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Absolute Cross Sections for the Photoionization of He($n^{1,3}P$) Atoms*

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A molecular-beam apparatus is used in conjunction with a tunable laser to determine absolute cross sections for the photoionization of helium 3^1P, 4^1P, 5^1P, 3^3P, 4^3P, and 5^3P atoms excited optically from the $2^{1,3}S$ metastable levels. The photoionization cross section for each of these states is determined at the wavelength used for its excitation from the metastable state. The results are compared with those of recent unpublished calculations.

Photoionization of excited states of helium plays an important role in the transfer of radiation through laboratory plasmas and hot stellar atmospheres. Experimental[1] and theoretical[2-5] studies of the photoionization of He(2^1S) and He(2^3S) metastable atoms have already been reported. The present paper reports measurements of the absolute cross sections for photoionization of helium atoms in the 3^1P, 4^1P, 5^1P, 3^3P, 4^3P, and 5^3P levels and compares these results with those obtained theoretically.[6,7]

The apparatus used in these measurements has been described in detail elsewhere[1,8] and only a brief description is included here. A mixed beam of helium $2^{1,3}S$ metastable atoms is produced by electron-impact excitation. The He(2^1S) metastable atoms in the beam may be selectively quenched using a helium discharge lamp; the separate effects appropriate to each metastable species are then obtained from observations with the helium lamp alternately on and off. After removal of any charged particles the metastable beam is irradiated by the output beam of a pulsed laser. Ions so formed are extracted by an electric field and detected by a Johnston particle multiplier. A gating technique is used to distinguish these ions from those liberated at surfaces by scattered laser light and those arising from chemi-ionization in collisions between metastable atoms and residual gas in the system. Knowledge of the metastable-atom beam flux,[9] cross section, and velocity distribution[10] then allows the determination of the number density N_m of the metastable atoms in the interaction region.

The wavelengths of the radiation required to excite the transitions to the various P levels are listed in Table I. This radiation is generated by a nitrogen-pumped tunable dye laser system operating at 10 pulses per second.[11] The wavelengths above 3600 Å are generated directly while the shorter wavelengths are obtained by frequency doubling.[12] The present laser system provides linearly polarized radiation with an intensity of several megawatts per square centimeter per angstrom at the interaction region. The width and shape of the laser pulse is determined using a fast biplanar diode and oscilloscope. The pulse width is of the order of 4 nsec full width at half-

TABLE I. Summary of results.

TRIPLET SYSTEM

n	excitation wavelength Å	lifetime μ sec	oscillator strength for He 2^3S-n^3P transitions	cross section cm^{2a} present experiment	quantum defect calculation[b]	close coupling calculation[c] length approx[II]	close coupling calculation[c] velocity approx[II]
3	3888.6	1.05	6.7×10^{-2}	$8.7 \pm 2.0\ 10^{-18}$	7.9×10^{-18}	7.5×10^{-18}	
4	3187.7	1.98	2.3×10^{-2}	$2.1 \pm 0.4\ 10^{-18}$	2.1×10^{-18}	1.8×10^{-18}	2.1×10^{-18}
5	2945.1	3.41	1.14×10^{-2}	$8.6 \pm 1.8\ 10^{-19}$	8.8×10^{-19}		

SINGLET SYSTEM

n	excitation wavelength Å	lifetime n sec	oscillator strength for He 2^1S-n^1P transitions	cross section cm^{2a} present experiment	quantum defect calculation[b]	close coupling calculation[c] length approx[II]	close coupling calculation[c] velocity approx[II]
3	5015.6	1.7	1.5×10^{-1}	$1.0 \pm 0.2\ 10^{-17}$	1.1×10^{-17}		
4	3964.7	4.1	5.07×10^{-2}	$2.2 \pm 0.4\ 10^{-18}$	2.4×10^{-18}	2.4×10^{-18}	2.5×10^{-18}
5	3613.6	7.8	2.21×10^{-2}	$8.8 \pm 1.8\ 10^{-19}$	9.7×10^{-19}		

[a] The experimental cross sections are obtained using linearly polarized light. The calculated cross sections with which they are compared are therefore of the form $Q = 4\pi^2 \alpha a_0^2 (3df_S/dE + 1.2\, df_D/dE)$.

[b] Ref. 7.

[c] Ref. 6.

maximum. A thermopile of known absolute sensitivity is used to measure the mean power and hence mean photon flux in the laser beam.[1]

The experimental method may be understood as follows. If the laser output is tuned to one of the absorption frequencies of the mixed metastable beam, for instance that for $2^3S \rightarrow 3^3P$, transitions of the type

$$\mathrm{He}(2^3S) + h\nu\ (\lambda = 3888.6\ \text{Å}) \rightarrow \mathrm{He}(3^3P)$$

will occur. Some atoms in the 3^3P state will then be subsequently ionized during the remainder of the same laser pulse,

$$\mathrm{He}(3^3P) + h\nu\ (\lambda = 3888.6\ \text{Å}) \rightarrow \mathrm{He}^+ + e;$$

and thus the ion count per pulse, S (typically 0.1 to 0.005), is related to the photoionization cross section Q by the relation

$$S = Qlk \int_{\text{pulse}} F(t) N_P(t)\, dt, \qquad (1)$$

where $F(t)$ is the instantaneous photon flux, $N_P(t)$ the instantaneous density of P atoms, and l the path length of the photons through the beam. k, the efficiency with which the signal ions are detected, is not measured but is taken as 0.95 ± 0.05.[13]

$N_P(t)$ is not directly measurable but its value may be inferred when a sufficiently intense laser beam is used because saturation of the transition will occur essentially instantaneously. For linearily polarized radiation, only $\Delta m_l = 0$ transitions are allowed and at saturation the number density of helium atoms in the P state will be equal to the density in the lower S state. If the natural lifetime of the P state is long compared to the duration of the laser pulse, as is the case for the $n\,^3P$ levels studied (see Table I), $N_P(t)$ may be assumed constant at half the original metastable atom density,

$$N_P(t) = \tfrac{1}{2} N_m(0),$$

whereupon (1) reduces to

$$S = Qlk\, \tfrac{1}{2} N_m(0) \int_{\text{pulse}} F(t)\, dt$$

and $\int_{\text{pulse}} F(t)\, dt$ is simply the total photon flux per pulse. The $n\,^1P$ levels studied, however, have very short natural lifetimes, and the value of $N_P(t)$ is not constant during the laser pulse but decreases because of spontaneous decay as

$$N_P(t) = N_P(0) e^{-t/2\tau},$$

where $N_P(0) = \tfrac{1}{2} N_m(0)$ and τ is the natural lifetime. The factor 2 arises because the loss of atoms from the ^1P state is partly compensated by the excitation of further ^1P states from the metastable level in order to maintain saturation of the

transition. The ^1P atom density $N_P(t)$ may therefore be obtained as a function of time during the laser pulse. Since both the laser pulse shape and total energy per pulse are known it is also possible to determine $F(t)$, thereby permitting the integral in (1) to be evaluated and Q determined.

The foregoing analysis assumes that saturation of the transition occurs instantaneously. Calculations showed that this should indeed be the case with the present laser powers, but it was also demonstrated experimentally for each upper level by observing that the ion production rate was proportional to the photon flux F over a wide range of laser powers. Had saturation not been achieved, the number density of P states produced would be proportional to F and the ion production rate therefore proportional to F^2. The observed saturation of the transition implies that coherence effects, which may result in an oscillatory behavior of the populations in each state, are unimportant.[14] This is to be expected because the laser output power varies from pulse to pulse and because wave-front distortions within the laser cavity result in an output beam containing radiation of many relative phases. Thus even if such coherence effects were present, they would in fact average to the assumed case over many laser pulses.

The oscillator strengths for transitions to upper $n^{1,3}P$ states decrease with increasing n (see Table I), and with the present laser it is possible to saturate transitions only to the levels reported here.

The experimental results are listed in Table I. Systematic uncertainties in the measurements arise primarily from two sources. The first results from the difficulty in accurately determining the width and shape of the laser pulse which are required to derive $F(t)$. The second major uncertainty arises in the determination of N_m. The metastable flux is determined by measuring the current of secondary electrons ejected from a stainless-steel surface whose absolute secondary-electron ejection coefficient γ is known from a previous *in situ* measurement using a gas-cell technique similar to that described elsewhere.[9] It has, however, recently been observed in this laboratory that the measured value of γ for He(2^3S) atoms [but not for He(2^1S) atoms] depends slightly on the gas employed in the cell. As a result a larger uncertainty is ascribed to the triplet γ than to the singlet γ. All anticipated sources of uncertainty are summarized in Table II and are seen to lead to an rms error in the cross section of ~ 20%.

TABLE II. Summary of experimental errors.

	Singlet	Triplet
Correction for P-state decay during the laser pulse	±10%	0
Uncertainties in γ	± 8%	±15%
Velocity determination	± 5%	± 5%
Absolute power determination	± 8%	± 8%
Ion detection efficiency	± 5%	± 5%
Statistical error	±10%	±10%
rms error	±20%	±21%

The experimental conditions are such that atoms are excited and subsequently ionized by linearly polarized light before the polarization transferred to them upon excitation is altered by collisional relaxation, external magnetic fields, or, in the case of triplets, spin-orbit interactions. As a consequence the measured cross sections must be identified not with the cross sections appropriate to unpolarized light and atoms, viz.,

$$Q = 4\pi^2\alpha^2 a_0^2 (df_S/dE + df_D/dE),$$

but rather with[15]

$$Q = 4\pi^2\alpha^2 a_0^2 (3\,df_S/dE + 1.2\,df_D/dE),$$

where α is the fine structure constant, a_0 is the Bohr radius, and df_S/dE and df_D/dE are the S and D partial-wave oscillator strengths for photoionization from the unpolarized $n^{1,3}P$ states. Values of Q determined using the partial-wave oscillator strengths recently calculated by Hartquist and Lane[7] using a quantum-defect procedure and by Jacobs[6] using a close-coupling calculation are included in Table I. Excellent agreement between experiment and both theories is observed.

It is hoped to extend the present measurements to higher P levels and to improve the precision of the data by the use of a more powerful, narrower-linewidth laser system. Use of a circularly polarized laser beam would also allow the unique determination of the D-wave contribution to the total cross section for photoionization of P states.[15] It should also be possible to excite transitions of the type He(2^3S) → He(2^3P) using light from a helium resonance lamp, and then to study the photoionization of the He(2^3P) atoms as a function of wavelength.

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Renormalization-Group Calculations of Divergent Transport Coefficients at Critical Points

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Recursion relations correct to lowest order in $\epsilon = 4 - d$ are obtained for simple models representing the critical dynamics of a binary liquid, a Heisenberg antiferromagnet, superfluid helium, and related systems. The exponents for the divergences of the transport coefficients satisfy the "scaling" relations predicted by mode-mode coupling theory. The viscosity of the binary liquid is predicted to diverge as $\kappa^{-\epsilon/19}$ near $d = 4$, which is a new result.

Recently the renormalization-group method[1] has been employed to calculate dynamic critical exponents for time-dependent Ginzburg-Landau models with Hermitian Liouville operators.[2] As pointed out by Kawasaki and by Kadanoff and Swift,[3] the transport coefficients in such models cannot diverge; on the other hand, more general models, with non-Hermitian Liouville operators, may possess divergences in their transport coefficients, brought about by the nonlinear couplings of their hydrodynamic modes near the critical point.[4,5] We have constructed two simple models of the latter type—the first representing the phase-separation point of a binary fluid or the critical point of a pure fluid, and the second a "planar ferromagnet," a Heisenberg antiferromagnet, or superfluid helium. Analysis of these models using the renormalization-group method and the ϵ expansion[1,2] yields divergent transport coefficients and scaling relations, which agree in most respects with the predictions of the mode-mode coupling approach of Kawasaki,[4] Kadanoff and Swift,[5] and others. In addition we have obtained values for the exponents of the transport coefficients in the binary liquid near four dimensions, which are new results.

Model I is defined by the equations

$$\frac{\partial \psi}{\partial t} = \lambda_0 \nabla^2 \frac{\delta F}{\delta \psi} - g_0 \nabla \psi \cdot \frac{\delta F}{\delta \vec{j}} + \theta, \tag{1a}$$

$$\frac{\partial \vec{j}}{\partial t} = \left(\eta_0 \nabla^2 \frac{\delta F}{\delta \vec{j}} + g_0 \nabla \psi \frac{\delta F}{\delta \psi} + \vec{\zeta} \right)_{TP}, \tag{1b}$$

$$F = \int d^d x \left(\tfrac{1}{2} r_0 \psi^2 + \tfrac{1}{2} |\nabla \psi|^2 + u_0 \psi^4 + \tfrac{1}{2} j^2 \right), \tag{1c}$$

where $\psi(\vec{x}, t)$ is a scalar order parameter, $\vec{j}(\vec{x}, t)$ is the transverse part (TP) of a d-component vec-