Experimental and Theoretical Studies of the Argon Pre-Breakdown Discharge.

Ian McKellar Stewart B.Sc.(Hons.) (UWA)

A thesis submitted for the degree of Doctor of Philosophy of the University of New England.

August 1995

Abstract.

This work concerns the use of an optical absorption technique to measure the concentration of metastable excited argon atoms within a 'chopped' prebreakdown or Townsend discharge. A diode laser tuned to the argon $1s_5 - 2p_9$ transition at 811.5 nm was used as the light source. The absorption was measured with a spatial resolution of less than 2 mm and a time resolution of as low as 0.1 ms. The reduced diffusion coefficient ND_m of the 1s₅ state was determined by measurements of the decay of the discharge current in the afterglow. A value of $(1.57 \pm 0.05) \times 10^{-18}$ cm⁻¹s⁻¹ was returned, which is consistent with other published values. An approximate value of the efficiency with which the 1s5 atoms eject secondary cathodic electrons was determined from spatially-resolved measurements of the quasi-steady-state concentration of these atoms. The resulting value of 85% is broadly consistent with published values. The primary ionisation coefficient α_i/N was determined at 454 Td of reduced electric field by spatiallyand time-resolved measurements of the 1s5 concentration in the first millisecond after the discharge was switched on. The measured value of 6×10^{-17} cm² is about 25 % lower than the previously accepted value. Measurements were also made from 36 to 82 Td, using just the current passed by the discharge: these values were about 10% lower than the canonical values. The 1s₅ excitation coefficient α_m/N was measured at the same pressure and field using both the quasi-static technique and the time-resolved technique, returning values of 8.0 and 7.3×10^{-18} cm² respectively. The only previously published data report a value of 27×10^{-18} cm² for this quantity.

In the accompanying theoretical sections, a boundary condition for the metastable-particle diffusion equation is derived; the occurrence of complex exponents in the Molnar analysis is investigated; a simple relation is derived which describes the saturation of the absorption coefficient for a narrow-band light source restricted to a thin beam; the electron diffusion equation is solved for the case of a spatially extended source of cathode current, leading to useful techniques for extracting electron transport coefficients from optical absorption measurements; a spatially resolved model of the steady-state distribution of metastable particles is developed; and techniques are described for the analysis of data comprising multi-exponential decays plus noise.

"Nec dubitamus multa esse quae et nos praeterierint; homines enim sumus et occupati officiis." Pliny, Preface, Historia Naturalis.

Acknowledgments:

I have found nearly all the staff of the Physics Department to be, at all times, both friendly and helpful, so I would like to extend a general "thank-you" to all members. There are, however, several people whose advice, example or support deserve, I feel, a more particular recognition. In alphabetical order, these are: Noel and Julie Adams, for being the kindest of neighbours, but also for setting an example in other ways; Ian Craven, for unstinting help, in both material and information; Allan Ernest, for his constant friendship, and for much enjoyable fat-chewing over many beers; Vernie Everett, for warming my winters, hardening my hands and cosseting my computers; my supervisor, Matthew Fewell, for his unfailing support, but perhaps particularly for his perennial willingness to listen to half-baked ideas; Frank Hibberd for much unsolicited help in the hunt for jobs; Colin Sholl for a great deal of fatherly advice and, together with Brian Seppelt, for allocating to me a scandalously small amount of "officiis"; Kuni Tachibana, for his generous loan of equipment, and for showing me a more interesting research direction; Michael Van der Valk, for being a good friend and a mine of technical lore of all kinds, but particularly for his patient and generous loan of tools, time and bench space; and, last but not least, Gerry Woolsey for his constant encouragement, and for generally taking an interest in my progress above and beyond the call of duty.

Finally, I would particularly like to thank my wife, for putting up with me, and for not being too much of a crustacean herself.

Abstract.....iii Acknowledgments.....iv 2.2.2. Generation of secondary electrons by the flux of excited atoms 2.2.3. Models of metastable-particle number densities in the steady 3.2.2. New results obtained using the method of separation of variables.41 3.2.2.2. General solution of the time-dependent diffusion equation after the primary current is extinguished......43 3.2.2.4. Complex components in Molnar's analysis......54

Table of Contents.

4. Laser perturbation of the discharge60
4.1. Energy levels and collisional coupling in argon
4.1.1. Free decay, no laser perturbation
4.1.2. Steady state, no laser perturbation
4.1.3. Laser perturbation of the steady-state discharge65
4.2. The absorption coefficient, and new results concerning saturation of the
absorption under the present experimental conditions
4.2.0. Introduction
4.2.1. Steady-state, uniform discharge perturbed by spatially uniform
laser light69
4.2.1.0. The weak-field limit72
4.2.1.1. Saturation of absorption in the absence of cross-
relaxation74
4.2.1.2. Saturation of absorption in the case of significant cross-
relaxation75
4.2.2. Steady-state, uniform discharge perturbed by a narrow beam78
4.3. Modifications to the free-decay theory
4.4. Conclusion
5. Theoretical background to optical measurements of electron transport properties
in a discharge with a non-point source of current96
5.1. Properties of the Hankel transform
5.2. Discharge geometry and processes
5.3. The electron diffusion equation; boundary conditions100
5.4. A new method of solving the diffusion equation with an extended source of
current102
5.4.1. General solution
5.4.2. Effects of the variation of α_i near the cathode106
5.4.3. Approximate solutions109

6. A new model of the steady-state distribution of metastable particles in a dis	charge
with cylindrical symmetry.	114
6.1. Principal equations and methods of solution	115
6.2. Finite-difference approximation.	117
6.3. Conversion of the metastable-particle diffusion equation to integral	
form	119
6.4. An integral equation in the metastable-particle flux density $\phi_{\rm m}(r,0)$	120
6.5. Fourier-Bessel expansion.	122
6.6. The effect on the growth of current of a radial variation in γ_m	131
6.7. Conclusion.	137
7. Experimental apparatus	139
7.1. The vacuum system and ionisation chamber.	139
7.2. Signal production and acquisition	144
7.2.1. Measurements of discharge current	144
7.2.2. The laser absorption experiment	147
7.2.2.1. Laser control circuitry.	147
7.2.2.2. Detection of the absorption signal	155
8. Methods of digital signal processing.	157
8.1. Removal of 'droop'.	157
8.2. Improving the signal-to-noise ratio.	159
8.2.1. An outline of a new, iterative, nonstationary, distortionless fi	lter.161
8.3. Curve-fitting techniques	163
9. Measurements of the discharge current.	165
9.1. Measurement of the primary and secondary ionisation coefficients	165
9.2. Time-dependent measurements.	175
9.2.1. Difficulties with the measurements	176
9.2.2. The diffusion and quenching coefficients	184
9.3. Electrical breakdown in argon	186
10. Optical absorption measurements	189
10.1. Saturation of the absorption coefficient	190

10.2. Measurement of the diffusion and quenching coefficients	194
10.3. Steady-state concentrations of metastable atoms	197
10.4. Rates of production of metastable atoms	201
11. Conclusion	206
Appendix A. Optimum depth of absorption features in a wavelength reference	211
Appendix B. The curve-fitting program.	212
Bibliography	217

Tables.

2.1. Values of γ_p	18
2.2. Values of $\gamma_{\rm m}$	19
2.3. Values of γ_i	20
3.1. Alternative expressions for the coefficient β	31
4.1. Characteristics of the argon 1s levels	94
4.2. The ratios C_{ji}/C_{ij}	94
4.3. Rates of destruction of the argon 1s levels at 1 Torr, 300 K	95
4.4. Some characteristics of selected 1s - 2p transitions in argon	95
9.1. Values of the diffusion coefficient of argon 1s5 metastables at 300 K	186