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NEW AUTOIONIZING ATOMIC ENERGY LEVELS IN He, Ne, AND Ar[†]

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The NBS 180-MeV electron synchrotron has been used as a continuum light source for absorption spectroscopy in the 180-470Å region. Two-electron transitions to states which autoionize have been observed in He; transitions to autoionizing states have also been observed in Ne and Ar.

Autoionization may occur when neutral atoms are excited to energy levels which lie above their first ionization limit. The strength of the interaction of these states with the continuum states determines the lifetime of the discrete levels. This interaction results in an interference phenomenon which imparts an unusual shape to the absorption in the region of the discrete level. Transitions to or from autoionizing levels have not previously been observed spectroscopically in He or Ne. Beutler[1] observed autoionized lines in absorption by Ar between the $^2P_{3/2}{}^\circ$ and $^2P_{1/2}{}^\circ$ edges of the first ionization limit near 780 Å. There has been no previous spectroscopic observation of such levels in Ar at higher frequencies. Autoionization has been observed in absorption for several elements, for example, in alkaline earth metals by Garton and Codling[2] and by Ditchburn and Hudson.[3] Previous workers[4-9] studying the photoionization cross section in He, Ne, and Ar in this wavelength region have been able only to suspect the presence of autoionized levels since emission line sources were used.

Autoionization in He is of special interest, since a transition to discrete levels lying above the first ionization limit requires that both electrons be excited. Evidence for the existence of such states was obtained by Whiddington and Priestley[10] and more recently by Silverman and Lassettre[11] from discrete electron-energy losses in the forward scattering of electrons by He. Using different approximations, the autoionization probability of these levels has been calculated by Wu[12] and by Bransden and Dalgarno[13] with more than an order of magnitude disagreement. A general theory of the shape of autoionized absorption lines has been formulated by Fano,[14] who has shown that the data of Silverman and Lassettre are compatible with the theory.

The light source used in the present experiments was the 180-MeV electron synchrotron at the National Bureau of Standards. The light radiated by the radially accelerated electrons is confined to a narrow cone in the forward direction of the electrons and is continuous in wavelength. The usable intensity extends from the infrared down to below 100 Å. Detailed characteristics of the radiation from this synchrotron will be described in another paper.

A 3-meter grazing incidence vacuum spectrograph, designed to withstand the vibrations attendant to the synchrotron application, was constructed. This instrument is being used with a grating having 15 000 lines per inch to produce a dispersion of 1 Å per mm in the 200Å region at a resolution width somewhat less than 0.1 Å. An aluminum foil filter is used with the instrument to keep light of wavelength less than 170 Å and greater than 800 Å from entering the spectrograph. Commercial tank gas samples are admitted to the spectrograph at pressures ranging up to several hundred microns of Hg. This gas can leak from the spectrograph through the entrance slit, requiring a differential pumping system to maintain the synchrotron doughnut pressure below 5×10^{-6} mm Hg. The gas-sample pressure is measured with a McLeod gauge.

The absorption spectrum obtained for helium

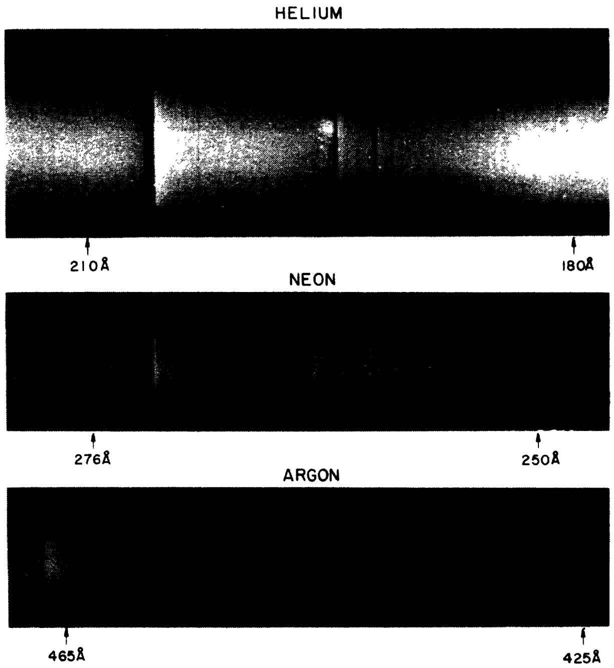

FIG. 1. Extreme ultraviolet spectra showing discrete anomalies in the photoionization continuum absorption of He, Ne, and Ar (increased blackness indicates increased absorption). The anomalies in the He absorption spectrum (top) are due to the mixing of two-electron excitation states with the continuum. In Ar (bottom) this mixing results in discrete windows in the absorption continuum.

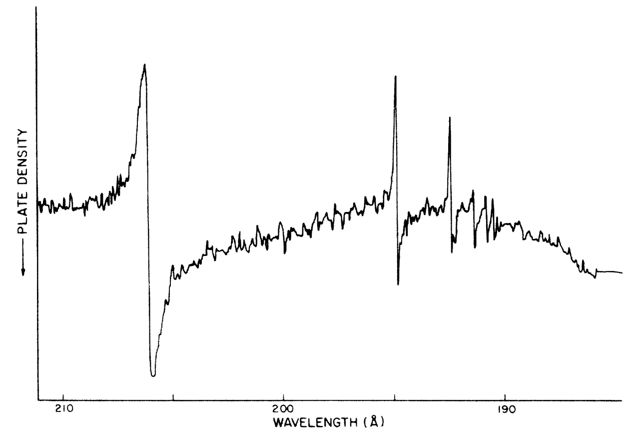

FIG. 2. Densitometer trace of the absorption spectrum of He as shown in Fig. 1 (top). The anomalies are of the Beutler-Fano shape. The absorption is enhanced on the low-frequency side and reduced on the high-frequency side.

in the 180-210Å region is shown at the top of Fig. 1 for a pressure of 0.3 mm Hg and a path length of approximately 83 cm. This region of the spectrum lies 35-40 eV above the first ionization limit for helium (24.58 eV). From Fig. 1, it can be seen that a discrete structure is superimposed on the continuous photoionization absorption. The first two members of the prominent series are at approximately 60.1 and 63.6 eV and correspond well to the electron loss lines as measured by Silverman and Lassettre.[11] The other members of the series (eight are observed on the original plate), which were not observed by electron scattering, converge on the $2s$ or $2p$ state of He II at 65.4 eV. The intensity distribution of these autoionized lines is best indicated by the densitometer trace for this plate given in Fig. 2. Here it can be seen that the photoionization absorption is enhanced on the low-frequency side of each resonance and is decreased on the high-frequency side. It is also apparent that the breadths of the anomalies decrease with progression to the higher series members. The first member of the series (60.1 eV) can be associated with the lowest allowed transition to a two-electron excitation level, namely, $1s^2\,{}^1S_0$-$2s2p\,{}^1P_1{}^\circ$. The exact position of this level can be theoretically predicted only after an evaluation of the mixing of this state with the neighboring configurations.

There are two series of energy levels important in determining the position of the remaining autoionized lines seen in He—in particular, $2snp$ and $2pns$. Both of these series will have $2s2p$ as the first member, and both converge to 189.6 Å (65.4 eV). From theoretical considerations, Cooper, Fano, and Prats[15] have determined that these two series mix strongly and in approximately equal amounts. They have further estimated that the probability of excitation is quite different for the two series which result from the mixing. From Fig. 1 (top) and Fig. 2, it would seem that only a single series has been observed.

As can be seen from Fig. 2 the intensity distribution in these absorption anomalies is in qualitative agreement with shapes first seen by Beutler,[1] and subsequently interpreted by Fano[14] for levels interacting with a single continuum. A critical study of the distribution of oscillator strengths in these lines is now in progress.

The middle spectrum in Fig. 1 indicates the autoionized lines which have been observed in Ne in the region 250-280 Å. (The first ionization potential of Ne is 21.56 eV.) This spectrum was obtained using 0.1 mm Hg of Ne and an absorption path length of approximately 95 cm.

The prominent series of Beutler-Fano shaped autoionized lines converges closely to 256 Å (48.5 eV) and most probably corresponds to the transitions $2s^2 2p^6\,{}^1S_0$-$2s2p^6np\,{}^1P_1{}^\circ$. Nine members of this series are observable on the original plates.

Many other discrete anomalies in the absorption spectrum of Ne can be seen in Fig. 1, and they differ considerably in character. The energy levels associated with these lines have not, as yet, been identified. However, it is reasonable to expect that many of these are two-electron excitation levels.

Some of the autoionized lines which have been observed in Ar are shown in the bottom spectrum of Fig. 1 for the 420-470Å region. (The first ionization potential for Ar is 15.76 eV.) An Ar pressure of 0.01 mm Hg, and an absorption path length of approximately 105 cm were used. The prominent series of autoionizing lines in this spectrum, due probably to transitions $3s^2 3p^6\,{}^1S_0$-$3s3p^6np\,{}^1P_1{}^\circ$, is different in character from the prominent series in Ne and He. The Ar series has the appearance of an emission series. These lines must be thought of as discrete decreases in absorption at the line positions, there being no accompanying region of pronounced increase in absorption. Fano[14] has indicated that such shapes are allowable from his theory as a special case of the general interference phenomena when the wave functions and interaction parameters have appropriate values.

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CLASSIFICATION OF TWO-ELECTRON EXCITATION LEVELS OF HELIUM

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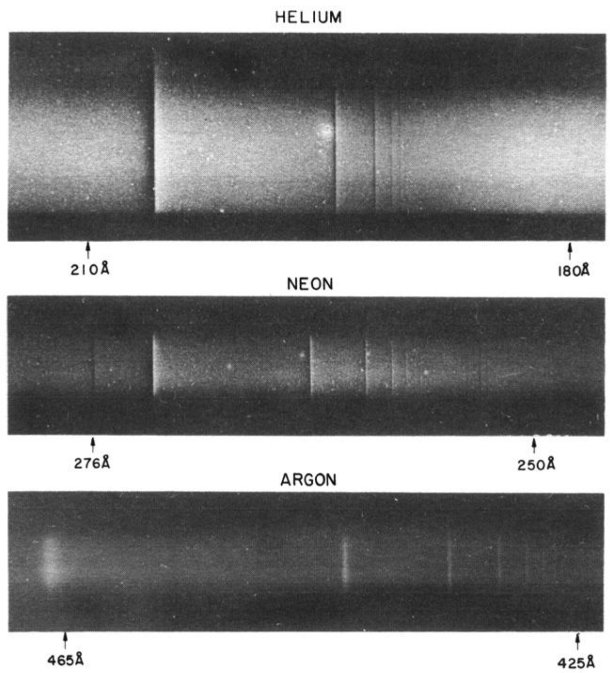
Madden and Codling[1] describe a series of autoionizing levels of He, in the 60- to 65-eV range, which converges to the $n=2$ level of He^+. The classification of these levels as ${}^1P$ appears obvious. However, since the $n=2$ level of He^+ is degenerate ($2s$ and $2p$), two separate He series might be expected to converge to this limit, namely, $2snp$ and $2pns$. These series have a common lowest level with $n=2$. The probabilities of dipole excitations from the ground state $1s^2$ to $2snp$ or $2pns$ should be comparable for reasons indicated below. Therefore the observation of a single series indicates that the classifications $2snp$ and $2pns$ do not constitute an appropriate zero-order approximation.

Because the states $2snp$ and $2pns$ are nearly degenerate, the electron-electron interaction will, if sufficiently strong, remove their degeneracy in the zeroth step of perturbation treatment and replace the symmetrized independent-electron wave functions $u(2snp)$ and $u(2pns)$ with the pair[2]

$$\psi(2n\pm) = \{u(2snp) \pm u(2pns)\}/\sqrt{2}. \qquad (1)$$

Indeed, a calculation with screened hydrogenic wave functions shows that the electron interaction matrix element $(2s3p\,|\,V\,|\,2p3s) \sim 1$ eV, whereas $E_{2s3p} - E_{2p3s} \sim 0.1$ eV.

This Letter points out the following properties



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