Comment on "Photoionization of helium atoms irradiated with intense vacuum ultraviolet free-electron laser light. Part I. Experimental study of multiphoton and single-photon processes"

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(Received 2 February 2006; published 6 September 2006)

The reported observation of two-photon ionization of He by 13 eV photons of the TESLA Facility in T. Laarmann, A. R. B. de Castro, P. Gürtler, W. Laasch, J. Schulz, H. Wabnitz, and T. Möller, Phys. Rev. A **72**, 023409 (2005) is ambiguous, as it is not supported by the experimental data presented. We discuss the lack of evidence for the claimed two-photon ionization.

DOI: 10.1103/PhysRevA.74.037401 PACS number(s): 42.50.Hz

In a recent publication [1], Laarmann *et al.* assert to have measured two-photon ionization (TPI) of He, utilizing the 13 eV photons of the TESLA Facility. This conclusion is ambiguous. In nonresonant TPI of He [2–6], like in any two-photon process obeying lowest order perturbation theory (LOPT), the log of the measured ion yield as a function of the log of the ionizing intensity should be a straight line with a slope of 2 (the slope is equal to the nonlinearity order) [3–6] up to the saturation of ionization. Note that the intensities in Refs. [3–6] span from 10^7 (W/cm^2) to more than 10^{11} (W/cm^2) and the measured slope in all works was essentially 2. In a more recent experiment [7] a slope of 2 has been measured at intensities as high as 10^{12} (W/cm^2). A slope of 2 is entirely missing in [1], although the experiment is in the perturbative regime. All measured slopes (with exception of one of the measured points) in [1] are close to unity, indicative for the observation of some single-photon ionization (SPI). The authors declare an observation of TPI and state that "The theoretical prediction for the two-photon absorption rate in this range of intensity is that the transition probability is far from the perturbative regime and close to saturation." In the case of saturation the measured slope should be around 1.5, due to volume effects, and not 1 as measured in [1]. Our calculations [8] show that the ion yield perfectly follows LOPT and given the 10^{-51} cm^4 s generalized cross section of the TPI of He [2] at the photon energy used and the assumed 100 fs duration of the ionizing pulse, the saturation intensity of He ionization is 2×10^{14} W/cm^2, which is more than two orders of magnitude higher than the highest intensity employed in [1]. Although not published our calculation results in a slope of 2 up to intensities approaching 10^{14} W/cm^2. Moreover, for an intensity interval exceeding two orders of magnitude, a slope close to unity is measured [Fig. 6(a)]. If this would be because saturation sets in already at the lowest measured intensities of Fig. 6(a), the authors should have observed He double ionization at the highest measured intensity of the same graph, which coincides with the intensity of Fig. 3. However, in Fig. 3 there is no evidence for double ionization. Finally it is worth noting that the calculated local minimum in Fig. 9, which is attributed by the authors to some (nonvisible to us) "interference effect" and could affect the ionization intensity dependence, appears at 10^{13} W/cm^2 and thus is outside the intensity range of the measurement. From the above we conclude that in the experimental study of Ref. [1] there is no evidence whatsoever for a TPI process.

The observed H_2O^+ yield in combination to the absence of TPI of He in [1] could serve for the estimation of an upper limit of the ionizing intensity and thus of a lower limit of the pulse duration. For this estimation the ratio of the partial pressures of H_2O and He is needed, which cannot be extracted from the information given in the manuscript. All other required parameters, i.e., the photon energy, the single- and two-photon ionization cross sections, and the correction factor for the different TPI and SPI volume are known.

Concluding, in contrast to previous work utilizing harmonics [3–6], in [1] there is no evidence of the observation of a TPI of He. However, the experimental data of [1] can be used by Laarmann *et al.* for an experimental estimation of the pulse duration of the TESLA Facility.

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