# Signatures of direct double ionization under xuv radiation

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In anticipation of upcoming experiments on two-photon double ionization of atoms and particularly helium, under strong short-wavelength radiation sources (45 eV), we present quantitative signatures of direct two-photon double ejection, in the photoelectron spectrum (PES) and the peak power dependence, that can be employed in the interpretation of related data. We show that the PES provides the cleanest signature of the process. An inflection (knee) in the laser power dependence of double ionization is also discernible, within a window of intensities that depends on the pulse duration and cross sections

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The issue of direct versus sequential double ionization has in the last few years emerged in a new context, namely, two-photon double ionization of helium under xuv radiation, and in particular photon energies of about 45 eV [1–7]. Although until very recently, sources of radiation in that wavelength range, mostly synchrotrons, could not provide the needed intensity (more than $10^{12}$ W/cm$^2$), the situation has now changed. It is conceivable that further developments and optimization of high-order harmonic generation (HOHG) might succeed [8]. The upcoming second phase of the free electron laser (FEL) xuv source at DESY [9], however, is expected to easily satisfy that requirement. Thus it is a matter of probably a short time that the first experimental data on this process will be obtained. When that happens, it is important to have available unequivocal and quantitative signatures of the process and this is the purpose of this paper.

What is it that makes this process interesting? Recall that single-photon double ionization, especially in helium, has been studied in great detail, both theoretically and experimentally [10]. It is basically well understood, although interesting details, especially near the threshold, keep coming up [11]. It could also be argued that this process is fundamentally two step, in the sense that the single available photon can only interact with one of the electrons and it is only through electron-electron correlation that double ejection is possible. As often said, correlation in either the initial or the final state is necessary [12]. To stress the point, let us note that the process would be impossible for noninteracting electrons. The same is true for the other extreme case of double ionization, namely, long wavelength (~780 nm) high intensity, and short pulse duration. The mechanism for that process, other than the sequential, is also explicitly understood as two step [13]. Specifically, the theoretical interpretation rests on the physical picture of one electron pulled out by the strong oscillating field, set into oscillation and liberating the other electron—with a probability depending on the intensity—as it is driven back to the vicinity of the nucleus. Despite the enormous number of photons streaming through the atomic diameter, at those intensities, these photons cannot act simultaneously and separately on each of the electrons, with a probability of any significance. The reason is screening, even in a two-electron system, which does not allow the long-wavelength photons to "see" both electrons at the same time; i.e., before one electron leaves and the other relaxes. Obviously, the same is true for atoms with more electrons, such as the rare gases, where double ionization has been observed under such conditions and interpreted similarly. This two-step process is referred to as nonsequential, to distinguish it from the sequential double ionization, in which one electron is ionized by the field with the subsequent ionization of the ion, through a second high-order process. The term "direct" has also been used for instead of nonsequential. We will, however, need to reserve here the term direct for a somewhat different process, when it comes to the interaction with photons of energy above ~40 eV and below 80 eV where the single-photon channel opens.

We shall be even more specific and consider the photon energy range ~40–54 eV, with emphasis at ~45 eV, for reasons that we explain now. Inspection of the energy level structure of helium, with a first ionization threshold at ~25 eV and the lowest doubly excited (autoionizing) state at ~56 eV, shows that the absorption of a photon of 45 eV raises the system to a virtual state within the single-electron continuum, detuned by ~10 eV from the nearest discrete state. The absorption of a second photon, assuming sufficient intensity (i.e., flux of photons) can, among other things, lead to double ejection, through a direct and obviously nonsequential process in which both electrons are acted upon by the photons independently. The reason is that, because of the short wavelength, screening is of no significance. Moreover, this two-photon double ionization would occur even if the two electrons were noninteracting particles. It is in order to stress this feature that we would propose to reserve the term direct for processes of this type, which are possible even in the absence of interaction, which entails correlation. In addition to the direct double ejection, producing $He^{2+}$, other processes that will take place, with the respective branching ratios, are as follows (a) Single-photon ionization producing $He^+(1s)$; this is by far the strongest channel, with a cross section $\sigma_a = 2.4 \times 10^{-18}$ cm$^2$. (b) Two-photon ionization of $He^+$, producing $He^{2+}$, with a generalized cross section $\sigma_b = 1.0 \times 10^{-53}$ cm$^4$ s. (c) Two-photon ionization of He [above

 

threshold ionization (ATI)], leading to $He^+(1s)$ or even excited states. (d) An, in principle, infinite sequence of higher-order processes, which owing to the combination of (short) wavelength and intensity of interest are of no significance in the context of this paper. The quantities $\sigma_a$ and $\sigma_b$ can be calculated very accurately. The cross section $\sigma_2$ for the direct process involves uncertainties as discussed below.

Let us now define the context. We have in mind possible observations with sources such as those mentioned above. To the best of our knowledge and estimate, one cannot expect at present more than $10^{13}$ or possibly $10^{14}$ W/cm$^2$ from HOHG sources. To be generous, however, let us say $10^{16}$. On the basis of what we know, $10^{15}$ would be a hopeful intensity, at least for the initial operation of the next FEL phase. The relevant pulse durations could range from a few tens of femtoseconds (for HOHG) to around 100 fs for the FEL. First note that, at $10^{16}$ W/cm$^2$ and photon energy 45 eV, the ponderomotive energy of the electron is ~0.5 eV which is 1% of the photon energy. This means that ATI beyond the first peak and related nonperturbative effects can be ignored. In addition, even a pulse duration of 1 fs is very much longer than one cycle (~0.1 fs) of the field, which means that a transition rate is extremely well justified. Obviously, the above conditions also imply that recollision processes, which are crucial at 780 nm, are completely insignificant here, which is another way of saying that whatever double ionization is observed will come either from the direct, as defined above, or the sequential process. The transition probability per unit time obtained through lowest-order perturbation theory is therefore meaningful, but it requires an accurate structure calculation for He, including the double continuum in the final state, for the direct process. The single-active-electron approximation, valid for long wavelength, is totally inappropriate here. In performing the summation over intermediate states, since the first photon reaches into the continuum, a pole is involved, which, however, can be handled (with care) using appropriate techniques [3,14]. For our purposes here, and the needs of interpretation of experimental data, in this context, it is mainly processes (a) and (b), in addition to the direct, that are of relevance. The role of processes (c) is discussed later on. It is important to note that, for photon energies around 45 eV, process (b) requires two photons, which makes the sequential process of third order, compared to the second order of the direct [1]. This feature, favoring the direct process quantitatively and spectroscopically as we shall see below, is lost above ~54 eV. Obviously, when the intensities reach values considerably higher, time-dependent solutions of the Schrödinger equation, such as those already presented in Refs. [2], among others, must come into play.

This is not the place to elaborate on the relevant theoretical techniques. It should suffice to say that the transition rate for the direct process has been calculated by a number of authors [1–3,5,7]. Strictly speaking, in some cases, it is the ionization yield, in a time-dependent approach, that has been reported, from which a generalized two-photon cross section can be readily extracted, since the conditions of validity are satisfied. Although the results have not yet reached the degree of accuracy found in single-photon double ionization, it can be reasonably said that the cross section is known well to within one order of magnitude. Actually, most calculations agree to within a factor of 2, but we shall try to be on the safe side.

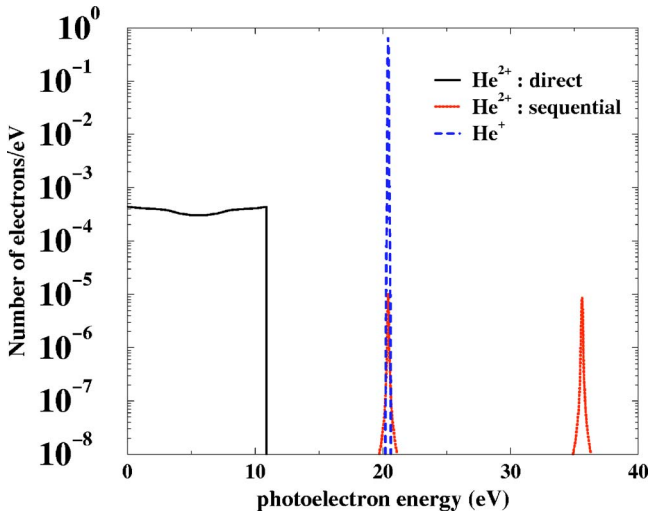


FIG. 1. (Color online) Two-photon PES of helium under irradiation with a Gaussian pulse of photon energy 45 eV, peak intensity $10^{14}$ W/cm$^2$, and 30 fs duration. Only the ionization paths (a) and (b) have been considered, in addition to the direct one, with $\sigma_2 = 8.1 \times 10^{-52}$ cm$^4$ s.

Perhaps the cleanest signature of the direct process is to be found in the angle-integrated photoelectron spectrum (PES). Its general outline was presented in [1] together with an estimate of the relevant cross sections. What is needed now is a quantitative picture, based on up to date information on the cross sections and laser parameters expected to be available. Thus, employing the approach in [3] and a rather generous value for $\sigma_2$, we present in Fig. 1, an example of the PES calculated under the conditions of intensity and pulse duration given in the caption. Note that the figure provides a quantitative result for the differential ion yields. The yield will scale according to the power of the intensity for the corresponding process and proportionally to the duration. In addition, the widths of the narrow peaks will change somewhat. The structure of the PES is due to the direct process plus (a) and (b) and will not change for different combinations of intensity and pulse duration. In particular, the relative position of the peaks, and the energy separation of the direct from the dominant peak (a) will not change appreciably. This is what makes this double-ionization process very special. Obviously, all electrons with kinetic energy less than ~11 eV, originate from the direct. Initial but somewhat limited theoretical studies [4] have given hints of rather unexpected behavior of the photoelectron angular distribution and its connection to correlation, as a function of the partition of the excess energy between the two ejected electrons. These would surely provide further signatures, although much more demanding experimentally. For the moment, it is safer to focus on the requirements for the detection and analysis of the angle-integrated signal. Perhaps the most serious challenge to a measurement of the spectrum comes from the high but narrow peak at ~20 eV, which, although well separated energetically, could mask the signal from the direct process. It should in addition be noted that processes

(c) would produce a series of peaks, between the edge of the direct process and (a), but of much smaller and diminishing height. We understand that electron-electron coincidence measurements [15] may be necessary to cope with that "noise," an issue well beyond our expertise. On the basis of the approach outlined here, one can readily calculate spectra for a variety of experimental conditions, which would, however, be meaningful only in relation to the specifics of the contemplated experiment.

Turning now to a second possible signature, we consider the ion yield, for both $He^+$ and $He^{2+}$, as a function of the laser power. This has in fact been the basic tool in the identification and study of nonsequential double ionization in the long-wavelength regime [16]. Can it also serve equally well in this case of the very different nonsequential process? Needless to say, detecting the ions is considerably less demanding experimentally. The question is whether it can provide sufficient information to at least identify the presence of the direct process. The direct process does of course contribute at any intensity and pulse duration, with a branching ratio determined by the atomic and source parameters. The former being fixed, it is the latter that will decide whether its contribution may be detectable or hopelessly beyond reach.

To evaluate its relative importance, we have considered the set of differential equations [17] governing the rate of production and destruction of the three species, namely, He, $He^+$, and $He^{2+}$ under a pulsed source of prescribed parameters. Let $N_0$, $N_1$, and $N_2$ be the number of He, $He^+$, and $He^{2+}$ in the interaction volume. Their evolution during the pulse obeys the equations

$$\dot{N}_0 = -\sigma_a F(t) N_0 - \sigma_2 F^2(t) N_0,$$

$$\dot{N}_1 = \sigma_a F(t) N_0 - \sigma_b F^2(t) N_1,$$

$$\dot{N}_{2,dir} = \sigma_2 F^2(t) N_0,$$

$$\dot{N}_{2,seq} = \sigma_b F^2(t) N_1,$$

with $N_2 = N_{2,dir} + N_{2,seq}$ and $F(t)$ the photon flux. Typical results about the expected dependence of the respective yields, in log-log plots, for the set of parameters indicated in the captions, are shown in Figs. 2(a) and 2(b). In Fig. 2(a), the value of the cross section for the direct process has been chosen such as to agree with the majority of the calculations published thus far, although one calculation [7] has given a somewhat lower value. It is evident that for peak intensity below about $10^{13}$ W/cm$^2$, double ionization is several orders of magnitude smaller than single ionization. Once double ionization begins making a relatively significant contribution, say above $10^{13}$, it is dominated by the direct process, by several orders of magnitude. In fact up to $10^{14}$, sequential double ionization is practically insignificant, becoming a non-negligible part of double ionization, eventually taking over, arround $\sim 10^{15}$, depending of course on the value of $\sigma_2$. In Fig. 2(b), the value for the cross section of the direct process has been chosen larger by a factor of 8. That is because an early calculation by two of us [3] had given such an optimistic value, which turned out to disagree with calculations by others that followed, including some by us [18]. Nevertheless, the results of Fig. 2(b) are shown here as perhaps an upper bound. As could have been expected, the overall behavior is similar to that of Fig. 2(a), but shifted to lower intensities The pulse duration of 30 fs chosen for the calculations is somewhere in the middle of the range of durations expected for HOHG sources, on the one hand, and FEL, on the other. Changing that value up to say 100–150 fs, or down to $\sim 10$ fs, would not change the overall behavior much. However, the contribution of the direct process will, at a certain peak intensity, begin increasing as the pulse duration becomes shorter; i.e., when the saturation of $He^+$ does not have the time to drain the neutral species. If we were to summarize the message of these two figures, it appears that an intensity of at least $10^{13}$ W/cm$^2$, and preferably considerably more, is necessary for a relatively significant presence of the direct process, and that more than $10^{15}$ or $5 \times 10^{15}$, depending on the cross sections, would hinder its observation. It is conceivable that our reading of the message of these two figures may seem incomplete to an experimentalist. In any case, here they are for the information and use of those interested.

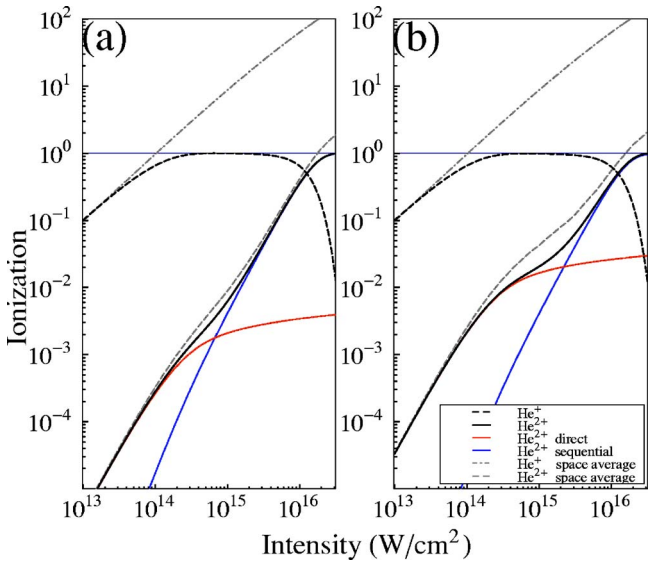


FIG. 2. (Color online) $He^+$ and $He^{2+}$ yield obtained from a Gaussian laser pulse of 30 fs (full width at half maximum). $\sigma_2$= (a) $1 \times 10^{-52}$ and (b) $8.1 \times 10^{-52}$ cm$^4$ s.

An additional aspect in such plots, which is fairly important, is the spatial distribution of the intensity in the interaction volume. That is because, due to focusing, as the peak intensity rises, an increasing part of the atomic beam begins contributing to the ionization species. A quantitative assessment of that effect requires the specifics of the spatial distribution of the radiation for a given experiment. Experience has shown, however, that a typical form [19] of the distribution provides useful insight into the overall effect. We have performed such a sample calculation for the set of parameters employed in Figs. 2(a) and 2(b), with the result plotted on the same figure. As expected, there is no change for lower intensities (up to $\sim 10^{14}$). Beyond that, the amounts of both $He^+$ and $He^{2+}$ continue growing, as larger portions of the atomic beam produce significant signal. In all of the figures,

with or without spatial integration, the curve for $He^{2+}$ exhibits an inflection (referred to as a "knee" in experiments at long wavelength [16]). Its presence is a signature of nonsequential (whether from recollision or direct) double ionization, while the degree of its prominence is here seen to depend on the relative magnitude of the cross sections. Unlike the long-wavelength case, where *ab initio* yields are practically impossible to come by, here we have a quantitative picture which, given sufficient information about the experimental parameters, can be directly related to the cross sections.

Although we have chosen a specific photon energy, as indicated earlier, the overall behavior is not expected to change much for photon energies up to about 54 eV. Finally, our particular choice of atom and order of the process should not be interpreted as a unique context for the study of these aspects of double ionization. In principle, they can be sought in a variety of atoms, in the appropriate range of wavelengths, one such example having been discussed in Ref. [20].

*Note added*: Recently, experimental results pertaining to two-photon double ionization of helium by photons of energy 42 eV were published by Nabekawa *et al.* [21]. Although that photon energy is not too far from the 45 eV employed in our work, a quantitative comparison with our results requires a calculation at the exact frequency of the experiment which we expect to report on elsewhere.

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