

Method for High-Resolution Frequency Measurements in the Extreme Ultraviolet Regime: Random-Sampling Ramsey Spectroscopy

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Ramsey-like schemes have been recently introduced in combination with high-order laser harmonic sources for high-resolution spectroscopic studies in the extreme ultraviolet (XUV). Here we demonstrate a novel method, combining measurements only in a limited subset of randomly chosen time-sampling intervals, which lead us to perform the first high-resolution XUV spectroscopy of atomic argon with a simple split-pulse setup. Providing an experimentally simple and convenient solution to the problem of performing high-resolution absolute frequency measurements in the XUV, our approach will help paving new roads into this challenging spectral territory.

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High-order laser harmonics are a convenient source of coherent radiation in the extreme ultraviolet regions of the electromagnetic spectrum, where investigations of simple atomic species and highly developed calculations promise new accurate tests of bound-state electrodynamics [1]. The use of high-order laser harmonics as a spectroscopic tool in the extreme ultraviolet (XUV) has been recently proven possible by means of Ramsey-like techniques [2–7], based on the idea first introduced by Ramsey in 1950 [8] and relying on the extension of the effective interrogation time by using pairs (or sequences) of phase-coherent [9–11], time-delayed XUV ultrashort excitation pulses [12–15]. Low-order laser harmonics [16,17] and broadband XUV pulses [18] have been also used in the same context. Just like in standard Fourier transform spectroscopy (FTS), the effective spectral resolution of these methods scales inversely with the maximum time delay between the exciting pulses, and the achievable resolving power corresponds to the number of optical periods contained in such a delay. This is closely related to the intuitive rule according to which, in order to increase the accuracy of a frequency measurement, one has to correspondingly increase the measuring time so as to count more and more oscillation periods.

An experiment performed on high-lying bound states of Argon [14] has recently demonstrated a potential resolving power higher than the best currently available in synchrotron facilities by measuring Ramsey quantum interferences in the excitation signal at delays larger than 100 ps. Indeed, if a Michelson-type interferometer is used to introduce the interpulse delay, and in the case of XUV radiation with wavelengths in the 30–100 nm range, just a few mm of mirror displacement are sufficient to obtain a resolving power larger than 10^5 . However, if accurate measurements of transition frequencies are to be performed, all the atomic quantum interference fringes have to be accurately

followed over a long time interval. While this is the norm for standard FTS in the visible and ir regions, following this approach for Ramsey spectroscopy in the XUV is far from straightforward and technically almost impossible. In fact, since these measurements are normally performed in an ion-electron counting regime, they are intrinsically very slow and one has to face long acquisition times that pose severe constraints on the overall system stability.

Here we demonstrate a novel method, theoretically proposed by our group [19], to overcome these constraints and make measurement times substantially shorter, by acquiring fringes only over a limited subset of randomly chosen delay intervals in the whole delay range T that is needed for a given target spectral resolution. By significantly reducing the experimental difficulty of XUV Ramsey-like spectroscopy, we are able to perform the first high-resolution absolute measurement of an XUV atomic transition frequency with a simple pulse-splitting interferometer. The method is nevertheless very general and can thus be advantageously exploited also in more sophisticated schemes and/or in other spectral regions.

A Ramsey-like experiment consists in exciting an atomic system by a first pulse and then probing the induced coherence with a second pulse delayed by t and with a fixed phase relation to the first pulse. Any observable related to the atomic excitation will exhibit Ramsey oscillations with a period given by the inverse of the atomic transition frequency. By scanning the delay t between successive excitations, the effects of such oscillations can be transferred to state populations and subsequently detected. It is worth examining in some detail a few different cases, assuming that we are dealing with a single resonant atomic transition of negligible linewidth compared to the inverse of the time delay. The simplest case is depicted in Fig. 1(a) where the atomic interference fringes are acquired while spanning the delay t in a single time window between t_0

and $t_0 + \delta t$. The square modulus of the Fourier transform (FT) of the Ramsey signal is a sinc^2 -shaped curve centered at the atomic transition frequency and with a spectral width $\approx 1/\delta t$ [right column of Fig. 1(a)]. If the Ramsey signal is acquired while the delay t of the second pulse is sequentially spanned in two δt -long windows starting at t_0 and $t_0 + T$ (T being accurately measured on the scale of the atomic oscillation period), the above spectral curve is further modulated by a \cos^2 term of period $1/T$ [see Fig. 1(b)]. This high-frequency modulation opens the possibility of high-resolution spectroscopy. However, if $T \gg \delta t$, many different spectral maxima are eligible as the real atomic frequency and the identification is highly ambiguous. This ambiguity can be solved by sampling the Ramsey fringes while scanning the delay of the second pulse in several time windows of width δt , either regularly spaced, or with a random sampling of the delay intervals. It can be shown (see [19]) that the random approach has various advantages in this sense, as different sinusoidal modulations with incommensurable periods cancel the

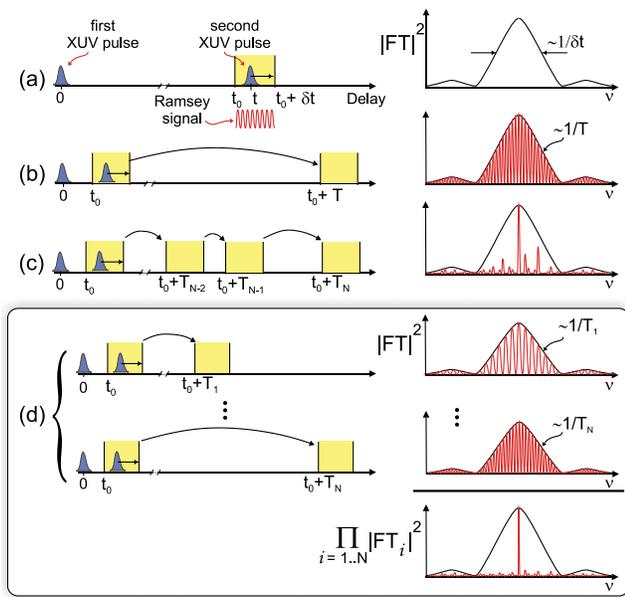


FIG. 1 (color online). Schemes of different options for scanning the delay (left panels) in a Ramsey spectroscopy experiment with short excitation pulses, and corresponding retrieved spectra (right panels). (a) The acquisition of Ramsey fringes in a single time window of width δt provides a spectral resolution proportional to $1/\delta t$. (b) Measuring the Ramsey signal in two short time windows delayed by T provides a potential spectral resolution of $\approx 1/T$ but may lead to ambiguities in the peak determination. (c) A sequence of N randomly spaced Ramsey acquisitions results in an effective cancellation of the satellite peaks and leads to the correct identification of the transition frequency in a fraction of the acquisition time. (d) Finally, a series of two-time window Ramsey acquisitions at random delays T_i 's can be combined by taking the product of the individual spectra.

satellite comb peaks [see Fig. 1(c)], leading to a substantial reduction of the acquisition time. Moreover, the width of the surviving tooth, corresponding to the atomic resonance, approaches the theoretical resolution limit ($1/T$) also in this case.

In principle, this scheme imposes to scan and accurately measure the delay t over all the measurement windows. Anyhow, as far as one is concerned with a well-isolated atomic resonance, a remarkable experimental simplification can be obtained by just accurately monitoring the relative delay of distinct pairs of measurements. In other words, the basic two-time-window acquisition schematically depicted in Fig. 1(b) can be repeated several times for a set of random delays spanning the whole range up to T , as in Fig. 1(d). Then, the square moduli of the FTs corresponding to different pairs can be simply combined by taking their product. The explanation of this procedure is quite straightforward if we recall that the FT for any two-time-window measurement always has a peak at the resonance frequency, while the positions of other peaks depend on the delay; multiplying several spectra at different delays thus performs a sort of logical AND operation which allows only the central peak to survive. Combining the information acquired in different measurement runs further reduces the experimental complexity and opens interesting and realistic possibilities for widespread high-resolution spectroscopy in the XUV.

The experimental setup is sketched in Fig. 2. The single-photon XUV transition at 87 nm from the ground to the $3s^23p^5(2P_{1/2})3d$ excited level (having a lifetime of 3.2 ns) in atomic argon is investigated. The upper level lies close to the first ionization threshold of the atom and excited electrons are promoted to the continuum by a further absorption of a single ir photon. The produced argon ions

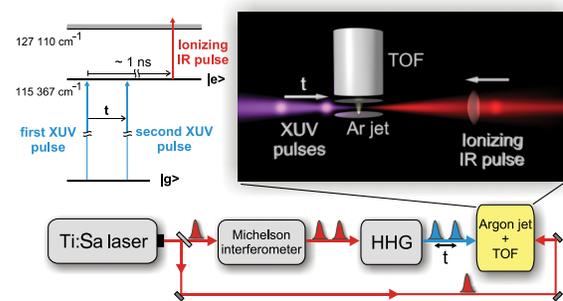


FIG. 2 (color online). Schematic view of the experimental setup and of the Ar level scheme. An ultrastable Michelson interferometer is used to split the ir pump pulses from the laser into two time-delayed replicas; HHG indicates the process of high-order harmonic generation and the selection of the ninth harmonic order at 87 nm; pairs of identical XUV pulses delayed by time t are thus produced and used to excite the Ar transition in a gas jet. Further excitation by a counterpropagating ir pulse ionizes the atoms and the produced ions are finally detected by a TOF spectrometer.

are detected by a time-of-flight (TOF) mass spectrometer; varying the delay between the first and the second XUV pulse results in evident oscillations in the ionization signal, as described in detail in [14]. The measurement procedure consists of different steps: first, the oscillating number of Ar^+ ions is recorded over a short (and slow) delay scan of about 5–8 fs (the δt time window), starting from $t_0 \approx 100$ fs; then the delay is rapidly swept by several hundreds of femtoseconds (T), while accurately monitoring it with subwavelength accuracy with the interferometric calibration signal from a copropagating cw He-Ne beam. Finally, starting at $t_0 + T$, another short scan δt of the delay is performed while recording the Ramsey interferences. The squared modulus of such a two-time-window Ramsey signal is shown at the bottom of the main plot in Fig. 3 for a representative value of the delay $T \approx 0.35$ ps. Unless precise preliminary spectroscopic data are available, it is difficult to identify the correct resonance frequency under the wide ($\approx 4000 \text{ cm}^{-1}$ full width at half maximum) sinc^2 shape (also shown in Fig. 3) corresponding to the average duration ($\delta t \approx 8$ fs) of the single-window Ramsey acquisition. The whole measurement process is then repeated several times for different delays T_i [see Fig. 1(d)]. The effect of increasing the number of random-delay acquisition pairs in different time spans ($\approx 0.5, 1,$ and 4 ps, respectively) is evident in the upper plots of Fig. 3. Increasing the number of terms in the product of spectra we obtain the expected progressive cancellation of the satellite peaks, which finally leads, in the top plot, to a single spectral peak corresponding to the atomic resonance. The error in the measured transition frequency is around 0.3 cm^{-1} compared to the NIST published frequency [20], corresponding to an accuracy of 3.8×10^5 . Its width (1.6 cm^{-1} full width at half maximum) is more than 3 orders of magnitude smaller than that of the sinc^2 curve of the single acquisition.

The advantage of the present approach is that this high spectral resolution is achieved by combining a very limited number ($N = 20$, in this case) of short (5–8 fs long) acquisition pairs at random but accurately-known relative delays, instead of performing a very demanding interferometrically calibrated single acquisition of Ramsey fringes over a long delay of several picoseconds. This corresponds to a reduction of the acquisition time by a factor of about 20, while preserving the final spectral resolution and with the fundamental experimental advantage of combining data from independent measurement runs. In the bottom inset of Fig. 3 a zoom of the frequency region around the resonance is shown for each of the 20 pairs of Ramsey measurements. After a few acquisitions the resonance peak is correctly identified and its width becomes progressively narrower as more pairs at longer relative delay are added. Errors in the absolute frequency determination are mainly due to the error in the accurate absolute measurements of delays. Currently, we estimate

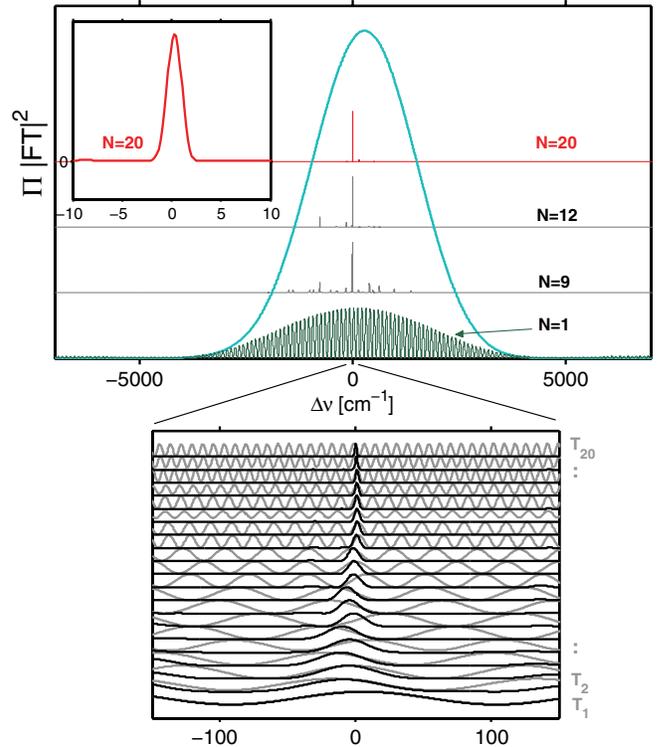


FIG. 3 (color online). Products of the $|FT|^2$ of N pairs of randomly sampled Ramsey acquisitions. The curves are vertically displaced for presentation purposes and the origin of the frequency axis is taken as the NIST frequency for the investigated transition [20]. From bottom to top, successive plots show that increasing the number N of randomly sampled Ramsey acquisition pairs leads to the progressive cancellation of the satellite spectral peaks and to the identification of the atomic resonance (plot $N = 20$, also shown in detail in the top inset). The wide sinc^2 profile corresponding to a mean of the squared FTs of single time window acquisitions is also shown in the main plot. The bottom inset shows a detail of the spectrum around the origin, where the $|FT|^2$ of each experimental Ramsey pair is plotted (after rescaling) and compared to the product of the previous ones (black lines). It is worth noting that the different Ramsey pairs were accumulated in experimental runs extending over several days.

an error of ≈ 10 nm on the Michelson path difference, which is nearly independent on the delay, due to the fact that there is no error in counting whole interference fringes from the reference He-Ne laser. For a delay of $T = 0.35$ ps this corresponds to a frequency error of $\approx 10^{-4}$ while, for the measurement at $T = 4.2$ ps, the error reduces to 8×10^{-6} . Thanks to the constancy of the absolute delay error, increasing the precision in absolute frequency measurements only involves the use of longer delays between the pairs of Ramsey acquisitions. Although scaling up the current performances to much higher spectral resolutions and accuracies will require moving to delays in the range of tens of nanoseconds (corresponding to meters of arm displacements), several solutions to the related technical

problems can be readily adopted in the line of those developed along the years for conventional FTS.

In conclusion, we have demonstrated high-resolution absolute frequency measurements of atomic transitions in the XUV by means of ultrashort high-order laser harmonics in a novel Ramsey spectroscopy scheme. The original idea at the base of our work, i.e., to make spectroscopy of an oscillating observable by sampling its status at random times, has important advantages: it avoids the introduction of the sampling frequency in the signal, and it keeps the measurement time orders of magnitude smaller than the full-sampling approach, while still reaching the maximum achievable resolution. In our experiment, this method led to an excellent frequency accuracy by using only very limited scans of the moving arm of a Michelson interferometer. In principle this method can also be applied to more sophisticated spectroscopic setups based on the use of frequency combs as a way to precisely control the delay between pairs of XUV pulses [15]. However, these schemes are intrinsically limited to states with lifetimes longer than the inverse of the comb repetition rate, or about 10 ns in the case of Ref. [15]. For shorter level lifetimes the approach to XUV Ramsey spectroscopy based on the Michelson pulse splitting is therefore the only viable solution. It should be noted that the recently attained up conversion of laser frequency combs [21–23] might represent an alternative to these Ramsey-like approaches for high-resolution XUV spectroscopy, as soon as an XUV comb structure is demonstrated. The comparison of our results to the performances of current synchrotron radiation sources is also striking. Here, a relatively simple tabletop apparatus and just a few mm of mirror displacement are sufficient to achieve the same resolution of the best available synchrotron monochromators [24].

We believe that the possibility of performing high-resolution XUV Ramsey spectroscopy with a relatively simple tabletop setup, and an efficient, and experimentally affordable, measurement scheme, will open new perspectives toward a more widespread application of high-order harmonic sources in a rich and almost unexplored spectral region.

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