PHYSICAL REVIEW A **83**, 041402(R) (2011)

Improving Ramsey spectroscopy in the extreme-ultraviolet region with a random-sampling approach

R. Eramo,[1,2] M. Bellini,[1,2] C. Corsi,[2] I. Liontos,[2] and S. Cavalieri[2,3]
[1]*Istituto Nazionale di Ottica (INO-CNR), Largo E. Fermi 6, I-50125 Florence, Italy*
[2]*European Laboratory for Non-linear Spectroscopy (LENS), I-50019 Sesto Fiorentino, Florence, Italy*
[3]*Department of Physics, University of Florence, I-50019 Sesto Fiorentino, Florence, Italy*
(Received 30 September 2010; published 15 April 2011)

Ramsey-like techniques, based on the coherent excitation of a sample by delayed and phase-correlated pulses, are promising tools for high-precision spectroscopic tests of QED in the extreme-ultraviolet (xuv) spectral region, but currently suffer experimental limitations related to long acquisition times and critical stability issues. Here we propose a random subsampling approach to Ramsey spectroscopy that, by allowing experimentalists to reach a given spectral resolution goal in a fraction of the usual acquisition time, leads to substantial improvements in high-resolution spectroscopy and may open the way to a widespread application of Ramsey-like techniques to precision measurements in the xuv spectral region.

DOI: 10.1103/PhysRevA.83.041402 PACS number(s): 32.80.Qk, 82.53.Kp, 32.30.Jc, 42.62.Eh

Ramsey-like experiments [1–4], probing the dynamical evolution of an atom between two interaction times, are a direct evolution of the method of separated oscillatory fields [5], with the main difference being that the excitation and the probing pulses are separated in time rather than in space. In brief, and for the simplest case (a two-level system with infinite lifetime), a first pulse induces a coherence oscillating at the transition frequency ν_o. The second pulse, delayed by t, can enhance or destroy the residual excitation, depending on the phase of the oscillations. As a result, any observable $f(t)$ (ionization, fluorescence) monitoring the residual excitation at the end of the second pulse exhibits interference fringes versus the delay t between the two pulses, with the resonance frequency appearing encoded, not just in the amplitude of the excitation signal as with conventional spectroscopy, but also in its phase. This experimental approach has been demonstrated to be very promising for high-resolution spectroscopy in the vacuum and extreme-ultraviolet (xuv) spectral regions by using low- [6,7] or high-order harmonics (HOH) [8–11] of femtosecond laser pulses. The fundamental interest in high-resolution xuv spectroscopy lies in its potential for a new generation of accurate tests of bound-state electrodynamics [12]. The main experimental difficulty in extending the Ramsey technique to the xuv spectral range is the generation of a pair of pulses with a precisely known phase difference. Two experimental approaches have been followed until now: one is based on the amplification to the millijoule level of a pair of subsequent pulses from an infrared (ir) frequency-comb (FC) laser with a double-pulse parametric amplifier [11,13]. The other approach [8] is to split the ir pulse from an amplified mode-locked laser in a Michelson interferometer. In both cases, the two ir pulses are then up-converted into the xuv spectral range, using HOH generation in a gaseous medium, and used for one-photon excitation of the transition under investigation. The two approaches appear complementary, as the FC setup is intrinsically limited to states with lifetimes longer than the comb repetition period, which is currently around 10 ns. Different from more traditional single-pulse spectroscopy with longer up-converted pulses strongly affected by chirp [14,15], the measurement of an xuv transition frequency using the Ramsey technique has the advantage of being essentially insensitive to the exciting pulse shape. This is because distortions of the pulses due to amplification or harmonic generation have no influence on the measurement, provided they are common to both the pumping and probing pulses. However, as expected on the basis of the time-frequency indetermination principle, a high-resolution Ramsey measurement requires one to follow the evolution of $f(t)$ for the longest measurable time span T, with an accuracy that is of the order of a fraction of the transition period. These constraints are particularly severe in the case of Ramsey spectroscopy in the xuv spectral range. In fact, the very short wavelengths involved imply an attosecond-scale measurement and control of the interpulse delays; and, even more importantly, a perfect long-term overall system stability is essential, since these measurements are intrinsically very slow, being performed in a charge-count regime that forces experimentalists to face very long acquisition times. Overcoming some of these constraints would open the way to a widespread application of Ramsey-like spectroscopy to new spectral regions. In particular, any approach aiming to shorten the acquisition time for a given measurement time span T, determined by the resolution goal, would be very appealing.

Here we propose a random subsampling methodology that holds promise for overcoming the above stability constraints, thus giving access to a new range of precision measurements in the xuv spectral range. The basic idea is that of substantially reducing the acquisition time by only sampling N random intervals of duration $\delta t \ll T$, instead of extending the measurement to the whole delay range T. In this way, the acquisition time $N\delta t$ can be orders of magnitude shorter, still preserving the same accuracy in frequency determination.

To formalize, let us assume that the measurement of the observable f is made in N time intervals of width δt, centered in t_m $(m = 1, \ldots, N)$,

$$F(t) = f(t)S(t), \tag{1}$$

where S is the sampling function, given by the sum of N rectangle functions,

$$S(t) = \sum_m [\theta(t - t_m - \delta t/2) - \theta(t - t_m + \delta t/2)], \tag{2}$$

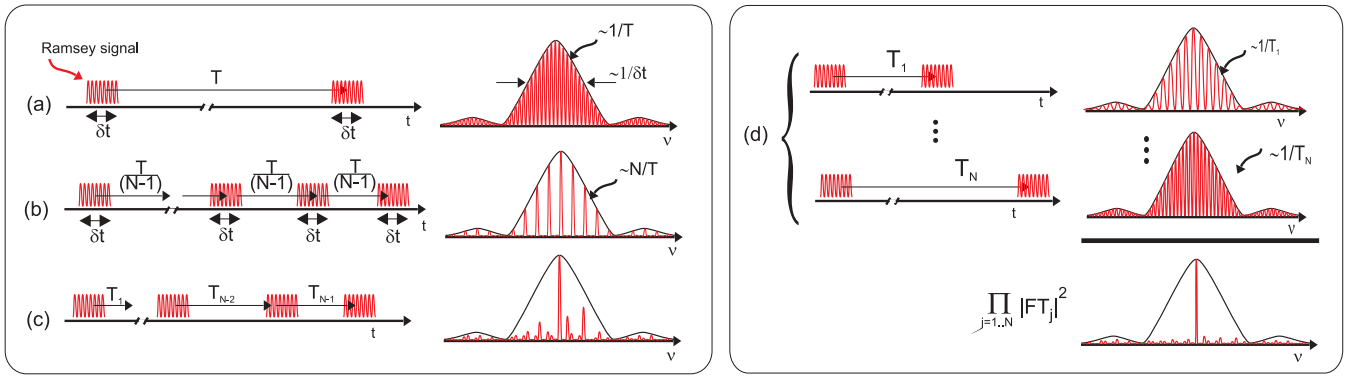

FIG. 1. (Color online) Delay scan schemes and corresponding retrieved spectra for a Ramsey spectroscopy experiment. (a) Two short-time windows of width δt delayed by T provide a potential spectral resolution of $\approx 1/T$, but lead to ambiguities in peak determination if $\delta t \ll T$. (b) N equally spaced acquisitions produce a comblike structure in the spectrum. (c) N randomly spaced acquisitions result in an effective cancellation of the satellite spectral peaks. (d) N two-time window acquisitions at random delays T_i can be combined by taking the product of the individual spectra. The schemes (c) and (d) are good candidates for experiments in xuv spectral regions.

with θ being the unitary step function. From Eq. (1), we obtain a Fourier transform as the convolution product between the Fourier transforms of the observable and of the sampling function, which is given by

$$\tilde{S}(\nu) = \delta t\, \mathrm{sinc}(\nu \delta t) Z(\nu), \tag{3}$$

$$Z(\nu) = \sum_m e^{-2\pi i t_m \nu}, \tag{4}$$

with $\mathrm{sinc}(t) = \sin(\pi t)/\pi t$ being the normalized sinc function. For the simplest monochromatic oscillator, $f(t) = e^{2\pi i \nu_o t}$, the convolution gives

$$\tilde{F}(\nu) = \tilde{S}(\Delta), \tag{5}$$

where $\Delta = \nu - \nu_o$. Thus the convolution kernel $\tilde{S}$ is the Fourier transform of the ideal monochromatic fringe signal with a shifted origin. For simplicity, in the following we will neglect the distinction between $\tilde{S}$ and $\tilde{F}$; however, it is tacitly understood that the results of our Rapid Communication concern the function $\tilde{S}$, which in a Fourier-transform spectroscopy context is known as *instrumental function* of the setup. In a real experiment, the simplest fringe signal can be modeled by $f(t) = e^{2\pi i \nu_o t} g(t) + \mathrm{c.c.}$, where $g = e^{i\phi(t)}$ is a stochastic function representing the sources of broadening and shifting of the resonance, whose control is the main task of the experimentalist.

The fact that $\tilde{S}$ is not a Dirac δ function in frequency space (as it would be for an acquisition on the whole temporal axis) means that the measuring procedure always introduces some measurement-related artifact in the final signal. The main results of this Rapid Communication concern the interferences between the terms contained in the exponential sum Z, leading to the possibility of performing spectroscopy with a resolution well below the sinc envelope limit, $\Delta_{\mathrm{fwhm}} = 0.886/\delta t$, where fwhm represents full width at half maximum.

Let us consider, for example, the case of two sampling intervals,

$$\tilde{F} = 2\delta t\, \mathrm{sinc}(\delta t \Delta) e^{-2\pi i \langle t \rangle \Delta} \cos(\pi T \Delta),$$

where $\langle t \rangle = [t_o + t_1]/2$ is the mean point of the sampled range, and $T = t_1 - t_o$. The envelope of $|\tilde{F}|^2$ is the single-sample result and corresponds to the sinc^2 behavior associated to the sampling window δt. The inner modulation $\cos^2(\pi T \Delta)$ is the result of the interference between the measurements in the two intervals and has a period $1/T$ [see Fig. 1(a)]. The fundamental characteristic of this function is that the envelope and the modulation are characterized by two independent frequency scales, $1/\delta t$ and $1/T$, respectively. For $T > \delta t$, the higher frequency of the modulation opens the possibility of high-resolution spectroscopy with wideband sampling intervals, and is at the base of Ramsey spectroscopy [5]. We observe that in the case of $T \gg \delta t$, the sinc envelope is nearly flat for the modulation peaks around the atomic resonance, leading to an experimental ambiguity for the resonance identification.

Now suppose that the sampling times are equally spaced, so that $t_m = t_o + m\delta t_s$, where $m = 0, \ldots, N-1$, and δt_s is the sampling interval, which is assumed to be greater than the common interval width δt, i.e., $\delta t_s \geqslant \delta t$. In this case, the N monochromatic terms in the interference sum have commensurable periods and a characteristic constructive recurrence peak appears each time they all get in phase, giving rise to a characteristic frequency comb [see Fig. 1(b)], extending over a frequency range $\approx 1/\delta t$ around the atomic frequency, with a tooth spectral separation given by the sampling frequency $1/\delta t_s$,

$$\tilde{F} = \delta t\, \mathrm{sinc}(\delta t \Delta) e^{-2\pi i \langle t \rangle \Delta} \frac{\sin \pi T \Delta}{\sin \pi \delta T \Delta / N},$$

where $T = N\delta t_s$, and $\langle t \rangle = [t_o + t_{N-1}]/2$. It is easy to show that in order to avoid the peak ambiguity, one should have $N\delta t \approx T$.[1]

Let us now turn to the case of a random sampling. If we want to sample the signal in N intervals randomly selected

[1]In order to observe the recurrence peaks, the width of the sinc term must be at least as large as this period: $1/\delta t > 2/\delta t_s$, i. e., $\delta t/\delta t_s = \delta t N/T < 1/2$. The ambiguity arises in the opposite limit, when $\delta t/\delta t_s \ll 1$, and one gets the order–of–magnitude condition $\delta t \approx \delta t_s = T/N$. Thus, the sampled range cannot be too sparse, i.e., the subsampling approach in this case cannot reduce significantly the acquisition time.

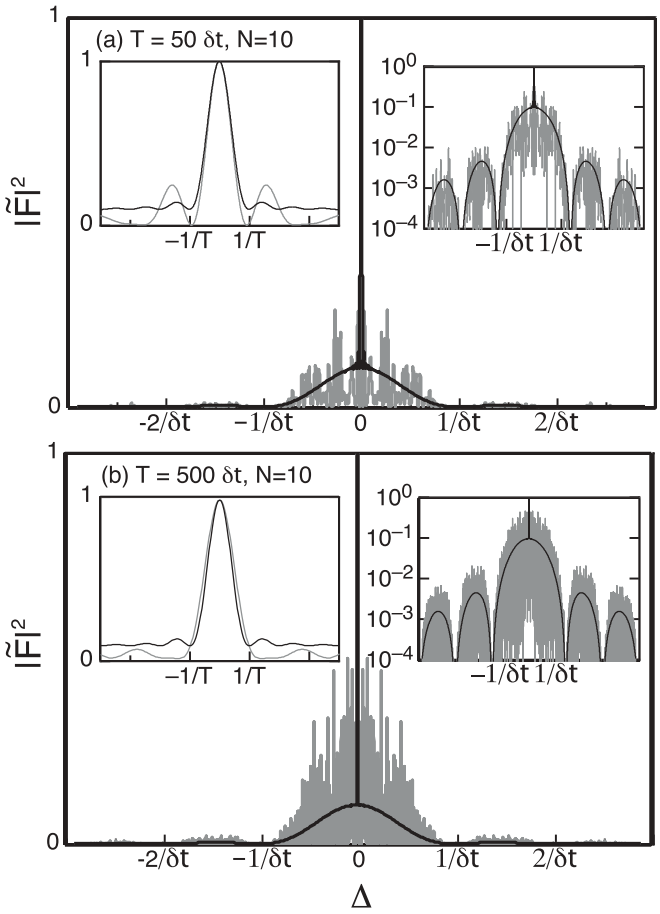

FIG. 2. Squared magnitude of $\tilde{F}$ for a random sampling in the condition (a) $T \gg \delta t$ ($N = 10, T = 50\,\delta t$, gray curve), and (b) $T \gg N\delta t$ ($N = 10, T = 500\,\delta t$, gray curve). The mean value, given by Eq. (6), is also shown (in black). In the inset on the left, a zoom around the resonance frequency shows that the random extraction is well represented around the origin by the asymptotic mean value, reaching the theoretical maximum resolution. The resonance frequency is unambiguously determined also in the case of sparse sampling. The semilog plots in the right insets show that the mean value is also indicative of the behavior in the wings of the curve.

in the range $[t_{\min}, t_{\max}]$, then the function $Z(\Delta)$ will show a complex pattern in which details depend on the particular random extraction [see Fig. 1(c)]. Nevertheless, the main peak of this function is at zero frequency and its width is $\approx 1/T$, with $T = t_{\max} - t_{\min}$ being the explored range.

This can be demonstrated by considering the interval centers as independent random variables, with a flat probability distribution, and taking the mean value. The result is

$$\langle |\tilde{F}|^2 \rangle = \delta t^2 \,\mathrm{sinc}^2(\delta t\,\Delta)\{N + N(N-1)\mathrm{sinc}^2(T\,\Delta)\}. \quad (6)$$

The linear term in N is the incoherent part, corresponding to a frequency flat background, whereas the quadratic term is the coherent interference peak and has the width corresponding to the whole explored range, $\Delta_{\mathrm{fwhm}} = 0.886/T$; this value determines the resolving power of the method. In Fig. 2(a), we present a plot for a random sampling in the condition

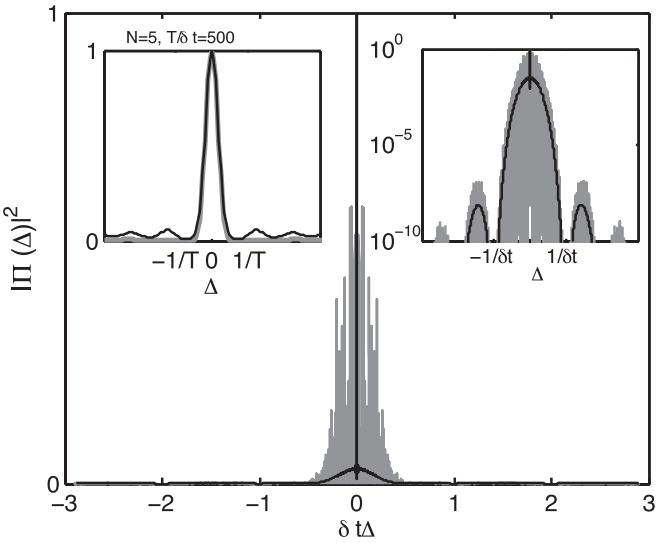

FIG. 3. Plot of the squared magnitude of the product of the Fourier transform of N couples [Eq. (7)] at random delays, and of the mean value [Eq. (8)]. Here $N = 5$ and $T = 500\delta t$. In the inset on the left, the mean value is barely discernible from the random extraction.

$T \gg \delta t$, showing that while in the wings Eq. (6) is just a mean-value representation of the frequency spectrum, the random extraction is quite stable around the frequency origin where the coherent part prevails, and this happens also with a limited number of samples ($N = 10$ in the examples).

Therefore, the resolution limit obtainable by a measurement in the complete delay range can be effectively reached also with the random-sampling approach. Going back to Eq. (4), it is evident that for a regular sampling, the measurement-induced artifacts given by $\tilde{S}$ result in a beat note at the sampling frequency, which appears in the spectrum together with its harmonics. For a random sampling, on the other hand, the introduced artifact is a noise that, once averaged, is spectrally flat and with infinite bandwidth. The advantage with respect to the equally spaced sampling case becomes particularly evident in the case of sparse sampling ($T \gg N\delta t$), when the resonance assignment can still be done, as clearly shown in Fig. 2(b). It is worth noting here that, with a much smaller acquisition time, we obtain the same resolution of a full sampling of the range T: the speed-up factor is about 50 in this example.

From an experimental point of view, the above results for the random-sampling case require that all the measurements are made in a phase-coherent sequence, with a time resolution smaller than the observable oscillation period. This can be achieved by using a reference laser interferometer monitor in the pulse-splitting approach, or by using frequency combs [6,11]. Ideally, in both approaches, the reference laser, assumed to be related to a standard of frequency, is thus translated to the spectral region under investigation. This is in a complete analogy with frequency-comb spectroscopy, where the standard of frequency is transferred from the radio domain to the optical region.

While the scheme of Fig. 1(c) is certainly feasible, a great advantage to the experimentalist would consist of relaxing the assumption of complete coherence among the samples and just measuring the delay T_i between pairs of them [see

Fig. 1(d)]. In this case, for each pair we have a Fourier transform proportional to

$$\tilde{F}_i(\Delta) = \mathrm{sinc}(\delta t\,\Delta)\cos(\pi T_i \Delta).$$

By measuring N pairs and taking the product, we have

$$\Pi(\Delta) = \Pi_i \tilde{F}_i(\Delta) = \mathrm{sinc}^N(\delta t\,\Delta)\Pi_i \cos(\pi T_i \Delta). \quad (7)$$

Considering T_i as independent random variables, uniformly spread in the interval between 0 and T, and calculating the cosine product average, we obtain

$$\langle|\Pi(\Delta)|^2\rangle = \mathrm{sinc}^{2N}(\delta t\,\Delta)\left[\frac{1+\mathrm{sinc}(2T\Delta)}{2}\right]^N. \quad (8)$$

Figure 3 shows a plot of the square of Eq. (7) for the case $N\delta t \ll T$, together with the mean-value estimate, given by Eq. (8): the random extraction is well represented both around the origin and in the wings by the asymptotic mean value, even with extremely sparse sampling ($N\delta t = T/100$, the speed-up factor is thus about 100 in the example). Due to the power function, this nonlinear resonance appears narrower than $1/T$, and a numerical calculation gives $\Delta_{\mathrm{fwhm}} \approx 0.92/T\sqrt{N}$. This width should be interpreted as the error on the oscillator frequency measurement and, as expected for the standard deviation of a mean value, it scales as $1/\sqrt{N}$. It should not be confused with the resolution of the method, i. e., its capability to resolve two oscillators with the same amplitude and different frequencies, which is given in this case by $1/\delta t$. Therefore, while this combination of N two-time window acquisitions is very effective in determining the absolute frequency of isolated resonances, it is only able to distinguish lines with frequency separations larger than $1/\delta t$. On the other hand, it should be noted that the results for the more experimentally demanding, randomly sampled coherent sequence are also valid for more than one oscillator, provided that the entire series of random delays is accurately known.

In conclusion, we have shown that a random subsampling approach can bring unexpected advantages in the context of Ramsey-like or Fourier-transform spectroscopy. This concept is particularly useful in the sparse-sampling conditions by allowing one to overcome the ambiguities in the peak frequency identification with a substantial reduction of the measurement time, opening a realistic possibility of very-high-resolution measurement in the xuv region. We have also demonstrated that significant experimental advantages can be obtained by simply combining multiple measurements of different phase-correlated acquisition pairs [Fig. 1(d)]. A narrow and accurate frequency measurement is obtained in this case even without interferometric coherence among all acquisitions, which permits the progressive accumulation of data even across different measurement runs. We believe that this approach can be of great relevance for high-resolution absolute-frequency measurements in situations where conventional techniques suffer great limitations. Furthermore, we think that the use of Ramsey-like techniques to the spectroscopy in the extreme-ultraviolet spectral region in connection with high-order harmonic sources is just one of such situations. An experimental application of this method is currently in progress.

[1] M. M. Salour, Rev. Mod. Phys. **50**, 667 (1978).
[2] R. R. Jones, C. S. Raman, D. W. Schumacher, and P. H. Bucksbaum, Phys. Rev. Lett. **71**, 2575 (1993).
[3] M. Bellini, A. Bartoli, and T. W. Hänsch, Opt. Lett. **22**, 540 (1997).
[4] S. Cavalieri and R. Eramo, Phys. Rev. A **58**, R4263 (1998).
[5] N. Ramsey, Phys. Rev. **76**, 996 (1949).
[6] R. T. Z. S. Witt, W. Ubachs, W. Hogervorst, and K. S. E. Eikema, Science **307**, 400 (2005).
[7] R. T. Zinkstok, S. Witte, W. Ubachs, W. Hogervorst, and K. S. E. Eikema, Phys. Rev. A **73**, 061801 (2006).
[8] S. Cavalieri, R. Eramo, M. Materazzi, C. Corsi, and M. Bellini, Phys. Rev. Lett. **89**, 133002 (2002).
[9] A. Pirri, E. Sali, C. Corsi, M. Bellini, S. Cavalieri, and R. Eramo, Phys. Rev. A **78**, 043410 (2008).
[10] I. Liontos, S. Cavalieri, C. Corsi, R. Eramo, S. Kaziannis, A. Pirri, E. Sali, and M. Bellini, Opt. Lett. **35**, 832 (2010).
[11] D. Z. Kandula, C. Gohle, T. J. Pinkert, W. Ubachs, and K. S. E. Eikema, Phys. Rev. Lett. **105**, 063001 (2010).
[12] M. Herrmann *et al.*, Phys. Rev. A **79**, 052505 (2009).
[13] D. Z. Kandula, A. Renault, C. Gohle, A. L. Wolf, S. Witte, W. Hogervorst, W. Ubachs, and K. S. E. Eikema, Opt. Express **16**, 7071 (2008).
[14] K. S. E. Eikema, W. Ubachs, W. Vassen, and W. Hogervorst, Phys. Rev. A **55**, 1866 (1997).
[15] S. Bergeson *et al.*, Phys. Rev. Lett. **80**, 3475 (1998).