Kinetics of He(2 ^1S) Using Resonance Ionization Spectroscopy*

M. G. Payne, G. S. Hurst, M. H. Nayfeh,† J. P. Judish, C. H. Chen,† E. B. Wagner, and J. P. Young
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
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We have demonstrated that quantum selected populations of He(2^1S) can be converted completely to ionization by single-photon excitation to He(3^1P) followed by collision-induced associative ionization. For helium pressures between 15 and 100 Torr, saturation of ionization was observed whenever a tunable dye laser produced more than 0.1 J/pulse. Absolute numbers of He(2^1S) per ion pair formed from proton interaction and the decay rates of He(2^1S) support a recent energy-pathways model.

Excited states play decisive roles in many physical phenomena; consequently, several methods, such as emission and absorption spectroscopy, surface ionization, and Penning ionization, have been devised for their detection. Recently[1] some of us introduced a new method (resonance ionization spectroscopy, RIS) in which quantum selected populations are converted completely to ionization with a pulsed dye laser.[1]

In that earlier work,[1] complete conversion of excited-state populations to ion pairs was accomplished with a photoionization process involving the absorption of two photons, one of which was resonant with an intermediate state. Thus, the well-known two-photon ionization process, when saturated, can be used as a basis for absolute and very sensitive measurements of quantum-selected (including ground-state) populations. For example, if He(2^1S) is excited at low pressure (0.6 Torr) and irradiated with a pulsed dye laser tuned to 5015 Å, He(2^1S) is promoted to He(3^1P) as the intermediate state. This intermediate state is photoionized with a cross section of approximately 4.4×10^{-18} cm^2 at 5015 Å; thus a laser pulse of 1 J converts nearly all of the He(2^1S) to ionization via the He(3^1P) state.

It is shown here that saturated ionization can also be accomplished by single-photon absorption followed by collisions with ground-state atoms. This technique requires less laser energy per pulse than does the two-photon method.[1] In this paper we develop this version of the RIS technique and apply it to kinetic studies of He(2^1S) over a wide range of helium pressures.

For helium, the states lying above the 3^3P can be converted to ionization by an associative process (known as the Hornbeck-Molnar reaction). The intermediate state He(3^1P) is converted to ionization with a two-body cross section of about 3×10^{-16} cm^2,[2] and to He(3^1D) with a cross section[3] of about 30×10^{-16} cm^2; He(3^1D) is associatively ionized with a cross section of about 20×10^{-16} cm^2.[2] Effectively, the He(3^1P) is converted to $(He)_2^+$ at a rate of about 10^7P, where P is the helium pressure in Torr and the rate is in inverse seconds. However, a competing process is collisional conversion of 3^1P to 3^1S (with $\sigma=4.5\times10^{-16}$ cm^2)[3] which is not associatively ionized but, instead, radiates (3×10^7 sec^{-1}) to 2^1P. Return of He(2^1P) to He(2^1S) occurs at the rate of $1.9\times10^6+(6.4\times10^4\ \text{Torr}^{-1})P$ sec^{-1}, completing a cycle.[4,5] During the cycle there is a loss from the He(2^1P) state due to resonance radiation transport to the wall at a rate of 0.9×10^6 sec^{-1} and due to excimer formation at a rate of (90 Torr^{-2})P^2 sec^{-1}. These losses are not important over a wide range of pressure since the chance that an atom promoted to the He(3^1P) intermediate state will enter the return part of the cycle (where the losses occur) is small. As the pressure increases, associative ionization dominates over photoionization. At 15 Torr, photoionization contributes a few percent at an energy of about 1 J/pulse.

Pulses (15 nsec) of 2-MeV protons were injected into a parallel-plate ionization chamber filled with rapidly flowing 99.9998% pure helium. After a suitable delay, a laser tuned to 5015 Å provided a photon flux which bathed the region of the chamber surrounding the proton path. Recordings were made of the electron currents due to (1) the direct proton ionization and (2) laser-induced ionization of the selected excited state. Important laser-pulse specifications included these: (1) The beam was reasonably uniform over its 10 mm diam, (2) the energy per pulse reached 0.7 J, (3) the photon spectrum was 30 Å full width at half-maximum (FWHM), and (4) the pulse duration was about 300 nsec. The output spectrum of each laser pulse was recorded with an optical multichannel analyzer.

Data were collected by using a transient recorder in order to store the signal which was

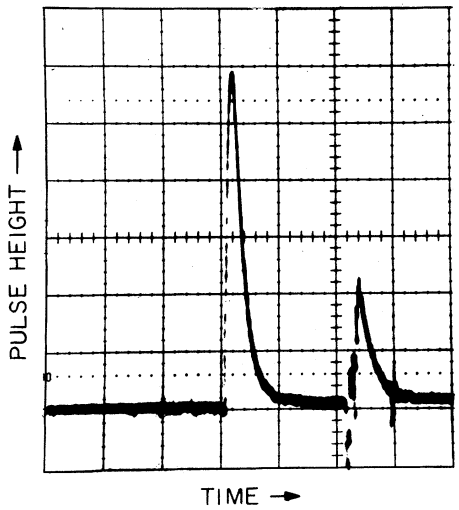

FIG. 1. Electron signals due to direct proton interaction and resonance ionization of the singlet metastable He(2^1S) with a dye laser tuned to 5015 Å. Helium at a pressure of 15 Torr was excited with pulses of protons followed with laser pulses after 10 μsec.

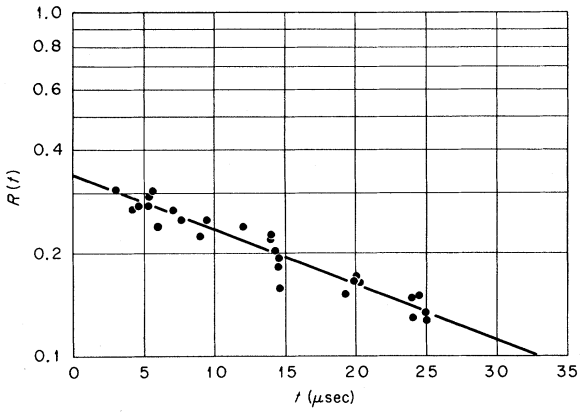

FIG. 2. The absolute population of He(2^1S) states per direct ion pair as a function of time, $R(t)$, after 2-MeV proton excitation of helium at 100 Torr.

generated across a few-kilohm resistor following the passage of a pulse of protons through the ionization chamber and the subsequent passage of a dye laser pulse. When the laser described in the previous section was tuned to 5015 Å and delivered at least 0.1 J/pulse, the signals like those in Fig. 1 were obtained. When the laser was detuned from 5015 Å but delivered 0.7 J/pulse, the second peak was observed to disappear, i.e., there was little background. Since the number of ion pairs produced per centimeter of path by the proton pulse is $\lesssim 10^7$ ion pairs/cm and almost all of the He(2^1S) population is ionized as a consequence of the passage of the laser pulse, the number of He(2^1S) states per ion pair is given by the ratio of the second peak height to the first. The production of more than 10^8 ion pairs/cm leads to space charge effects which were avoided.

Saturation of the ionization of He(2^1S) as a consequence of the laser pulse (0.7 J at 5015 Å) was checked by attenuating the beam by a factor of 3 and observing that $R(t)$, the ratio of the second to the first peak height, was unchanged within experimental error. The insertion of a lens system which increased the beam radius by a factor of 1.5 also gave no observable change in $R(t)$. Whenever associative ionization occurs at a rapid rate (i.e., higher pressure), a very modest energy (i.e., 0.1 J) per pulse is required to saturate the ionization, even when the spectrum is wide (30 Å FWHM). This experimental proof was confirmed with appropriate rate-equation calculations.

Measurements were carried out under saturation conditions for a large number of delay times at each of the following pressures: $P = 15.3$, 30, 60, and 100 Torr; the decay curve for 100 Torr is shown in Fig. 2. At times greater than 2 μsec following excitation, we find that the He(2^1S) population decays exponentially with decay constant $\beta(2^1S)$ given by

$$\beta(2^1S) = (220\ \text{Torr}^{-1})P + (1.4\ \text{Torr}^{-2})P^2\ \text{sec}^{-1}.$$

At low pressures this rate agrees well with Phelps,[6] and at pressures above 100 Torr it is nearly the same as that of the 601-Å, collision-induced emission of He(2^1S).[4,5]

We now show how the absolute number of excited states was used to verify a key process in a recent energy-pathways model.[5] The population of He(2^1S) at times greater than 1 μsec is due not only to the initial excitation of this state but also to excitation of higher states followed by radiative cascade and collisional conversion processes[4,5] which ultimately lead to He(2^1S). Initially, the He(2^1P) population makes up about 50% of all the excited state population, and it is about a factor of 5 more numerous than He(2^1S). However, it is observed[5] that He(2^1P) decays exponentially with time with a decay constant $\beta(2^1P) = 2.8\times10^6 + 6.4\times10^4P + 90P^2$, with $\beta(2^1P)$ in units of sec^{-1} and P in Torr. The 2.8×10^6/sec term represents a rate of spontaneous emission to the 2^1S state (1.9

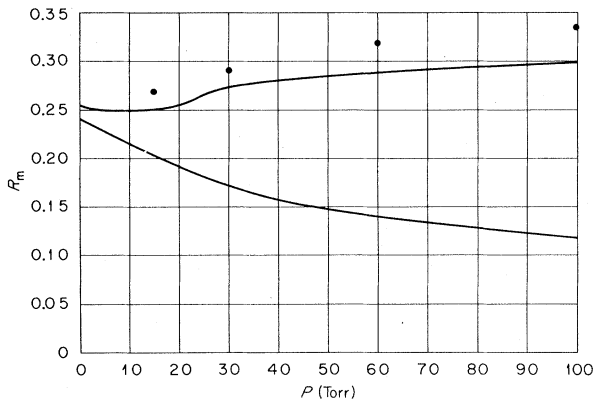

FIG. 3. The number of He(2^1S) states per direct ion pair (R_M) as a function of helium pressure. Circles, RIS measurements. Collisional conversion of He(2^1P) to He(2^1S) is assumed to obtain the upper curve. The lower curve is the calculated ratio assuming that collisional destruction of He(2^1P) does not lead to He(2^1S).

$\times 10^6$/sec) plus a rate of escape of resonance photons to the cell wall ($\sim 9\times 10^5$/sec). A theoretical argument[5] suggests that the 6.4×10^4P term represents a rate for the process He(2^1P) + He(1^1S) → He(2^1S) + He(1^1S). The $90P^2$ term presumably represents the rate of formation of the $B\,^1\Pi_g$ and $D\,^1\Sigma_g^+$ dimers. If the linear term does represent collisional conversion of He(2^1P) to He(2^1S), then at $P \simeq 100$ Torr about 89% of the initial He(2^1P) population is converted to He(2^1S); otherwise, only about 20% of the He(2^1P) population would become He(2^1S). Figure 3 shows the experimental $t=0$ intercepts (R_M) of the curves of $R(t)$ versus t as a function of pressure. The upper solid curve is based on the assumption that the 6.4×10^4P term represents collisional conversion of He(2^1P) to He(2^1S), while the lower solid curve is based on the assumption that the only conversion of He(2^1P) to He(2^1S) is due to radiative transitions. The theoretical excited state populations per ion pair formed were taken from Refs. 4 and 7. Figure 3 strongly suggests that two-body conversion of 2^1P to 2^1S does indeed occur.

In conclusion, we have shown that the RIS technique can be extended to situations where the final step is collision induced. Associative ionization is one collision mechanism; Penning ionization of an impurity and charge transfer to electronegative impurities may provide other useful mechanisms. Utilization of these collision processes in resonance ionization spectroscopy is very important because it makes possible the use of lasers having rather modest energy per pulse.

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Surface Magnetic Confinement*

A. Y. Wong, Y. Nakamura B. H. Quon, and J. M. Dawson
Department of Physics, University of California, Los Angeles, California 90024
(Received 28 April 1975)

We discuss advantages of surface confinement such as plasma stability, direct injection of ion beams, electrostatic confinement, negligible synchrotron loss, and "geometric mirror." Preliminary experimental results are presented.

We describe a new class of large-scale surface-magnetic-confinement devices (Surmac)[1] with characteristic dimensions of 10 m or larger, which hold promise as confinement systems for fusion plasmas. There are a number of configurations of the surface fields, e.g., the closed