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Proton Excitation of the Argon Atom*

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Studies of the excitation of argon with 4-MeV protons have been carried out by making use of a vacuum ultraviolet scanning monochromator and a 6-MeV Van de Graaff accelerator. Protons were directed through the gas and then into a Faraday cup in an arrangement in which the relative intensity of emitted light could be studied over a wide range of gas pressure (1 to 1500 Torr). Four continua near 2100, 1900, 1300, and 1100 Å, respectively, as well as escape radiation originating from the 1048 Å resonance line, were studied as a function of pressure.

Studies of the intensity of the four main continua (per unit of proton power dissipation) as a function of pressure led us to the conclusion that each continuum has a separate atomic precursor. We suggest that the 1300 Å continuum and the 1100 Å continuum are dissociative diatomic continua and originate from the $^1P_1$ (11.83-eV) and the $^3P_1$ (11.62-eV) resonance atomic states, respectively. We tentatively suggest that the continua near 2100 Å and near 1900 Å are recombination spectra involving the formation of argon excimers with binding energies of about 4 eV.

The 1300 and 1100 Å continua have been observed in gas discharges, but these were interpreted as a single continuum originating from the $^3P_1$ state. We believe that the present experimental method, which makes possible gas kinetic observations at spectroscopically defined photon energies, is indeed a powerful tool for the study of atomic and molecular structure and processes.

INTRODUCTION

Continuous emission was observed in helium discharges by Hopfield[1] and reported in 1930. This remarkable work in which a differentially pumped gas-discharge source, vacuum ultraviolet spectrograph, and photographic plates were used to study continuous emission and continuous absorption, has played a classic role in the development of the field of vacuum ultraviolet spectroscopy. Subsequently, continuous emission has been observed in nearly all of the rare gases, and this information has been used to develop continuous light sources for the vacuum ultraviolet region.[2] Reference 2 contains an excellent bibliography of the basic papers as well as a discussion of some of the practical aspects of gas-discharge sources.

In the case of argon a continuum is observed to lie between 1100 and 1600 Å, which we refer to as the 1300 Å continuum. This continuum has been reported by Tanaka[3] using repetitive condensed discharges and quite recently by Wilkinson[4] using microwave excitation. Wilkinson's paper is the first effort to comment in detail on the mechanism

of the 1300 Å continuum produced in argon gas discharges.

Another very interesting line of investigation has made use of radioactive sources of charged particles to excite and ionize noble-gas atoms. Ionization measurements have been made in nearly all of the noble gases by allowing α particles to be completely absorbed in the gas, and by collecting the total number of ion pairs produced by the particles. It is found in all carefully examined cases that when small traces of molecular impurities are present the number of ion pairs is substantially increased. Impurity enhanced ionization was first studied quantitatively by Jesse and Sadauskis[5,6] for the case of He. Thus the general effect is known as the Jesse effect, although a similar effect in gas discharges is known as Penning ionization.

Following the detailed observations of Jesse effects in argon,[7] spectroscopic studies of charged particle gas luminescence were carried out in an effort to obtain further information on the nature of the Jesse effect. Thus Strickler and Arakawa[8] used an α particle source and observed continuous emission in the region of 2100 Å and in the region of 1300 Å. Their new continuum near 2100 Å played a major role in supporting the proposal[7,9] that at least two long-lived excited states are created by charged particles. Continuous emission in the 2100 Å region produced by α-particle excitation of argon has recently been reaffirmed.[10] Thus studies of Jesse effects in argon have motivated spectroscopic investigations, and continuous emission has been observed which tend to support the basic idea that Jesse effects are due to long-lived excited states. However, it has not been proven that there is direct connection between the observed Jesse effects and the observed continuous emission.

In the present series of investigations, protons from a 6-MeV Van de Graaff accelerator are used to excite argon under conditions which allow good estimates of energy losses to be made as a function of gas pressure. We[11] have confirmed the continuous emission in argon around 1300 Å and around 2100 Å and have been able to resolve other continua at around 1900 Å and near 1100 Å. These continua, as well as the region around the 1048 Å resonance line, have been studied as a function of gas pressure.

EXPERIMENTAL METHOD

Figure 1 shows the experimental arrangement. Protons from a 6-MeV Van de Graaff accelerator are brought through an entrance foil (0.0002-in. Havar) into the gas cell and are finally collected in a Faraday cup, which is isolated from the gas cell by means of an exit foil (0.0002-in. Havar). This arrangement permits gases to be used over a pressure range from about 1 to 1500 Torr. Light emission is analyzed with a $\frac{1}{2}$-m scanning monochromator of the Seya geometry which uses a reflecting diffraction grating with 600 lines per mm, over-coated with magnesium fluoride and blazed at 1500 Å. The dispersed light strikes a sodium salicylate converter and is then detected photoelectrically using an electron multiplier tube type EMI 6256-S.

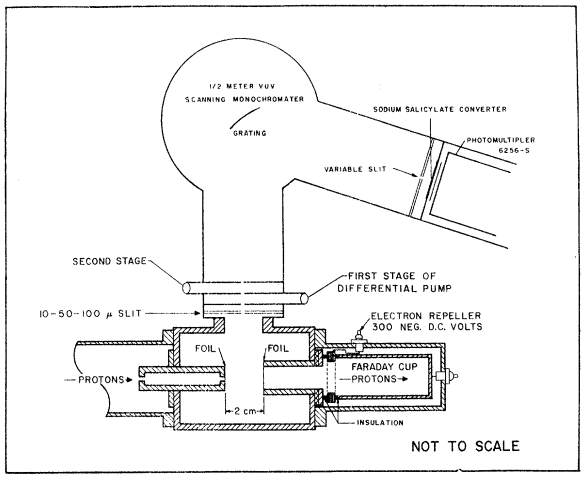

FIG. 1. Schematic of experimental arrangement used to study proton excitation of argon gas.

The gas cell is isolated from the vacuum ultraviolet monochromator with a LiF window (2 mm thick) which will transmit down to about 1100 Å. As shown in Fig. 1, the LiF window can be removed in which case a differentially pumped slit is used to isolate the gas cell from the monochromator. In the region of 1300 Å, the conversion efficiency of sodium salicylate appears to be nearly independent of wavelength[12] and thus the main factor governing the wavelength response of the instrument is the reflection efficiency of the grating which for unpolarized light is the average of the two curves shown in Fig. 2.[13] In Fig. 2 R_p' and R_s' are the absolute reflectances of the grating for incident flux polarized with the electric field vector parallel and perpendicular to the plane of incidence, respectively.

In the present geometry the relative amount of energy lost under the slit of the monochromator can be estimated easily. Each proton looses an amount of energy equal to ΔE, where

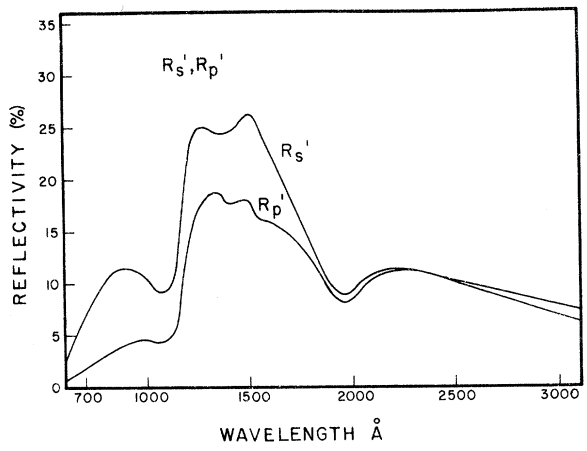

FIG. 2. Reflectivity of grating as a function of wavelength.

$$\Delta E = \int_0^l (dE/dX)(P/P_0)dl\,, \qquad (1)$$

in which dE/dX is the stopping power of protons in the gas at the pressure P_0, P is the pressure, and dl is an element of proton path which is seen by the monochromator grating. In the case of argon and when the proton beam is 4 MeV or greater dE/dX is nearly constant throughout the path even when P_0 is as large as 1500 Torr. Thus

$$\Delta E = (dE/dX)(P/P_0)l\,, \qquad (2)$$

where l is the effective path length seen by the monochromator. The rate of energy dissipation is given by

$$\dot{E} = (dE/dX)li_p(P/P_0)\,, \qquad (3)$$

where i_p is the proton current in particles per second. A knowledge of the rate of energy absorption is essential for kinetic interpretations, thus excitation by heavy charge particles offers considerable advantage over more conventional spectroscopic sources.

EXPERIMENTAL RESULTS

We found that reproducible results are obtained most easily by flowing pure argon (Matheson's Gold Label) through the gas cell. When the gas is allowed to stand in the system a rapid decrease in light intensity results.

Figure 3 shows two scans of the spectrum between 1000 and 2300 Å both made with 4-MeV protons exciting argon at a pressure of 100 Torr. Five regions of the spectrum are of particular interest in this paper: (1) the region near the resonance line at 1048 Å, (2) the continuous spectrum near 1100 Å which blends onto the 1068 Å resonance line, (3) the strong continuum near 1300 Å, (4) the continuum around 1900 Å, and (5) the continuum around 2100 Å. The latter is not clearly visible at the 100-Torr pressure used to obtain Fig. 3. Scan A shown in Fig. 3 is a normal scan with the monochromator tank evacuated, while scan B applies to the case when the monochromator tank is filled with argon at cell pressure. Here we note that argon absorbs its own emission when the wavelength is less than 1090 Å. The spectra shown in Fig. 3 and the figures to follow have not been corrected for instrument response which is determined largely by the grating reflectivity shown in Fig. 2.

A very interesting and complicated behavior is noted (in Fig. 4) of the 1048 Å region where we see that both the intensity and the peak wavelength depend on pressure. These data are at first astonishing; however, clarifying comments will be made in the next section.

Because of the absorption limit at 1090 Å in argon the intensity of region 2 will be plotted as the intensity at 1100 Å where no appreciable reabsorption occurs even when a one-meter absorption path (the path of the light in the monochromator tank) is used. Figure 5 shows a plot of the relative phototube current per unit of power dissipated by the 4-MeV protons under the slit of the monochromator as a function of gas pressure P. As explained in the section on experimental method this quantity is essentially I/P where I is the relative

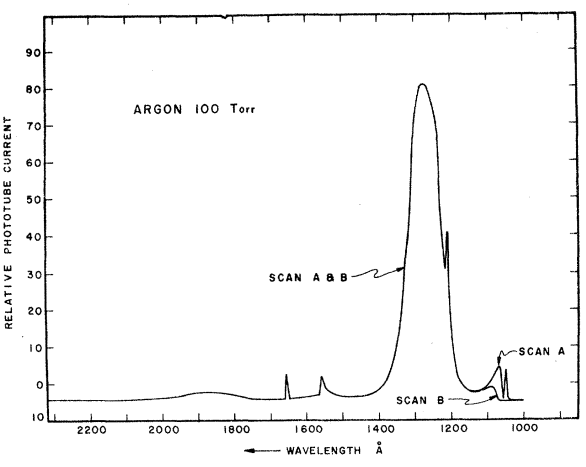

FIG. 3. Relative phototube current as a function of wavelength for 4-MeV protons in argon at 100 Torr. (Proton current = 0.75 μA, entrance and exit slits = 100 μ, 100 divisions of phototube current = 3×10^{-9} A. Scan A applies to evacuated spectrometer; scan B applies to 100 Torr of argon pressure in the spectrometer tank.)

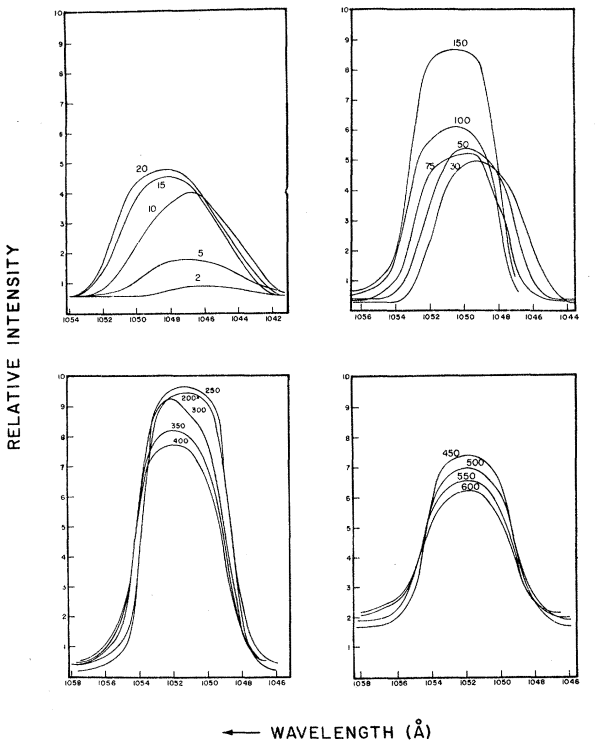

FIG. 4. Relative intensity of the 1048 Å region as a function of wavelength at various pressures indicated (in Torr).

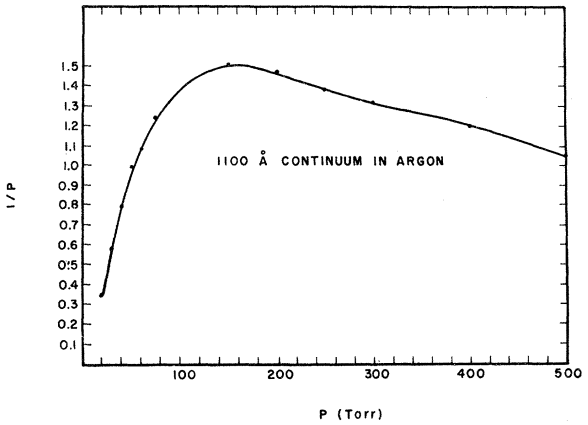

FIG. 5. I/P versus P for the 1100 Å continuum in argon.

phototube current. Thus the data in Fig. 5 indicate that the fraction of the proton energy lost in producing photons at 1100 Å rises with pressure, reaches a maximum at about 150 Torr, and then declines. Obviously this implies that other mechanisms are consuming some of the energy lost by the protons, otherwise the curve in Fig. 5 would be a horizontal line.

A plot of the function I/P versus P for the 1300 Å continuum, shown in Fig. 6, is nearly constant in the pressure region between 400 and 1500 Torr. In this case a LiF window was used and the pressure could be increases to about 2 atm. We find that the spectral distribution of the 1300 Å continuum is pressure-independent; while is gas discharge work[2] this is not the case. Furthermore, the corresponding functions I/P versus P are much more complex in the case of discharges.[2]

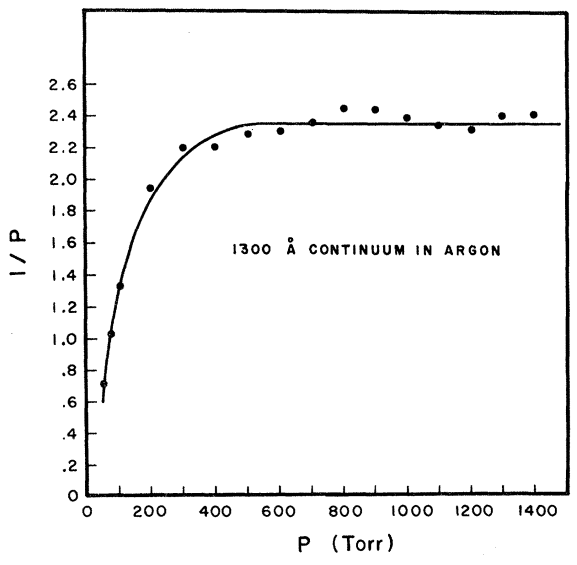

FIG. 6. I/P versus P for the 1300 Å continuum in argon.

Because the edges of the continua near 1900 Å and near 2100 Å are overlapping, we reproduce in Fig. 7 a section of the stripchart between 1700 and 2200 Å, where this region is shown at various pressures. Were this taken as a single continuum, its interpretation would be most difficult. However, a plot of I/P versus P for the resolved continua are shown in Fig. 8, and are well behaved.

INTERPRETATION

We first comment on the 1100 and 1300 Å continuous regions of the spectrum, leaving until later the more difficult 1048 Å resonance line region. Figures 5, 6, and 8 strongly suggest that the four continua originate from separate excited states of the argon atom. Thus since the function I/P versus P represents the fractional utilization of atomic excited states in producing photons over a continuous range of frequencies in the respective regions, and since these functions are unique to a given region, the implication of unique atomic states is strong.

It is well known[14] that continuous spectra can be produced from the interaction of atoms in discrete excited states with identical atoms in their ground states. The spectra which result are a consequence of Franck-Condon transitions in the diatomic collision complex. The spectra have been classified by Herzberg as dissociative spectra and recombination spectra, as shown in Fig. 9, depending on the stability of the molecular final

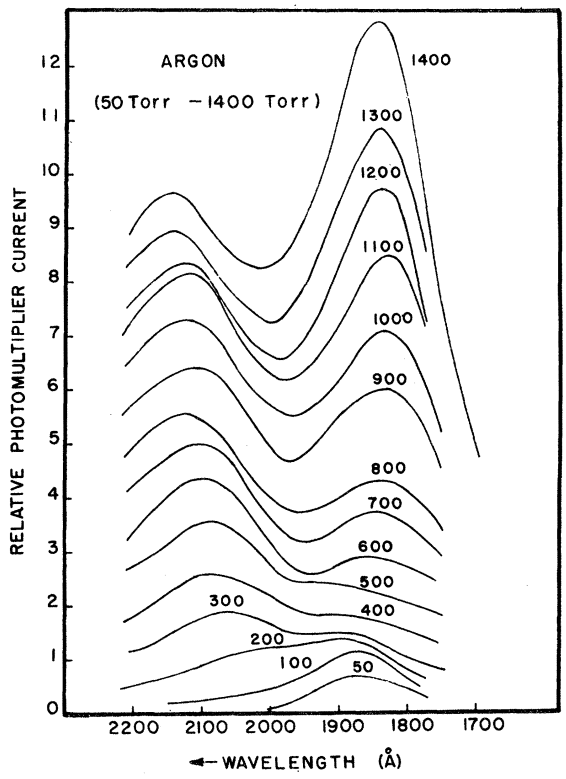

FIG. 7. I versus wavelength for the 1700–2200 Å region in argon.

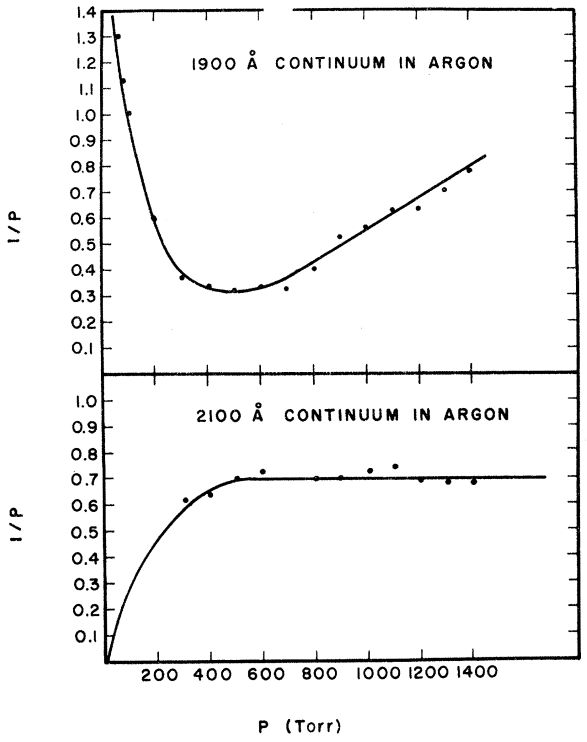

FIG. 8. I/P versus P for the 1900 and 2100 Å continua in argon.

state. In both cases, the two particles collide with total energy E and kinetic energy at infinite separation equal to ΔE. At the classical turning point, r_0, the relative motion of the two particles vanishes and they separate again to $r=\infty$, unless emission occurs. Franck-Condon emission takes place, as in diagram A, with greater probability near the classical turning point, and is least likely near the minimum of the upper interaction potential surface $U(r)$ where the kinetic energy of the relative motion of the two particles is equal to D (the dissociation energy of the upper state) plus ΔE. When the final state is repulsive a considerable amount of kinetic energy T appears in the outgoing particles in their ground state. The total energy equation

$$E = h\nu(r) + T(r) \tag{4}$$

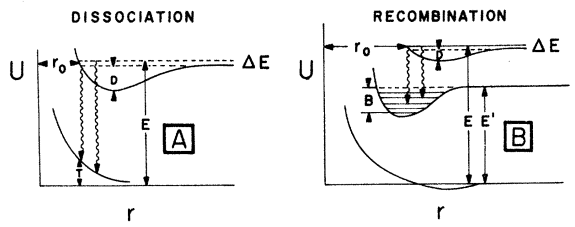

FIG. 9. Schematic explaining dissociative and recombination continua.

is satisfied for various values of r, hence $h\nu(r)$ is a continuous function, and the function $I(\nu)$ will also be continuous. The spectrum of emitted light intensity $I(\nu)$ will depend, therefore, on the shape of the upper as well as the lower potential surface.

Recombination spectra are produced (see diagram B in Fig. 9) when Franck-Condon transitions occur from an unbound state at total energy E to a bound vibrational state whose total energy is less than E. When the spread in energy ΔE is comparable to vibrational level spacing in the lower bound state emission spectra appear continuous. Again, the spectrum $I(\nu)$ depends on both the initial and the final state.

Since the 1100 Å continuum blends smoothly onto the 1067 Å resonance line we believe it to originate from the $^3P_1$ (11.62 eV) atomic resonance state from the process

$$\mathrm{Ar}(^3P_1) + \mathrm{Ar}(^1S_0) \rightarrow \mathrm{Ar}(^1S_0) + \mathrm{Ar}(^1S_0) + h\nu\,. \tag{5}$$

As mentioned in the previous section, kinetic arguments based on the function I/P versus P favor the argument that the 1100 and 1300 Å continua arise from different atomic states. Therefore in spite of previous conclusions[4] that the 1300 Å continuum also originates from the $^3P_1$ state we suggest that it originates from the $^1P_1$ (11.83-eV) atomic resonance state in the process

$$\mathrm{Ar}(^1P_1) + \mathrm{Ar}(^1S_0) \rightarrow \mathrm{Ar}(^1S_0) + \mathrm{Ar}(^1S_0) + h\nu\,. \tag{6}$$

It is interesting that the metastable states $^3P_0$ (11.72 eV, 1058 Å) and $^3P_2$ (11.55 eV, 1074 Å) do not appear in the spectrum of Fig. 3. Commenting further on the above assignment of the atomic precursors of the 1100 and 1300 Å continua, we note that the 1100 Å continuum is connected onto its precursor line 1067 Å, while the intensity of the 1300 Å continuum drops to a very low value in a wide region above its precursor line at 1048 Å. This behavior implies that the well depth D in diagram A of Fig. 9 is much greater for the 1048 Å resonance state than the corresponding value for the 1067 Å resonance state. Such differences are not unexpected[15] in view of the fact that the oscillator strength for the $^1P_1$ state is four times greater than that of the $^3P_1$ state.[16] Evidently the classical turning point r_0 is somewhat greater in the case of the $^3P_1$ state, since the wavelength for maximum intensity is closer to the resonance line. While kinetic arguments based on the behavior of I/P versus P strongly favor the above assignments, other support may be obtained by comparing the relative intensities of the two continua. After correcting for grating reflectivity, it is found that the ratio of emitted energy in the 1300 Å continuum to the emitted energy in the 1100 Å continuum, at pressures where it appears that essentially all of the energy of the atomic states is utilized in molecular emission, is close to the ratio of oscillator strengths.

Having thus connected the 1100 and 1300 Å continua with resonance states, we must comment further on the collision processes [Eqs. (5) and (6)], and we must attempt to explain the data shown in

Figs. 5 and 6. It is especially important to realize that the effective lifetimes of the excited states involved in processes 5 and 6 are greater than natural lifetimes, due to resonance photon trapping,[17,18] i.e.,

$$\mathrm{Ar}^* \rightleftharpoons \mathrm{Ar} + h\nu\,, \tag{7}$$

where the arrow to the left indicates photon reabsorption by another atom. Continuous emission is then viewed as a detuning process in which

$$\mathrm{Ar}^* + \mathrm{Ar} \rightarrow \mathrm{Ar} + \mathrm{Ar} + h\nu_c \tag{8}$$

and the Franck-Condon photon $h\nu_c$ is not retrapped. With this mechanism [Eqs. (7) and (8)] one may show that the functions I/P versus P are given by

$$I(P)/P = \mu\sigma n v/(\sigma n v + 1/\tau') \tag{9}$$

in which μ is constant, σ is the cross section for process 8, v is the mean relative velocity of Ar* and Ar, n is the number density of Ar atoms, and τ' is the lifetime of a resonance photon in a small volume of gas defined by the geometry of the monochromator input slit. When the gas density increases sufficiently $I(P)/P$ approaches the constant μ, since τ' is not a decreasing function of n. Thus Eq. (8) explains the general rise to an asymptotic limit as seen in Fig. 6. The decline in Fig. 5 at high pressure may be due to a depopulation of the $^3P_1$ resonance state by three-body collisions, namely

$$\mathrm{Ar}(^3P_1) + \mathrm{Ar}(^1S_0) + \mathrm{Ar}(^1S_0) \rightarrow \mathrm{Ar}(^3P_{0,2}) + \mathrm{Ar}(^1S_0) + \mathrm{Ar}(^1S_0)\,, \tag{10}$$

in which spin is conserved and in which $\mathrm{Ar}(^3P_{0,2})$ represents one or both of the metastable states of very low oscillator strengths.

We now comment on the complicated data shown in Fig. 4 for the 1048 Å region. From the fact that the peak wavelength shifts by about +5 Å as the pressure is increased from 2 to 600 Torr, we suggest that the spectrum in this region is due to two components: (1) a self-reversed resonance line at 1048 Å, and (2) a small piece of the 1300 Å dissociative continuum, corresponding to Franck-Condon transitions at large values of the internuclear separation distance, r, in Fig. 9, diagram A. This region of the spectrum is further complicated by absorption in argon. We may, however, proceed one step further in the analysis. Let us call the relative frequency integrated intensity in this region $I(1048) + I(1048^+)$, corresponding to the two components listed above, and plot the ratio

$$[I(1048) + I(1048^+)]/I(1300)$$

as a function of P^2, as shown in Fig. 10. Thus at the higher pressures, where none of the 1048 Å peak will be seen, the ratio should be an exponential whose negative slope is related to the absorption coefficient in argon. In other words the ratio $I(1048^+)/I(1300)$ should be a constant value if molecular absorption did not occur and if the 1048^+ radiation is, in fact, connected to the 1300 Å continuum. Figure 10 shows the anticipated exponential behavior at high pressure, with the slope agreeing roughly with data published by Wilkinson.[4] At low pressures the ratio plotted in Fig. 10 increases appreciably indicating some leakage of the self-reversed 1048 Å resonance radiation. This leakage, however, is not spectacular because of photon scattering in the resonance region. In view of the complexity of Fig. 4, it is remarkable that a simple analysis leads to the well-behaved function shown in Fig. 10.

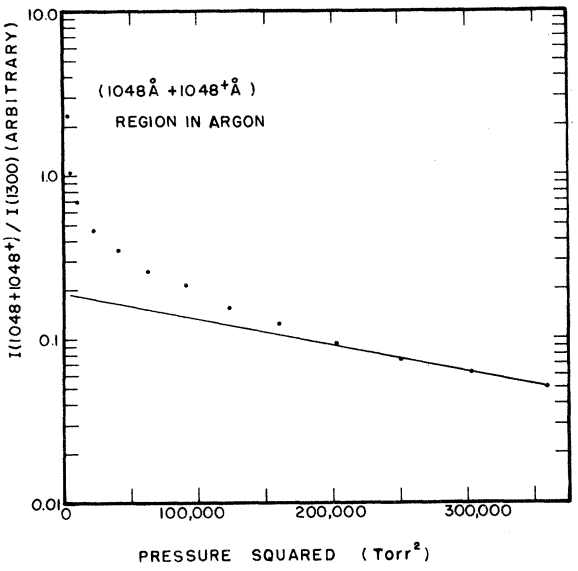

FIG. 10. Frequency integrated intensity in the 1048 Å region divided by the intensity in the 1300 Å region as a function of the square of the argon pressure.

The 1900 and 2100 Å continua are most likely recombination continua and involve a final state having a well depth $B \geq 3$ eV (see Fig. 9, diagram B.) As yet we are unable to suggest either the initial or the final states; however, several possible resonance states with the required energy have been found by electron-impact experiments.[16] The complicated behavior of $I(1900)/P$, Fig. 8, may be due to depopulation of a resonance state by three-body collisions, producing an excited state which cascades to the ground state at intermediate pressures. In spite of cascade processes, two-body collisions at still higher pressure could again produce Franck-Condon transitions which would cause an increase in the intensity at very high pressure.

In Fig. 11 we show one set of potential energy surfaces which are consistent with the emission continua observed at 1100, 1300, 1900, and 2100 Å, as well as the absorption continuum for wavelengths less than 1090 Å. The assignment of the 1900 and 2100 Å continua must involve a final state with a very deep well. For example, for the 1900 Å continuum the final-state energy must be less than 8.5 eV, [ionization potential of argon-$h\nu$ (1700 Å)]. Therefore we suggest that the final states in these continua involve the $^1P_1$ (11.83 eV) atomic

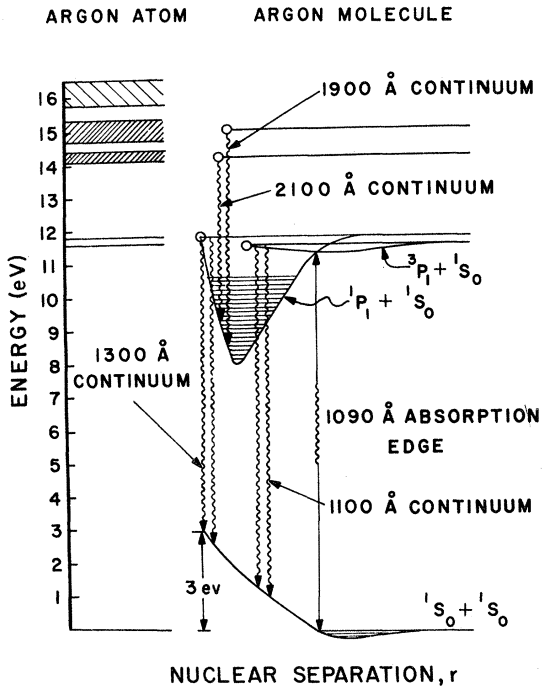

FIG. 11. Possible potential energy surfaces in the argon molecule.

state interacting with the $^1S_0$ ground state producing a potential well having a minimum depth of 3.3 eV. Considerable oscillator strengths are found[16] at levels near 14.3 and 15 eV (see the shaded areas in Fig. 11); therefore, these groups of levels contain likely initial atomic states for the 1900 and 2100 Å continua, thus making about 4 eV a more realistic minimum well depth. In Fig. 11 the position of the van der Waals attractive interaction in the ground molecular state is so chosen to account for continuous absorption (at wavelengths less than 1090 Å) into either the attractive $(^3P_1 + {}^1S_0)$ or the $(^1P_1 + {}^1S_0)$ molecular states.

We have found evidence for resonance states, in the energy regions suggested previously,[9] to explain Jesse effects in argon; however, we have not proven that these states are responsible for Jesse effects. Because of new evidence presented in this paper, and in spite of continuing implication[19] to the contrary, the role of atomic metastable states in explaining Jesse effects in argon is questionable.

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