Laser-peak-intensity calibration using recoil-ion momentum imaging

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We exploit a method to precisely calibrate the peak intensity of intense laser pulses by measuring the momentum transferred to the ion in single ionization by circularly polarized light. Using this approach, we show how the experimental data can be used to get information about the absolute ionization rate at a specific local laser-peak intensity within the focal volume. The results of this method are compared to other results obtained with different methods of laser-peak-intensity calibration described in the literature.

I. INTRODUCTION

Strong field ionization of atoms and molecules has been thoroughly investigated in recent years both experimentally and theoretically. Extensive studies dealing with ionization, dissociation, and fragmentation of atoms and molecules in laser fields lead to a better understanding of the dynamics for the field-dressed atomic and molecular states. Several theoretical models have been developed as an effort to fully understand the behavior of atoms and molecules when exposed to the laser field. In addition to the laser pulse duration, the value of the laser peak intensity stands as a crucial parameter when applying these models to describe and interpret the experimental observations. Different methods such as normalizing the experimental yields to (ADK) calculations or calculating the laser intensity from the laser's parameters have been widely used to calibrate the laser intensity and compare experimental findings with theory. However, these methods are known to have large errors in determining the laser intensity. In this paper we use a method based on recoil momentum measurements which is both accurate and "*in situ*" in determining the laser peak intensity.

An additional use of our approach would allow us to measure the local ionization rate without having to integrate over the focal volume of the laser. Recent experiments have shown that in focused laser beams some physical observations could be hidden due to the spatial intensity variation that cannot be resolved when integrating over the entire focal volume [1,2]. Several efforts have been made to overcome these volume effects. One major technique is to use a small aperture in front of a time-of-flight (TOF) or mass spectrometer of smaller dimension than the Rayleigh range such that the collection of ions or electrons is limited to a slice orthogonal to the laser propagation direction [3–5]. This restriction on the spatial variation of the intensity was helpful in observing several phenomena in multiphoton ionization of atoms such as charge-state depletion and coherence effects in resonant ionization [1–5]. Nonetheless, measurements of the absolute ionization rates independent of the volume effects are still rare. A valuable application of the recoil momentum method described here would be to extract information about the local experimental absolute ionization rate. Our method would avoid the necessity to do a volume integral.

In this work we use an experimental approach supported by a simple analytical treatment to study the dependence of the ionization yield on the laser-peak intensity. We used cold target recoil-ion momentum spectroscopy (COLTRIMS) to produce high-resolution momentum measurements for the products of the strong field ionization reaction. We *in situ* calibrate the laser peak intensity by measuring the drift momentum that an ion gains when ionized by circularly polarized light. Unlike the ionization by linearly polarized light, where the value of the phase at the time of ionization is essential to determine the laser peak field [6], in circularly polarized light the size of the momentum of the recoil ion produced in the single ionization process provides a direct measurement of the laser peak field independent of the phase value at the time of ionization [7,8]. The method we use was, to our knowledge, proposed and used by Litvinyuk *et al.* [8]. In this paper we focus on how to establish an accurate laser peak intensity scale. Measurements of absolute ionization rate using this approach will be presented in an upcoming publication.

II. ANALYTICAL APPROACH: DISTRIBUTION OF DRIFT MOMENTUM

The temporal-spatial intensity distribution of a pulsed laser can be approximately expressed by a Gaussian beam as

$$I = \frac{I_0 w_0^2}{w^2(z)} e^{(-r^2/w^2)} e^{(-4\ln 2)t^2/\tau^2}. \tag{1}$$

Here we choose cylindrical coordinates with z is along the laser beam direction. The laser is circularly polarized in the x-y plane. I_0 is the peak intensity, and w_0 is the waist size. The z-dependence of the waist is given by $w(z) = w_0\sqrt{(1+z^2/z_0^2)}$, where z_0 is the Rayleigh range given by $z_0 = \pi w_0^2/\lambda$ and τ is the laser pulse duration. The volume encompassed at or below a given laser intensity I at time t can be obtained from Eq. (1) as [9,10]

$$V = \pi z_0 w_0^2 \left[\frac{4}{3} \left(\frac{I_0 f(t) - I}{I} \right)^{1/2} + \frac{2}{9} \left(\frac{I_0 f(t) - I}{I} \right)^{3/2} \right.$$
$$\left. - \frac{4}{3} \tan^{-1} \left(\frac{I_0 f(t) - I}{I} \right)^{1/2} \right], \tag{2}$$

with

$$f(t) = e^{(-4 \ln 2)t^2/\tau^2}. \tag{3}$$

Now the yield of singly charged ions per laser pulse N can be written as

$$N = \eta n_g \int W(I) dV dt = \eta n_g \int W(I) \frac{dV}{dI} dt dI. \tag{4}$$

Here n_g is the gas target density, η is the detector efficiency, and $W(I)$ is the ionization rate. Since the laser intensity we are interested in here is below the saturation intensity, we ignore depletion.

Doing the time integral first, we define a function $F(I)$ as

$$F(I) = \int \frac{dV}{dI} dt = -\int \frac{\pi z_o w_o^2}{3I^{5/2}} [2I + I_o f(t)] \sqrt{I_o f(t) - I} dt \tag{5}$$

the ionization yield at given laser intensity can then be expressed as

$$\frac{dN}{dI} = \eta n_g W(I) F(I). \tag{6}$$

The expression obtained for $F(I)$ in Eq. (5) assumes a Gaussian temporal-and spatial-distribution, but this analysis will still hold for any other distribution that could be observed experimentally when diagnozing the optical properties of the laser beam.

What can be directly measured in the experiment is dN/dp^2 (or, equivalently, dN/dp) where p is the drift momentum acquired by the ion (electron), and so it is important to express the yield as a function of drift momentum in order to compare with the experiment.

When circularly polarized light is used, there is a one-to-one correspondence between the momentum p with which the ion emerges from the interaction and the local peak intensity I at which the ion was created. This relationship is $p = E/\omega$ or $I = (p\omega)^2$, where E and ω are the electric field amplitude and the frequency of the circularly polarized light, respectively. This result comes from the classical equation of motion for an electron released at rest in circularly polarized light of peak intensity I [7,8].

The relationship between the observed momentum spectrum dN/dp^2 evaluated at some momentum p, and the absolute value of the local single ionization rate $W(I)$, evaluated at the corresponding value of I, can be obtained as follows:

After substituting $(p\omega)^2$ for I in Eq. (6), we get

$$\frac{dN}{dp^2} = \omega^2 \eta n_g W(I) F(\omega^2 p^2) \text{ or} \tag{7}$$

$$W(I) = \frac{dN}{dp^2} \frac{1}{\omega^2 n_g \eta F(\omega^2 p^2)} \tag{8}$$

in terms of

$$p, \ F(\omega^2 p^2) = -\int \frac{\pi z_0 w_0^2}{3\omega^2 p^5} [2p^2 + p_0^2 f(t)] \sqrt{p_0^2 f(t) - p^2} dt. \tag{9}$$

This expression can be evaluated numerically as a function of p for any p_0 and $f(t)$. From the measured dN/dp^2, the ionization rate can be directly obtained without any volume integration. The momentum spectrum is directly related to the ionization rate and the value of drift momentum is directly related to the laser-peak intensity. By conducting the experiment with a single I_0, one can get the ionization rate over a range of intensities. In this paper we do not exploit this possibility, but only point out that it could be done.

III. EXPERIMENT

We have used the COLTRIMS technique [11,12] to measure the momentum of H_2 molecules when ionized by linear and circular polarized light over a wide range of laser intensity. The Ti:sapphire laser system produces 3 mJ, 25 fs pulses at 1 kHz repetition rate. A linear optical density filter was used to control the power going into the chamber. The laser was focused by a 10 cm focal length spherical mirror onto a tightly collimated supersonic jet. The ions were extracted onto a time-and position-sensitive channel plate detector by a uniform electric field which was varied between 1 and 30 V/cm. The channel plate detector was followed by a multihit anode to measure the position of the extracted ions. From the times-of-flight and the positions, the full momenta of these ions were obtained. In the case of linear polarization, the direction of the laser polarization was set by an achromatic half-wave plate parallel to the extraction field of the spectrometer and perpendicular to the jet direction. An achromatic quarter-wave plate placed in the way of the laser beam was rotated by 45° to switch to circular polarization.

In the direction transverse to the jet (the time-of-flight direction), the temperature is less than 0.01 K, and the momentum of the jet in this direction is negligible compared to the drift momentum gained in the single ionization process by circular polarized light. The cold target is essential for the use of momentum imaging to calibrate the laser-peak intensity. In the case of warm targets the momentum gained in the laser field will be masked by the momentum spread due to the temperature of the target.

Figure 1 shows typical time-of-flight spectra for H_2 in the case of linear (upper panel) and circular polarized light (lower panel). In the linear polarization case, the peaks correspond to single ionization and other well-known dissociation channels such as bond-softening (BS), above threshold ionization, and enhanced ionization (EI) [13,14]. For the circular polarization case, the single ionization showed a double-peak structure which results from the drift momentum gained in the laser field. In the following discussion we show how to extract the laser peak intensity from this structure.

IV. RESULTS AND DISCUSSION

In our experiment, the momentum p_x (in the time-of-flight direction) can be measured with much higher precision than
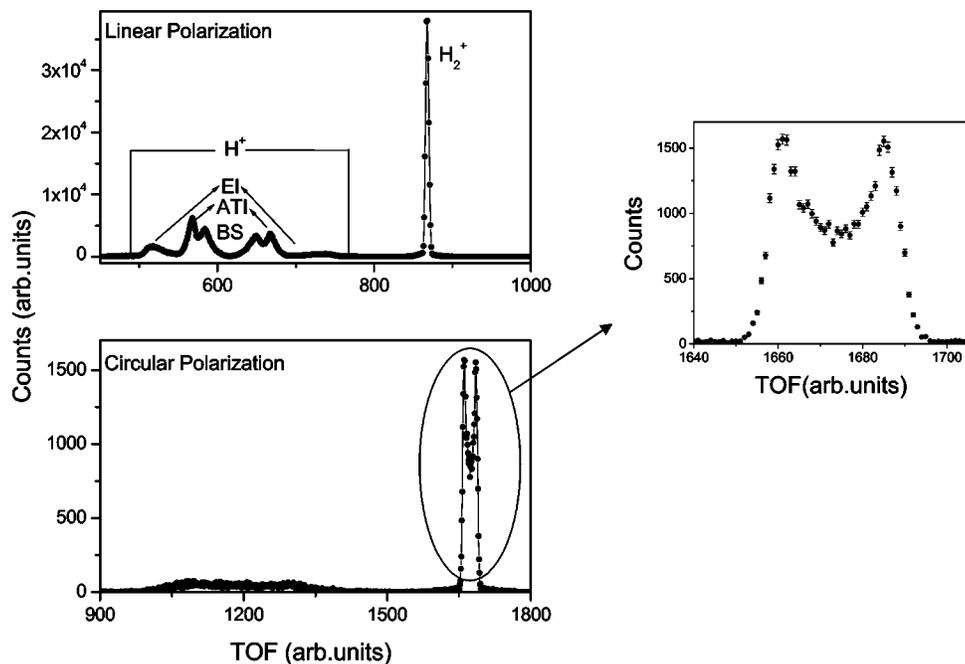
FIG. 1. Time-of-fight spectra for H_2 in the case of linearly and circularly polarized light. In addition to the single ionization peak, other fragmentation channels are shown in the spectra. The double-peak structure of the single ionization by the circularly polarized light is an indication of the drift momentum gained by the ion in the laser field.

p_y (the momentum along the jet direction). This is the case because the jet is much colder along x than along y. To cancel the y-dependence and minimize the uncertainty in the momentum measurements, we integrate the ion yield along the y-direction. After integration, the measured ion yield as a function of momentum transfer along x-direction can be expressed as

$$\frac{dN}{dp_x} = \frac{\eta \omega^2 n_g}{\pi} \int W(\omega^2 p^2) F(\omega^2 p^2) dp_y \qquad (10)$$

with

$$p = \sqrt{p_x^2 + p_y^2}.$$

In Eq. (10), the shape of $F(\omega^2 p^2)$ depends only on p_0 or the peak laser intensity ($p_0 \equiv \sqrt{I_0}/\omega$). To evaluate the expression in Eq. (10) we assume $W(I)$ to be an ADK-like ionization rate

$$W(I) \propto W_{ADK}(I) \propto I^{C_1} e^{-C_2/\sqrt{I}} \qquad (11)$$

with C_1 and C_2 being constants which can be obtained from molecular tunneling ionization model [15] or by using them as fitting parameters as will be shown below.

Figure 2 shows the calculated dN/dp_x versus the normalized momentum (p_x/p_0) as obtained from Eq. (10) for different peak momentum p_0 in the case of H_2 (upper panel) and He (lower panel) targets. As shown in the graph, the value of the momentum at which the yield maximizes is different from the value of the peak momentum p_0 which corresponds to the peak intensity. This difference becomes more obvious as the value of the peak momentum (peak intensity) increases, such that ionization yield maximizes at the leading edge of the laser pulse before reaching the peak value of the intensity. This is expected due to the fact that both $W(I)$ and $F(I)$ together determine the value of dN/dp_x. For higher intensities the rate $W(I)$ does not increase significantly as the

value of the intensity approaches the peak value (saturation), and $F(I)$ is larger for low intensities. For that, the resultant ionization yield dN/dp_x will maximize at intensities lower than the peak intensity. These results are well exhibited when comparing H_2 and He targets which have different ionization potentials and require different values of p_0 to reach a value where the ionization rate does not increases rapidly.
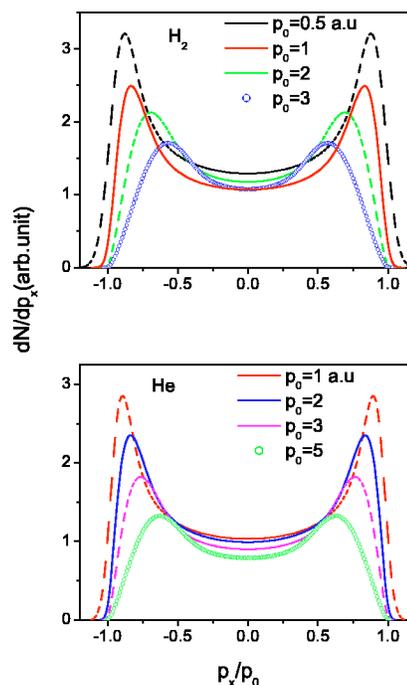

FIG. 2. (Color online) dN/dp_x for H_2 and He targets obtained using Eq. (10). The value of the momentum at which the yield maximizes is different for the two targets due to the difference in their ionization potentials.
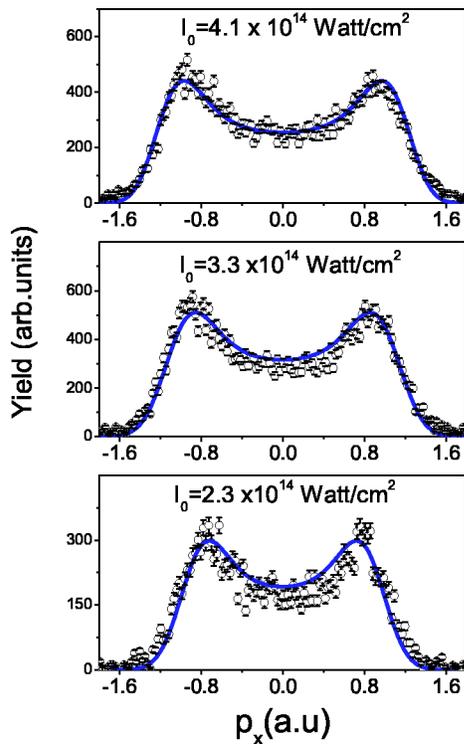
FIG. 3. (Color online) A comparison between the measured momentum spectra (points) and the calculations (curve) in circularly polarized light which is used to find the laser-peak intensity.

FIG. 4. H_2^+ ion yield as a function of peak intensity. By fitting the experimental data with molecular ADK predictions, an independent intensity calibration scale can be obtained.

It is important to note here that we apply this method for laser intensities where the ionization rate changes rapidly before reaching the laser-peak intensity (i.e., below saturation).

We now analyze the time-of-flight, or p_x, spectra and show how to deduce the absolute laser intensity scale from these spectra. Figure 3 shows the momentum spectra of H_2^+ for different laser powers in circularly polarized light. The scattered points represent the measured data and the curve represents the results obtained from Eq. (10) with a given peak intensity I_0. We fit the experimental data with Eq. (10) by the following two methods: (1) using I_0 as a parameter and $W(I)$ from static molecular ADK rate with $C_1 = -0.44$ and $C_2 = 0.809$; (2) using both I_0 and C_2 as parameters. We found that the value of C_2 obtained from fitting the experiment is essentially the same as the theoretically predicted one for higher laser intensity. For lower intensity the fitted C_2 is smaller than 0.809, which is consistent with the prediction by Perelomov *et al.* [16]. Although the C_2 may be different in the two methods, the results for the peak laser intensity are in agreement within 5%. Thus, we only present the theoretically predicted spectra by using the molecular ADK rate. We have also fitted the experimental data by using p_0, C_1, and C_2 as parameters. Since C_1 and C_2 are not linearly independent, we cannot get a unique solution of C_1 and C_2. Despite that, I_0 is very stable in the fitting procedure. This can be explained as follows: The ionization rate increases exponentially as a function of laser intensity, and the ionization mainly happens at the peak intensity. The volume integration [Eq. (5)] decreases slowly near the peak intensity.
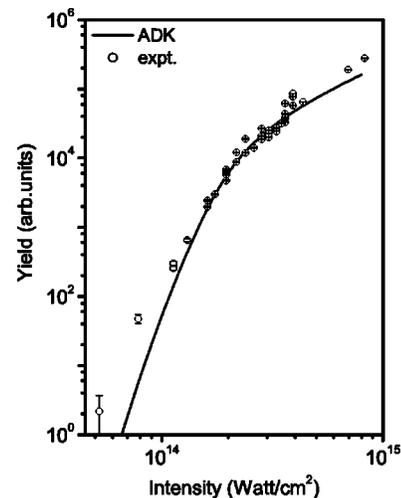
So, the measured spectra are sensitive to the peak intensity, and insensitive to the detailed form of the ionization rate.

We have also fitted the measured spectra with different temporal profiles for $f(t)$, namely, $\tau^2/(t^2 + \tau^2)$ and $\sin^2(t/\tau)$. The obtained laser peak intensity I_0 is found not to be sensitive to the temporal profile. Therefore this process for calibration of the laser peak intensity is robust.

Once the peak intensity is found for each power used in the measurements, we get a linear correlation between the power measured by the powermeter and the laser-peak intensity in the form $I = kP$, where I is the peak intensity in W/cm^2, P is the power in watts measured by the power meter, and k is the calibration constant which was found to be $1.25 \pm 0.12 \times 10^{16}$ cm^{-2}. Using this calibration we can determine the laser-peak intensity for any process of interest and compare with other methods of calibrations.

To compare the calibration scale obtained by the method above with another method, we measured the single ionization of H_2 at different laser powers in linearly polarized light and we fitted it with the molecular ADK predictions [15]. The fitting was done by integrating the calculated ionization probability over the pulse duration and the focal volume [17,18]. Using this fitting another intensity calibration scale was found, for which the calibration constant $k = 1.65 \pm 0.78 \times 10^{16}$ cm^{-2}. Figure 4 shows the results of this fitting. Generally, in getting this fitting there is no unique procedure for scaling the yield and the intensity axes to adjust the experimental data to the calculations, and so, we believe this method of calibration is not expected to give very accurate results. However, the results obtained by the fit presented in Fig. 4 agree within 30% with the drift momentum measurements. The value of k obtained is also consistent with that expected from the values of the beam-spot, pulse duration, and laser energy, but within a large error bar.

Being the simplest molecule for theorists and well-accessible for experimentalists, H_2 plays an important role in understanding the behavior of diatomic molecules in strong laser fields. Models such as bond softening (BS) [13,19],
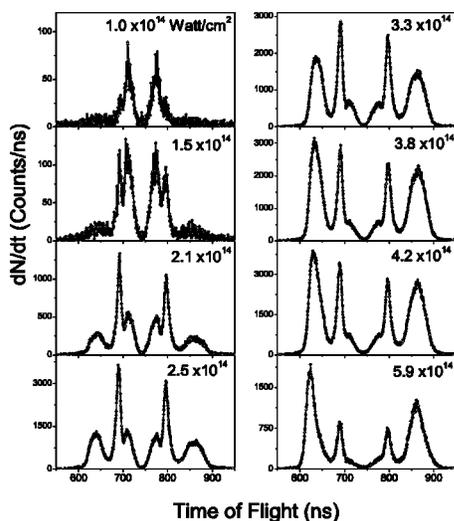
FIG. 5. Time-of-flight spectra of H_2 at different laser peak intensities in linearly polarized light. The branching ratios of the different fragmentation channels can be used as a guide to assign the laser peak intensity.

enhanced ionization (EI) [14,20,21], rescattering double ionization [22–25], and sequential ionization [26] have been widely used to describe the dissociation and fragmentation of H_2 when exposed to strong fields. The interpretation of these models is strongly influenced by the value of the laser-peak intensity for a given reaction. Therefore, an accurate laser-peak intensity calibration scale is very important to monitor the branching ratios for the different fragmentation channels of H_2 as a function of the laser-peak intensity.

Figure 5 shows the time-of-flight spectra for H_2 at different laser-peak intensities assigned using the intensity-power correlation obtained by the circular polarized light. To enhance the visibility of the different fragmentation channels labeled in Fig. 2 the single ionization peak is not shown in these spectra. The relative contributions of the different fragmentation channels are very sensitive to the laser-peak intensity as presented in Fig. 5, and previously observed by Thompson *et al.* [27]. Our intensity scale is a factor ~ 2 lower than what has been reported in Ref. [27], where the intensity dependence of the H_2 fragmentation was obtained using an intensity scale achieved by comparing the ADK predictions with the experimental data of Xe^+ ion yield. We believe that it is more accurate to rely on the results obtained by the present method due to the relative small error in determining the intensity. Using the results of Fig. 5 the hydrogen TOF spectrum can serve as a ratiometric approach [18,28] to precisely assign the peak intensity for a given experiment whether H_2 is the main target or a residual gas in the background.

V. CONCLUSION

In conclusion, we have presented a simple and accurate method to calibrate *in situ* the laser-peak intensity by means of high-resolution momentum spectroscopy. Using this method the value of the peak intensity can be determined to within 10%. We have also shown that hydrogen's time-of-flight spectrum can be used to assign the laser-peak intensity for a given reaction, whether it is the main target or a residual gas in the background. We point out that the momenta of the electrons, rather than the recoil ions, could have been used in exactly the same way to determine the peak intensity using circularly polarized light. Finally, we have shown that this method can be used to get the absolute ionization rate without any volume integration and can be applied in future experiments to get the experimental absolute ionization rate directly with no further approximations.

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