Interaction of resonance radiation with atomic beams

by Steven Charles Seitel

A thesis submitted to the Graduate Faculty in partial fulfillment of the requirements for the degree of DOCTOR OF PHILOSOPHY in Physics
Montana State University

Abstract:
A dual atomic beam device for investigating the resonance transitions in the rare gases is described. The device is useful wherever low beam densities or windowless light paths are desired. A simple model is developed for the frequency distribution of a resonance line excited by electron bombardment in an atomic beam light source. The model is used to interpret the observed absorption and scattering of the 1048 å (3Pi) and 1067. å (3Pi) argon resonance lines. The ratio of the oscillator strengths of these lines is measured by a new method with the result f(1048)/f(1067) = 3.99+0.55 in agreement with values given in the literature, The electron impact excitation functions for these lines are observed for the first time and compared to a recent theoretical treatment.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

ABSTRACT

A dual atomic beam device for investigating the resonance transitions in the rare gases is described. The device is useful wherever low beam densities or windowless light paths are desired. A simple model is developed for the frequency distribution of a resonance line excited by electron bombardment in an atomic beam light source. The model is used to interpret the observed absorption and scattering of the 1048 Å ($^1P_1^o$) and 1067 Å ($^3P_1^o$) argon resonance lines. The ratio of the oscillator strengths of these lines is measured by a new method with the result f(1048)/f(1067) = 3.99±0.55 in agreement with values given in the literature. The electron impact excitation functions for these lines are observed for the first time and compared to a recent theoretical treatment.

I. INTRODUCTION

Atomic first resonance transitions are of interest for the information they provide about the lowest-lying excited states. The oscillator strengths are intimately related to the lifetimes[1] and to the small-angle inelastic electron-scattering cross sections.[2] In the rare gases, the resonant transitions occur in the vacuum ultraviolet spectral region[3] where special optical techiques are required.

Two types of vacuum ultraviolet light sources have recently been described in the literature: Verkhovtseva, et. al.,[4] employ an ultrasonic gas jet excited by a high-energy electron beam; Govertsen and Anderson[5] use a collimated atomic beam excited by electron bombardment to produce narrow spectral lines. Because of the low atom densities and the ability to operate near threshold energies, the latter source is uniquely suited to an investigation of resonant transitions.

A feature common to sources of this type is the presence of "background" atoms in the region where the beam is excited. If these atoms are of the same species as the beam atoms and have large velocity components in the direction of observation, the background emission takes the form of a broad spectral line superimposed upon the narrow beam line.[6] The presence of stray atoms is particularly troublesome in the case of resonant transitions; long path lengths and the extremely large resonant cross sections[1] result in substantial self-absorption, even if background densities are reduced by efficient pumping techniques.

The process of excitation by electron bombardment involves a transfer of momentum from the electron to the excited atom. The spectral line emitted by the beam is broadened as a result. These "recoil-broadening" effects are not well understood. An early analysis (for H_α) by Mack and Barkofsky[7] predicts a broadening mechanism which is essentially non-Gaussian in character. A more recent calculation by Korolyov and Odintsov[8] has yielded theoretical widths for several lines in the singlet spectrum of helium which agree with the observed widths. Details of the broadening mechanism unfortunately are not presented. Larson and Stanley[9] have observed substantial broadening in He II; they comment that the source profiles were "mainly" Gaussian in character. Recoil-broadening effects should be less important with heavier atoms.

In this work, a simple model for the frequency distribution of a resonance line excited by electron bombardment in an atomic beam light source is developed.

The model is used to interpret the observed absorption and scattering of the 1048 Å ($^1P^\circ_1$) and 1067 Å ($^3P^\circ_1$) fine-structure components of the first resonance transitions in argon (figure 1). A new method for measuring relative oscillator strengths is used to determine the ratio f(1048 Å)/f(1067 Å). The electron impact excitation functions for these lines are observed and compared to a recent theoretical calculation.[10]

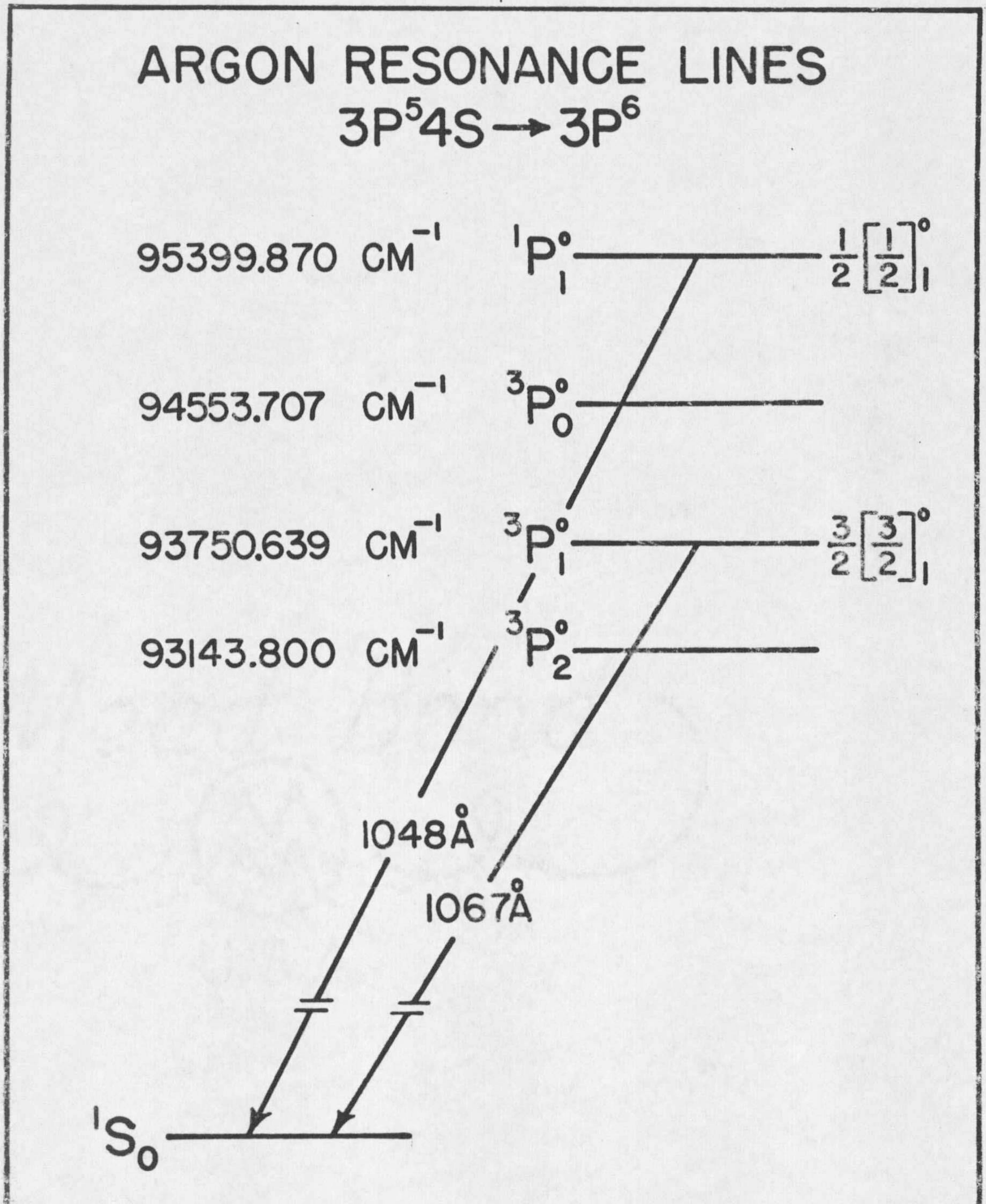
ARGON RESONANCE LINES
3P⁵4S → 3P⁶
95399.870 CM⁻¹
¹P°₁
1/2 [1/2]°₁
94553.707 CM⁻¹
³P°₀
93750.639 CM⁻¹
³P°₁
3/2 [3/2]°₁
93143.800 CM⁻¹
³P°₂
1048Å
1067Å
¹S₀

FIG. 1

II. THEORY

An optical transition of an atom between an excited state and the ground state is called a resonant transition; the radiation emitted or absorbed in the process is called resonance radiation. If resonance radiation corresponding to a particular transition propagates in the $\hat{x}$-direction from the point of emission x=0 through an absorbing vapor of density n(x), the total intensity reaching a point x=L is

$$I(L) \propto n(0)\int_{-\infty}^{\infty} d\omega E(\omega)\exp\{-\int_{0}^{L} dx\, n(x)\sigma(x,\omega)\} \quad . \tag{1}$$

The emission profile $E(\omega)$ is the frequency distribution of the radiation emitted; $\sigma(x,\omega)$ is the cross section for resonant absorption, or absorption profile, near x. $E(\omega)$ is determined by the distribution of the $\hat{x}$-components v_x of the velocities[11] of the excited-state atoms near x=0, and $\sigma(x,\omega)$ is determined by a similar distribution for ground-state atoms near x, both according to the Doppler relation

$$v_x = \frac{c}{\omega_o}(\omega-\omega_o) \quad . \tag{2}$$

Here ω_0 is the separation in angular frequency of the ground and excited states, and c is the speed of light.

The distribution of atomic velocities throughout a gas in thermal equilibrium is Maxwellian. The corresponding absorption profile is independent of x:

$$\sigma(\omega) \propto \frac{f}{\gamma} \exp\{-(\frac{\omega-\omega_0}{\gamma})^2\} ,$$

(3)

$$\gamma = \frac{\omega_0}{c}\sqrt{\frac{2kT}{M}} .$$

The absolute temperature T of the gas and the mass M of an individual atom determine the absorption width γ. Boltzmann's constant is denoted by k. The oscillator strength f of the resonant transition appears because of the normalization requirement[1]

(4) $$\int_{-\infty}^{\infty} d\omega\sigma(\omega) = \frac{\pi e^2}{mc^2} f$$

where m is the mass and e the charge of an electron.

It is shown in Appendix A that an atomic beam produced by effusion through a small aperture and collimated with a circular opening downstream exhibits an absorption profile of the form (3), provided T is interpreted as an effective beam temperature Θ. This effective temperature is a measure of the geometric collimation of the beam and is in general less than the temperature of the gas in the source. The absorption width γ is correspondingly reduced.

The velocity distribution in an atomic beam undergoing excitation by electron bombardment is altered by momentum transfer from electron to atom. The beam emission profile differs in form from the absorption cross section as a result. The distribution of atomic recoil momenta along a direction perpendicular to the axis of a perfectly collimated atomic beam (0°K effective temperature) can be calculated from the differential cross section for inelastic electron scattering. This is done in the first Born approximation in Appendix B. The corresponding emission profile is

$$E(\omega) \propto \int_{a}^{\pi-a} d\theta \, \frac{\operatorname{ctn}(\theta/2)}{\sqrt{\sin^2\theta-\zeta^2}} \,, \tag{5}$$

$$\zeta = \frac{\omega-\omega_0}{\omega_m-\omega_0} \,, \quad a = \sin^{-1}|\zeta| \,.$$

The angular frequency $\omega_m - \omega_o$ corresponds to the maximum recoil velocity commensurate with energy-momentum conservation. The function is strictly zero for values $|\zeta|>1$ (recoil cutoff).

If the beam is not perfectly collimated, the emission profile is a convolution of forms (3) and (5):

$$E(\omega) \propto \frac{1}{\Gamma}\int_{-1}^{1} d\zeta' \exp\{-(\frac{\zeta-\zeta'}{\Gamma})^2\}\int_{b}^{\pi-b} d\theta \frac{\mathrm{ctn}(\theta/2)}{\sqrt{\sin^2\theta-(\zeta')^2}} \tag{6}$$

$$\Gamma = \frac{\gamma}{\omega_m - \omega_o}, \quad b = \sin^{-1}|\zeta'| \quad .$$

$E(\omega)$ can be evaluated numerically with the aid of the first program in Appendix C. The results for several Γ are shown in figure 2.

The parameter Γ depends through γ and ω_m upon the atomic properties, the beam temperature, and the incident electron energy E:

$$\Gamma = \sqrt{\frac{Mk\Theta}{m(E-h\omega_o)}} \quad . \tag{7}$$