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The research conducted under this project is in the general area of atomic and molecular collisions. The optical method has been a very successful tool for measuring electron-impact excitation cross sections. The experiments outlined in the preceding paragpaph provides a new approach for studying excitation into the resonant levels. To determine the cross sections for exciting a resonant level, one must detect the radiative transitions from the resonant level to the lower non-resonant levels as well as to the ground level and measure their optical emission cross sections.

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Final Report
(November 1, 1998 through October 31, 2001)

"Interactions and Collisions of Electrons with Atoms and Molecules"
AFOSR Grant F49620-99-1-0001

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The research conducted under this project is in the general area of atomic and molecular collisions. The specific work carried out during the reported period include the following sections.

1. The optical method has been a very successful tool for measuring electron-impact excitation cross sections. Here an electron beam passing through a gas excites the atoms to higher energy levels and the subsequent emissions from the excited atoms are detected and utilized to determine the excitation cross sections. Because the emissions are very weak, photomultiplier tubes are generally used to detect the radiation. This limits the spectral range of the emitted radiation on account of the unacceptably low sensitivity of the photomultiplier tubes in the infrared. Since many important emission lines (or emission bands in the case of molecules) in electron excitation experiments are in the infrared spectral region, the use of photomultiplier tubes places a limitation on the optical method. This limitation is especially serious in the context of measurement of electron-impact excitation cross sections. In the optical method one measures the total radiation intensity emitted by level-i that was excited by the electron beam. This gives the apparent excitation cross section of level-i which includes the contribution from direct electron-impact excitation of level-i and from cascade from the higher-lying levels that were excited by the same electron beam. To obtain the direct excitation cross section one must measure the cascade radiation into level-i from the higher levels. However, most of the cascade emissions are in the infrared region and do not lend themselves to photomultiplier detection. Thus for a long time we could only measure the apparent excitation cross sections for the excited levels of the rare-gas atoms but not the cascade cross sections. Without the latter it was not possible to determine the direct excitation cross section which is the quantity of fundamental importance.

   The advent of the commercially available weak emission Fourier Transform spectrometer makes it possible to use solid-state detectors for infrared emission in electron excitation experiments. In our laboratory we have acquired two Fourier Transform weak emission spectrometers. We have adapted it to our electron-beam excitation apparatus and used it successfully to measure the electron-impact optical emission cross sections for infrared lines. However, many of the infrared emission lines in electron-beam excitation experiments are weak. Thus it is important to optimize the sensitivity of the Fourier Transform spectrometer. Since the infrared radiation of our interest is usually a small signal imbedded in a large background, we use lock-in detection to separate the signal from the background. Removal of the background is most effective if the lock-in detection is done at a very low frequency. For this purpose we used a NICOLET Fourier Transform spectrometer with step-scanning capabilities so that the lock-in modulation rate can be made as low as practical. This apparatus plays a key role in our research project.

2. We have used the combined photomultiplier and Fourier transform spectrometer detection techniques to study electron excitation of the $2p^53p$ configuration of the Ne atom. These are ten levels within this configuration (called 2p in Paschen's notation). For each of the ten levels we have measured the apparent excitation cross sections (photomultiplier detection) and the cascade cross sections (Fourier transform spectrometer detection) from which we obtain, for the first time, the direct electron-excitation cross sections for incident electron energy from threshold to 200 eV. The ratios of the cascade to the apparent excitation cross sections for the ten levels is found to vary from almost zero to about 50% differing greatly from the estimates based on theoretical cross sections.

   Our measurements of the cascade radiations into the $2p^53p$ levels also provide the apparent excitation cross sections for the levels of the $2p^54s$ and $2p^53d$ configurations. Each of these configurations contains a number of levels (four for $2p^54s$ and twelve for $2p^53d$) with total angular momentum J from 0 to 4. The energy dependence of the apparent excitation cross sections was found to exhibit interesting correlations with the value of quantum number J.

3. A similar study has been made for the krypton atom. The main interest is on the electron-impact excitation of the ten levels of the $4p^55p$ configuration, also designated as the 2p levels in Paschen's notation. The apparent excitation cross sections of the ten 2p levels were measured by means of photomultiplier detection. Again the use of Fourier Transform spectroscopy makes it possible to measure the cascade cross sections so that we can obtain, for the first time, the direct excitation cross section for the ten 2p levels. The Fourier Transform detection technique also allows us to access the higher levels from the $4p^56s$, $4p^57s$, $4p^54d$, and $4p^55d$ configurations and obtain their apparent excitation cross sections and the energy dependence.

4. The new excitation cross section data on neon and krypton described in the two preceding sections are analyzed in conjunction with the cross sections of argon and xenon measured previously in our laboratory. This provides a comprehensive study of the excitation of the heavy rare-gas atoms (neon, argon, krypton, xenon). For all four atoms we have

measured the direct excitation cross sections for the ten 2p levels (Paschen's notation), i.e., the ten levels of the Ne ($2p^5 3p$), Ar ($3p^5 4p$), Kr ($4p^5 5p$), and Xe ($5p^5 6p$) configurations. We examine the variations of the cross sections from one level to another and correlate the magnitude of the cross section with the quantum numbers of the level. For all four atoms the cross sections for the levels with even values of J (the total angular momentum) are generally larger than the cross sections for levels with odd values of J at high incident electron energies. This is a direct consequence of the multi-pole picture of electron-impact excitation developed in our laboratory. However, we have observed one important feature that is different for Ne, Ar versus Kr, Xe. In the case of Kr and especially Xe, the cross sections for the upper four levels ($2p_1$, $2p_2$, ..., $2p_4$), as a group, are smaller than the cross sections for the lower six levels ($2p_5$, ..., $2p_{10}$), but this is not so for Ne and Ar. We explain this on the ground that the ten levels in Kr (and even more so in Xe) separate into an upper group of four levels and a lower group of six levels with very different ionization energies in contrast to the case of Ne and Ar where all ten levels have more or less the same ionization energy. Excitation into the upper set of levels with smaller ionization energy (larger electron orbit) is generally less favorable than into levels of higher ionization energy (the lower set). Thus for Kr and Xe the cross sections of the ten 2p levels are dictated not only by the quantum number J but also the ionization energy.

Another quantity of special interest is the sum of the direct excitation cross sections of all ten 2p level which corresponds to excitation into the entire 2p manifold. This "2p-manifold cross section" is found to increase from Ne to Xe. To quantify the variation, we write the 2p electron configuration of the 2p levels for the four atoms as $n'p^5(n'+1)p$ with $n'=2$ for Ne, $n'=3$ for Ar, $n'=4$ for Kr, and $n'=5$ for Xe. A plot of the 2p-manifold cross sections versus n' shows a good linear relation. This result may provide some insight toward understanding the fundamental nature of electron excitation processes.

5. For a long time a very puzzling observation in the study of electron excitation of rare-gas atoms is that the excitation cross sections of the non-resonant levels exhibit quite significant dependence on the gas pressure. While the resonant levels are known to have pressure-dependent excitation cross sections due to reabsorption of the resonant radiation, the pressure-dependence of the cross sections of the non-resonant levels is quite surprising. For several years we have conducted an extensive study on the origin of this anomalous pressure dependence. Our experiments showed that the observed pressure dependence for the non-resonant levels is due to cascades from the higher resonant levels which are known to be pressure dependent (through reabsorption). Thus the apparent excitation cross sections (the sum of direct excitation cross sections and cascades) of the non-resonant levels should have some pressure dependence. With our Fourier Transform spectroscopic technique we were able to measure directly the cascade cross sections into the non-resonant levels and found that such cascade cross sections are indeed pressure dependent. Interestingly when we subtract the cascade cross sections from the apparent excitation cross sections, the resulting direct excitation cross sections for the non-

resonant levels are indeed independent of pressure as they should be. We first conducted this study on Ar and Xe. Our more recent experiments on Ne and Kr further confirm it. Thus we have completely resolved this long-standing puzzle of anomalous pressure dependence.

6. The problem of anomalous pressure dependence has motivated us to conduct a quantitative study of the effects of reabsorption of the resonance radiation on the measured cross sections. Specifically we consider a resonant level (α) which can radiatively decay partly into the ground level (0) and partly into some non-resonant levels (β). The photons from the $\alpha \rightarrow 0$ transition can be re-absorbed by a neighboring ground-state atom generating atoms in the level-α and providing a second chance for the $\alpha \rightarrow \beta$ emission. The new $\alpha \rightarrow 0$ photons can be re-absorbed again and the cycle continues. The result is to increase the $\alpha \rightarrow \beta$ emission at the expense of the $\alpha \rightarrow 0$ emission. Since re-absorption is more likely at higher gas pressure, the measured emission cross section of the $\alpha \rightarrow \beta$ transition should increase with pressure ultimating reaching a asympotic limit of complete reabsorption where the $\alpha \rightarrow 0$ emission is all converted into $\alpha \rightarrow \beta$ transitions. We have measured the optical emission cross sections for a number of the $\alpha \rightarrow \beta$ transitions of the Ne, Ar, and Kr atoms at different gas pressures. The observed pressure dependence is in good agreement with calculations based on the theoretical model of Heddle.

The experiments outlined in the preceding paragraph provides a new approach for studying excitation into the resonant levels. To determine the cross sections for exciting a resonant level, one must detect the radiative transitions from that resonant level to the lower non-resonant levels as well as to the ground level and measure their optical emission cross sections. Transitions to the non-resonant levels are generally in the near ultraviolet to near infrared region therefore can be detected readily by photomultipliers. On the other hand the $\alpha \rightarrow 0$ transition for the rare-gas atoms is in the vacuum ultraviolet region where radiometry is very difficult. For this reason, there is a scarcity of the excitation cross sections of the resonant levels. Now we see, from the preceding paragraph, that by increasing the gas pressure we can "transfer" the $\alpha \rightarrow 0$ transition into the $\alpha \rightarrow \beta$ transition through reabsorption of resonant radiation. At the "high pressure" limit, the troublesome $\alpha \rightarrow 0$ emission completely disappears so that the excitation cross sections for the level-α can now be determined by just measuring the $\alpha \rightarrow \beta$ emissions with photomultipliers. In practice, however, it is not possible to perform the excitation experiment at the truly "high pressure" limit because of the poor performance of the electron beam at high pressures. Instead we measure the $\alpha \rightarrow \beta$ emission cross sections at different pressures in the "low" and "intermediate" pressure regions and then use the theoretical model to extrapolate these cross sections to the "high pressure" limit. In this manner we have been able to determine, for the first time, the excitation cross sections for a number of resonant levels of the Ne, Ar, and Kr atoms.

7. The electron-impact excitation cross sections measured in our laboratory not only provide the basis for understanding the fundamental electron-atom interactions but also find applications in other areas. For instance researchers in other laboratories are using our cross sections to study the electron energy distribution in a plasma. We have collaborated with a group in Germany (Essen) and completed an experimental determination of quenching coefficients in a hydrogen RF discharge by time-resolved spectroscopy.

8. An on-going effort in our laboratory is the development of ultracold trapped atoms as targets for electron collision experiment. The trapped atoms are all inside a very small cloud (0.5 mm). Thus it is easy to maintain a substantial fraction of the trapped atoms in an excited level with uniform density throughout the target using laser irradiation because all the trapped atoms are well within the laser beam. This makes the trapped-atom target very well suited for electron collisions with excited atoms. With laser irradiation we have produced a trapped Rb atom target containing 40% atoms in the 5P excited level and 60% in the 5S ground level. We passed an electron beam through the trapped Rb atoms and measured the ion yield using a 5P-5S mixed trapped atom target and a pure 5S trapped atom target. This allows us to determine for the first time the electron-impact ionization cross sections of the excited Rb(5P) atoms.

9. Publications

(a) "Measurement of Electron-Impact Excitation into the $3p^5 3d$ and $3p^5 5s$ Levels of Argon using Fourier-Transform Spectroscopy" by J.E. Chilton and C.C. Lin, Physical Review A **60**, 3712 (1999).

(b) "Electron-Impact Excitation Cross Section of Neon", by J.E. Chilton, M.D. Stewart, Jr., and C.C. Lin, Physical Review A **61**, 052708 (2000).

(c) "Cross Sections for Electron-Impact Excitation of Krypton", by J.E. Chilton, M.D. Stewart, Jr., and Chun C. Lin, Physical Review A **62**, 032714 (2000).

(d) "Electron-Impact Ionization Cross Section Measurements Out of the 5^2P Excited State of Rubidium", by M.L. Keeler, L.W. Anderson, and C.C. Lin, Physical Review Letters **85**, 3353 (2000).

(e) "Determination of Quenching Coefficients in a Hydrogen RF Discharge by Time-Resolved Optical Emission Spectroscopy" by T. Gans, C.C. Lin, V. Schulz-von der Gathen, and H.F. Döbele, Journal of Physics D: Applied Physics **34**, L39 (2001).

(f) "Use of Radiation Trapping for Measuring Electron-Impact Excitation Cross Sections for Higher Resonance Levels of Rare-Gas Atoms", by M.D. Stewart, Jr., J.E. Chilton, J.B. Boffard, and C.C. Lin, Physical Review A **65**, 032704 (2002).