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ADVANCED INTERDISCIPLINARY RESEARCH IN THE DEFENSE APPLICATIONS OF LABORATORY ASTROPHYSICS

A. V. Phelps, et al

Joint Institute for Laboratory Astrophysics

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JOINT INSTITUTE FOR LABORATORY ASTROPHYSICS

UNIVERSITY OF COLORADO

REPORT

NATIONAL BUREAU OF STANDARDS

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SEMIANNUAL TECHNICAL STATUS REPORT

August 1972 through January 1973

Research Sponsored by

Advanced Research Projects Agency

ARPA Order No. 492

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I. MODELING OF IONIZED GAS SYSTEMS

Program Leader - Dr. A. V. Phelps

Our studies of the modeling of ionized gas systems include a) the theoretical prediction of the conditions for the growth of a constriction in a weakly ionized gas, such as found in a molecular gas laser; b) the development of theories of radiation transport and hydrodynamics applicable to a wider variety of real situations, such as expanding and turbulent plasmas in geometries other than the usual planar geometry; c) the testing of radiative transport theories under controlled laboratory conditions; and d) the experimental determination of the laws of radiative scattering and absorption appropriate to radiative transport problems. The overall objective of this program is to supply experimentally tested understanding and computational techniques required for the analysis and prediction of the role of a) the generation and transport of radiation in plasmas and b) the transport and reactions of charged particles in weak to fully ionized gases. Because of its concern with a number of simultaneous atomic processes and with relatively large numbers of interacting atoms or molecules, this program also provides an important link between the experimental and theoretical determinations of cross sections and rate coefficients which make up much of the remainder of the program and the technology of ionized and excited gases which this work is designed to support.

Stability of Discharges in Weakly Ionized Gases (Dr. E. F. Jaeger, Dr. L. Oster and Dr. A. V. Phelps)

The objective of this theoretical project is the quantitative investigation of factors which control the growth of instabilities in weakly ionized gas discharges, such as used in high power molecular lasers. The present phase of this project is concerned with instabilities which result from a decrease in gas density and the accompanying increase in the ratio of the electric field to the gas density E/N and in the rate of ionization.

The charged and neutral particle equations describing the growth of ionization and the flow of charged and neutral particles during the transient following the application of voltage to a weakly ionized gas have been formulated and programmed for solution on a high speed computer. In their present form, the program solves four simultaneous equations of which three describe the energy, momentum and number balances for the neutrals and one describes the number balance for the electrons and positive ions. Infinite cylindrical geometry is assumed and the axial electric field is allowed to vary as expected for a constant external resistance in series with a fixed voltage supply. The electron transport and ionization rate coefficients are assumed to be determined by the ratio of the instantaneous axial electrical field to the local gas density as expected for high pressure, molecular gas lasers. Solutions obtained thus far have been for a gas with the properties of helium as given in a previous investigation of glow discharge stability by Ecker et al.[1] so as to make possible comparisons with the earlier

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[1] G. Ecker, W. Kröll, K. H. Spatschek and O. Zöller, Phys. Fluids 10, 1037 (1967).

results. More realistic transport and rate coefficients for helium and molecular gas mixtures are currently being generated under the program discussed in Section II of this report and will be applied to this problem during the next report period.

Results obtained using this computer program for helium at 1000 torr are illustrated in Figs. 1-2. These curves show the effect of a change in the radial distribution of the initial ionization and the effect of a change in the external series resistance on the growth of the electron density at the axis of the discharge. Figure 2 shows representative spatial distributions for the electron and gas densities. Figures 1 and 2 show results for an initial field strength of 1500 V/cm, a series resistance of 1000 ohm per cm of discharge length and an initial ionization distributed according to a Bessel function with zero density at the tube wall and a density of 10^{10} cm^{-3} on axis. Figure 1 shows that the electron density rises to a nearly constant value in about 20 μsec. The magnitude of this quasi-steady state electron density is determined largely by a balance between the ability of the circuit to supply current and the steady state current density vs E/N relation characteristic of the gas under study. This steady state current density or input power density per molecule vs E/N relation can be calculated for any weakly ionized gas mixture in which the electron loss by diffusion and electron production by cumulative ionization are small provided the appropriate electron-ion recombination coefficient is known. The electron density distribution curves $n_e/n_e(r=0)$ of Fig. 2 show that during this quasi-steady state period the electron density becomes relatively independent of position. Also, the neutral atom density curves (not plotted for these early times) show

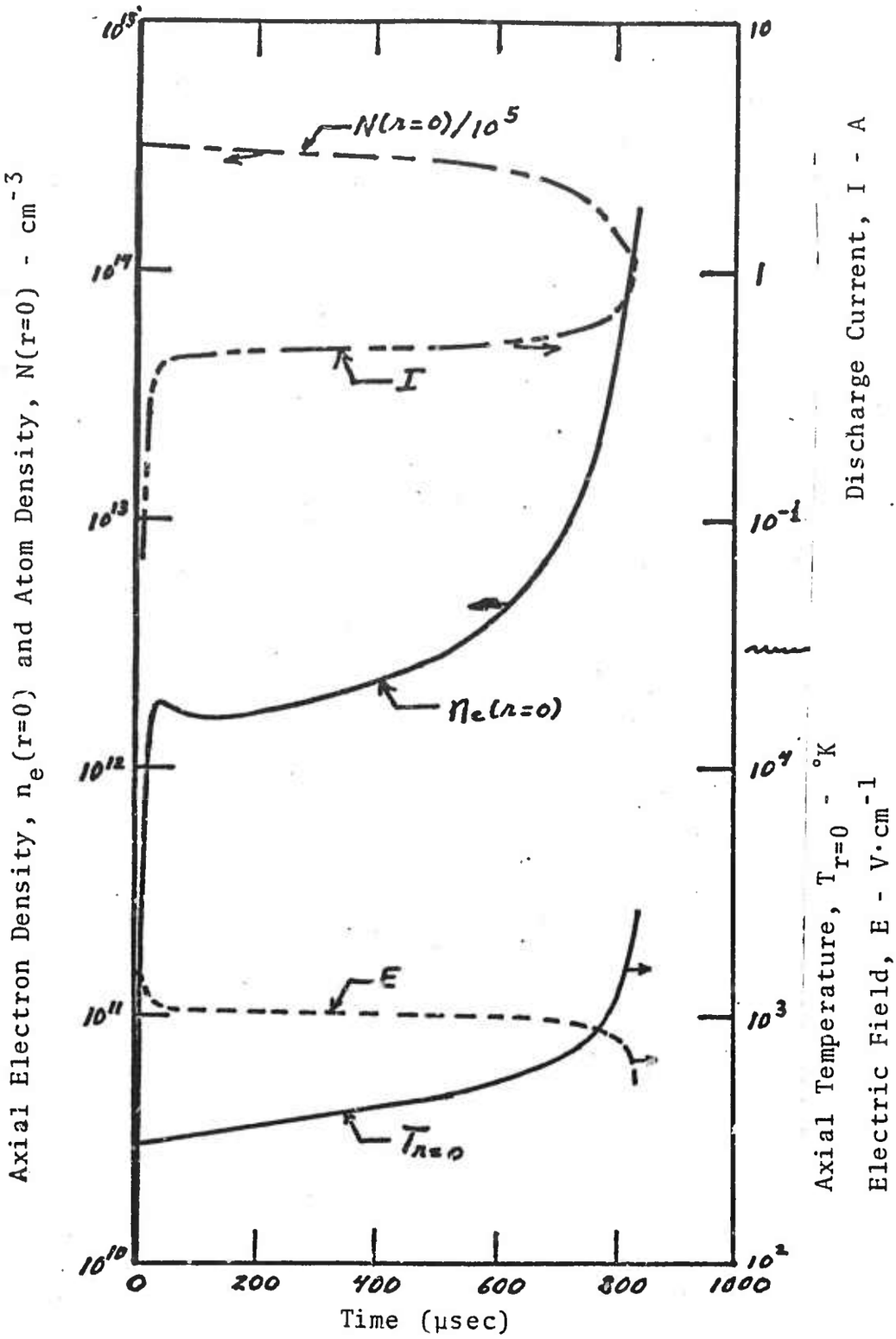

Fig. 1. Time variation of axial gas temperature T(r=0), electron density $n_e(r=0)$, and atom density and of discharge electric field E and current I. The initial electric field strength is 1500 V/cm, the initial helium pressure is 1000 Torr at 300°K, and the series resistance is 1000 ohm per cm length of discharge. The initial electron density is 10^{10} electron/cm^3 at the axis and is distributed as a Bessel function.

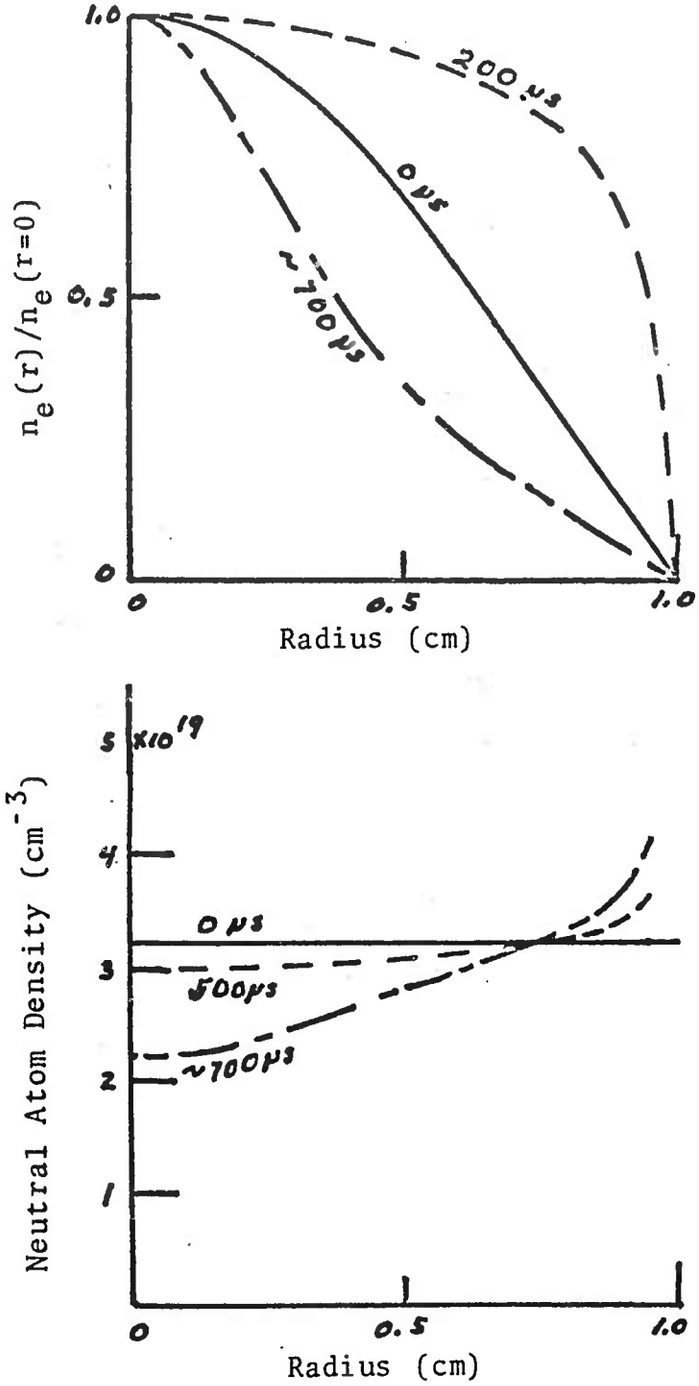

Fig. 2. Spatial variation of the electron and neutral atom density at various times during the transients of Fig. 1.

relatively little change from their initial constant value.

After the quasi-steady state current (about 0.5 A) flows for about 500 μsec the energy input (about 200 J/ℓ) is sufficient to raise the gas temperature significantly. The resultant outward flow of gas raises the E/N and ionization rate near the axis and lowers the E/N and ionization rate near the wall. The discharge thus constricts toward the axis as shown in Fig. 2. For the discharge conditions used in this example the discharge continues to constrict until the current channel becomes too narrow for accurate calculations. The curves shown for 700 μsec are only roughly correct because of the use of a large artificial viscosity term inserted into the equations in order to reduce the computer time for the longer runs.

A few calculations have been made for cases in which the initial ionization is restricted to a channel of much smaller diameter than the distance to the tube wall as is expected for TEA lasers with pin type cathodes. The general behavior of the discharge is similar to that shown in Fig. 2, i.e., an initial expansion of the discharge to about twice the initial diameter is followed by a collapse to a narrow channel. However, when all other parameters are unchanged the time scale for the rise in gas temperature is about an order of magnitude shorter when the diameter initial ionization is reduced by an order of magnitude. Although the total energy input decreases with radius there is some tendency for the energy input per unit volume prior to a given temperature rise to increase with decreasing radius of initial ionization and with decreasing external resistance.

As reported in the Quarterly Management Report for October 1 through December 31, 1972 these theoretical studies of discharge stability

have led us to propose that the stability of a pulsed discharge in CO_2 can be significantly improved by the addition of a small amount of a detaching gas such as H_2 or CO, i.e., the addition of a gas which detaches the electron from O^- ions formed in the dissociative attachment process in CO_2. The high degree of sensitivity of a diffuse discharge in an attaching gas to the growth of instabilities is the result of the very rapid change in the net ionization frequency with E/N which occurs at the E/N appropriate to low and moderate current densities in a recombination dominated discharge (see above). We have tested this hypothesis by experiments carried out under another contract. Thus, we found discharges in mixtures of He, CO_2 and H_2 in the ratios of 90:9:1 to be much more reproducible than were discharges in 90% He and 10% CO_2. These measurements were carried out at atmospheric pressure with axisymmetric Rogowski shaped electrodes spaced at about 2 cm and with an applied voltage of about 11 kV/cm.

All of the preceding numerical results should be considered preliminary pending further checks of the computer code and more extensive variation of the discharge parameters.

Scattering and Transport of Resonance Radiation (Dr. A. V. Phelps, Dr. J. E. Jenkins and Dr. R. L. Smick)

The measurements of the collision broadening of potassium resonance lines by potassium and of the specular reflection at the potassium-glass interface have been completed and were presented at the Conference on Spectral Lines. Soon afterwards Dr. Jenkins returned to England and Dr. Smick began assembling apparatus for the measurement of the spectral distribution of scattered radiation using laser excitation. The long-awaited cw dye laser was delivered by the manufacturer. As expected, the line width of the laser output was significantly greater than desired for our experiments. Accordingly, some effort has been devoted to the development of a fixed etalon for use inside the laser cavity and to the assembly of a rapidly swept, tunable etalon for monitoring the laser operation. At present we are able to obtain reasonably stable laser operation with effective line width of about 0.01 μm. This value is comparable with the resolution of our spectrometer and should be satisfactory for our present needs. A new oven for the Na vapor cell has been built with windows for the incident and reflected light beams and for observation of the diffuse scattering. The data recording equipment has been revised to make it compatible with an available multichannel scaler. It is expected that this system will be operational during the next report period.

Radiation Hydrodynamics (Dr. D. G. Hummer, Dr. J. I. Castor and Mr. P. Kunasz)

Line formation in static spherical geometry. A very general and efficient code for solving line transfer problems in an extended static spherical cloud has been completed and extensively tested. This code utilizes the non-linear iteration technique (the so-called variable Eddington factor method) used previously for continuum problems,[1,2] generalized to the case of non-gray opacity with coupling between frequencies. An account of this work is being prepared for publication.

To illustrate the effects of geometry upon the radiation field, we present some results for a rather artificial model that allows comparison with the standard plane-parallel case. We consider a spherical shell with inner radius R_c and outer radius R surrounding a hollow core. The mean optical thickness of the shell is T and the electron temperature and density are constant. The opacity is regarded as a general function of radius and frequency; for the present purpose we choose $k(\nu,r) = K\phi(\nu)r^{-2}$, where $\phi(\nu)$ is the Doppler profile normalized to unity. Figure 3 shows how the radial behavior of the source function (which is proportioned to N_2/N_1) is modified as R/R_c increases from unity, the plane-parallel limit, in an atmosphere of fixed optical thickness T, here taken to be 1000. Two cases $\varepsilon \equiv C_{21}/C_{21} + A_{21} = 10^{-2}$ and $\varepsilon = 10^{-4}$ are considered. In the former case, in which the product $\varepsilon T \gg 1$, most of the photons created are destroyed in the gas (energy returned to electron gas), so that at the inner parts of the cloud

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[1] D. G. Hummer and G. B. Rybicki, Monthly Notices Roy. Astron. Soc. 152, 1 (1971).

[2] J. P. Cassinelli and D. G. Hummer, Monthly Notices Roy. Astron. Soc. 153, 9 (1971).

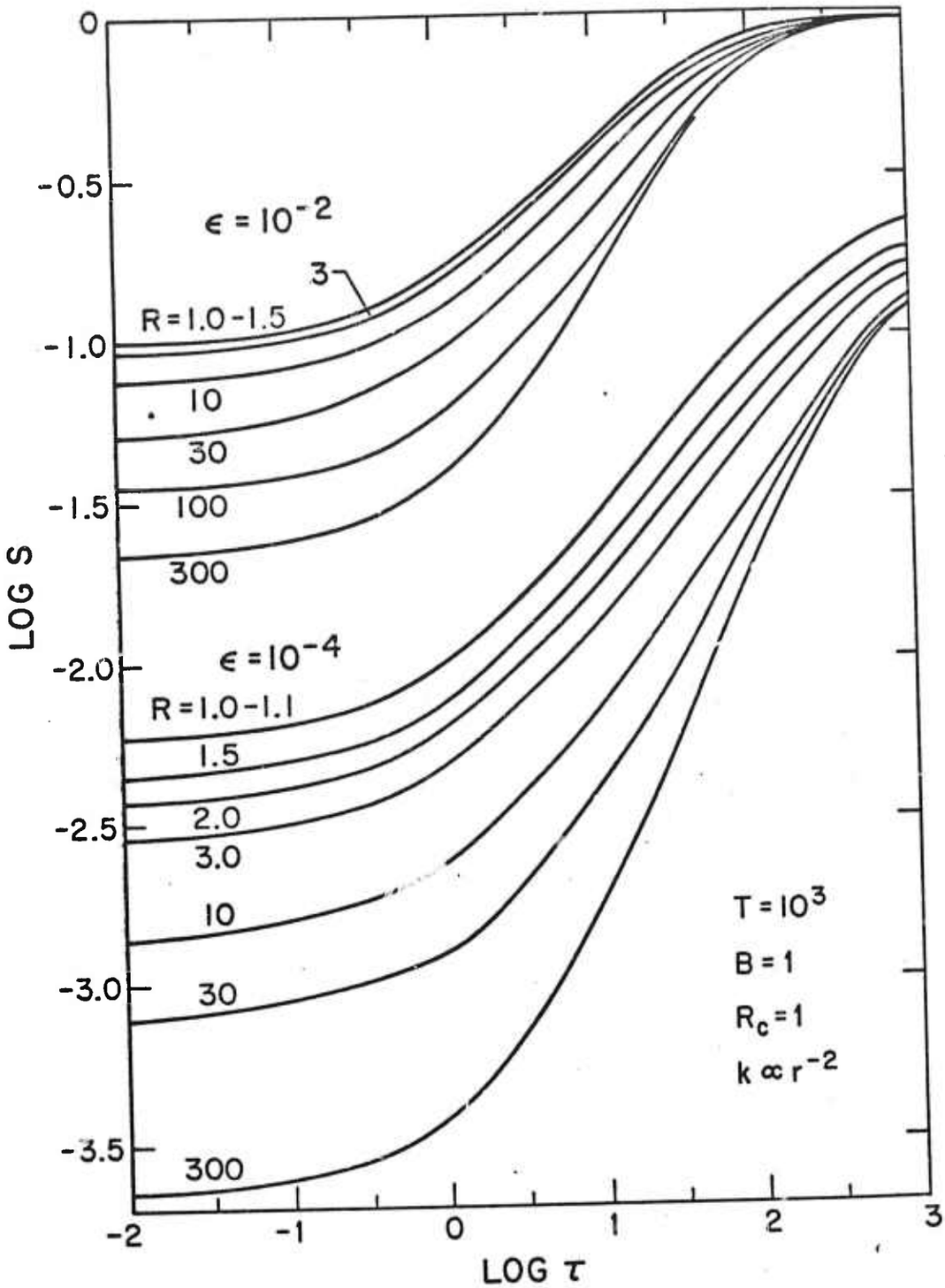

Fig. 3. The source function vs. optical depth for spherical shells with inner radius $R_c=1$ and outer radius R as shown on the curves. The total optical thickness of the shell is 10^3, the temperature is constant throughout and the opacity is proportional to r^{-2}. The upper family of curves corresponds to $\varepsilon \equiv C_{21}/C_{21} + A_{21} = 10^{-2}$ and the lower family, to $\varepsilon = 10^{-4}$.

the excitation is essentially independent of geometry. In the outer part of the cloud, the excitation decreases steadily as the size of the cloud increases. For $\varepsilon = 10^{-4}$, $\varepsilon T \ll 1$, so that most of the photons escape. In this case, the level of excitation throughout the cloud depends on geometry quite strongly.

In the special case of a homogeneous sphere (constant density), the integral for the source functions can be transformed into a form that is amenable to solution by the kernel approximation method.[3] This calculation is very easy and straightforward to program, and gives results that should be accurate to better than one percent. While these results were first obtained in order to check the more general code, they are of some interest in themselves, for a comparison of the source functions with those for plane-parallel slabs whose optical thickness is equal to the optical diameter of the sphere show the following result:

$$S_{sph}(\tau) = CS_{pp}(\tau)$$

when $C \lesssim 1$ and is practically independent of τ, i.e. depends only on ε, T and the frequency-dependence of the opacity. The smallest value of C encountered in a very exhaustive survey of the parameter space for both Doppler and Lorentz profiles was $C_{min} \approx 0.44$. A similar result was obtained by Avery, House and Skumanich[4] for finite cylinders from Monte Carlo calculations.

Some light on this result is obtained from the consideration of the probability that an excited atom will emit a photon that escapes the cloud with no

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[3] E. H. Avrett and D. G. Hummer, Monthly Notices Roy. Astron. Soc. 130, 294 (1965).

[4] L. W. Avery, L. L. House and A. Skumanich, J. Quant. Spectrosc. Radiat. Transfer 9, 519 (1969).

further scattering. We have shown, for any distribution of density with radius, that the probability $P_{sph}(\tau)$ is related to the corresponding quantity for the plane-parallel case for $P_{pp}(\tau)$, by the relation

$$P_{sph}(\tau) = P_{pp}(\tau)Q(r)$$

where Q(r) is an integral containing the radial-dependence of the density and satisfies the following inequalities:

$$1 \leq Q(r) \leq 2 \ , \quad \text{Doppler profile}$$

and

$$1 \leq Q(r) \leq 3/2 \ , \quad \text{Lorentz profile.}$$

Thus the escape probability depends primarily on the shortest path to the surface and only very weakly on longer paths. It appears that this circumstance might provide the basis for a systematic calculational procedure for handling radiative transfer problems in complicated geometries.

For the results shown in Fig. 3 in which the level of excitation does depend strongly on the size of the sphere, some other factor must be operative. This seems to be a dilution effect, since the power input goes as R (for $k(r) \sim r^{-2}$), while the available volume goes as R^3. This point is now being examined in detail.

Line formation in expanding spherical geometries. While the variable-Eddington factor procedure can be generalized to handle radiative transfer in spherical atmospheres with velocity fields, a number of problems have to be overcome. In order to obtain accurate solutions that can be used to check the results from the variable-Eddington factor procedure, we have devised a less general non-iterative technique that can treat velocities up to a few

times larger than the mean thermal velocity. This method depends on a generalization of Rybicki's[5] rearrangement of the standard Feautrier[6] procedure for solving the discrete form of the radiative transfer equations. Although the matrix of the system is very large, all but one of the submatrices are tri-diagonal or have some other useful special form, so that the amount of computing required is surprisingly small. This procedure has been coded and is now nearly debugged. In the limit of zero velocity, it should yield a good check on the Eddington-factor results for the static case, since it is a completely independent procedure.

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[5] G. B. Rybicki, J. Quant. Spectrosc. Radiat. Transfer 11, 589 (1971).

[6] P. Feautrier, C. v. hebd. Seanc. Acad. Sci. Paris 258, 3189 (1964).

Plasma Statistics (Dr. W. E. Brittin and Dr. W. R. Chappell)

Work was done on the problem of formulation of fundamental theories designed to treat interacting composite particles, such as electrons, protons, atoms, ions, etc. Although no completely satisfactory theory was discovered through this work, it was felt that this work provided part of the basis for subsequent work which appears to be satisfactory. Basically, the general idea was an attempt to treat various composite particles as though they were elementary particles with internal structure. In essence what was sought was a generalized quantum mechanical transformation which would allow one to pass from the standard representation which describes the elementary particles comprising the system to a new basis described in terms of composite particle quantities.

II. TRANSPORT COEFFICIENTS AND REACTION RATES

Program Leader - Dr. S. J. Smith

The prediction of the performance characteristics of ionized gas devices, such as the efficiency, power output and stability of high power lasers, requires that one be able to calculate accurate values of electron transport and reaction rate coefficients for electrons and ions. In order to meet some of these requirements we are engaged in: a) a determination of sets of electron collision cross sections consistent with measured electron transport and ionization coefficients and with recent electron beam measurements of excitation cross sections for the upper excited states of gases found in molecular lasers; b) measurement of cross sections for molecular dissociation; c) evaluation and compilation of low energy electron, photon, and ion collision cross section data; and d) determination of negative ion characteristics, such as single and multiple photodetachment cross sections, negative ion stability and low electron energy scattering parameters.

Electron Transport and Ionization Coefficients (Dr. G. E. Chamberlain, Dr. L. J. Kieffer and Dr. A. V. Phelps)

For the past six months we have continued in our effort to summarize and analyze the techniques for measuring electron transport and ionization coefficients. We are compiling a complete set of these types of data for N_2 in order to:

(a) compare the data taken by differing techniques

(b) look for systematic errors

(c) be able to test against the data our criteria for data selection and estimates of systematic errors as these criteria and estimates develop.

We have concentrated on measurements in N_2 since we believe they exemplify most of the problems that occur in the measurement of transport and ionization coefficients and because of the importance of N_2 in molecular gas discharge lasers. We have put together, in first draft form, several components of our review.

(A) A summary of the principal techniques (drift tube with electrical shutters and without shutters) for measuring drift velocity and a discussion of the corrections to the primary data that are necessary to account for effects of diffusion, ionization, absorbing boundaries, and high gas densities.

(B) A summary of the Townsend-Huxley technique for measuring the characteristic energy (diffusion/mobility) by observing the radial spread of a swarm. The high sensitivity of the method of geometric alignment, space charge, and attaching gas impurities are noted. To this is to be added a discussion of systematic effects in the Townsend-Huxley method due to the assumed boundary conditions. An additional section is to be written on diffusion/mobility values obtained from drift gap measurements, although the technique involved is already covered in the discussion of drift velocities

obtained by the drift gap technique.

(C) We have begun a discussion and made some estimates of the effects of space charge with and without ionization on the measured values of transport and ionization coefficients. Drift velocities are at least susceptible to space charge; no experimental evidence has been found for a change in measured drift velocity due to high electron densities. Ionization and diffusion measurements are certainly sensitive to space charge and a simple model has been considered. Additional material in the literature exists and will be added to the space charge review. Our current efforts are directed toward completing the review of diffusion measurements, initiating and completing a review of ionization measurements, and augmenting the review and modeling of space charge effects.

Electron transport and rate coefficient bibliography scheme. The indexing scheme for maintaining the Information Center's bibliography of data on electron transport and rate coefficients is defined by the list of index terms and descriptors shown in Table 1. The bibliography will be sorted on the index terms, the descriptors will be attached to each citation of the paper as supplementary information on the type of paper and type of measurement.

The data categories are meant to be general categories indicating the type of coefficient. For example, measurements of (diffusion coefficient)/(mobility), or characteristic energy, are to be found under Diffusion. The Collision Frequencies category has been added to cover the numerous types of elastic and inelastic collision frequencies that occur in the literature. Our coverage is expanded to include energy relaxation measurements which will be found under Excitation, or Collision Frequencies as appropriate to the specific paper.

We have dropped the Conductivity category. Papers with measurements of

conductivity from which drift velocity or collision frequency values can be obtained will be indexed in the appropriate category.

The Analysis descriptor has been added to identify papers in which electron collision cross sections have been determined by making comparisons with electron transport and rate coefficient data.

Table 1

Index Terms and Descriptors for Bibliography of Data on Electron Swarm-Gas Interactions

INDEX TERMS	DESCRIPTORS
(1) Data Categories	
Drift Velocity	Experimental
Diffusion	Theoretical
Excitation	Analysis
Ionization	
Attachment	Drift
Detachment	Afterglow
Recombination	Discharge
Collision Frequencies	Breakdown
Distribution Function	
(2) Species	
Atoms	
Molecules	
Mixtures	
(3) Citation Number	

JILA Information Analysis Center (Dr. L. J. Kieffer)

In addition to our continuing to keep current bibliographies on low energy electron collision cross sections and photoabsorption and ionization cross sections and compiling the data for these processes, we have initiated a new project on electron transport and rate coefficients. A current bibliography and data compilation will be maintained. This is a follow-up to a project which was carried on for many years by Professor J. Dutton and which is being terminated by a critical data compilation mentioned in previous reports. We have devised a new indexing scheme (see Table 1) and expanded the coverage. This project is being carried out by Dr. G. E. Chamberlain of the Information Center Staff.

We continue to help Dr. A. V. Phelps by preparing best cross section sets for discharge laser modeling. Some revisions in the currently used nitrogen cross section have been made but more work will be done.

Work has begun in preparing a set of atomic hydrogen cross sections for use in modeling electrical discharges in H_2.

Molecular Dissociation Processes (Dr. L. J. Kieffer)

No experimental work was done under this project during this reporting period. Data taken last July (see previous semiannual report) were analyzed. Two manuscripts are being prepared for publication.

Electron Transport in Mercury Vapor (Dr. A. V. Phelps)

As the result of numerous inquiries from laser development groups we have assembled and reviewed most of the available data on electron transport and ionization coefficients in mercury vapor. We have also reviewed many of the available references dealing with the spectra emitted by the first few excited states of mercury and with the collision processes leading to the production of these states.

The available electron transport coefficient data for mercury consist primarily of measurements of electron mobility and electron "temperature" as determined from measurements in the positive columns of low pressure discharges. There is only one measurement of electron drift velocities using the drift tube technique[1] and this author's results show a large scatter from one graph to another. Two groups have measured Townsend ionization coefficients, but there appears to be no measurement of the ratio of the diffusion to mobility coefficients. It is strongly recommended that any attempts to determine a set of electron collision cross sections consistent with the positive column data be carried out with a computer code capable of including the effects of electron-electron scattering on the electron energy distribution. Unfortunately, our code is for weakly ionized gases only and does not include the effects of electron-electron scattering.

At present there seems to be no completely consistent model of radiation sources and collision kinetic which will describe the available data on fluorescence in mercury vapor. It is therefore recommended that appropriate spectroscopic and kinetic experiments be undertaken.

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[1] E. W. McCutchen, Phys. Rev. 112, 1848 (1958).

Three experiments which seem particularly valuable are a) the determination of the molecular constants of the states of Hg_2 formed from the Hg $^3P_1$ and $^3P_0$ levels of Hg using laser induced fluorescence of higher Hg_2 states as proposed by Hessel and Smith,[2] b) the measurement of far wing fluorescence following optical excitation of the $^3P_1$ state and c) the measurement of far wing absorption at high mercury temperatures, i.e., a modern version of the Kuhn and Freudenberg[3] experiment. With the results of this combination of experiments one should be able to construct a set of potential energy curves and transition probabilities appropriate to the excited states of mercury of laser interest.

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[2] M. M. Hessel and E. W. Smith, proposal to Air Force Weapons Laboratory.

[3] H. Kuhn and K. Freudenberg, Z. Physik 76, 38 (1932).

Negative Ion Kinetics (Dr. W. C. Lineberger, Dr. J. Hall, Dr. H. Hotop, and Mr. T. A. Patterson)

During the present contract period we have completed our studies of metallic negative ions of gold, platinum, copper, and silver. Two manuscripts have been prepared[1,2] which describe these results; these papers will be published in the Journal of Chemical Physics. Further studies of metallic negative ions using our new sputtering ion source will be pursued when the laser photoelectron spectroscopy apparatus is available.

The studies of the electron affinities of the noble metals suggest that we should be able to obtain relatively accurate estimates of the elements in the three long series: (K→Cu, Rb→Ag, Cs→Au) by means of horizontal isoelectronic extrapolations. In particular, Charkin and Dyatkina[3] and Zollweg[4] have shown that the energy difference between the two configurations $d^k s^2$ and $d^k s$ (k=0,...,10) increases nearly linearly with k. Since it is well established that the ground state configuration of negative ions in the three long periods is $d^k s^2$, one can estimate the electron affinities of these atoms if those for K, Rb, Cs on the one side and those for Cu, Ag, and Au on the other side are known, because thereby the slope of the straight line representing the $d^k s^2 \rightarrow d^k s$ energy differences between the negative ion and the $d^k s$ neutral atom state is fixed. The results of such an extrapolation are presented in Table 2, and probably represent the best present estimates of the electron affinities of the elements in the three long series.

For this extrapolation we had relied upon the alkali electron affinities

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[1] H. Hotop, R. A. Bennett and W. C. Lineberger, J. Chem. Phys. (March 1, 1973).

[2] H. Hotop and W. C. Lineberger, J. Chem. Phys. (March 1, 1973).

[3] O. P. Charkin and M. E. Dyatkina, J. Struct. Chem. 6, 397 (1965).

[4] R. F. Zollweg, J. Chem. Phys. 50, 4251 (1969).

Table 2

Electron affinities of the elements in the three long series [eV]

	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu
Clementi[a]	0.92	-----	-0.14	0.40	0.94	0.98	-1.07	0.58	0.94	1.28	1.80
Charkin and Dyatkina[b]	[0.50]	-1.62	-0.39	0.15	0.6[illegible]	0.85	-1.19	0.11	0.72	1.10	1.35
Hotop et al.[c] (Zollweg[d] method)	[0.47]	-1.62	-0.80	-0.11	0.51	0.85	-1.19	0.14	0.65	1.13	[1.23]
	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag
Charkin and Dyatkina[b]	[0.60]	-0.50	0.30	1.04	1.32	1.28	0.96	1.44	1.34	1.40	1.48
Hotop et al.[c] (Zollweg[d] method)	[0.42]	-1.74	-0.76	0.08	0.77	0.86	0.63	1.04	1.12	0.40	[1.30]
	Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au
Hotop et al.[c] (Zollweg[d] method)	[0.39]	-0.54	0.44	-0.78	-0.05	0.98	0.09	1.10	1.58	2.12	[2.31]

[a] Ref. 6

[b] Ref. 3

[c] Ref. 1

[d] Ref. 4

calculated by Weiss.[5] While we would like to measure the electron affinities of the alkali metals with our tunable laser apparatus it is not currently possible for us to make 2 μm tunable light, which would be required to see the photodetachment thresholds. However, recent calculations by Norcross and Moores have shown that, at the thresholds for the first excited state channels in the alkalis, there should be a pronounced cusp-like feature. The location of this cusp would thus provide an accurate measurement of the electron affinities of the alkalis. Further these cusps will appear in an energy region accessible to our tunable laser apparatus. Consequently, we have constructed a double charge transfer alkali negative ion source. Our preliminary measurements on sodium show that this cusp is in fact present and imply that EA(Na) = 0.542 ± 0.010 eV. Similar measurements on other alkalis are currently in progress.

During the present reporting period, we have been very fortunate to have several JILA visitors working on the megawatt pulsed nitrogen laser. Professor V. Ehlers has completed the testing and performance evaluation of the laser and a manuscript describing the laser will soon be submitted to the Review of Scientific Instruments. Dr. R. Keller has successfully used the nitrogen laser to pump dyes which laze in the 8,000-9,000 Å region. By use of several dyes as excitation transfer agents, he has been able to achieve fairly efficient conversion (greater than 5%) from 3371 Å to 8500 Å. This infrared laser will be used to determine the electron affinity of atomic oxygen.

While waiting for machine shop modifications to the new charge transfer source system, some existing data have been analyzed to yield electron affinity information for the ions NH_2^-, NH^-, OH^-, SO_2^-, and S_2^-. The resul-

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[5] A. W. Weiss, Phys. Rev. 122, 1826 (1961); 166, 70 (1968).

[6] E. Clementi, Phys. Rev. 136, A980 (1964).

tant affinity values cannot lay claim to such high precision and absolute accuracy as our careful measurements and analysis of NO^- and O_2^-. However, the errors associated with the experimental data are only $\lesssim$ 10 meV, the primary uncertainties being in the interpretation of rotational effects. All of the data have been taken relative to O^- and have been expressed in absolute units using the O^- result of Hotop, Patterson and Lineberger. Thus we report $EA(NH_2) = (0.779 \pm 0.037)$eV, $EA(OH) = 1.829^{+0.010}_{-0.014}$ eV, $EA(NH) = (0.38 \pm 0.03)$eV, $EA(SO_2) = (1.097 \pm 0.036)$eV, and $EA(S_2) = (1.663 \pm 0.040)$eV.

Additionally the angular distributions were measured for OH^- and NH_2^- to yield the anisotropy parameter β at 4880 Å as $\beta(NH_2^-) = 0.027 \pm 0.012$ and $\beta(OH^-) = -0.993 \pm 0.040$. Our data show no $OH(1,0)OH^-$ peak in the photodetachment spectrum larger than 0.0006 of the strong peak $OH(0,0)OH^-$. Thus, using arguments given by Branscomb[7] we can calculate an upper limit on the change in internuclear separation upon photodetachment. We find $\Delta r_e \leq 3 \times 10^{-4}$ Å formally, although the theoretical model probably loses validity before this level is reached. This work is in the final stages of preparation for publication.

During the next report period we hope to survey a large number of interesting atomic negative ion photodetachment spectra, using the new "Coulatron" universal positive ion source and a charge transfer system which should soon be operational.

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[7] L. M. Branscomb, Phys. Rev. 166, 11 (1966).

III. ENERGY LOSS AND RADIATION PRODUCTION BY ELECTRON COLLISIONS

Program Leader - Dr. E. C. Beaty

Interaction of Electrons with Ions (Dr. G. H. Dunn and Dr. F. L. Walls)

The measurement of the energy dependence of electron-molecular ion recombination cross sections is being carried out with an ion trap technique illustrated in Fig. 4. During this report period a new trap fabricated from molybdenum to high tolerances was installed and tested. Figure 5 shows an ion peak of mass 18 (NH_4^+), and demonstrates achievable mass resolution and signal to noise with the new trap. Indications are that ion lifetimes and signal-to-noise ratios are now adequate to pursue recombination measurements as planned. Since September 1, 1972 this work has been supported by another agency.

TRAPPING OF IONS

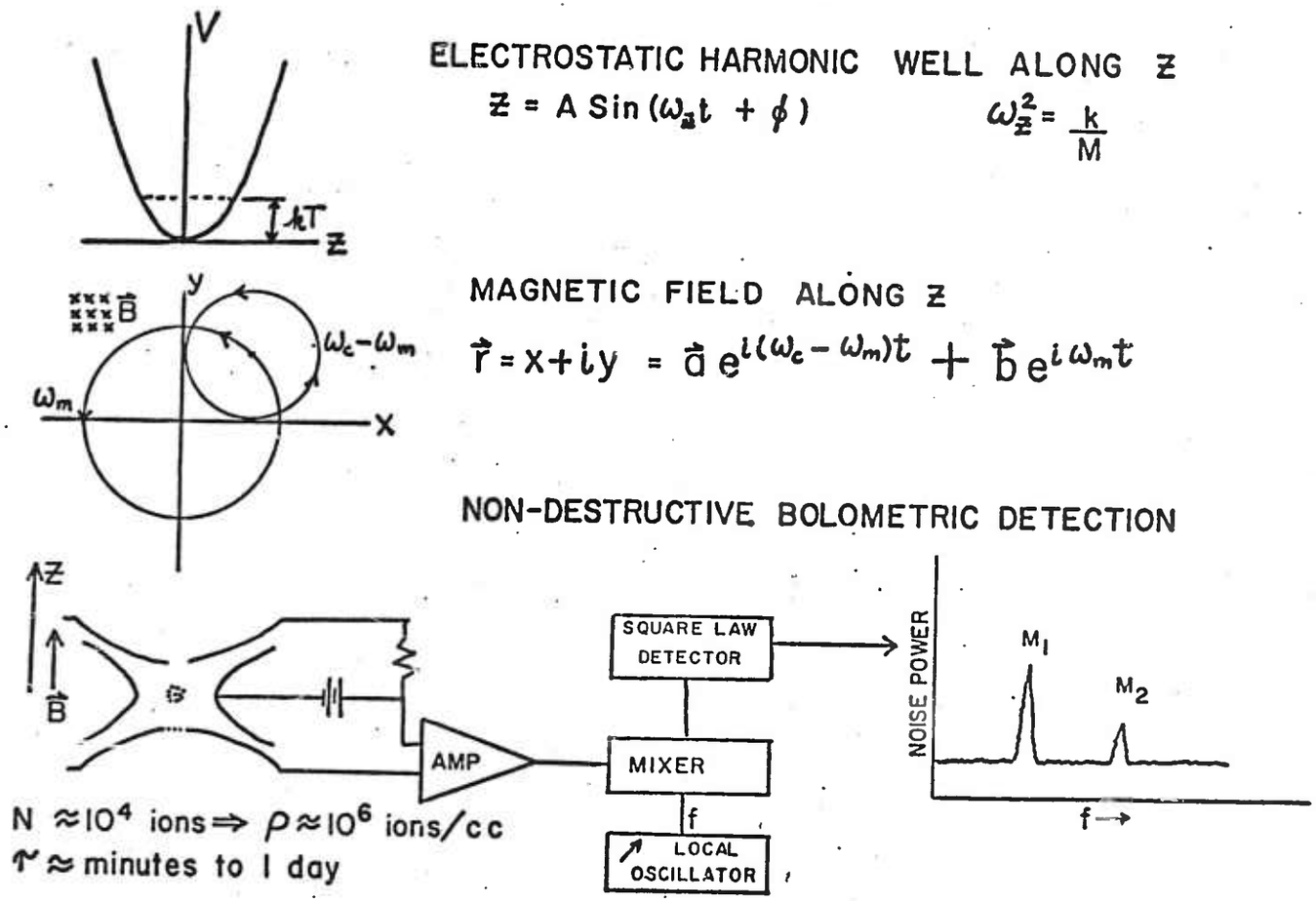

Fig. 4. Top of figure demonstrates trapping and harmonic motion of ions in the Z direction. Middle section illustrates B field trapping in the r-θ plane and shows motion at frequency near the cyclotron frequency ($\omega_c - \omega_m$) and at the precession frequency ω_m. Lower portion of figure schematically shows the trap and electronics. The noise power spectrum illustrated on the right shows the resonances occurring at $\omega_z/2\pi$ for ions of two different masses.

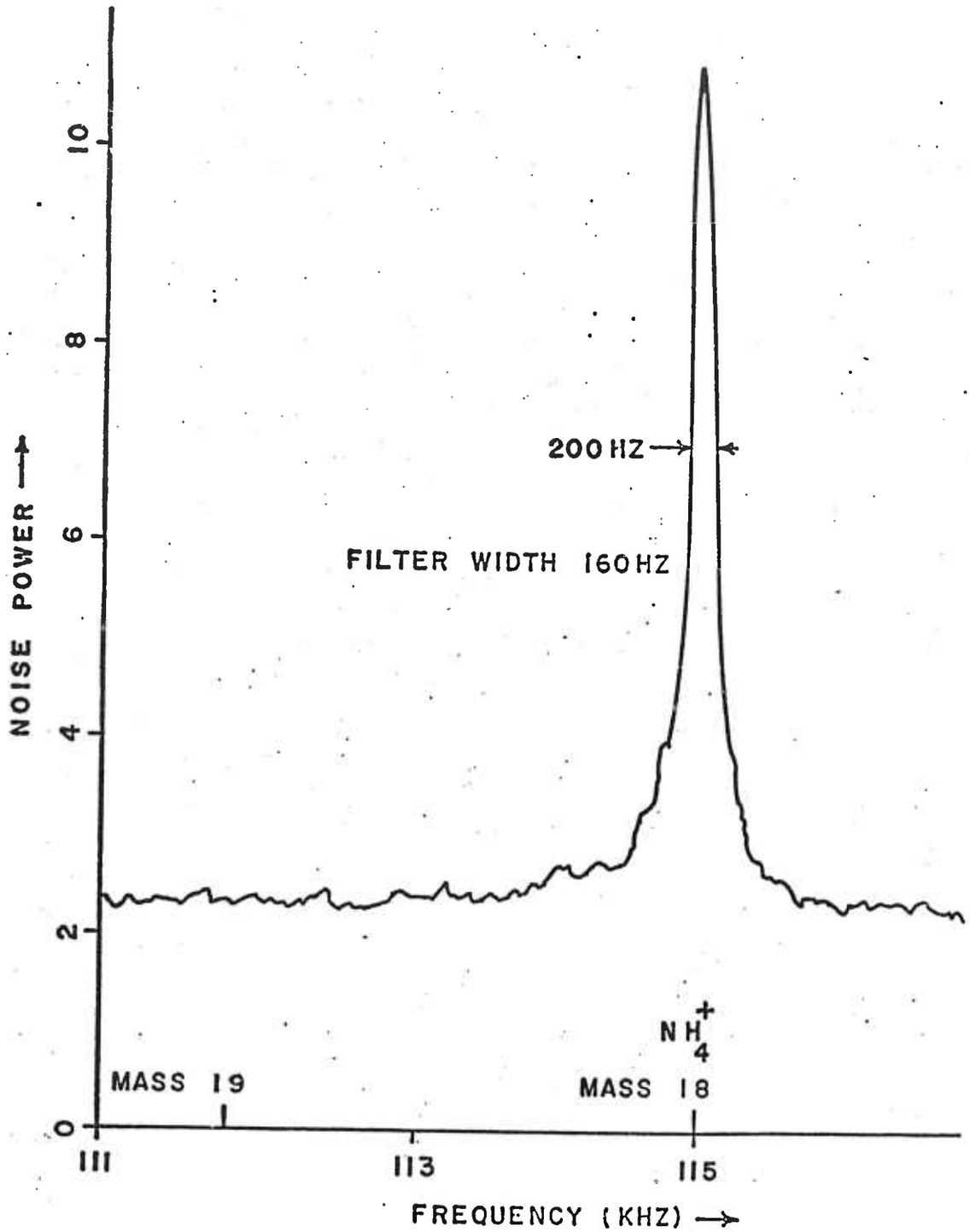

Fig. 5. Noise power spectrum showing trapped ion resonance at $f = \omega_z/2\pi$ for mass number 18 (NH_4^+). Distinction from H_2O^+ has been made by cyclotron resonance.

Theoretical Atomic Physics (Dr. D. G. Hummer and Dr. D. W. Norcross)

Electron excitation cross sections for atmospheric ions. In this period, a considerable amount of effort was expended in improving the atomic structure code to allow more accurate atomic models to be obtained. The wave functions for C III, N IV and O V are significantly better than those obtained in our early calculations and seem to be comparable to those obtained by other very sophisticated procedures. The calculated oscillator strengths agree quite well with the results of beam-foil spectroscopy (generally to within 30%).

The Distorted Wave Code is also being modified to provide better accuracy, greater convenience in use and substantially increased efficiency, in preparation for the larger calculations that we are planning for the immediate future. We expect to begin production within the next two months.

Atomic and Radiation Interaction Theory (Dr. Sydney Geltman)

Free-free absorption coefficients. We have completed the work on the calculation of atomic free-free absorption coefficients and it has been accepted for publication.[1] The coefficients have been calculated for the electron-neutral atom systems involving He, C, N, O, Ne, Ar, Kr, and Xe. Some angular distributions are presented and thermal averages have been evaluated in the ranges of λ = .5 to 20 μm and T = 500 to 20000°K. The computer codes remain available for application to any neutral atom of interest.

Angular distributions for ionization. The "Coulomb-projected Born" approximation is being applied to the high energy electron impact ionization of the hydrogen and helium atoms. This method has proved useful in describing the large-angle differential cross sections for proton-hydrogen charge transfer[2] and electron impact excitation of hydrogen and helium,[3] where the conventional Born approximation breaks down completely. The mathematical problems of evaluating the ionization amplitude are much more severe than in the case of excitation, but progress is being made.

Interaction of atoms with intense radiation fields. The usual form of perturbation theory which is applied to the calculation of absorption coefficients breaks down at radiation intensities of the order of 10^{14} W/cm^2, where the interaction energy between electron and field is comparable to the binding energy between electron and nucleus. Since laser intensities of order 10^{19} W/cm^2 are now routinely available, it is urgent that the absorption mechanism of such intense radiation be thoroughly understood. We are inves-

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[1] S. Geltman, J. Quant. Spectrosc. Radiat. Transfer (in press).

[2] S. Geltman, J. Phys. B 4, 1288 (1971).

[3] S. Geltman and M. B. Hidalgo, J. Phys. B 4, 1299 (1971); 5, 617 (1972).

tigating alternate theoretical procedures in which the electron-nuclear interaction is regarded as the perturbation, while the electron-field interaction is taken to be part of the unperturbed problem. The method is being applied to the N-photon absorption in bound-bound, bound-free and free-free hydrogenic transitions, and the results will be compared with other theoretical approaches.[4]

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[4] H. Reiss, Phys. Rev. A 1, 803 (1970); G. J. Pert, J. Phys. A 5, 1221 (1972).

IV. GENERATION AND ABSORPTION OF RADIATION

Program leader - Dr. A. C. Gallagher

Alkali-Inert Gas Line Broadening and Intermolecular Potentials (Dr. A. C. Gallagher and Dr. C. G. Carrington)

We have measured the pressure dependence of the Rb-Xe molecular-continuum radiation to determine the rate of formation of bound Rb^*-Xe molecules:

$$Rb^* + 2Xe \rightarrow RbXe^* + Xe.$$

This step following electron excitation of the atom is an important part of the association laser dynamics. Combined with the equivalent measurements of the formation rates for Na^* and Li^* that are planned, this should allow us to understand how this process depends on well depth, vibrational spacing, and other parameters of possible association laser molecules.

Quantum mechanical calculations of Frank-Condon factors are being carried out to determine the validity of the classical model we have proposed for calculating the optical properties (e.g., gain) of association laser molecules. Preliminary results for the bound-free and bound-bound transitions are shown in Fig. 6. In the spectral region of primary interest for association lasers this indicates about 10% accuracy of the classical approximation. In some other regions such as the far red edge of the spectrum there are major differences.

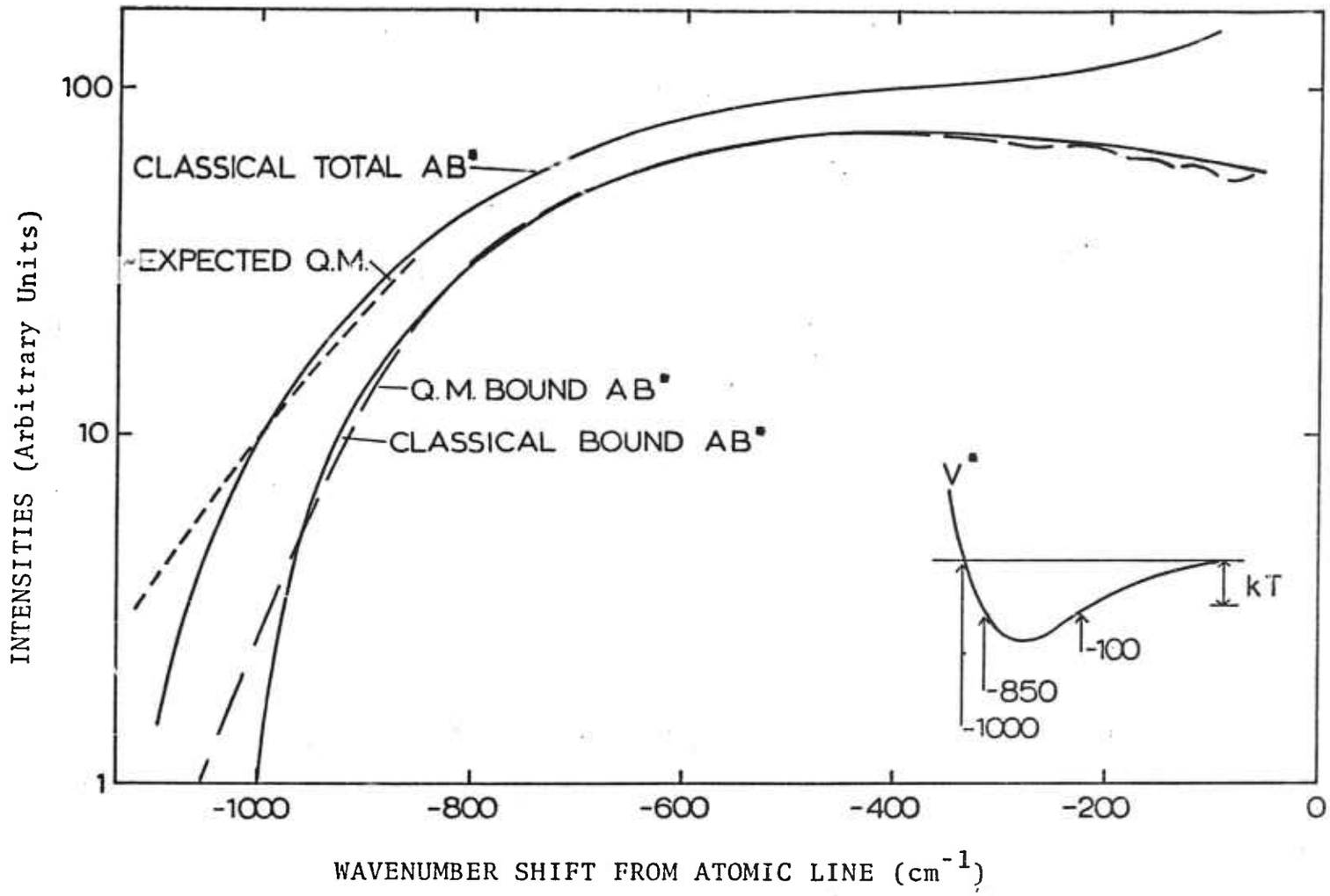

Fig. 6. Classical vs quantum-mechanical emission spectrum for radiatively dissociating molecules. The potentials are modeled after the Cs-Ar system; the temperature is half the well depth. The bound AB* curves represent the portion of the total radiation due to bound excited molecules. The relation between the spectrum and the AB* potential is shown in the insert.

Excitation of Metal Resonance Lines (Dr. A. C. Gallagher and Mr. D. Leep)

We have made preliminary measurements of the excitation cross section of the Li resonance line. The Li-rare gas molecules are expected to have the best interaction potentials of the various alkalis for use as an association laser molecule. Such a laser would utilize electron excitation of the lithium resonance line, with efficiency dependent on this cross section.

Manuscripts Involving ARPA Funds

Listed below are papers submitted for publication during the period covered by this report.

S. Geltman, "Free-free radiation in electron-neutral atom collisions," to appear in J. Quant. Spectrosc. Radiat. Transfer.

H. Hotop, R. A. Bennett and W. C. Lineberger, "Laser photodetachment studies of Cu, Ag, Au, and Pt negative ions," to appear in Bull. Am. Phys. Soc. [Abstract]

H. Hotop, R. A. Bennett and W. C. Lineberger, "The electron affinities of Cu and Ag," to appear in J. Chem. Phys.

H. Hotop and W. C. Lineberger, "Dye-laser photodetachment studies of Au^-, Pt^-, PtN^-, and Ag^-," to appear in J. Chem. Phys.

H. Hotop, T. A. Patterson, and W. C. Lineberger, "Dye-laser photodetachment of OH^- and OD^-," to appear in Bull. Am. Phys. Soc. [Abstract]

E. F. Jaeger, J. T. Mariska, L. Oster, and A. V. Phelps, "Numerical study of the diffusion controlled positive column," to appear in Bull. Am. Phys. Soc. [Abstract]

L. Oster and A. V. Phelps, "Analytical study of the diffusion controlled positive column," to appear in Bull. Am. Phys. Soc. [Abstract]

T. A. Patterson, H. Hotop and W. C. Lineberger, "Photodetachment of Na^-," Bull. Am. Phys. Soc. 17, 1128 (1972). [Abstract]

A. V. Phelps, "Tunable gas lasers utilizing ground state dissociation," JILA Report No. 110, September 15, 1972.

R. J. VanBrunt and L. J. Kieffer, "Electron energy dependence of the energy and angular distributions of O^- from dissociative ion pair formation in O_2," Bull. Am. Phys. Soc. 17, 1139 (1972). [Abstract]