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FINAL TECHNICAL REPORT, 1992 19 January 1993

Grant No. AFOSR-89-0256 The University of Chicago

Penning Detachment: A New Frontier

Principal Investigator: R. Stephen Berry Department of Chemistry and the James Franck Institute The University of Chicago

This Project brought to a threshold of productivity an attack on a kind of physical process that has essentially never been investigated, but that has now been introduced--conjecturally--to interpret several puzzling phenomena. The Project fell behind its original calendar because we were late in starting the project's work; we requested and received a no-cost extension to give the project enough time to yield its first publishable scientific results. These dealt with two-photon photodetachment, a process closely related to the main objective of Penning detachment. As a result of subsequent events, we have delayed publication of the first results concerning twophoton detachment, in order to get to Penning detachment as fast as possible.

Penning detachment is the name we have given to the process in which an excited species M*, atom or molecule, acts as an energy donor when it collides with a negative ion X⁻, also atomic or molecular, transfers its excitation energy and thereby releases the extra electron of the negative ion:

in direct analogy with the well-known and thoroughly studied process of Penning ionization,

$M^* + X \rightarrow M + X^+ + e^*$.

By contrast with Penning ionization, there is essentially no literature--two papers--on Penning detachment^{1,2}. (One brief abstract was also presented³.)However it has been invoked to interpret kinetic processes in plasmas behind shock waves and may be important in electric discharges and other ionized gas conditions.

To guote from our last Technical Report,

"Our early, crude calculations had indicated that the cross sections for Penning Detachment would be large¹, and that, as a result, the process might be important in plasmas, shocks, discharges and other conditions in which electronically excited neutrals and electronegative species would both be present in moderately high concentration. Exploration of this process was the first motivation to start the project. As the ideas were explored, some interesting possible scientific consequences emerged which could be achieved from Penning detachment; these included exploration of optically forbidden states of neutrals and perhaps a means to prepare

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beams of neutral clusters in which all the clusters are of the same size (monodisperse cluster beams)."

The efforts of this Project were concentrated on apparatus and on preliminary experiments, including a) developing a reliable, high-intensity negative ion source, b) setting up the laser photodetachment system and electron collection and counting system for the two-color, two-photon photodetachment experiments, and c) carrying out the photodetachment experiments with O^{-} , CI^{-} and CN^{-} . The negative ion source was developed to the point of yielding high, stable currents of those atomic ions, which are meant to be the first in both the photodetachment and Penning detachment studies. This objective was achieved after a lengthy series of modelling computations of the ion optics and experiments with various designs. In its present form, the duoplasmatron yields steady currents of several nanoamperes of O^{-} and as much as 35 nA of CI^{-} at about 1 kilovolt, corresponding to steady-state ion densities as large as have been previously achieved or larger. A sketch of the entire apparatus is Appendix 1; we are considering publishing the design and operating conditions of the negative ion source as a note in a suitable journal.

The results of the two-photon photodetachment experiments were: a) achievement of two-photon detachment from all three ions in 1-color experiments, all with 563 nm radiation, with only very rough measurements of the electron energies, only enough to be consistent with the expected electron spectra but no more, and with electron intensities quadratic in the laser intensity; some results for CN⁻ are shown in Appendix 2;

b) achievement of two-photon detachment in two-color experiments with CI⁻ and marginally with CN⁻, with no energy resolution;

c) measurement of a dependence of the electron energy on the ponderomotive effect of the laser field. These results were presented at the 1992 Meeting of the Division of Atomic, Molecular and Optical Physics of the American Physical Society in May, 1992.

The laser source for the two-photon detachment work as well as for the resonant two-photon excitation of the energy donors, now provides pulses of two tunable colors in synchrony with a variable delay between pulses. This source has been tested and used. The system consists of a doubled Nd-YAG pump and two dye lasers, one commercial and the other, home-made. The electron counting system is based on a multichannel plate (MCP) operating in counting mode. The electronics are now operated in a gated mode so that the MCP only counts during a window around the time of the laser pulse. The system is now operated with both beams, laser and ion, modulated, so that counting will be done under four conditions: (laser) on, (ion beam) on; on, off; off, on; and off,off. The average of the sum of signals from the first and fourth, minus the second and third, gives only the true on-on signal without the noise from the other three conditions. The same mode of operation will be used at first for the Penning detachment experiments. However for the best noise reduction in the Penning experiment, the laser, neutral and ion beams must all be modulated and the signal extracted as follows. If we indicate the three beams in that sequence and call,

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for example, (o-o-o) the average number of counts in the selected interval from the allon condition and (x-x-x) the all-off counterpart, the desired net signal is

1 = (0-0-0) + (0-x-x) + (x-0-x) + (x-x-0) - (0-0-x) - (0-x-0) - (x-0-0) - (x-x-x).

This will be the mode of data handling used for the three-beam system. To handle this operation, we have linked the electron time-of-flight energy analysis to a CAMAC-based data-handling system that has been used with other experiments. The detection of neutrals is also computer-controlled, through a separate PC.

We expect to continue the two-photon photodetachment work by carrying out measuremenis of the two-color, two-photon photodetachment of the chloride ion, to determine the dependence of the transition probability on ΔE , the difference in energies of the two photons (getting a two-dimensional map of transition probability as a function of total energy and ΔE) and on the relative polarization of the two photons. We will also include in the two-photon detachment study diatomic negative ions, at first continuing our work on CN⁻. Both this work and the Penning detachment study require considerably higher resolution of electron times of flight than we achieved during this Project. Since the Project's termination, we have constructed an new "magnetic bottle" electron spectrometer that is expected to give a resolution a resolution of order 10-20 meV, enough to resolve vibrational states but not rotational states, even of very light molecules. We also have constructed a detection system for neutrals which will enable us to measure approximate values of the total cross sections for both two-photon detachment and Penning detachment.

The apparatus is now almost ready for the first Penning detachment experiments, which will be done with Cl⁻ colliding with excited alkaline earth atoms.

Per: Jnnel

The personnel who worked on this project, in addition to the Principal Investigator, were a postdoctoral associate, Dr. Ming Zhou, and a graduate student, Mr. Richard Niedziela. These are the two people who have been with the project from its inception. Dr. Zhou is now working at the Jet Propulsion Laboratory, Pasadena, California. After the AFOSR Project concluded, support was received from the University of Chicago to continue the construction of the apparatus, and from the National Science Foundation for the support of a foreign visitor, Dr. Zdenek Herman, of the Heyrovsky Institute, Prague, (then) Czechoslovakia, to participate in the experiments. Dr. Herman will arrive in Chicago in March, 1993.

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ABSTRACT

The main objective of this project is a first exploration of a process not heretofore studied systematically, the detachment of electrons from negative ions when the ions collide with electronically excited neutral atoms or molecules. A steppingstone experiment, the two-color, two-photon photodetachment of the electrons from negative ions, is a part of the project as it is now being done this report describes the completion of the apparatus for the steppingstone experiment and the first photodetachment results, and outlines precisely what the goals for the next few months will be, based on the success of the ion source and the lasers. The steppingstone experiment is being conducted with Cl⁻⁻ ions to determine the dependence of the detachment probability on the total energy and the difference in energies of the two photons; when this is complete, the ion optics and target cell for the collisional detachment experiment will be completed while the photodetachment work is extended to the study of the CN⁻⁻ ion.

APPENDIX 1

The Apparatus for Two-Photon Photodetachment





The Photodetachment Signal from CN-





