RESEARCH IN LASER PROCESSES

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In order to provide data necessary for predicting the operating characteristics of proposed alkali atom-rare gas excimer lasers, measurements have been made of the fluorescence spectral intensity from the $NaXe_2$ molecule. At Xe pressures of 20 atm, the $NaXe_2$ fluorescence exceeds the NaXe fluorescence for wavelengths greater than 720 nm. New techniques have been developed for the calculation of the growth of instabilities in laser type discharges and for the analyses of measurement of rate coefficients for the electron excitation of molecules. The collisional quenching of Na atoms in (continued)

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the 3^2P states has been observed and appears to be the result of excitation transfer to Na_2 molecules.

SEMIANNUAL REPORT

This Semiannual Report contains descriptions of work carried out under ONR Contract No. N0014-76-C-0123 and ARPA Order No. 2683, Amd. 2, and covers the period from 1 August 1975 to 31 January 1976. Section I is the Semiannual Report Summary while Sections II-V are more detailed descriptions of work carried out under the four projects supported by this contract.

I. SEMIANNUAL REPORT SUMMARY

The four projects being carried out in the area of Laser Processes under this contract are summarized below. More detailed discussions are given in Sects. II through V of this report.

1) Generation and Interpretation of Molecular Continuum Radiation

Many metal-noble gas molecules are viable candidates for high-efficiency, high-power excimer lasers. Since the spectra and potentials of most of these molecules are unknown, the merits of different metal-noble gas combinations can only be assessed from detailed measurements of the kind reported here. The alkali-noble gas molecules, which we have studied in detail under this and previous contracts, are candidates for excimer lasers in the 0.7-1 μm wavelength region. Since the proposed laser systems operate best at several atmospheres of buffer gas, we are measuring the increases in the stimulated emission coefficients which occur when the alkali atom-rare gas excimers are perturbed by a second rare gas atom. Thus, for the first time measurements have been made of the optical properties of a triatomic excimer molecule formed from a metal atom and rare gas atoms, i.e., $NaXe_2$. These molecules contribute a large part of the observed fluorescence and predicted stimulated emission in the red wing of the sodium resonance line when the Xe pressure is 20 atm. In particular, for wavelengths greater than 720 nm at pressures of 20 atm. the stimulated emission spectrum is dominated by the $NaXe_2$ spectrum. Using this data and our previous fluorescence and potential energy data, we will be able to test models of triatomic excimer molecules so as to make possible predictions of the stimulated emission coefficients for other metal vapor-rare gas laser systems.

2) Stability of Discharges in Weakly Ionized Gases

The technical problem being investigated is the optimization of the use of electrical energy for the production of excited molecules in electrically excited gas lasers. In particular, the efficient use of electrical excitation in gas lasers requires that the discharge remain diffuse as the density of the excited molecules is raised as high as possible, i.e., that the discharge not form a constricted channel or arc. During the period of this report we have obtained solutions for the electron and positive ion densities and for the electric field in a geometry which allows the propagation of constricted regions of the discharge in the direction of the electric field. The ability to predict the conditions for the rapid propagation of such instabilities is crucial to the use of models for the scaling of lasers to the high powers and large dimensions of ARPA interest.

3) Electron Excitation of Molecular Metastables

The electron excitation of molecular radiators in an electrical discharge can be an efficient means of producing high power densities in large lasers without excessive losses caused by amplified spontaneous emission. We have developed and are applying a new technique for the measurement of the efficiency of electron excitation and quenching rate coefficients of those molecules which radiate slowly and yet are important donors in proposed high power laser systems, e.g., the $O_2(a^1\Delta_g)$ - $I(^2P_{1/2})$ system or systems using the $N_2(A^3\Sigma_u^+)$ state. Measurements have been made of the rate coefficients for the electron excitation and collisional quenching of the $O_2(b^1\Sigma_g^+)$ state. These indicate that previous excitation cross section data are significantly in error.

4) Scattering and Transport of Resonance Radiation in Gases

The technical problem considered here is the measurement of the transport and scattering of resonance radiation emitted by metal vapors so as to obtain the rates of radiative and non-radiative energy loss by excited atoms in excimer lasers, etc. During the period of this report the first measurements have been made of the time dependent decay of the resonance radiation emitted by alkali metal atoms (Na) at the high vapor densities characteristic of proposed high power excimer laser systems. These data show a previously suspected but unmeasured collisional quenching of the sodium atoms in the 3^2P state. This quenching appears to be caused by Na_2 molecules and may result in excitation of these molecules to states of interest for high power laser applications.[1]

II. GENERATION AND INTERPRETATION OF MOLECULAR CONTINUUM RADIATION (Drs. R. Scheps (to 10/75), H. Rothwell (from 10/75), and A. Gallagher).

Proposed excimer laser systems utilizing metal vapor-rare gas mixtures are normally modeled and operated at rare-gas pressures of 10-100 atm. At these pressures the gain and absorption coefficients of the medium are primarily determined by the diatomic excimers (AX) where X is the noble gas and A may be another rare gas atom or any atomic specie. However, the triatomic excimer AX_2 can also make a major contribution to the gain and absorption coefficients at these high X pressures, and in some spectral regions it may be the dominant contribution. While it is apparent from simple arguments that this must occur, the actual magnitude and shape of the AX_2 gain coefficient does not follow from the AX gain coefficient as it depends on three-body potential surfaces.

We report here the first measurements of the optical properties of such a triatomic excimer molecule. Further, we have already measured

the spectra and interaction potentials of the associated diatomic specie, so this data will also test the accuracy of various approximation schemes for predicting the AX_2 spectra from knowledge of the AX spectra. As examples, the simpliest model for the AX_2 potential surface is based on adding the two long-range A-X pair potentials. The next level of sophistication is to use the three pair-potentials A-X, A-X, and X-X. Still more correct is a rediagonalization of the Hamiltonian corresponding to the long-range A-X interaction, etc. The various pair-potentials are known so that we can now test these various approximations against the data. While the results will have important implications for all excimer laser systems, the particular Na + Xe case studied gives a good example of the importance of the AX_2 contributions. This is outlined in the following paragraph.

The measured NaXe and $NaXe_2$ flourescence spectra are given in Fig. 1. These are reported in units which can be directly converted to gain and absorption coefficients using standard formulas.[1,2] Note that the 20 Atm composite spectrum is dominated by the $NaXe_2$ contribution at $\lambda \geq 720$ nm. Since the stimulated-emission/absorption ratio increases rapidly with increasing λ, net gain is very easily achieved on this long-wavelength $NaXe_2$ band. Since the A-X band of Na_2 has a large gain coefficient at 800 nm, the $NaXe_2$ gain at this wavelength will be valuable in the Xe buffered Na_2 laser we have previously modeled.[1] In addition to contributing to the net gain at 800 nm, the $NaXe_2^* \rightarrow NaXe_2$ transition can rapidly ($\sim 10^{-10}$ sec) convert the entire Na^* stored energy into a brief laser pulse. Without this effect, only a small fraction of the stored, energy is available at one time from the $Na_2^* \rightarrow Na_2$ transition.

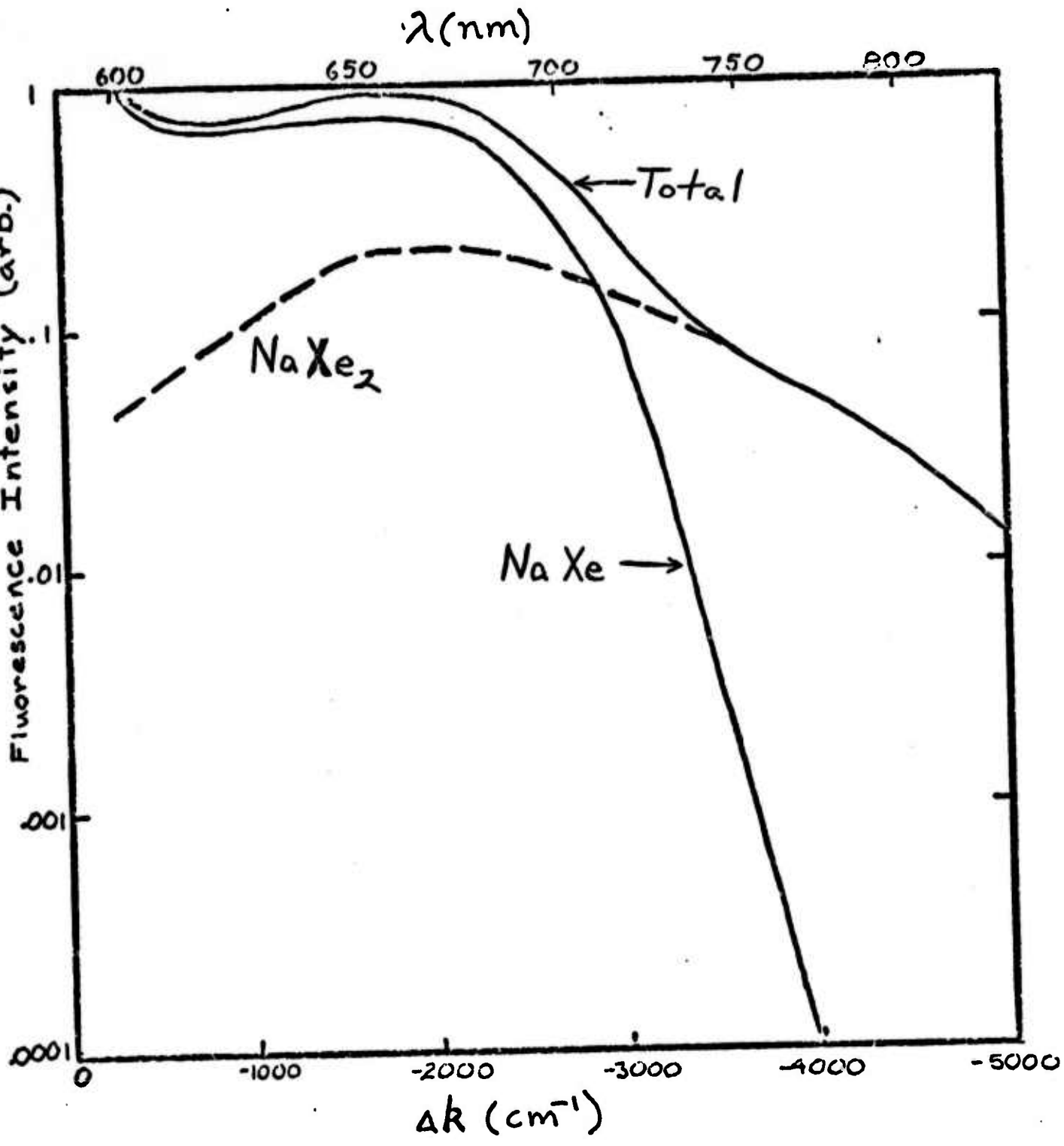

Figure 1

Fluorescence intensity of the NaXe and $NaXe_2$ A-state excimer molecules for a Xe pressure of 20 atm. The emission intensity per excited $Na^*(3p)$ and per cm^{-1} is shown. These two contributions to the total intensity were separated on the basis of their linear and quadratic dependence on Xe pressure.

We expect an increased red-shift of the AX_2 band relative to the AX band to occur frequently. Consequently it can have an equally profound effect on the gain and power available in many laser systems. The present emphasis is thus on comparing the data to models in order to improve our predictive capability for other mixtures.

III. STABILITY OF DISCHARGES IN WEAKLY IONIZED GASES (Drs. H.-C. Chen and A. V. Phelps).

The objective of this theoretical investigation of the growth of instabilities in weakly ionized gas discharge is to make more quantitative techniques for the prediction of conditions for formation of the arcs which limit the power output from high power laser systems. The availability of such techniques would greatly and the optimization of high power lasers currently under development and would greatly improve the reliability of the design of sealed-up versions of these lasers.

During this report period we have obtained solutions for the radial and axial variations in electron and positive ion density and in electric field under conditions appropriate to the development of an electrical discharge in a high pressure gas, such as might be used with a metal vapor-rare gas laser. A unique feature of our model is the inclusion of the effects of electron density gradients on the electron energy distribution so as to avoid what we believe are false increases in ionization caused by large radial space charge fields.[3] As is often the case in numerical solutions of coupled non-linear equations, we are continuing to work on improved methods of avoiding the numerical instabilities which limit the length of time which can be modeled.

Our earlier work on the growth of instabilities in cylindrical geometry has been accepted for publication in the Physics of Fluids.[4]

IV. ELECTRON EXCITATION OF MOLECULAR METASTABLES (Drs. S. A. Lawton and A. V. Phelps).

The objective of this project is the measurement of excitation and destruction rate coefficients for metastable states of molecules which serve as upper states of lasers or which make possible efficient excitation of such laser states. Our current measurements are for the $b^1\Sigma_g^+$ state of O_2. Some other molecular systems of potential use for high power visible lasers include: the transfer of energy from the $a^1\Delta_g$ metastable state of O_2 to the $^2P_{1/2}$ state of I; the transfer of energy from the $A^3\Sigma_u^+$ metastable state of N_2 to other molecules, such as NO and halide compounds; and the indirect excitation of molecules, such as KrF, following electron excitation of atomic metastables, such as those of Ar or Kr.

Because of large (50 to 300%) discrepancies between predicted and measure excitation rate coefficients for the $O_2(b^1\Sigma_g^+)$ state, we have been inproving the measurement technique by a) the development of a fast rise time amplifier and recording system capable of separating the electron and negative ion components of the current through the drift tube in the presence of an attaching gas and b) the installation of a radiance standard for the calibration of the detection system for the few photons emitted by the metastables. The measurement of the electron and negative ion components of the charge crossing the drift tube not only makes possible an essential correction to the excitation rate coefficients measurements but also provides electron attachment coefficient data for the gas under investigation. This facility is particularly important for attaching gases other than O_2, where there are large uncertainties in electron attachment coefficients.

In addition to the improvement of the experiment, one of us (AVP), has developed an improved technique for analyzing the excitation rate coefficient data in terms of electron excitation cross sections. The technique has been applied to our previously measured electron excitation coefficient data for the 4.3 μm band of CO_2 so as to obtain the energy dependent cross section for excitation of the 001 and 011 levels while adjusting the cross sections for excitation of the 010 and 100 levels to fit the small amount of data available for these two levels. Previous attempts to fit the electron scattering data and the electron transport data had led to a significant underestimate of the 4.3 μm band (001 and 011 levels) excitation. The technique used is to plot measured rate coefficients vs measured characteristic electron energies and to adjust cross sections so that calculated values agree with experiment. This approach makes the 001 and 011 level cross section adjustment much easier than when the rate coefficient is plotted against the electric field to gas density ratio E/N because it reduces the dependence of the error on the cross sections for the 010 and 100 levels. The resultant set of vibrational excitation cross sections[5] is shown in Fig. 2. Unofrtunately, there is still considerable uncertainty in the relative cross sections for electron excitation of the 010 and 100 levels.

V. SCATTERING AND TRANSPORT OF RESONANCE RADIATION (Drs. T. Fujimoto and A. V. Phelps).

The objective of this project is to determine the rates of radiative and non-radiative energy loss from resonance states of a typical metal vapor atom (Na). Such data is essential to the accurate modeling of proposed high power metal vapor-rare gas excimer lasers operating at near visible wavelengths.

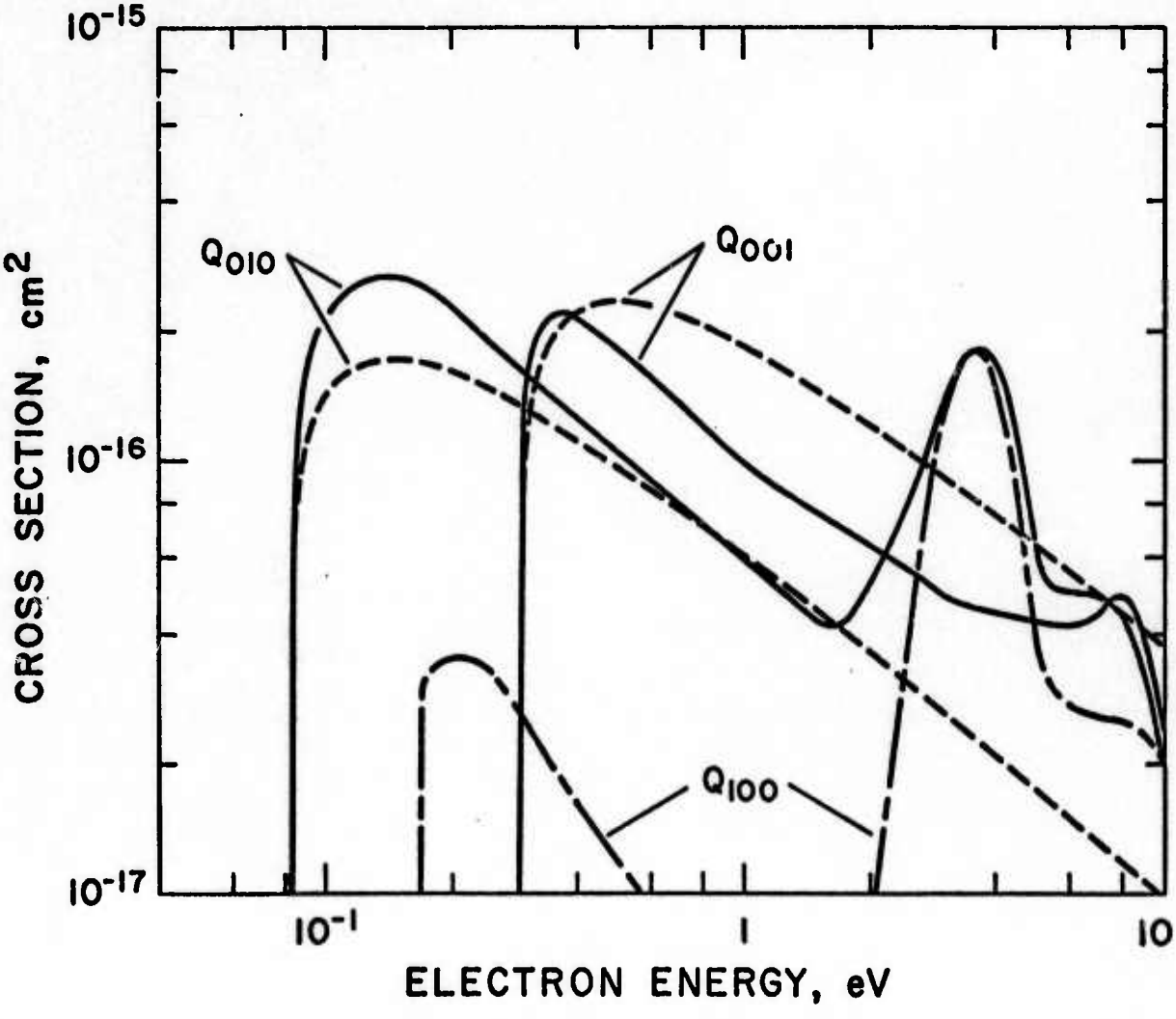

Figure 2

Cross sections for excitation of lowest vibrational levels of bending (010), symmetric stretch (100) and asymmetric stretch (001) modes of CO_2. The solid curves are derived from analysis of this experiment as discussed in the text. The short dashed curves are calculated from the transition probabilities of Penner and Olfe and the Born approximation formulas of Takayanagi. Note that the curves of this figure may be changed prior to submission for publication.

During the early part of this reporting period comparisons were made of theoretically calculated and neasured spectral intensity of scattered white light in the vicinity of the 589.0 and 589.6 nm resonance lines of Na. These comparisons showed good agreement at Na densities below about 5×10^{14} atoms/cm^3, but showed that the experimental values of the scattered intensity were much too low at higher Na densities. As indicated by the curves of Fig. 3 with $Q > 0$, the lower experimental intensities are roughly those expected if there were collisional quenching of the excited Na by Na_2 molecules.

Since the scattering experiments are less sensitive to collisional quenching than time dependent "afterglow" experiments, a collaborative experiment was set up with other interested groups at JILA and NBS-Boulder in which a N_2 laser pumped dye laser was used to excite Na vapor. Measurements were made of the decay of intensity at wavelengths near the resonance lines of Na. Preliminary results show the expected radiative loss of excited atoms at Na densities below about 10^{15} atom/cm^3 and an apparent collisional loss at high Na densities. Although the measured constants of the exponential decay vary with temperature in approximately the manner expected for collisional transfer of energy from excited Na to Na_2, the density dependence is slower than expected. This anomoly will have to be resolved before the destruction mechanism can be definitely assigned to the collisions between excited atoms and ground state molecules.

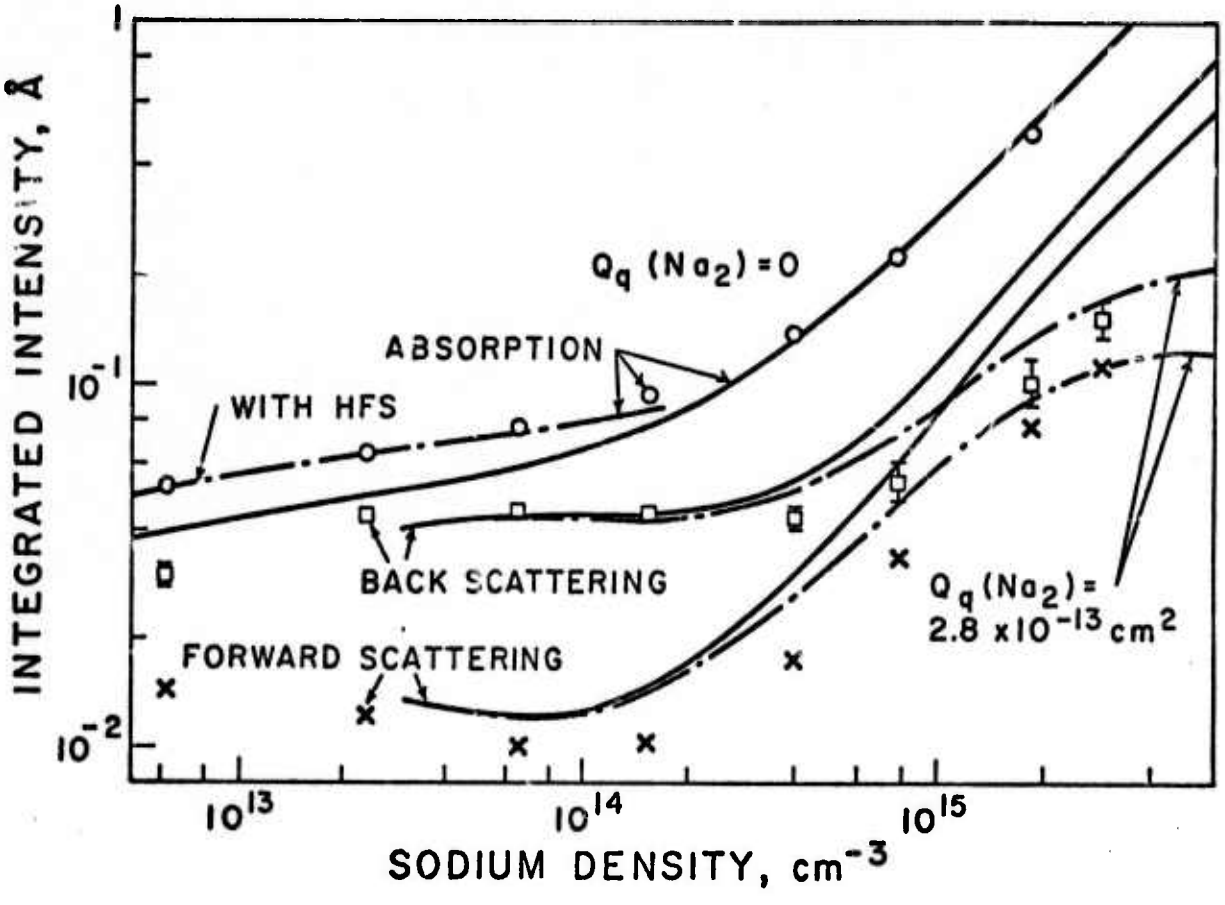

Figure 3

Integrated absorption and integrated back and forward scattering of white light in Na near D_2 line. The cell thickness is 0.113 cm. The points are experimental data and the lines are calculations using the theory of Hummmer and Kunasz.[6]

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