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## Self-calibrating method for measuring local multiphoton-ionization yields as a function of absolute intensity

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Very recently, Walker, Hansch, and van Woerkom [1] described a method for measuring multiphoton-ionization (MPI) ion yields resolved for intensity. In their method, the experimental problem of spatial averaging is circumvented by detecting only a slice of the focal region, and applying a deconvolution technique to the measured signal, making certain assumptions on the laser beam profile that limit the applicability of their method. In this Brief Report we will briefly describe a more elegant method, that does not try to “circumvent” the problem of spatial averaging in MPI experiments, but simply eliminates it, and is of general applicability.

In general, the ion (or electron) yield signal measured in pulsed-laser high-intensity MPI experiments is in fact a spatiotemporal average of particle yields over the source volume of the spectrometer that is used and the specific temporal pulse shape of the laser. Although the high order of many processes that are studied may help to reduce the effects of spatial averaging in specific cases, there is a fundamental experimental problem facing us here. In principle, one ideally would like to measure ion yields as a function of peak intensity for a given pulse shape, without any spatial averaging. As clearly explained by Lambropoulos [2], temporal effects (i.e., caused by the rising and falling of the laser pulse during its temporal evolution) cannot be neglected in high-intensity experiments. However, temporal effects are interesting in the sense that the dynamics of the system is in-

involved. Spatial intensity variations, on the other hand, just lead to blurring of the intensity dependence of the signal to be measured, and tend to obscure the underlying physics. In mathematical terms, when the source volume of the ion spectrometer is unrestricted (i.e., its typical dimensions are much larger than the typical structures in the spatial beam profile), the experimentally measured ion signal  $N_{\text{expt}}^{\text{unr}}(E)$  as a function of total laser pulse energy  $E$  is given by the spatiotemporal integral

$$N_{\text{expt}}^{\text{unr}}(E) \propto \frac{1}{\Delta V} \int \int \int_{\text{all space}} d\mathbf{r} \int_{-\infty}^{\infty} dt \eta(\mathbf{r}) R(I(E, \mathbf{r}, t)), \quad (1)$$

where  $R(I)$  (the quantity of main interest here) is the ion rate as a function of intensity,  $\eta(\mathbf{r})$  is the local collection efficiency of the spectrometer [ $0 \leq \eta(\mathbf{r}) \leq 1$ ], and  $\Delta V = \int \int \int d\mathbf{r} \eta(\mathbf{r})$  is the effective volume of the region in space the spectrometer accepts particles from (the source volume). In Eq. (1), we have explicitly written the dependence of the intensity on space and time. As Eq. (1) shows, ions are collected from all points in space  $\mathbf{r}$  where  $\eta(\mathbf{r}) \neq 0$ , and a corresponding range of intensities  $I(E, \mathbf{r}, t)$  contributes to  $N_{\text{expt}}^{\text{unr}}(E)$ . For an unrestricted source volume, this range of contributing intensities is so large that the intensity dependence of  $R(I)$  is “smeared out.” Furthermore, the relative contribution of a specific intensity interval depends on the spatial volume where this intensity range is found. This is what is generally called the volume effect. In this Brief Report we will briefly describe a method that enables us (i) to detect particles from a volume in space with fixed and

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known dimensions that is so small that  $\eta(\mathbf{r})$  can be effectively replaced by a  $\delta$  function located at a specific point  $\mathbf{r}_0$  in space; (ii) to position this point  $\mathbf{r}_0$  at any desired position in space; and (iii) to measure the peak intensity at this point in space [i.e., the absolute maximum  $I_0(E, \mathbf{r}_0)$  of  $I(E, \mathbf{r}_0, t)$  for a given pulse energy  $E$ ] in an absolute and unambiguous way. Using this method, we immediately obtain the intensity dependence of MPI ion yields by simply moving  $\mathbf{r}_0$  through the focal volume. The authors of Ref. [1] demonstrate that their method can be applied for a Gaussian beam profile. However, the feasibility of their deconvolution method depends very critically on the intensity distribution, and thus their method has a rather limited applicability in practice. Our method, on the other hand, does not rely on (possibly speculative) assumptions on the beam profile inside the focal volume, nor does it need (possibly cumbersome) mathematical procedures based on such assumptions to be invoked. On the contrary, the method we propose even allows detailed analysis of the intensity distribution in the focal volume and thus is an important tool for beam diagnostics. Because in our so-called confined-volume method the source volume of the spectrometer has fixed and known small dimensions ( $\sim 2 \mu\text{m}$  in the critical direction has been achieved [3]), it does not suffer from any volume effect. The ion yields measured with this method are given by the time integral

$$N_{\text{expt}}^{\text{conf}}(E, \mathbf{r}_0) \propto \int_{-\infty}^{\infty} dt R(I_0(E, \mathbf{r}_0) F(t)), \quad (2)$$

where we have explicitly factorized out the temporal profile of the laser pulse  $F(t)$ , normalized according to  $0 \leq F(t) \leq 1$ . The peak intensity can be varied by varying  $E$  and/or  $\mathbf{r}_0$ . However, to vary the laser energy  $E$  one typically would put some filter in the laser beam, and this may lead to unwanted changes in the intensity distribution. Therefore variation of  $\mathbf{r}_0$  is the preferred way to vary intensity. Briefly, the confined detection volume of the five-grid spectrometer [4] is realized as follows. A laser beam passes between the electrically grounded entrance slit of the spectrometer and a repeller at kV voltage at a few mm distance. The entrance slit geometrically cuts off the detection volume in two dimensions. In the third direction, parallel to the spectrometer axis, ions are labeled by the electrostatic equipotential surface they are created on, and the kinetic energy they have when entering the grounded spectrometer entrance has a one-to-one relation to their initial position. The five-grid spectrometer accepts ions only from a limited region in space by acting as an energy bandpass filter. By changing the potentials of the five grids, the characteristics of this bandpass filter are changed, and thus the detection volume is moved in space. The feasibility of our method is illustrated by Fig. 1, where we show the spatial distribution of  $\text{H}_2\text{O}^+$  ions (circles) inside the focus of a 170-fs Ti:sapphire laser beam ( $\lambda = 790 \text{ nm}$ ). In these measurements, the confined volume was  $400 \mu\text{m}$  wide along the propagation direction of the laser, and only  $\sim 20 \mu\text{m}$  wide in both other (critical) directions [3]. The beam waist with a diameter of about  $170 \mu\text{m}$  was scanned by moving the confined volume in steps of typically 5 to  $10 \mu\text{m}$ . The differences between the left and the right wing show that the beam profile is particularly asymmetric, and for this case a deconvolution method would be unrealis-

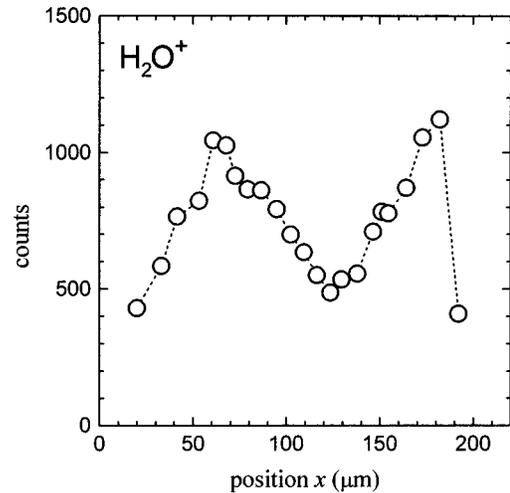


FIG. 1. Dependence of the  $\text{H}_2\text{O}^+$  yield on the radial position  $x$  of the confined volume inside the beam waist of a Ti:sapphire laser ( $\lambda = 790 \text{ nm}$ , pulse duration 170 fs). The intensity in the center ( $x \approx 125 \mu\text{m}$ ) is of the order of  $10^{16} \text{ W cm}^{-2}$ . The confined volume was moved through the focus in steps of typically 5 to  $10 \mu\text{m}$ .

tic. The reduction of the  $\text{H}_2\text{O}^+$  yield in the center, where the intensity is about  $10^{16} \text{ W cm}^{-2}$  (estimated by conventional methods) is caused by photodissociation.

To measure absolute intensities inside the focal region, the five-grid spectrometer only needs to be extended by a very simple time-of-flight electron spectrometer. Using a suitable gas, such as He, one can deduce the intensity at any position in space by performing electron-ion coincidence measurements. In that case, a repeller with a slit must be used, to allow electrons to enter the electron spectrometer, and the repeller voltage must be pulsed, so that the electrons can drift away under field-free conditions. A very slow 1-eV electron would travel the typical distance of 1 mm between the laser focus and the repeller in 1.7 ns, so a delay of a few ns between the laser pulse and the voltage pulse on the repeller is sufficient. The He ions, on the other hand, have thermal velocities of about  $1.1 \mu\text{m ns}^{-1}$  at room temperature, so they are essentially frozen in space during this ns delay, and the confined volume remains well defined. In this way, a kinetic energy spectrum can be recorded for electrons originating exclusively from the confined volume. As was demonstrated in Ref. [5], the redshift in a He above-threshold ionization (ATI) spectrum is proportional to the peak intensity of the laser pulse for intensities up to about the saturation intensity of  $(6.9 \pm 0.8) \times 10^{14} \text{ W cm}^{-2}$  (at 248.6 nm). This redshift is caused by the ponderomotive shift  $U_p$  of the ionization threshold given (in practical units) by

$$U_p(\text{eV}) = 93.4[\lambda(\mu\text{m})]^2 I(10^{15} \text{ W cm}^{-2}), \quad (3)$$

where  $\lambda$  is the wavelength and  $I$  the intensity. Thus, for every position  $\mathbf{r}_0$  of the confined volume, a recorded coincident He ATI spectrum will immediately reveal the absolute intensity. In practice, the He pressure must be optimized for maximum signal-to-noise ratio, and one has to correct for random coincidences, e.g., by correlating electrons from one laser pulse with ions from the next one. However, space does not permit a detailed discussion of such practical aspects, and we want

to stress the generality of the proposed method. Anyhow, the intensity measurements can be done once and for all, and only need to be repeated when the laser profile is changed. Since the ion yield measurements with a moving confined volume do not require the laser beam to be manipulated, this profile remains unaffected.

In summary, we have proposed a self-calibrating method for measuring MPI ion yields as a function of absolute intensity, that is free of volume effects, and is intensity re-

solved. The method is generally applicable, and does not require a specific laser profile. It is based on concepts that have already proved their applicability and reliability in the past. As a spinoff, our method allows absolute intensity distributions inside a beam waist to be measured with  $\mu\text{m}$  resolution, and thus is a valuable tool for laser beam diagnostics.

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