type, Ohta[14] includes diagrams containing a $\Delta(1236)$ resonance and obtains a rather large renormalization of g_P, which does improve the photon spectrum and presumedly changes α (cf., however, Beder[15]). Clearly other mesonic exchange contributions could be included, to the extent that they are not already included via an effective nuclear potential.

Another very interesting possibility results from the additional fundamental terms in the weak-interaction vertex which appear when the nucleons are off the mass shell. Such contributions should be small since they are proportional to the amount by which a nucleon is off shell. However for β decay some such terms,[16] when chosen in a natural way, reduce to nonrelativistic operators having one fewer powers of $1/m$ than some of the main terms. Thus some terms of this kind may be more important than one would *a priori* expect. A detailed study of such terms would clearly be a useful contribution.

The result of our theorem, namely that in the standard theory the entire interesting contribution to α is given by terms $O(1/m^2)$ which one *a priori* might have expected to be small, means that one should look rather carefully at a consistent calculation in the standard theory, which is being done, and at possible more exotic corrections, a few of which I have mentioned. I hope that the existence of this general result will stimulate further work on this problem.

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[1]L. DiLella, I. Hammerman, and L. M. Rosenstein, Phys. Rev. Lett. 27, 830 (1971); L. M. Rosenstein and I. S. Hammerman, Phys. Rev. C 8, 603 (1973).

[2]R. M. Sundelin and R. M. Edelstein, in *High Energy Physics and Nuclear Structure*, edited by S. Devons (Plenum, New York, 1970), p. 150.

[3]H. P. C. Rood and H. A. Tolhoek, Nucl. Phys. 70, 658 (1965).

[4]H. W. Fearing, Phys. Rev. 146, 723 (1966).

[5]E. Borchi and S. De Gennaro, Phys. Rev. C 2, 1012 (1970).

[6]See, e.g., K. Huang, C. N. Yang, and T. D. Lee, Phys. Rev. 108, 1340 (1957); J. Bernstein, Phys. Rev. 115, 694 (1959); G. K. Manacher and L. Wolfenstein, Phys. Rev. 116, 782 (1959).

[7]M. Hasinoff, private communication.

[8]G. K. Manacher, Carnegie Institute of Technology Report No. NYO-9284, 1961 (unpublished).

[9]R. Guardiola and P. Pascual, Can. J. Phys. 48, 1304 (1970).

[10]J. D. Bjorken and S. D. Drell, *Relativistic Quantum Mechanics* (McGraw-Hill, New York, 1964).

[11]L. L. Foldy and S. A. Wouthuysen, Phys. Rev. 78, 29 (1950).

[12]J. L. Friar, Phys. Rev. C 10, 955 (1974), and references cited therein.

[13]H. P. C. Rood, A. F. Yano, and F. B. Yano, Nucl. Phys. A228, 333 (1974).

[14]K. Ohta, Phys. Rev. Lett. 33, 1507 (1974).

[15]D. S. Beder, Rutherford Laboratory Report No. RL-75-003, 1975 (unpublished).

[16]J. Delorme and M. Rho, Nucl. Phys. B34, 317 (1971).

Saturated Two-Photon Resonance Ionization of He($2\,^1S$)*

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We have developed a photoionization method for complete conversion of a quantum-selected population to ionization, making possible sensitive and absolute measurement of the selected populations in a gas. Each photoionization involves the absorption of two photons (from a pulsed dye laser), one of which is resonant with an intermediate state. In this demonstration we measured the absolute number of He($2\,^1S$) states per ion pair following interaction of pulses of 2-MeV protons with He.

In noble-gas energy pathways research,[1] attempts are made to deduce the number of various excited species as a function of time after proton excitation. Photon-emission processes, when viewed over a range of gas pressure, are often so complex that unique kinetic models cannot be constructed even from time-resolved emission experiments.[1] In a search for more direct information, we conceived of a method in which each atom in a selected quantum state would be converted to an ion pair by the absorption of two photons, one of which is resonant with an intermediate state. Two-photon ionization processes are well known and have been used more recently

by a number of groups for isotope separation. Two-photon processes have been ingeniously employed in connection with photoionization cross-section measurement of He($n^1, ^3P$) atoms.[2]

Imagine the passage of charged particles through a gas in a short pulse at $t=0$. We wish to find the number of excited species in each quantum state as a function of t. To do this, we tune a pulsed dye laser so that resonance photons excite the selected quantum state to an intermediate state lying more than half the distance to the ionization continuum. With modest energy per pulse and with a laser linewidth of several angstroms, the intermediate state comes into quasiequilibrium with the excited state. Other photons from the same laser pulse photoionize the intermediate state.

To illustrate the above ideas let us consider He($2\,^1S$). For this state we can use He($3\,^1P$) as the resonance intermediate state; thus the laser is centered at 5015 Å. If the laser delivers a pulse of 0.7 J in a 1-cm^2 beam, a photon fluence of 1.8×10^{18} photons cm^{-2} is delivered at 5015 Å. The product of photon fluence and photoionization cross section [approximately 4.4×10^{-18} cm^2 for He($3\,^1P$) at 5015 Å] is considerably greater than unity; therefore, one may hope to saturate the ionization. If so, each excited state is converted to an ion pair.

Thus, resonance ionization spectroscopy (RIS), as we choose to call it, is an absolute and a very sensitive method for the measurement of excited-state populations. RIS can be extended to the analysis of certain ground-state species even with existing laser technology.

Figure 1 is a schematic of the RIS experiment used for the He($2\,^1S$) demonstration. Narrow pulses (15 nsec) of 2-MeV protons arrived at a beam deflector every 64 μsec. When the manual control was operated, one pulse of approximately 10^8 protons entered the ionization chamber and triggered a dye laser after a variable delay time. Each encounter between the proton pulse and the delayed laser pulse was recorded on a transient recorder. The proton pulse created about 5×10^7 ion pairs at 0.6 Torr, and this caused a current to flow through a signal resistor of a few kilohms. These pulses were amplified $\times100$ with a wide-band pulse amplifier and then recorded.

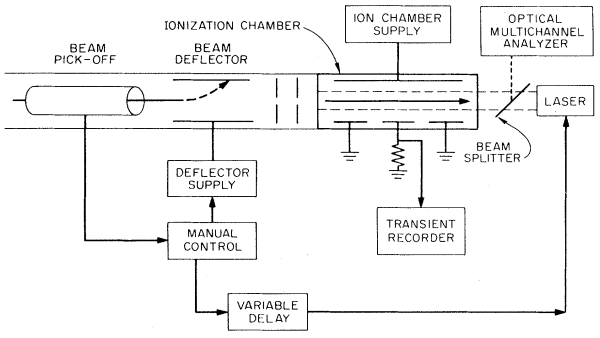

FIG. 1. Schematic of resonance ionization spectroscopy appled to excited states created by pulses of charged particles.

Several laser specifications are of first-order importance. After some modifications of a commercial (Phase-R Company) laser, we obtained (1) a beam of reasonable quality over a 10-mm diam, (2) an energy per pulse of about 0.7 J at 5015 Å, (3) a photon spectra of about 30 Å full width at half-maximum, (4) a pulse width of about 300 nsec, and (5) noise transients which we did not want.

Figure 2 shows a typical recording to 0.6 Torr He. With -50 V applied to the field plates spaced 2 cm apart, the positive-ion signals were collected in about 8 μsec, in agreement with ion mobility and diffusion data. The electron pulses were collected very quickly and are not resolved in the figure. It was shown that below 10^8 ion pairs the pulse heights are linearly related to the number of ion pairs. A simple ratio of pulse heights gives the number of quantum-selected atoms per ion pair.

The He($2\,^1S$) state at 0.6 Torr has a lifetime of the order of 10^{-3} sec in our apparatus. Because of this slow decay, we did not measure in detail the decay curve, but we measured its population at 20 and at 88 μsec after proton excitation.

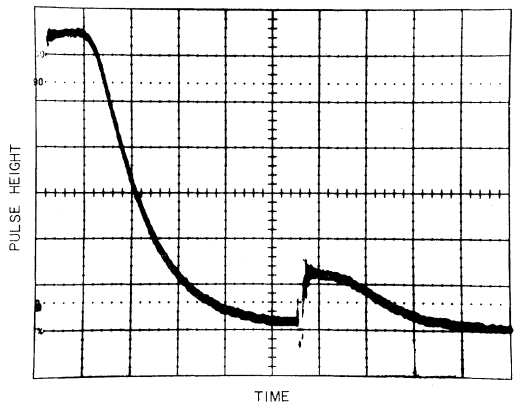

FIG. 2. An oscilloscope trace of the positive ionization as a function of time. The large pulse is direct ionization created by protons; the smaller pulse is created by the laser tuned to 5015 Å and delayed 22 μsec.

There was little observable change between the two delay times.

In the course of carrying out the measurements several checks were made to insure that we were actually observing ionization of the He($2\,^1S$) population. Firstly, the same procedure which yielded Fig. 2 was carried out except that a quartz beam viewer was used to block the proton beam from the ionization chamber. In the absence of the direct ionization and of the He($2\,^1S$) population, only the rf transient (see the sharp time dependence near the beginning of the second ion peak in Fig. 2) was observed when a 0.7-J pulse tuned to 5015 Å was fired into the 99.9998%-pure He. Secondly, the laser was detuned to 5060 Å (and to 4970 Å) and both the protons and a laser pulse of 0.7 J were fired into the cell. In these cases the peaks due to laser-induced ionization were absent. These observations, together with the pulse shapes (which indicate that the ions producing the signal are created nearly simultaneously near the cell center), lead us to believe that the second peak of Fig. 2 is due entirely to resonant two-photon ionization of He($2\,^1S$). Figure 3 shows the ratio of peak heights plotted against energy per pulse at 5015 Å. The fact that a decrease of a factor of 3 in energy per pulse decreases the ratio only by ~20% suggests that the ionization is close to saturation.

The above experiment was carried out with a laser which produces long and slowly changing pulses (duration $\simeq 0.3$ μsec). Also, the linewidth is very broad (i.e., full width at half maximum $\simeq 30$ Å) compared with the $3\,^1P \rightarrow 2\,^1S$ emission line of He. Further, collisional broadening effects destroy quantum coherence in times $\sim 10^{-9}$ sec at 0.6 Torr. For these reasons, rate equations can be used for purposes of estimating the percentage of the He($2\,^1S$) population converted to ion pairs by the laser pulse. Thus, one calculates that a 0.7-J pulse (duration 0.3 μsec and 30-Å spectral width centered at 5015 Å) causes about 80% conversion of He($2\,^1S$) to ion pairs at a pressure of 0.6 Torr. However, the yield of ion pairs may be higher than 80% since some photons are reflected back into the interaction region by a mirror at the end of the cell. About 92% of these ion pairs are created by two-photon resonance ionization, while 8% are due to associative ionization out of the $3\,^1P$ and $3\,^1D$ states. The $3\,^1D$ state comes into the picture because two-body collisions with ground-state atoms convert $3\,^1P$ to both $3\,^1D$ and $3\,^1S$.[3]

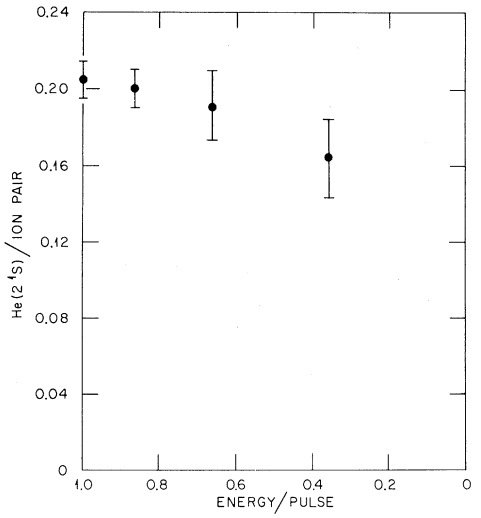

FIG. 3. The number of He($2\,^1S$) states per ion pair as a function of the laser energy per pulse (700 mJ = 1 on energy scale).

Since the calculation (and Fig. 3) suggests that the ionization is nearly saturated, we compared our ratio at 0.7 J/pulse with theory. From the work of Alkhazov[4] on electron excitation, Bartell, Hurst, and Wagner[5] estimated that about 1 μsec after proton excitation the He($2\,^1S$) population per direct ion pair is 0.27 at 0.6 Torr. This is in reasonable agreement with Fig. 3. We should point out that at times greater than 1 μsec the He($2\,^1S$) population includes not only its directly excited population but also much of the original populations of He($2\,^1P$), He($3\,^1S$), and He($3\,^1P$) which, because of radiation trapping, cascade preferentially to He($2\,^1S$). Cascade effects were included, of course, in estimating the 27%.

The RIS technique reported here shows that quantum-selected excited states (at a given time after excitation) can be converted to ionization with nearly unit efficiency. With present dye-laser technology, some of the elemental metal vapors (and some molecules as well) can be converted from their ground states to the ionization continuum with high efficiency. Furthermore, these resonance processes can be highly selective so that a few of one species can be measured without interference from much higher populations of other species. Since single ions can be detected with reasonable efficiency (even after mass analysis), the technique offers the possibility of unprecendented sensitivity for making absolute determinations of either ground-state or transient species.

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[1]For helium, see D. M. Bartell, G. S. Hurst, and E. B. Wagner, Phys. Rev. A 7, 1068 (1973); M. G. Payne, C. E. Klots, and G. S. Hurst, to be published. For argon, see N. Thonnard and G. S. Hurst, Phys. Rev. A 5, 1110 (1972).

[2]F. B. Dunning and R. F. Stebbings, Phys. Rev. Lett. 32, 1286 (1974).

[3]H. F. Wellenstein and W. W. Robertson, J. Chem. Phys. 56, 1072 (1972).

[4]G. D. Alkhazov, Ioffe Institute of Physics and Technology, Leningrad, Report No. FTI-108, 1968 (unpublished) (translation: Scientific Translation Service, STS Order No. 7926).

[5]D. M. Bartell, G. S. Hurst, and E. B. Wagner, Phys. Rev. A 7, 1068 (1973).

Strong New Emission Bands in Alkali–Noble-Gas Systems*

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Strong new visible emission bands are observed when mixtures of Cs of Rb with noble gases are excited with blue lines of an argon-ion laser. The bands in Cs are believed to be due to transitions between $7s\sigma$ bound excited states and dissociating $6s\sigma$ ground states of alkali–noble-gas molecules, and they may have potential for excimer laser systems.

The emission and absorption spectra of alkali–noble-gas systems have been the subject of detailed investigation over a period of many years. Therefore, we were very surprised recently to discover that when we illuminated Cs vapor in a few atmospheres of Xe with any one of several blue lines from an argon-ion laser we observed a striking yellow fluorescence which has apparently never before been observed. We at first mistook this to be resonance fluorescence from a sodium impurity. A check with a grating monochromator, however, revealed that a large fraction of the emission from the vapor in the entire spectral region from the violet to the near infrared (9000 Å), which includes the first two resonance doublets, was concentrated in a single band about 50 Å wide, peaked at 5723 Å, which did not correspond to any known feature of the Cs-Xe spectrum. Similar bands were observed at slightly different wavelengths with other noble gases. These bands are not present in the emission from pure Cs vapor with similar excitation and no corresponding feature has been observed in optical-absorption studies of similar Cs–noble-gas mixtures.[1] The existence and surprising intensity of the emission bands we observe may have significance for laser applications,[2] and a detailed analysis of their structure should provide extensive information about molecular potential curves and transition moments of previously unknown bound excited states.

Typical emission spectra, recorded with a scanning monochromator with a resolution of 1 Å and an extended S-20 photomultiplier tube, are shown in Fig. 1. To obtain these spectra, cells of alkali-resistant glass (Corning 1720) were

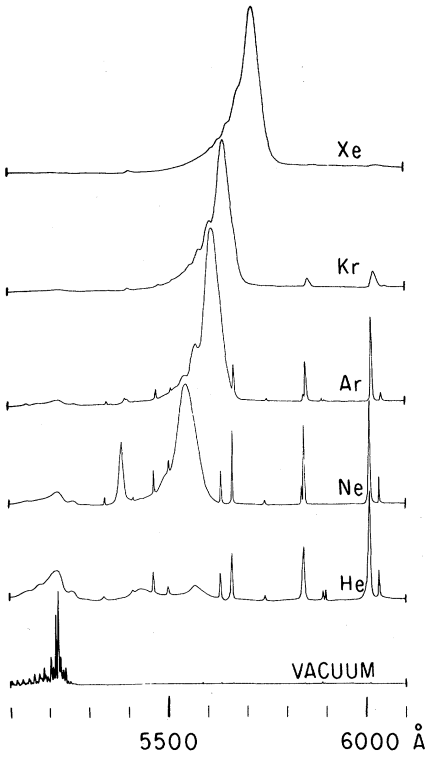

FIG. 1. Yellow emission bands of cesium in noble gases.