyzed by a TOF spectrometer. A switchable mirror (Sw) can redirect the harmonic field to the exit slit of the monochromator, where a spectrum of the pulse-pair field can be measured and used for the fine alignment of the Michelson interferometer. A phosphor/phototube pair, at the exit of the interaction chamber, monitors the harmonic field intensity.

amplified and then shaped by a fast constant-fraction discriminator, is finally sent to a fast multiscaler PC board, with 0.5 ns time resolution. The choice of the electron detection configuration, as compared to the ion-detection one, has the advantage of giving a more stringent monitor of the on-resonance condition through the electron spectrum; moreover, it allows a partial resolution of the ionization coordinate along the TOF axis, thus reducing the count average from 3D to 2D geometry, a considerable improvement when working in conditions where the signal depends critically on the phase-front superpositions of two pulses [11]. Finally, the spectrally unresolved harmonic field of the selected harmonic is observed downstream from the interaction region by a second phosphor/photomultiplier pair whose signal, recorded by a digital oscilloscope, gives a pulse by pulse monitor of the harmonic signal.

Figure 3 shows the interference fringes in the signal of the electrons, emitted in the direction of laser polarization, when the delay is smoothly varied around the 110 fs delay. As the $8s'$ and $6d'$ states have an energy separation which 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. The total time of the acquisition, about 1 min, is dictated by the contrasting necessities of fringe stability and count statistics. The simultaneous detection of the transmitted harmonic intensity does not show any modulation, thus verifying the absence of a residual optical interference between the harmonic pulses.

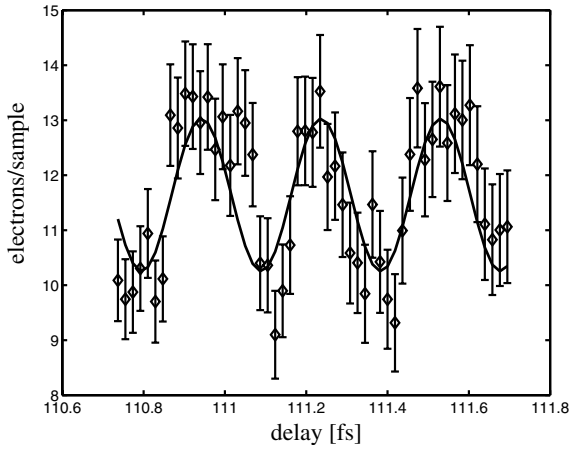


FIG. 3. Detail of the fringe pattern around the 110 fs delay. The fringe spacing corresponds to the 0.29 fs-period of the atomic transition. Experimental points result from summing over ten laser pulses and smoothing on a small fraction ($\approx 10\%$) of the fringe period. The data bars measure the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

The fringe contrast as a function of the pulse delay is shown in Fig. 4; the data are reported with the statistical error while the solid curve superimposed on the data points is a theoretical curve obtained by assuming an essential-state model for the interaction [7]. The model parameters, i.e., the state energies, energy widths, ionization constants, and the Fano parameters, are the ones reported in the synchrotron measurements of Ref. [12]. In the simulation, we assume the harmonic field to be formed by a pair of Fourier-limited Gaussian pulses, with a FWHM duration of 20 fs as derived from the measured 0.51 nm linewidth of the single-pulse harmonic spectrum. As we are in the perturbative regime, no coupling is taken into account between the two states, which thus evolve independently following a two-state dynamic in the Fano approach. The theoretical ionization probabilities for the two states are then simply summed to give the total ionization signal, whose fringe contrast is superimposed to the data points in Fig. 4. It is important to note that only a single parameter, not known from literature, namely, the branching ratio between the two continuum $8s'$ and $6d'$ states along the direction of detection, has been varied in the fit. The reported curve corresponds to an effective branching ratio $\epsilon(8s)/\epsilon(6d) = 1.4 \pm 0.3$. The ripple in the fringe pattern is due to the energy difference between the two states which gives rise to a beating with a period of 143 fs; the comparison with the experimental data shows a fairly good agreement.

At a first glance, the use of 20 fs-duration pulses in the simulation can be surprising. The ionizing harmonic pulses are indeed dispersed by the grating, and are thus assimilable to a superposition of wave trains of frequency $\omega$, with

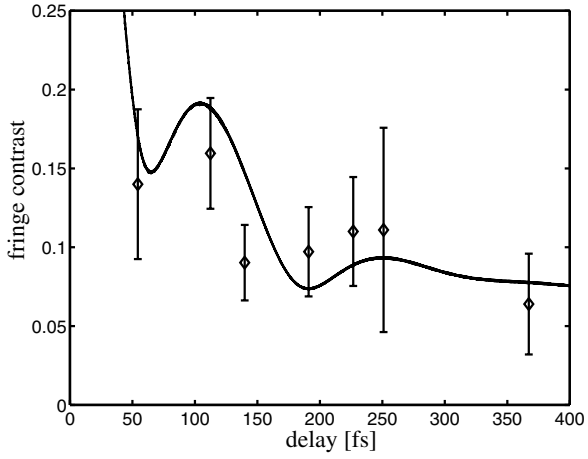


FIG. 4. Fringe contrast vs pulse delay. The solid curve superimposed to the data points is a theoretical curve, as explained in the text, where atomic parameters are obtained by the synchrotron radiation measurements of Ref. [12]. A single free parameter, corresponding to the ratio between the electron emission probabilities of the $8s'$ and $6d'$ states in the laser polarization direction, has been used in the fit.

wave vectors $k(\omega)$ slightly misaligned, and lasting for a time of the order of the inverse of the grating resolution, about 600 fs in our case. Anyhow, in our experimental conditions, where the photoelectrons due to each of these wave trains are collected with nearly the same efficiency, the ionization process is linear in the field intensity, and the dispersion $dk/d\omega$ is nearly linear in $\omega$, the collection average cancels the effect of the grating. As a result, the ionization due to the atom collection in the dispersed field is equivalent to that given by a single atom interacting with the undispersed short pulses.

In conclusion, we have tested the use of a Ramsey-type spectroscopic technique in conjunction with high-order harmonic radiation pulses. The results of the experiment, performed on krypton atoms in resonance condition with a couple of autoionizing states, show a clear quantum interference in the ionization signal versus the delay between the harmonic pulses. The fringe contrast behavior compares favorably with a simple essential-state model, whose parameters have been taken from the literature. We believe that the proposed spectroscopic tool, based on a relatively simple tabletop equipment, could become an attractive alternative to synchrotron radiation sources for selected applications in the XUV region.

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