Sequential ionization in ^{3}He with a 1.5-ps 1-μm laser pulse

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We have examined the laser ionization of ^{3}He using 1.053-μm 1.5-ps laser pulses. The use of ^{3}He rather than ^{4}He in the experiment enables the detection of the second charge state separately from a troublesome background contaminant H_2^+. In these experiments, the production of doubly charged ions is dominated by the sequential process rather than by the direct or simultaneous process.

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Helium and other atoms can, in principle, become doubly ionized when exposed to an intense laser pulse through two different channels: a direct process whereby both electrons are ejected simultaneously from the atom and a sequential process whereby the second charge state is created only after the first charge state. Crance and Aymar [1] calculated for He that if the number of photons required for direct ionization is less than the number of photons required for sequential ionization, then, depending on the duration and intensity of the applied field pulse, one or the other ionization channel may be favored. Their analysis used high-energy photons such that in the sequential ionization channel the removal of the first electron required one photon and the removal of the second required two. However, they suggested that the result should qualitatively hold for lower-energy photons where a large number are required for ionization. They predicted that the direct-ionization channel should become apparent in He at 1.06 μm for sufficiently short pulse durations. A rough estimate that they give [Ref. [1], Eq. (52)], when applied to typical ionization-field intensities for He, suggests that the pulse duration must be about 10 fs or shorter before direct ionization can occur significantly.

Lambropoulos [2] made a strong case that any neutral atom exposed to intensities much above 10^{14} W/cm^2 at near-infrared or higher frequencies cannot survive for a longer duration than about 100 fs. Helium has the highest ionization potential of any atom, but it also has a large difference in ionization potentials between the first and second charge states. Therefore the neutral atom would have to survive well past its first ionization threshold to allow for a simultaneous ejection of both electrons from the atom. If a laser pulse is too long, then by the time the field reaches sufficient intensity to produce doubly ionized helium, the neutral atoms are depleted, restricting ionization to the sequential channel.

L'Huillier *et al.* [3–5] performed a pioneering series of experiments studying direct versus sequential ionization of noble gases. They observed evidence for direct ionization in xenon at wavelengths of both 1.06 and 0.53 μm and a pulse duration of 50 ps. They also reported evidence in support of direct ionization in other noble gases, including helium, under these conditions. They extended [5] the work at 0.53 μm in xenon to include pulse durations from 5 to 200 ps and found a trend showing that a shorter pulse increases the likelihood of direct ionization.

Recently Fittinghoff *et al.* have observed direct ionization in helium with a 614-nm 120-fs laser pulse [6]. They suggest that the phenomenon is due to the first electron leaving the atom so quickly that the second electron can be left in an excited state, which then easily ionizes. Their calculations show that for pulses of 1 ps or longer, the process should no longer be significant.

We have studied the ionization of He with a 1.053-μm 1.5-ps laser. For our experimental conditions, the sequential-ionization channel was observed to be more probable than the direct channel by at least four orders of magnitude (our detection limit) in the range of intensity where the second charge state begins to appear. We found that the intensities at which the first and second charge states begin to appear have a separation of nearly an order of magnitude. Thus the neutral helium is ionized to He^+ with probability of virtually 1 before any He^{2+} appears.

In our previous ^{4}He ionization experiments, the sequential channel for ionization was shown to be preferred by at least a factor of 10^2 (Ref. [7], Fig. 1). This

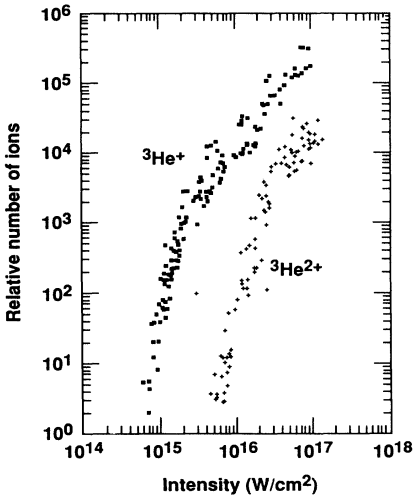

FIG. 1. The relative number of singly and doubly charged He ions produced as a function of peak laser intensity.

value was limited by a background contamination of H_2O in the laser focus. In a standard time-of-flight spectrometer, H_2^+ that comes from the water drifts at the same speed as $^4He^{2+}$ since their mass-to-charge ratios are nearly identical. As a consequence, when trying to observe low numbers of $^4He^{2+}$, the measurement can be overwhelmed by the background of H_2^+. Without considering this problem, one might conclude from the data that He has a strong direct-ionization channel under the previously stated laser conditions. We have circumvented this problem by measuring ionization in ^3He since both its singly and doubly ionized states produce unique signatures in the time-of-flight spectrometer. To our knowledge, this is the first time that this isotope of helium has been employed in a laser-ionization experiment.

The setup for this experiment was similar to that described in Ref. [7]. The laser was a Nd:glass chirped-pulse amplification system, which has been described in detail [8,9]. The repetition rate was one shot every 70 s with a nominal pulse duration of 1.5 ps and measured focal-spot area of 100 μm^2. The f number of the focused beam was ~ 3 with a 20-cm lens. The fluctuations in pulse duration and focal-spot area resulted in a 40% uncertainty in the relative intensity from shot to shot. The uncertainty in the absolute intensity was 50%. The laser was focused inside an evacuated chamber that had a background pressure of less than 10^{-8} Torr, and the chamber was back-filled with ^3He to pressures in the range of 10^{-7}–10^{-5} Torr. The laser intensity was continuously varied from a few times 10^{14} W/cm^2 to 2×10^{17} W/cm^2, and the resulting ions were detected with an ion time-of-flight spectrometer. The ions were accelerated by an electric field of 10^3 V/cm into a 0.6-m drift tube at the end of which a microchannel plate detected them at their arrival times. It was determined that for the range of pressure used, the number of ions arriving at the detector scaled linearly with pressure. At higher laser intensities, a low pressure was used to avoid detector saturation, and for lower intensities a higher pressure was used to enhance the signal. The experiment was repeated with ^4He, which gave good agreement with the ^3He experiment. It was found that if the background pressure in the tank was well below 10^{-8} Torr, the H_2^+ signal did not seriously affect the low-level $^4He^{2+}$ signal.

Figure 1 shows a plot of the relative number of ions produced versus the peak laser intensity, where each point represents a single shot. No smoothing has been applied to the data points. The nearly vertical linear portions of the curves in Fig. 1 show that the ionization is a strong function of intensity. The upper portions of the curves that bend over are due simply to a depletion of the atoms within the laser focus [3–5,10]. The distinct separation between the curves for $^3He^+$ and $^3He^{2+}$ shows that over nearly an order of magnitude in intensity from where the first charge state begins to appear there is no evidence for production of $^3He^{2+}$. The vertical separation between the two curves at the point the second curve begins to appear reveals that the sequential-ionization channel is at least 10^4 times more probable. If direct ionization were to be observed, it should evidence itself as a small deviation in the base of the second curve, which tapered off as the first charge state saturated with increasing intensity [3–6]. The broadening of the two curves is due to the temporal and spatial fluctuations of the laser, which were described above.

In experiments carried out previously under similar laser conditions, Augst *et al.* [11] found that the ionization of noble gases could be explained well by classical field ionization (barrier-suppression ionization). In this model the atom or ion becomes one state further ionized when the laser electric field reaches the value required to depress the potential barrier sufficiently so that the bound electron can escape "over the top." The model assumes that the bound-electron orbital frequency is much larger than the laser frequency so that the electron can escape within a single laser period. This classical ionization model therefore precludes the simultaneous ionization of two electrons. The results presented in the present paper are thus consistent with this model.

In summary, we have shown that the production of doubly ionized He in a 1.053-μm 1.5-ps laser pulse is dominated by the sequential channel with the direct channel being at least 10^4 less probable. This is consistent with the work of Augst *et al.*, [11], which showed that the ionization can be described well by a simple barrier-suppression model that allows for sequential ionization only. If the experiment is done in ^4He rather than ^3He, background contamination of H_2^+ can skew the He^{2+} measurement. Such an effect can be seen at the base of the $^4He^{2+}$ curve of our previously published data [7].

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