$-\vec{q}$ with q from (2) one can extract the initial momentum k_i of the electron from a measurement of $\vec{k}_f$. This is true since the dominant contribution to the cross section comes from $q \approx q_{min}$ which means that $\vec{q}$ lies in the beam direction. We detected electrons with k_i in the region 580 $Å^{-1} \lesssim k_i \lesssim 690$ $Å^{-1}$ which is approximately four times the Bohr momentum of a K electron for $Z = 87$. The measured spectrum can be explained by means of the K-shell momentum distribution of the quasimolecule.

Since the efficiency of the β spectrometer can be improved by mounting it axially to the beam and using a larger counter in the focal plane, the sensitivity of the setup would be sufficient for cross sections which are more than an order of magnitude smaller than those in the present measurement. Thus it might be possible to study the electronic properties of the U + U quasimolecule, as planned by a Heidelberg-Munich collaboration in order to study QED effects in high fields.[15,16]

[1]E. Merzbacher and H. W. Lewis, in *Encyclopedia of Physics*, edited by S. Flügge (Springer, Berlin, 1958), Vol. 34, p. 166.

[2]F. Folkmann, K. O. Groeneveld, R. Mann, G. Nolte, S. Schumann, and R. Spohr, Z. Phys. A275, 229 (1975).

[3]T. Huus and J. H. Bjerregård, Phys. Rev. 92, 1579 (1953).

[4]C. Župančič and T. Huus, Phys. Rev. 94, 206 (1954).

[5]H. M. Sakai, *Future of Nuclear Structure Studies* (International Atomic Energy Agency, Vienna, 1969), p.57.

[6]E. Moll and E. Kankeleit, Nucleonics 7, 180 (1965).

[7]V. Metag, D. Habs, H. J. Specht, G. Ulfert, and C. Kozhuharov, Hyperfine Interact. 1, 405 (1976).

[8]D. R. Bates and R. McCarroll, Proc. Roy. Soc. London, Ser. A 245, 175 (1958).

[9]J. S. Briggs, J. Phys. B 8, L485 (1975).

[10]M. R. C. McDowell and J. P. Coleman, *Introduction to the Theory of Ion-Atom Collisions* (North-Holland, Amsterdam, 1970).

[11]D. H. Jakubassa, Phys. Lett. 58A, 163 (1976).

[12]F. Rioux and P. Kroger, Am. J. Phys. 44, 56 (1976).

[13]D. H. Jakubassa, to be published.

[14]P. A. Amundsen, J. Phys. B 9, 971 (1976).

[15]A. I. Akiezer and V. B. Berestetskii, *Quantum Electrodynamics* (Wiley, New York, 1965).

[16]B. Müller, H. Peitz, J. Rafelski, and W. Greiner, Phys. Rev. Lett. 28, 1235 (1972).

Nanosecond Time-Resolved Spectroscopy of the n=2 Levels in a High-Pressure He Discharge

J. E. Lawler, J. W. Parker, and L. W. Anderson
Department of Physics, University of Wisconsin, Madison, Wisconsin 53706

and

W. A. Fitzsimmons
National Research Group Inc., Madison, Wisconsin 53705

(Received 19 July 1977)

Nanosecond time-resolved absorption spectroscopy is used to study the populations of the 2^1P, 2^3P, 2^1S, and 2^3S levels in a high-pressure He discharge. At high pressures, the population of the 2^1P level decays primarily through three-body molecular formation involving two ground-state He atoms at a rate of $(3.5 \pm 0.8) \times 10^{-31}$ cm^6 sec^{-1}. The population of the 2^3P level decays primarily as a result of mechanisms other than collisions involving ground-state He atoms.

We report an experimental investigation of the time dependence of the populations of the 2^1P, 2^3P, 2^1S, and 2^3S levels in the afterglow of a pulsed He discharge at pressures of 50–3000 Torr. The populations of the metastable 2^1S and 2^3S levels have long been recognized to play an important role in the kinetics of the He afterglow.[1] The populations of the 2^1P and 2^3P levels are also quite important in the afterglow of a fast pulsed discharge. The fast pulsed discharge is characterized by relatively high pressures, a short voltage rise time, and a high value of the electric field divided by the pressure at breakdown. This leads to high initial densities of excited states and free electrons in the afterglow.

Immediately following the pulsed discharge, the populations of all the $n = 2$ levels are comparable. Radiation trapping can maintain significant populations of the 2^1P and 2^3P levels for many radiative lifetimes. The kinetics of these levels is not well understood, but is interesting and important for many reasons. For instance, it has been

suggested that the 2^1P population is the precursor of some of the vacuum-ultraviolet molecular radiation from He.[2,3] The nanosecond time-resolved absorption spectroscopy reported in this Letter makes possible the mapping of absolute populations of the nonmetastable as well as the metastable levels on a time scale characteristic of the decay time of the nonmetastable levels.

We report data demonstrating that the 2^1P level is depopulated at pressures above 150 Torr primarily by three-body molecular formation involving two ground-state He atoms at a rate of $(3.5 \pm 0.8) \times 10^{-31}$ cm^6 sec^{-1}. We also report data indicating that the 2^3P level is depopulated by radiating to the 2^3S level and possibly by collisional deexcitation due to electron impact. There is no evidence for significant depopulation of the 2^3P level by collisions with ground-state He atoms at pressures up to 300 Torr.

Figure 1(a) is a schematic of the experimental apparatus. The heart of the experiment is a commercially available National Research Group Inc. twin-pulsed-discharge laser. This device has two energy-storage capacitors and discharge channels switched by a common spark gap. The jitter between breakdown in the two channels is less than 1 nsec. One channel contains pure He (99.995%); the other channel is used as a N_2 laser to pump a narrow-banded tunable dye laser.[4] The dye-laser-pulse duration of 3 nsec determines the temporal resolution of the experiment. The spectral bandwidth of the pressure-tuned, etalon-narrowed dye laser is 1.7 GHz. The output of the dye laser is collimated by a beam-expanding telescope, and reflected repeatedly in an optical delay in order to provide an adjustable delay of 0–500 nsec. After being delayed by a given time, the dye-laser pulse is attenuated, passed through the He discharge, and detected by a photomultiplier. Time-resolved absorption spectroscopy is performed during the first 500 nsec of the afterglow with 3-nsec temporal resolution.

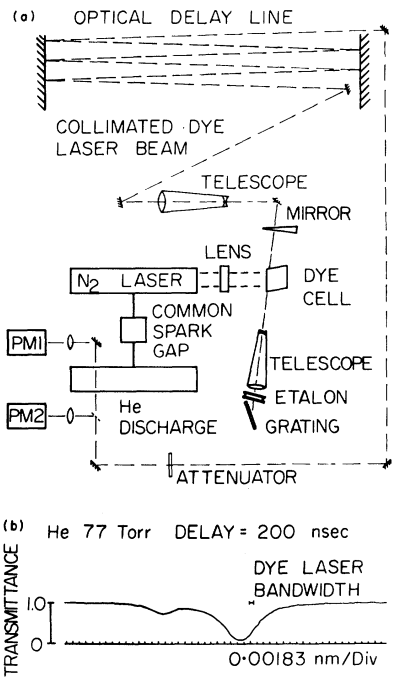

FIG. 1. (a) Schematic of the experiment. PM1 and PM2 are signal and reference photomultipliers, respectively. (b) Absorption spectrum at $\lambda = 447.1$ nm. The dye laser is pressure tuned across the line.

Figure 1(b) shows the experimental absorption profile of the $2^3P \rightarrow 4^3D$ transition recorded as the dye-laser wavelength was scanned across the line. The He lines are all pressure broadened at the pressures used in this experiment. As indicated in Fig. 1, the ratio of the width of the He line to the dye-laser bandwidth is large at 77 Torr; it is even larger at higher pressures. The population of a particular level is determined from the equivalent width of an absorption line using curves of growth calculated from experimentally determined line shapes and accurately known oscillator strengths.[5] The populations of the 2^1P, 2^3P, 2^1S, and 2^3S levels were measured using the 492.2-, 447.1-, 501.6-, and 388.9-nm lines, respectively. The upper levels of these transitions are relatively unoccupied due to rapid associative ionization.[6,7]

Figure 2 shows the observed populations of the various $n = 2$ levels of He as a function of time at a He pressure of 77 Torr. All absorption measurements are made along the same path through the discharge channel. The populations of all of the $n = 2$ levels decay exponentially as a function of the time except that of the 2^3S metastable level which increases initially with time after breakdown. Many nanoseconds after breakdown the population of the 2^3S level reaches a maximum and then decays very slowly. The decay of the 2^3S population is negligible on our time scale. Radiative cascade from higher states can be ignored in a first approximation at the pressures used in this experiment because of the rapid rate of associative ionization of atoms in all states above the $3S$ levels.[6,7]

The decay rates of the populations of the 2^1S, 2^1P, and 2^3P levels as a function of pressure and of pressure squared are shown in Figs. 3(a) and

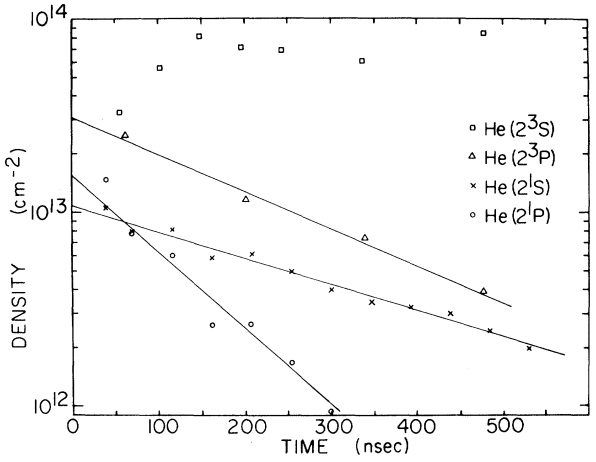

FIG. 2. Populations of the various $n = 2$ levels of He as a function of time after breakdown in 77 Torr of He. The discharge is approximately 1 cm thick along the absorption path.

3(b), respectively. The decay rates of the 2^1S population are consistent with the metastable spin-exchange reaction discovered by Phelps,[1]

$$\mathrm{He}(2^1S) + e^- \rightarrow \mathrm{He}(2^3S) + e^-. \quad (1)$$

The weak pressure dependence of the 2^1S decay rate may be due to a variation of the electron concentration with pressure. The rate of the metastable spin-exchange reaction has been determined by Phelps to be 3.5×10^{-7} cm^3/sec at an electron temperature of 300°K. If this rate is relatively independent of the electron temperature, the electron concentration can be inferred from the observed decay rates of the 2^1S population. The 2^1S decay rates indicate an electron concentration in the range $(1-3)\times10^{13}$ cm^{-3}.

The decay rate of the 2^1P population is strongly pressure dependent. The data of Bartell, Hurst, and Wagner[2] on the decay of the 2^1P population are also shown in Fig. 3(a). They described the pressure dependence of the 2^1P-population decay rate by a polynominal of the form $A + Bp + Cp^2$, where p is the pressure of the He gas. They identified the constant term, A, as resulting from radiative transitions to the 2^1S state at the natural decay rate of 2×10^6 sec^{-1} plus leakage of the highly trapped 58.4-nm resonance radiation. This leakage term represented a pressure-independent decay rate of approximately 1.0×10^6 sec^{-1} in their system.[2] The quadratic term in pressure was identified as resulting from three-body molecular formation,[2]

$$\mathrm{He}(2^1P) + 2\mathrm{He}(1^1S) \rightarrow \mathrm{He}_2(B \text{ or } D) + \mathrm{He}(1^1S). \quad (2)$$

In a later paper Payne, Klots, and Hurst[3] proposed that the linear term was caused by a two-body mechanism,[3]

$$\mathrm{He}(2^1P) + \mathrm{He}(1^1S) \rightarrow \mathrm{He}(2^1S) + \mathrm{He}(1^1S). \quad (3)$$

It has been suggested that this mechanism accounts for over 60% of the 2^1P decay rate in the 100–200-Torr range.[3,8] We do not observe the increase in the 2^1S population that would result if

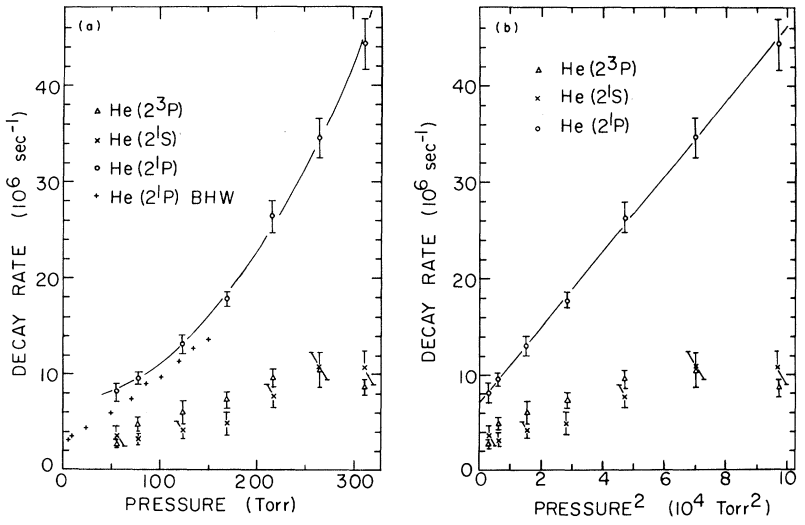

FIG. 3. Decay rates of the populations of the 2^1S, 2^3P, and 2^1P levels as a function of (a) pressure, and (b) pressure squared. BHW refers to the data of Bartell, Hurst, and Wagner (Ref. 2). The curves represent a least-squares fit with $A + Bp + Cp^2$.

most of the 2^1P states were converted into 2^1S states by Reaction (3), but instead observe only an exponential decay for the 2^1S state.

By fitting our data for the decay rate of the 2^1P population with the expression of the form $A + Bp + Cp^2$, we determined that $A = (7.0 \pm 2.0) \times 10^6$ sec^{-1}, $B = (2.2 \pm 28) \times 10^3$ sec^{-1} Torr^{-1}, and $C = 384 \pm 90$ sec^{-1} Torr^{-2}. Using the theory of Holstein,[9] we estimate that the leakage of 58.4-nm resonance radiation in our system represents a pressure-independent decay rate of 0.6×10^6 sec^{-1}. This value was calculated using the dimensions of our discharge channel (2.2 cm $\times$ 3.3 cm $\times$ 61.0 cm) and the self-broadening constant for the 2^1P level measured by Camm and Copley.[10] Radiative transitions to the 2^1S state represent a decay rate of less than 2×10^6 sec^{-1} as a result of radiation trapping. We cannot completely account for A with radiative processes; it is possible that collisional de-excitation due to electron impact also contributes to the pressure-independent term in the decay rate.

Our best value for B is 25 times smaller than that determined previously,[3,8] although the uncertainty is large. Primarily because there is no increase in the 2^1S population after breakdown and also because of the small value of B we conclude that the rate of Reaction (3) is at least a factor of 5 smaller than that reported previously.[3,8] The quadratic term is important for all pressures in the range 50–300 Torr and is dominant above 150 Torr. We interpret this term as a rate for Reaction (2). At the operating temperature of our system, 19°C, this term represents a molecular-formation rate of $(3.5 \pm 0.8) \times 10^{-31}$ cm^6 sec^{-1}. The possible pressure dependence of the electron concentration causes some uncertainty in this interpretation, but the pressure dependence of the decay rate of the population of the 2^1P level is so different from the pressure dependence of the other decay rates that the existence of the dominant quadratic term is clear.

The population of the 2^3P level is observed to decay at a rate less than the radiative-decay rate at low pressures. This indicates that the 2^3S metastable density in the afterglow is sufficiently high to cause trapping of the 1.08-μm radiation from the 2^3P level. At higher pressures the linewidth of this transition is broadened so that radiation trapping is reduced. The decay rate of the 2^3P population approaches the natural radiative-decay rate at high pressures. It is possible that variations in the electron concentration also contribute to the increase with pressure in the decay rate of the 2^3P population through inelastic electron collisions. The leveling off of the decay rate at high pressures indicates that conversion of 2^3P to 2^3S through ground-state collisions, as suggested previously,[3] is not a dominant mechanism.

In summary, the 2^1P population does not decay at high pressures primarily by conversion to the 2^1S state through ground-state collisions, but rather by molecular formation at a rate of $(3.5 \pm 0.8) \times 10^{-31}$ cm^6 sec^{-1}. The absence of any increase in the 2^1S population after breakdown and the quadratic pressure dependence of the decay rate of the 2^1P population support this conclusion. For pressures up to 300 Torr the 2^3P population does not decay by conversion to the 2^3S level through ground-state collisions. The leveling off at high pressures of the decay rate of the 2^3P population supports this conclusion. The 2^3P population decays primarily by radiating to the 2^3S level although inelastic electron collisions may play a role.

We acknowledge the generous loan of a twin laser by National Research Group, Inc. We also acknowledge research support from the University of Wisconsin Research Committee with funds from the Wisconsin Alumni Research Foundation.

[1]A. V. Phelps, Phys. Rev. 99, 1307 (1955).
[2]D. M. Bartell, G. S. Hurst, and E. B. Wagner, Phys. Rev. A 7, 1068 (1973).
[3]M. G. Payne, C. E. Klots, and G. S. Hurst, J. Chem. Phys. 63, 1422 (1975).
[4]J. E. Lawler, W. A. Fitzsimmons, and L. W. Anderson, Appl. Opt. 15, 1083 (1976).
[5]W. L. Wiese, M. W. Smith, and B. M. Glennon, *Atomic Transition Probabilities*, U.S. National Bureau of Standards, Reference Data Series—4 (U.S. GPO, Washington, D.C., 1966), Vol. I, pp. 11–14.
[6]J. A. Hornbeck and J. P. Molnar, Phys. Rev. 84, 621 (1951).
[7]H. F. Wellenstein and W. W. Robertson, J. Chem. Phys. 56, 1077 (1972).
[8]M. G. Payne, G. S. Hurst, M. H. Nayfeh, J. P. Judish, C. H. Chen, E. B. Wagner, and J. P. Young, Phys. Rev. Lett. 35, 1154 (1975).
[9]C. van Trigt, Phys. Rev. 181, 97 (1969).
[10]D. M. Camm and G. H. Copley, J. Quant. Spectrosc. Radiat. Transfer 13, 1251 (1973).