RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 49, NUMBER 1 JANUARY 1994

Ionization of helium by a short pulse of radiation: A Fermi molecular-dynamics calculation

Peter B. Lerner, Kenneth J. LaGattuta, and James S. Cohen
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 10 August 1993)

We describe the results of our quasiclassical calculations of the ionization probabilities of a helium atom, exposed to a short pulse of linearly polarized laser radiation. The technique known as Fermi molecular dynamics was applied. Of particular interest was our observation of the signature of a new process of double ionization, at relatively low laser irradiances. We interpret this as a signature of the simultaneous double ionization of helium. This new process was found to proceed in competition with sequential double ionization. The simultaneous double ionization of helium, as a distinct process at relatively long wavelengths, has just been discovered experimentally [D. Fittinghoff, P. Bolton, B. Chang, and K. Kulander, Phys. Rev. Lett. **69**, 2642 (1993)].

PACS number(s): 32.80.Rm

INTRODUCTION

We report here the results of our simulations of the interaction of a short pulse of linearly polarized, relatively-long-wavelength, laser radiation with a helium atom. The technique known as Fermi molecular dynamics (FMD) was applied. This was a method derived from nuclear physics [1], and adapted to atomic dynamics by Wasson and Koonin [2]. Our procedure is much the same as their's, and has been described by us in an earlier publication [3].

The relative ease with which FMD simulations can be performed makes them potentially useful in those cases where a complete quantum calculation is impractical. Presently, this means for any system containing two or more electrons. But the reliability of the FMD approach, for the simulation of real experiments, can always be questioned.

It is interesting, therefore, whenever a pronounced feature appearing in a new experiment is also found in the FMD simulation of this experiment. We refer here to an apparent signature of the simultaneous multiphoton double ionization (MPDI) of helium, discovered by Fittinghoff *et al.* [4]. Our FMD simulations of this experiment show a similar feature.

We will interpret these results as evidence for the existence of a distinct mechanism for the simultaneous MPDI of helium, at relatively long wavelengths, in FMD. By a distinct mechanism, we mean one that does not arise in the limit, as $\Delta t \rightarrow 0$, of a continuum of delays between successive ejections during a process of inherently sequential MPDI. The Fittinghoff *et al.* data clearly indicate two onsets for the production of He^{2+}, suggesting the existence of two distinct MPDI processes.

RESULTS

We have simulated the experiment of Ref. [4] using our FMD code [3]. The probabilities for the formation of both He^+ and He^{2+} were determined, for laser frequency $\omega = 0.1$ a.u. ($\lambda = 455$ nm), and for linear polarization, as a function of peak laser irradiance. The pulse shape (peak electric field strength vs time) was a pure $\sin^2$ wave, with a duration of 3300 a.u. (80 fsec). This choice of parameters differs somewhat from those of Ref. [4], where $\lambda = 614$ nm, with a pulse length of 120 fsec.

In Fig. 1 we plot the results of these simulations. The curve describing He^+ formation is smooth, implying the existence of a single-multiphoton-ionization mechanism. However, the curve for He^{2+} production contains a shelf on the low-irradiance side, while on the high-irradiance side, just a smooth curve appears. This is reminiscent of the experimental result [4]. (Error bars reflect our estimate of the statistical variance, due to Monte Carlo sampling. We assumed a binomial distribution of outcomes.)

We have also simulated the process $He^+ + \text{photons} \rightarrow He^{2+} + e^-$, for the same choice of laser parameters. Results appear in Fig. 2, where they are compared with the probability of He^{2+} production from Fig. 1. Interestingly, the shelf on the low-irradiance side is present in both sets of data. More significantly, however, the two curves cross.

At high irradiances, the probability of He^{2+} formation

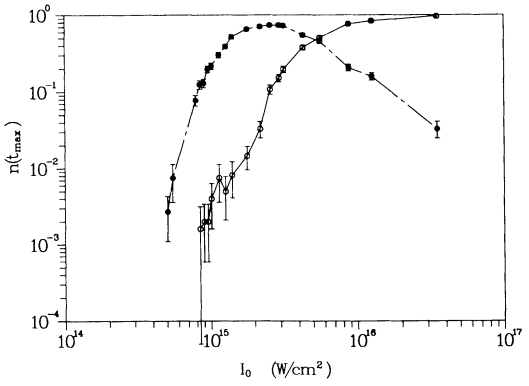

FIG. 1. Probability of He^+ production from He (solid dots), and He^{2+} production from He (open dots), for a $\sin^2$ pulse of duration 3300 a.u. (80 fsec), and with $\omega = 0.1$ a.u. ($\lambda = 455$ nm). Lines have been added to guide the eye.

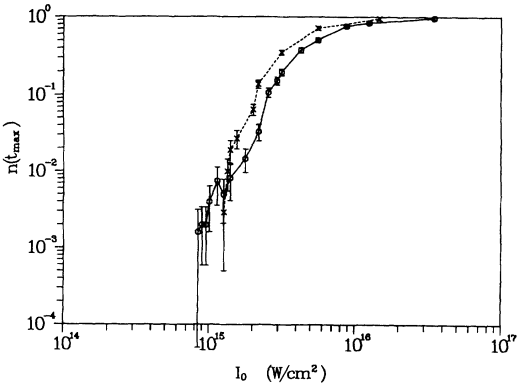

FIG. 2. Probability of He^{2+} production from He^+ (crosses), and He^{2+} production from He (open dots). Conditions as in Fig. 1.

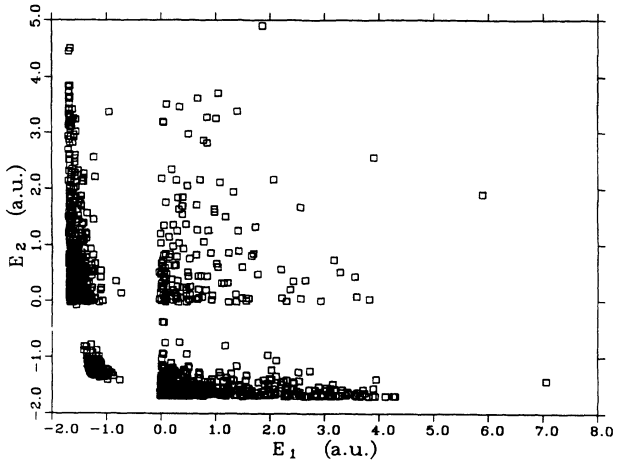

FIG. 3. E_2 vs E_1 for 1.55×10^{15} W/cm^2 $< I_0 <$ 2.95×10^{15} W/cm^2. Conditions as in Fig. 1.

from He^+ is seen to exceed the probability for its formation from neutral helium. In fact, at high irradiances, the product of the probabilities of formation of He^{2+} from He^+, and of He^+ from neutral He, is approximately equal to the probability of He^{2+} formation from He. This implies a dominant mechanism of sequential multiphoton double ionization, in the region of high irradiances. But, if sequential ionization is the dominant mechanism of double-electron ejection at all irradiances, then as the irradiance decreases, the two curves in Fig. 2 should move farther apart. Instead, they cross. We take this to be evidence of a mechanism of double ionization other than sequential; i.e., an inherently simultaneous process.

The assumption of two distinct mechanisms for MPDI will lead, through a rate-equation description of these phenomena, to curves that are similar in appearance to Fig. 1. The description of these processes in Ref. [4] was suggestive. However, it was based on an interpretation [5] of a theory [6] of double ionization by just one photon; i.e., ionization at relatively short wavelengths ($\lambda < 15$ nm). That interpretation [5] led to a picture in which simultaneous double ionization was seen as a special case of sequential double ionization, wherein a "shake-up" of the second electron became a "shake-off."

Other possibilities exist in the one-photon case. For instance, double ejection from two-electron bound states may appear as an inherently simultaneous process, mediated by the mechanisms of "ground-state correlation," and/or "inelastic internal collision," to use the language of Ref. [6]. But, for many-photon absorption, the implications of the work reported in Refs. [5] and [6] are not very clear. For the frequency employed in our simulations, a minimum of ten photons are required for single ionization and 30 for double ionization. Hence, we refer to this as the "relatively-long-wavelength" regime.

Measurements of the ejected-electron kinetic-energy spectrum were also reported in Ref. [5] (for double ionization via single photon absorption). The double-peak structure of this spectrum, with a minimum at the position of the average energy, arose through a sharing of the continuum energy between the two electrons. This sharing can be seen as the consequence of a delay Δt between the times of electron emission. The first electron to be emitted is screened by the second, still bound, electron. Consequently, the first electron is emitted with a relatively large kinetic energy. Then, the second electron to be emitted is exposed to the full nuclear charge, and appears in the continuum with a relatively low value of kinetic energy. As $\Delta t \rightarrow 0$, the dispersion in kinetic energies goes to zero. Hence, in this picture, simultaneous double ionization is seen as a special case of what is almost always a sequential process, and trajectories leading to double escape, with $E_2 = E_1$, appear with a minimum probability. (The "core rearrangement" of Ref. [6] describes a similar mechanism.) But this picture makes sense only if the pulse envelope is very slowly varying. For rapidly increasing envelopes, the opposite behavior can appear, since the last electron may emerge at a much higher irradiance. See the Discussion section for more information.

We have investigated this question somewhat further, at relatively long wavelengths, using the FMD approach. First, we recorded the spectrum of emitted electron kinetic energies at large values of I_0, such that only sequential MPDI occurs. Another record was formed at lower irradiances, such that both simultaneous and

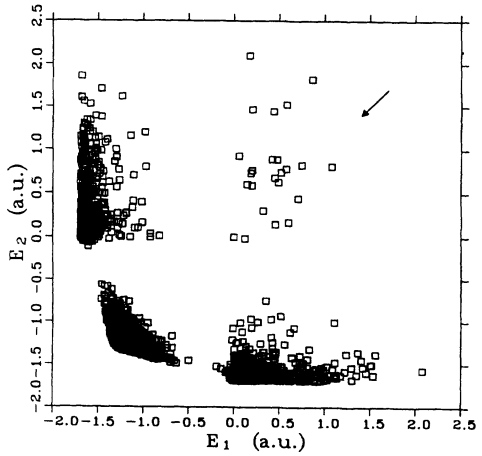

FIG. 4. E_2 vs E_1 for $7.9\times10^{14} < I_0 < 1.4\times10^{15}$ W/cm^2. Conditions as in Fig. 1.

sequential MPDI occur. In Fig. 3 we plot values of E_2 vs E_1 for 1.55×10^{15} W/cm^2 $< I_0 <$ 2.95×10^{15} W/cm^2. Based on examination of Figs. 1 and 2, we judge that these values of I_0 lead only to sequential MPDI. Indeed, in Fig. 3 we see no evidence of an *excess* of events for which $E_2 \approx E_1$.

At somewhat lower irradiances (7.9×10^{14} W/cm^2 $< I_0 <$ 1.4×10^{15} W/cm^2) we plot the corresponding data in Fig. 4. This is a region of irradiances in which, judging from Fig. 1, evidence of simultaneous double ionization should appear. Now we do see a tendency of data to cluster along the $E_1 = E_2$ line; see the following section for further information.

DISCUSSION

In order for the simultaneous process to appear, it is necessary that the laser pulse rise time be short enough, for a given peak irradiance, to prevent depletion of the neutral species by single ionization. In the work of Burnett [7], collective effects are predicted to occur during the photoionization of helium whenever the irradiance exceeds some threshold value. However, at very high irradiances sequential double ionization recurs for ramped pulses. In fact, in the work of Ref. [8] no hint of the presence of the simultaneous process was seen for a pulse length of 1.5 psec at $\lambda = 1$ μm. One may conjecture that the pulse was too long. In Ref. [9] an attempt was made to define the maximum pulse length for a given value of I_0. However, that work does not seem to have immediate applicability to ours, since it refers to double ionization by two or three photons at most.

For a given pulse length, if the peak irradiance is very high, then both electrons ionize during the rising portion of the pulse. In this case, double ionization is purely sequential. The first electron emerges at a lower irradiance, while being exposed to a reduced (screened) value of the nuclear charge. The second electron emerges at higher irradiance, while being exposed to a full nuclear charge of +2. Thus competing effects determine the form of the kinetic energy spectrum (E_2 vs E_1). Although over most of this domain either $E_1 > E_2$, or the reverse, there may be a limited region for which $E_1 \approx E_2$. On the other hand, if the peak irradiance is relatively low, then double ionization occurs only if both electrons emerge together at the peak of the pulse. Moreover, in order for double ionization to occur at all in this case, the probability for single ionization must also be low. Finally, since the electrons emerge simultaneously, one expects that $E_1 \approx E_2$. This is reminiscent of the behavior seen in Figs. 3 and 4.

Phenomena similar to those described here were reported earlier [2]. Those data pertained to the photoionization of helium by a laser which was turned on rapidly over a time of 20 a.u. (0.5 fsec), and then held at a constant irradiance for 830 a.u. (20 fsec). A second set of data was accumulated for short Gaussian pulses of duration 6 a.u. $< t_{\text{pulse}} <$ 200 a.u. The frequency was high (0.12 a.u. $< \omega <$ 1.5 a.u.), as was the irradiance. They found evidence for the existence of a process of "collective" ionization, as opposed to sequential ionization, for cases in which the ". . . ionization rate is comparable to the period of the applied field." They also noted that, "Collective ionization is characterized by the atom being left in an excited state when one electron ionizes." Presumably, this refers to cases for which the amount of double ionization is small, but not zero.

Indeed, we see some indication of this tendency to leave behind excited bound states of He^+, if the irradiance is just high enough to produce some double ionization, by the mechanism we have called simultaneous MPDI. Compare Figs. 3 and 4 in those regions of the E_2-vs-E_1 plots for which either $E_1 < 0$ and $E_2 \geq 0$, or the reverse. Importantly, it is also shown in Ref. [2] that the two-electron system always shows evidence of large amounts of electron-electron interaction for these cases of marginal double ionization. This observation supports the idea of an essential collectivity, which operates most efficiently over a small range of laser parameters. Among these parameters, the pulse length and shape were judged to be critical.

As pointed out by Corkum [10], the mechanism of inelastic internal collision [6] can be invoked to explain the simultaneous double-ionization process. In our case, the probability of double ionization P_{2+} can be related to the probability of single ionization P_+ and to the rate of electron collisional ionization R_{coll}, for a pulse length t_{pulse}, through $P_{2+} \approx P_+ R_{\text{coll}} t_{\text{pulse}}$, provided that $P_+ \ll 1$. We applied the formulas of Sampson and Zhang [11] for the collisional ionization cross section, assuming that the "projectile" electron was contained in a spherical volume of radius equal to one quiver amplitude (F_0/ω^2), and moved with a kinetic energy equal to three times its quiver energy [$3(F_0/2\omega)^2$]. We then obtained qualitative agreement with this relationship, for $I_0 = 1\times10^{15}$ W/cm^2, which is just above the threshold for ionization of the second electron at this wavelength. This threshold also is in accord with the lowest irradiance at which we see doubly charged ions in our simulations. A pronounced wavelength dependence of the threshold for ejection of the second electron has just been reported by experimentalists [12].

We call attention to other aspects of our simulations which seem to be of inherent interest. First we note that the onset of ionization is observed to occur, in all cases, at a value of the irradiance and frequency such that $\omega Z_{\text{eff}}/E_0 \equiv \gamma \approx 1$, where Z_{eff} is the effective positive charge to which the ionizing electron is exposed. That is, in all cases, the onset of ionization is observed to occur at the boundary of the so-called tunneling region, where γ is the conventional Keldysh parameter. (We use atomic units here. In these units, $I_0 = E_0^2$, where 1 a.u. of irradiance is equal to 3.51×10^{16} W/cm^2, and 1 a.u. of field strength is equal to 5.15×10^9 V/cm.)

For example, the process $He + \text{photons} \rightarrow He^+ + e^-$ has an onset near $I_0 = 5\times10^{14}$ W/cm^2 = 0.0142 a.u.; see Fig. 1. Since the binding energy of this first electron is 0.90 a.u., the effective charge is $Z_{\text{eff}} = 1.34$, and the value of the tunneling parameter is $\gamma = 1.1$. Referring to Fig. 2, the threshold for the process $He^+ + \text{photons} \rightarrow He^{2+} + e^-$ is observed to occur near $I_0 = 1.2\times10^{15}$ W/cm^2. Since the value of the effective charge, in this case, is

$Z_{\text{eff}}=2.00$, then the tunneling parameter is also $\gamma=1.1$. Finally, we consider the onset of the presumptive simultaneous double-ionization process (Fig. 1 or 2), which appears at an irradiance of $I_0=9\times10^{14}$ W/cm^2. We make so bold as to estimate the effective charge for this process of "two-electron tunneling" as the square root of the average of the squares of the effective charges for the two one-electron processes; i.e., as $Z_{\text{eff}}\approx1.70$. The corresponding tunneling parameter is then, again, $\gamma=1.1$. Of course, this last estimate is somewhat notional.

The onsets described here are thresholds into the tunneling region, in the further sense that, for $\gamma\approx1$, the ionization probability is observed to rise as I_0^N, where N is the minimum number of *photons* required to ionize; i.e., $N\approx Z_{\text{eff}}^2/2\omega$. For higher irradiances ($\gamma<1$), the rate of increase slows drastically.

It is perhaps coincidental that predictions of behavior based on the tunneling concept seem to have merit here. However, it has been suggested that, in classical systems, so-called "stochastic ionization" [13] may play a role analogous to that of tunneling at irradiances below the over-the-barrier threshold (OBT). By stochastic ionization, we refer to a process of forced diffusion through a continuous distribution of states of increasing energy. The OBT is the minimum value of applied field strength needed to bring the total electronic potential energy to a value equal to its unperturbed binding energy. For the cases examined here, the OBT values of irradiance were computed from $I_{\text{OBT}}=(Z_{\text{eff}}^3/16)^2$ a.u. to be 8.1×10^{14} W/cm^2 and 8.8×10^{15} W/cm^2 for the first and second electrons, respectively (during a sequential process). For the simultaneous process, the effective OBT value was 3.4×10^{15} W/cm^2. All of these values are well above the onsets observed in our simulations.

Finally, we remark that a rate-equation description of these processes fails to produce values of the ionization probability that agree quantitatively with our simulations, if a tunneling formula for the ionization rates [14] is employed. Moreover, no plausible adjustment of parameters in the tunneling formula led to significant quantitative improvement. Nevertheless, the qualitative behavior was reproduced, as in Refs. [4] and [8], by assuming the existence of a distinct second process leading to simultaneous double ionization.

SUMMARY

We have described the results of our FMD simulations of a process of one- and two-electron ionization of helium by a pulse of long-wavelength laser radiation. Evidence for the existence of two distinct processes leading to double ionization was presented. This was further construed as evidence for the existence of a process of simultaneous multiphoton double ionization, in FMD. This process has been referred to as "collective ionization" by Wasson and Koonin [2], and by Burnett [7]. We also pointed out coincidences of the observed onsets of relatively rapid ionization, in these quasiclassical calculations, with the boundary of the quantum tunneling regime.

ACKNOWLEDGMENT

This work was performed under the auspices of the U.S. Department of Energy.

[1] L. Wilets, E. Henley, M. Kraft, and A. Mackellar, Nucl. Phys. **A282**, 341 (1977).
[2] D. Wasson and S. Koonin, Phys. Rev. A **39**, 5676 (1989).
[3] P. Lerner, K. LaGattuta, and J. Cohen, Laser Phys. **3**, 331 (1993).
[4] D. Fittinghoff, P. Bolton, B. Chang, and K. Kulander, Phys. Rev. Lett. **69**, 2642 (1993).
[5] R. Wehlitz, F. Heiser, O. Hemmers, B. Langer, A. Menzel, and U. Becker, Phys. Rev. Lett. **67**, 3764 (1991).
[6] T. Chang and R. Poe, Phys. Rev. A **12**, 1432 (1975).
[7] K. Burnett, J. Mod. Opt. **36**, 925 (1989).
[8] J. Peatross, B. Buerke, and D. Meyerhofer, Phys. Rev. A **47**, 1517 (1993).
[9] M. Crance and M. Aymar, J. Phys. (Paris) **46**, 1887 (1985).
[10] P. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[11] D. Sampson and H. Zhang, Astrophys. J. **335**, 516 (1988).
[12] K. Kondo, A. Sagisaka, T. Tamida, Y. Nabekawa, and S. Watanabe, Phys. Rev. **48**, R2531 (1993).
[13] R. Jensen, S. Susskind, and M. Sanders, Phys. Rep. **201**, 1 (1991).
[14] F. Ilkov, J. Decker, and S. Chin, J. Phys. B **25**, 4005 (1992).