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METASTABLE ENHANCEMENT OF C^+ AND O^+ CAPTURE REACTIONS.

FINAL TECHNICAL REPORT
COVERING THE PERIOD JUNE 87 THROUGH SEPTEMBER 90

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TO DEPARTMENT OF ENERGY,
DIVISION OF CHEMICAL SCIENCES,
WASHINGTON DC 20874

by

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ABSTRACT

We have studied single electron capture by 10 to 500 eV singly charged C and O ions traversing targets of H_2 and H with emphasis on comparing cross sections for metastable species with those for the ground state. For an H_2 target cross sections are of the order 10 $Å^2$ and 20 to 30 times larger than for ground state species. Electron impact ion sources typically produce 5 to 30% of their output in the metastable state. Previous published work has largely ignored (or failed to detect) the presence of metastables and is incorrect by as much as an order of magnitude. Discrepancies between data sets have been resolved and a reliable data set provided for energies from 10 to 10^5 eV. Similar experiments for an atomic H target are underway. We propose to extend the program to similar studies with multiply charged projectile species.

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A. INTRODUCTION

The present work involves measurement of the charge transfer neutralization of carbon and oxygen ions in H_2 and H targets which can be described by the general equations:

$$C^+ \text{ (or } O^+) + H_2 \text{ (or H)} \rightarrow C^0 \text{ (or } O^0) + H_2^+ \text{ (or } H^+) \qquad (1)$$

We concentrate on impact energies from near threshold to a few hundred eV. This is an energy region where processes are expected to be adiabatic and described in terms of curve crossings of the projectile-target complex. The reactants represent species where adiabatic potential energy curves are available from molecular spectroscopy. Thus there is a good opportunity to formulate and test theoretical predictions. Apart from theoretical interest these processes are very important to the modelling and understanding of processes occuring in the sheath of a plasma such as a tokamak device. The choice of carbon and oxygen represents common plasma impurities and the target H_2 or H represents fuel that is found in the wall region. Our choice of energies represents the sheath potential of a typical plasma device where impurity ions emerging from a plasma acquire most of their energy.

The cross section for a process described by Eq. 1 is very sensitive to the internal excitation state of the incoming projectile. For example when C^+ is incident on H_2 the cross section for metastable impact is a factor of ten greater than for the ground state. Hints of such behaviour have long been in the literature (for example Moran and Wilcox[1]) but have been largely ignored. There are a number of confusing consequences. A typical ion flux, whether from an ion source or from the center of a plasma, will contain a significant fraction of ions in metastable states (up to 20 to 30 % in some cases). The apparent cross section for neturalization will then be dominated by the metastable component which is often undiagnosed and ignored. Different sources produce different metastable fractions which in turn gives rise to different apparent cross sections. Most of the recent experimental measurements ignore the presence of metastables. As a result data from different groups disagrees by factors of three to ten and is quite unreliable. Data are often described as being for ground state beams (which is incorrect) leading theoreticians to compare with ground state theoretical calculations; any conclusions are likely to be irrelevent. Use of any of this information for modelling fusion or other plasmas is likely to give misleading results.

The major objective of the present work was to study the charge transfer process of Eq. (1) separately for the ground and metastable states of the projectile. This should provide correct data for modelling purposes, resolve differences in data published by different groups and provide a firm basis for the comparison with theory.

The present work represents a study of the charge transfer process with two defined states of the incoming projectile; the metastable state and the ground state. Thus we capitalize on the opportunity to vary the pre-collision channel. This provides an opportunity to see how the input channel influences cross section. In the present study the transfer of a single electron from H_2 to a incoming metastable C^+ involves an energy change of only a fraction of an eV; if the resulting target H_2^+ is left in a vibrationally excited state the energy transfer can be essentially zero leading to an accidentally energy resonant situation. For low energy impact this is likely to give a large cross section and that, we shall see, is what is observed. By contrast the transfer of a single electron from H_2 to ground state C^+ involves an energy defect of many eV, it is certainly not resonant and the cross sections at low energy are likely to be small; again this is what we observe. As best we can tell there is not a single theoretical treatment of how a change in the incoming channel (e.g. by excitation) can alter the cross sections for a process. There are of course many studies of cross section as a function of the post collision channel (through measurement of excited state formation) but none on the prior collision channnel.

The work to date can be considered in two parts. First we have studied the charge transfer neutralization of C^+ and O^+ in H_2 separately measuring cross sections for ground and metastable excited projectile states. These studies are complete and have been published.[2,3] We briefly describe the results below and refer the reader to the published articles for details. Secondly, we have started studies of the same process but with an atomic H target. There is need for a change of technology here with the provision of an atomic H target in the form of a beam from an RF discharge device. Substantial work has been done on this project, a paper on design considerations has been published, but final data on the cross sections are not yet in hand. We will describe the present status of the work. Finally we will discuss briefly the work that is contained in a renewal proposal to DOE which proposes to extend these studies to multiply charge projectile ions.

B. METASTABLE AND GROUND STATE C^+ AND O^+ ON H_2

Under a present DOE grant we have performed a study of single electron transfer processes[2,3] for C^+ and O^+ at energies from 10 to 500 eV. Of particular interest is the case of an H_2 target where there is an opportunity for a near resonant transfer (energy defect of zero) and which is of practical importance in modelling the edge of fusion plasma devices.

Figure 1 gives the basic experimental arrangement. Ions are produced in a controlled energy electron impact source, extracted at a few eV energy, mass selected (with an RF quadrupole) accelerated to the desired energy (10 to 500 eV) focussed and directed through a cell containing the target gas. Ions that survive the transit of the target are accelerated to 5 keV and counted with a channeltron. For a two component (one metastable and one ground state) beam the flux of ions I surviving transit of the cell is given by

$$I = I_o(1-F)\exp(-n\,\sigma_{gs}\ell) + I_o F \exp(-n\,\sigma_m\,\ell) \qquad (4)$$

Here I_o is the transmitted current when target pressure is zero, n is the number density of target atoms in the cell of length ℓ. The cross section for charge transfer neutralization of ground state ions is σ_{gs} and that for metastables is σ_m. The fraction of the ion beam in the metastable state is F and the fraction of ground state ions 1-F. If I is measured as a function of n the resulting experimental data can be deconvoluted[2,3] to give the two cross sections σ_{gs} and σ_m as well as the fraction of metastables F. Calculation of effective absolute values for cell density n and cell length ℓ requires a correction for density gradients at the cell's entrance and exit apertures;

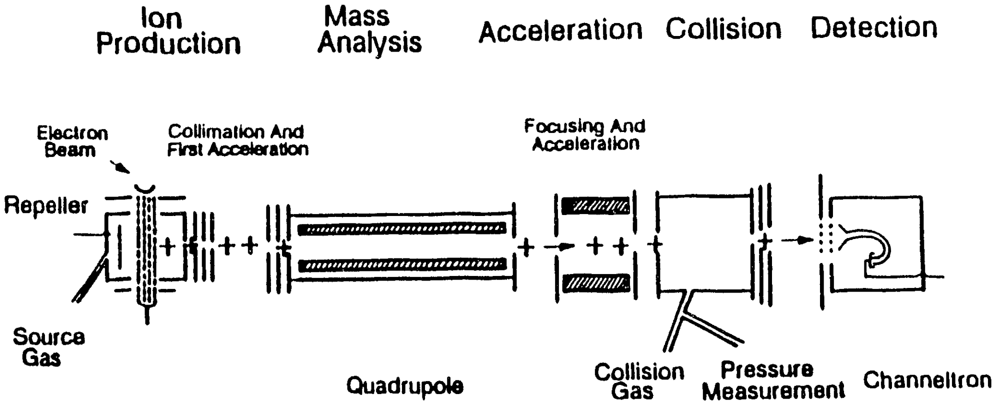

Fig. 1. Experimental arrangement used in the present study of C^+ and O^+ neutralization in H_2.

this was accomplished following a prescription by Van Zyl.[4] Precise control of electron energy and gas (CO) density in the ion source premits a reproducable metastable state fraction F. By choosing an electron energy between ground and metastable thresholds it is possible to create an ion beam of ground state ions only. In that case F = 0 and the data is represented by only a single exponential decay.

The apparatus described above has the great virtue of simplicity and through simplicity it operates reliably down to 10 eV energy, a level not previously described in the literature. A sample of the data for C^+ ions is shown in Figure 2.

The measured cross sections for metastables are about 10 $Å^2$ (see Fig. 2b) and vastly exceed the ground state cross sections of 1 $Å^2$ or less[2,3] (see Fig. 2a at low energies). We have demonstrated[2,3] that the earlier data of Moran and Wilcox at 500-2000 eV are correct and that the considerable body of data by Nutt et al.[5] are wrong. With the benefit of data from other groups[6,7] we can see that the metastable cross section for an H_2 target is about 10 $Å^2$ at 10 eV and remains virtually unchanged to 10^5 eV when it starts to fall. The ground state cross section is .3 $Å^2$ at 10 eV and rises with energy until it equals the metastable cross section from about 10^4 eV onwards.

We originally suggested[2,3] that the high cross section for metastables might be explained by the process being near resonant (energy defect of zero) if the target H_2^+ was left in a vibrationally excited state; near resonant processes are expected to have large cross sections. By contrast the ground state process will involve a large energy defect and should have a small cross section. Recently, however, we had access to unpublished data by Winter[8] on single electron capture by ground and metastable C^{2+} in H_2; this we show as Figure 3. Here again we see a metastable cross section ten times that of the ground state (at 500 eV) but for neither case is the reaction near resonant. It seems therefore that the concept of energy resonance will not explain the large differences in cross section.

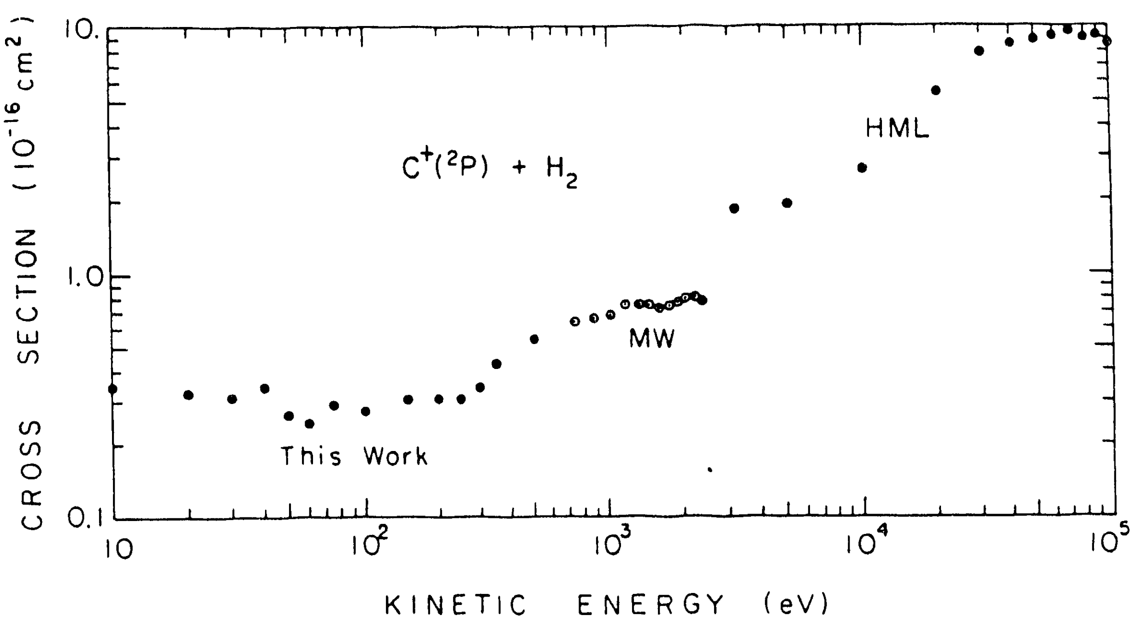

Fig. 2a. Charge transfer neutralization of ground state C^+ in H_2 as a function of energy. The present work is shown along with higher energy data. From Ref. 2.

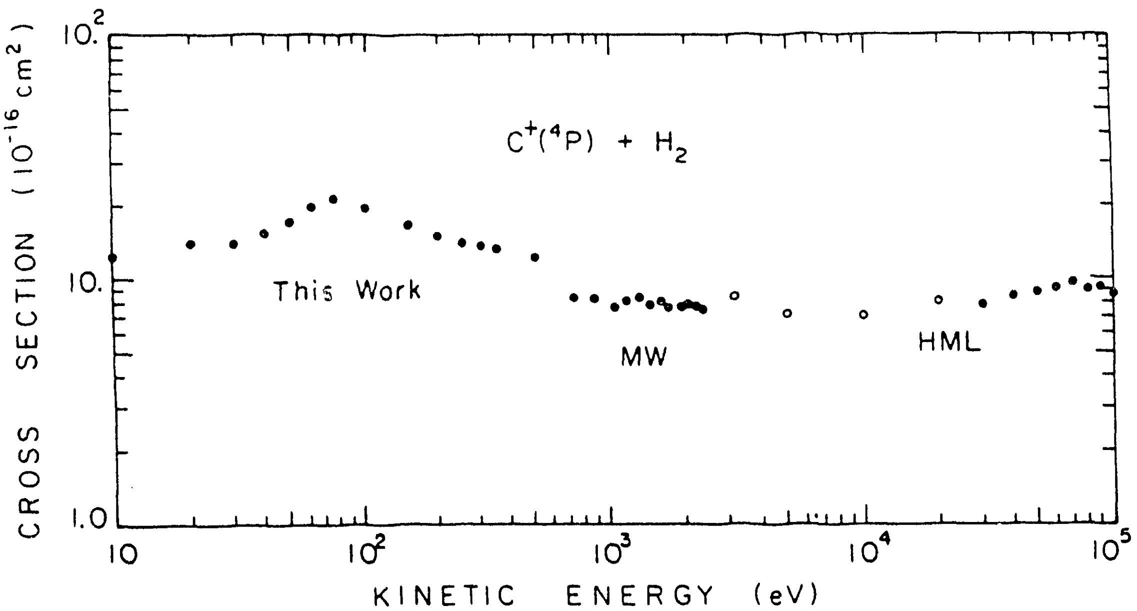

Fig. 2b. Charge transfer neutralization of metastable state C^+ in H_2 as a function of energy. The present work is shown along with higher energy data. From Ref. 2.

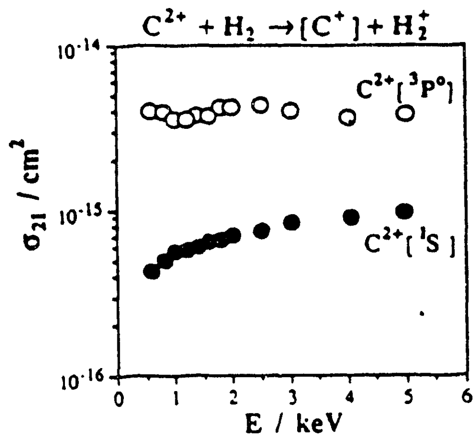

Fig. 3. Preliminary studies by Winter[8] on single electron capture by ground state (^1S) and metastable state ($^3P^o$) C^{2+} in H_2.

The general conclusion of the work is that the metastable state exhibits cross sections ten times that of the ground state at low energy. This is generally in accord with the observations of Moran and Wilcox[1] that have largely been ignored in the literature. All other studies at low energies have claimed that measured cross sections are for the ground state alone. We believe this to be incorrect and that the data are quite unreliable. For higher energies above 10 keV the cross sections for ground and metastable states become virtually the same and any neglect of metastable fractions in ions fluxes is pretty irrelevant. Discrepancies between data sets by different authors have now been resolved and we have established a reliable data set extending from 10 to 10^5 eV impact energy. The results of the work are now all fully published and the reader is referred to the relevant articles for full details.[2,3]

C. METASTABLE AND GROUND STATE C^+ AND O^+ ON ATOMIC H

We are currently performing the same type of single electron charge transfer studies on an atomic H target. The major technical change involves the provision of the atomic (rather than molecular) H source. The revised experimental system is shown in Figure 4. The H is produced as a beam by dissociation of H_2 in a Slevin type RF discharge source. We have recently analyzed the RF characteristics of this source.[9] Due to the tenuous nature of the target it is not feasible to use the attenuation technique of Eq. 4. Instead product ions are monitored and identified by a small inflection field magnetic spectrometer operating at 90^o to the incident ion beam. A small repeller voltage repells product ions out of the collision chamber and into the analyzing system. The system is rotatable through angles from 90^o to 0^o so that the incident ion beam can be monitored also as it is attenuated by the target gas. The H and H_2 components from our RF discharge source are determined using an electron beam (not shown in figure and which crosses the H/H_2 beam from the discharge) and the analyzing mass spectrometer system. Results are placed on an absolute basis by normalizing to the data for H_2 that we have recently published.[2,3] The experiment is greatly complicated by the fact that the Slevin type source produces only a 90% dissociation so that 10% of the target beam is H_2 for which cross sections can be very high. Thus one has a mixed beam of C^+ (or O^+) ground and metastable ions crossing a target beam that includes both H and H_2. By a process of elimination one can arrive at the desired cross sections for C^+ (or O^+) on H but the accuracy will inevitably be poor. We anticipate that the results will be greatly different from those of Nutt et al.,[5] since these workers normalized their H results to their H_2 data and that latter data is quite incorrect.

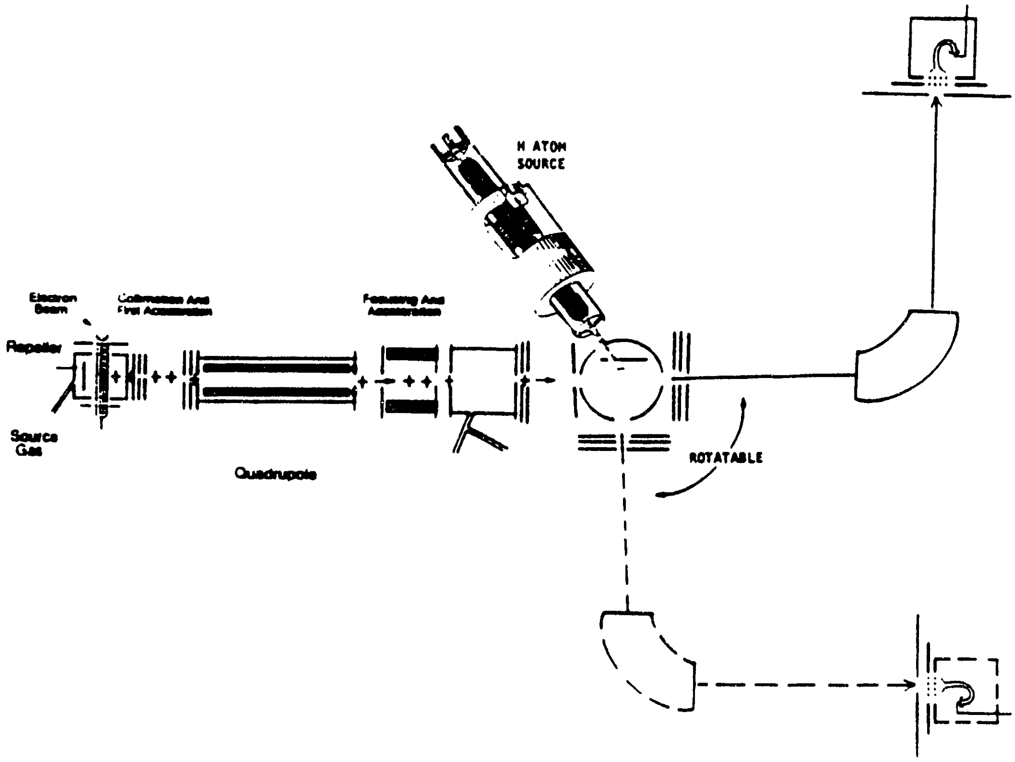

Fig. 4. Schematic of experimental apparatus used to measure charge transfer cross sections of ions with H atoms.

The apparatus is complete and preliminary data are being obtained. Some difficulties have been experienced in making the RF source produce the desired 90% dissociation of source gas (H_2) into H. Dissociation fraction of the emergent neutral flux is greatly dependent on the nature of the wall surface in the extraction capillary Various standard surface cleaning techniques have been employed and dissociation fraction of around 80% are now routinely obtained. An interesting feature is that the RF discharge seems to induce precipitation of pump oil inside the source shield. Due to the configuration of our source region that oil migrates to the extraction orifice and causes it to be periodically blocked. Other workers using this type of source seem to have experienced the same problem. Various strategies for eliminating the problem are being employed. These include altering the surface migration path and maintaining a continuous flow of source gas even when the apparatus is not in use. The phenomenon is not related to excessive oil in the vacuum system. Preliminary data for charge transfer of singly charged C and O ions on atomic H are now being obtained and the experiment should be completed during the next few months. A paper concerning the design of the RF cavity is in course of publication.

D. PROPOSED WORK.

A proposal for continuation of the work beyond the period of the present grant has been submitted to DOE. The general objective is to study the same processes of electron transfer but for multiply charged ions of C and of O; again targets will be H_2 and H. Again the emphasis is on studying the difference between ground and metastable excited species concentrating on low energies (10 to 500 eV) that are typical of a tokamak fusion plasma's edge. Experimental facilities will remain basically the same as use in the previous work reported here except that we will need to provide a source of multiply charged ions (rather than singly charged ions as at present).

As a source we propose a simple Electron Cyclotron Resonance (ECR) source following the general design of Sortais et al.[10] and Melin et al.[11] This is a very simple system designed to be added to accelerator terminals or to Molecular Beam Epitaxy systems. Part of our objectives is to diagnose the metastable fraction of the ions from such an ECR source. The literature provides very little indication of what excited state fractions might be expected from ECR sources.

The source has been fully designed and is now being constructed. It will be added to a Hitachi RMU-7L mass spectrometer that will provide both mass resolution and energy definition of the ion beam. This in a sense replaces the source-quadrupole part of the apparatus shown in Figs 1 and 4. All other parts of the experiement remain basically the same.

Very little prior work exists on the subject of how multiply charged ions in metastbale states pick up an electron by charge transfer. Most published work on this general area assumes that the ions from a source are all ground state and that metastable content is irrelevant; there is generally no evidence to support this contention.

In unpublished work Winters[8] has shown that 400 eV metastable C^{2+} in H_2

has a cross section of 40 Å^2, ten times that of the ground state. The cross section gap appears to be increasing towards lower energies. The experimental arrangement was almost the same as in our own work with the ions being produced in an electron impact source. Aumayr and Winters[12] also showed that 50% of a typical C^{2+} ion beam is metastable. Studies of neutralization of O^{2+} in H_2 by two different authors[13,14] show a difference of an order of magnitude in the few hundred eV energy region. Similar differences were apparent before in data for singly charged ions and explained by us[2,3] as due to differing levels of metastable content in the various experiments along with great differences between ground and metastable state cross sections. Beyond that,the literature contains a few standard expressions of concern but no information.

The general intent is to cover O and C ions with charge states from twice ionized to completely stripped, if at all possible. Some of these species (for example completely stripped ions and ions with only three electrons remaining) do not have metastable excited states and any measurements is clearly for a ground state species. For most of the other configurations metastable states exist, are expected to be present in ion beams, and may show cross sections greatly different from the ground state.

The experiment is now in course of assembly and the pursuit of this problem is covered by the proposed three year renewal of this project.

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F. PUBLICATIONS

The following two publications have fully described the results to data.

PHYSICAL REVIEW A VOLUME 41, PAGE 1408 1 FEBRUARY 1990

Charge-transfer reactions of ground-state C^+ (^2P) and metastable-state C^+ (^4P) ions with H_2 molecules

Yaodong Xu, T. F. Moran, and E. W. Thomas

Abstract: Cross sections for charge-transfer reactions of ground state C^+ (^2P) and metastable-state C^+ (^4P) ions with H_2 have been measured in the 10- to 500-eV kinetic energy range. Ground-state reaction cross sections range from 0.3 to 0.5 x 10^{-16} cm^2, and the corresponding values for metastable state C^+ (^4P) ions vary from 20 to 12 x 10^{-16} cm^2. both sets of cross-section values smoothly extrapolate to previously measured data at higher energies.

J. PHYS. B: AT. MOL. OPT. PHYS. VOLUME 23, PAGE 1235 APRIL 1990

Charge transfer reactions of ground O^+ (^4S) and metastable O^+ (^2D,^2P) ions with H_2 molecules

Yaodong Xu, E. W. Thomas and T. F. Moran

Abstract: Cross sections for charge transfer reactions of ground O^+ (^4S) and metastable O^+ (^2D,^2P) state ions with H_2 have been measured for reactant ions with 10 to 500 eV kinetic energies. Ground-state ion cross sections range from 0.5 to 0.9 x 10^{-16} cm^2 and metastable-state ion cross sections are approximately constant at 10 x 10^{-16} cm^2.

The following article describing the design feautres of the RF discharge source of atomic H has been submitted for publication and tentatively accepted subject to some revisions.

JOURNAL OF PHYSICS E (SUBMITTED)

Simple Method to Calculate the Operating Frequency of a Helical Resonator/RF Discharge Tube Configuration

R. F. Welton, E. W. Thomas, R. K. Feeney and T. F. Moran

Abstract: A practical technique is described to estimate the resonant frequency shift of a helical resonator due to addition of a coaxially mounted discharge tube. A simple formula is derived which relates this change in resonant frequency of the cavity to dimensions and dielectric properties of the discharge tube and its contents.

G. PERSONNEL

Edward W. Thomas of the School of Physics and Thomas F. Moran of the School of Chemistry have acted as co-PIs for the majority of the reporting period. The project was in part inspired by Mr. C. F. Barnett of the ORNL and an adjunct faculty person at Georgia Tech; Barnett was to have been co-PI with Thomas but passed away early in the period of the present grant.

Students Robert F. Welton (of Physics) and Yaodong Xu (of Chemistry) have been employed on the project for most of the reporting period. Both will use protions of this work as the basis for their PhD Theses; Xu is expected to graduate within the next twelve months and Welton in the year following.

The project has made use of technical service and shop facilities in both the schools of Physics and Chemistry as these were required.

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