IV. ATOMIC RESONANCE AND SCATTERING

Academic and Research Staff

Prof. Daniel Kleppner   Prof. Stuart S. B. Crampton   Dr. Theodore W. Ducas
Prof. David E. Pritchard   Dr. Richard R. Freeman

Graduate Students

Riad N. Ahmad   William E. Cooke   William D. Phillips
Jerome Apt III   Michael G. Littman   Naser S. Saleh
Gary M. Carter   Myron L. Zimmerman

RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH   JSEP

Our interests center on the structure and interactions of atoms and simple molecules, and on their interactions with radiation at optical and radio frequencies. Experimental methods involve colliding beam scattering, atomic and molecular beam resonance spectroscopy, and optical excitation by tunable lasers.

1. Optical Frequency Standard

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

David E. Pritchard

We are trying to develop a simple and economical "frequency meter" to be used with a tunable laser to preset the laser frequency, measure the laser frequency, or provide frequency marker pulses when the laser frequency is swept.

A prototype system capable of 50 MHz accuracy is nearing completion and should soon be ready for calibration.

2. New Methods for Radiation Detection

Joint Services Electronics Program (Contract DAAB07-74-C-0630)

Daniel Kleppner

The possibility of creating a gas of highly excited atoms in essentially unperturbed states offers new opportunities for efficient detection of optical, infrared, and microwave photons. We have undertaken a theoretical study of some properties of highly excited atoms relevant to this application. An atomic beam apparatus has been constructed and work has started on the preparation of atoms in high-lying states using stepwise excitation with pulsed lasers. We have performed spectroscopy and measured ionization characteristics of these high-lying states.   JSEP

3. Studies of Superradiance and Coherence

U. S. Air Force Office of Scientific Research (Contract F44620-72-C-0057)

Daniel Kleppner

The objectives of this work are to study superradiance in small systems and to investigate the nature of atomic systems in a radiation field.

We have extended the analysis of two-particle superradiance at small distances as manifested by the interaction between single-electron atoms. In small systems the energy shift which accompanies superradiance can play an important dynamical role, and may be used to bind two free atoms into a superradiant complex. Apparatus has been developed for creating these complexes by laser excitation in a high-density atomic beam, and for observing the fluorescent spectra and lifetimes.

We have demonstrated the feasibility of exciting a parity-forbidden dipole optical transition by applying a static electric field. The method has been used to excite the 6D and 7D states in C_S. Lifetimes and Stark mixing coefficients have been measured.

Work is in progress on quantum-beat spectroscopy on the 6D state in hydrogen.

4. Van der Waals Molecules

Daniel Kleppner, David E. Pritchard

The objective of this work is to study magnetic interactions between alkali and rare gas atoms by applying molecular beam resonance methods to alkali-rare gas van der Waals molecules. We have measured the spin-rotation constant and the hyperfine interaction in potassium-argon. The hyperfine interaction in potassium is shifted by the argon atom; the resulting hyperfine spectrum has resolved structure which yields a detailed picture of how the hyperfine shift varies with internuclear separation. Both positive and negative shifts are evident, reflecting two separate interaction mechanisms. The results provide a much more detailed check of theory than is provided by optical pumping pressure shift measurements.

Work is in progress on measuring the shift in the electronic g-factor between a free alkali and an alkali bound with a rare-gas atom.

A. ENERGY-TRANSFER STUDIES USING VELOCITY-SELECTED ATOMS AND MOLECULES

Jerome Apt III, David E. Pritchard

Collisional transfer of energy from one molecule or atom to another and/or among the sublevels of one molecule is important in many electronic devices, such as plasma devices, gas discharge lasers, and chemical lasers. The dependence of the collision cross sections for energy transfer on the relative velocity of the collision partners is, after the magnitude of the cross section, the single most important feature of the energy-transfer collision process, first because it governs the temperature dependence of the energy-transfer process, and second because it provides a sensitive indicator of the detailed nature of the molecular processes occurring in the collision. In spite of this, there have not been extensive experimental studies of the energy dependence of energy-transfer processes because the means of doing so experimentally are complicated (e. g., shock tubes) or do not work over a very wide range of collision velocities (e. g., they change the temperature of the apparatus).

We have recently proposed[1] a general method for achieving velocity selection in the study of energy-transfer collision cross sections involving atoms and molecules excited by a monochromatic laser. This method is based on the Doppler shift and promises to permit us to study collisions in which the velocities correspond to thermal energies from $300°$ K to roughly $2,000°$ K without changing the temperature of our apparatus. The method produces a velocity-selected excited-state population in the following manner. If the frequency v of a highly monochromatic laser beam is slightly offset from the resonance frequency v_0 of an atomic or molecular transition, then (to first order) only those atoms or molecules whose component of velocity along the beam is

$$V_{1z} = c(v-v_0)/v_0$$

will be Doppler shifted into resonance and hence excited.

During the last few months we have applied this method to the study of electronic excitation transfer between Na and Rb

$$Na^* + Rb \rightarrow Na + Rb^*,$$
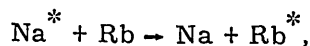
where the star refers to electronic excitation. We used a single-mode tunable dye laser (Spectra Physics Model No. 580 System) to excite the Na to either the $j = 3/2$ or $j = 1/2$ fine-structure level of its 3p state. Fluorescence from the $5p_{1/2}$ and $5p_{3/2}$ states of rubidium was collected by an f:1 collection lens and detected by a cooled RCA 31034 photomultiplier tube. The rubidium fluorescence radiation and the sodium fluorescence

radiation were distinguished by three-cavity interference filters and colored glass filters.

Velocity selection was achieved by offsetting the laser frequency from the center of the desired fine-structure transition line by an amount $\Delta\nu$, which yielded a specific relative velocity V_{rel}. The cross section for excitation transfer to a particular fine-structure level of Rb can then be determined from the ratio of the measured light intensity from the appropriate Rb fluorescence line, F_{Rb}, to that from the sodium, F_{Na}, by using the relation (valid in the absence of radiation trapping)

$$Q^*(V_{rel}) = F_{Rb}(\Delta\nu)/(n_{Rb}V_{rel}\tau_{Na}F_{Na}(\Delta\nu)),$$

where n_{Rb} is the density of rubidium, and τ_{Na} is the excited-state sodium lifetime. We have studied the dependence of Q^* upon V_{rel}, and have verified that Q^* is roughly independent of n_{Rb}, but have not yet attempted to measure the absolute magnitude of Q^*.

Our experimental chamber was a heated quartz cell connected to separately heated sidearms containing Na and Rb metal, as shown in Fig. IV-1. The pressure in the cell was maintained below 1 μm by a zeolite trapped oil diffusion pump. At this pressure, the time between collisions is roughly 10^3 times the decay time of the excited-state sodium. The biggest experimental problem proved to be scattered light from the cell walls, which limited our maximum energy to roughly 6 kT, in spite of the use of a phase detector in synchronism with frequency modulation of the laser on and off the desired
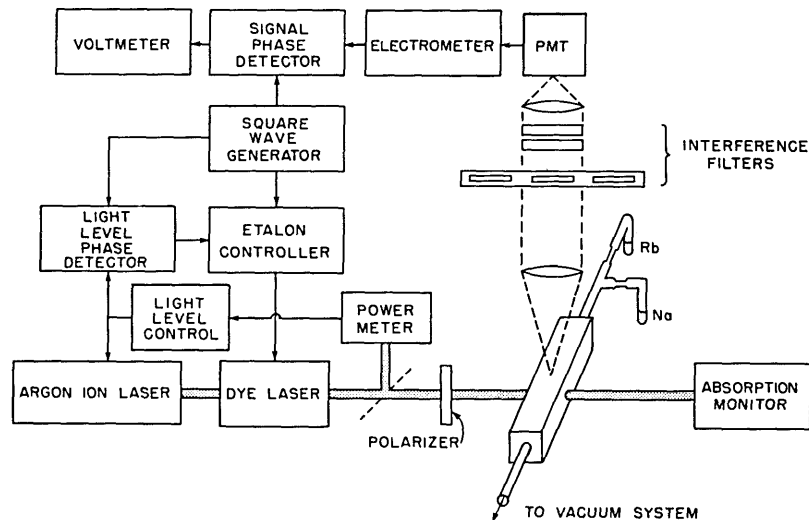

Fig. IV-1. Apparatus. Output from the light-level phase detector is applied to the etalon controller to equalize the laser intensity at the two frequencies between which it is chopped.
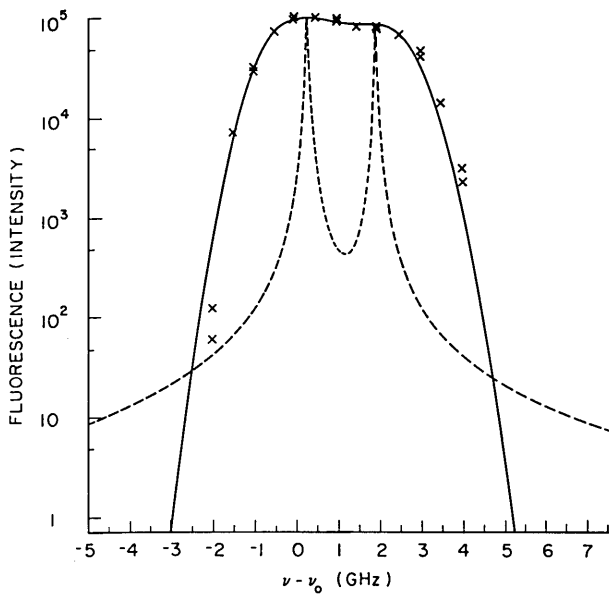
Fig. IV-2.

Comparison of observed sodium fluorescence signal (x) with Gaussian curve (solid line) and Lorentz curve (dashed line). The Lorentz curve is drawn under the assumption that the atoms have zero velocity along the laser beam.
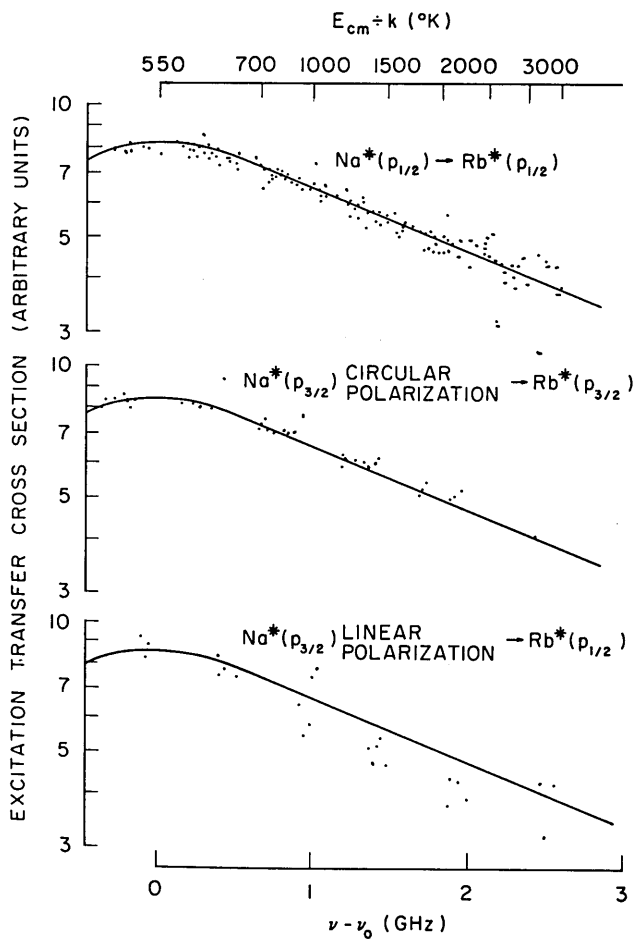

Fig. IV-3.

Dependence of excitation transfer cross sections on the center of mass energy of the collision, E_{cm}. The solid lines show a cross section which varies as $E_{cm}^{-1/2}$.

portion of the line.

Figure IV-2 shows the observed sodium fluorescence as a function of the laser offset from the line center (the double hump is caused by the presence of the two sodium ground-state hyperfine components). The solid curve is the fluorescence spectrum expected from Doppler broadening alone at the temperature of the cell (485° K) and the agreement of the data with this expectation is evidence that our velocity-selection method is working. The dashed curve shows the natural (Lorentzian) line shape of sodium atoms, with Doppler broadening ignored. This shows the ultimate limit of our technique. If we attempt to select atoms with energies higher than approximately 10 kT we shall be in the region where the Lorentzian line shape dominates: we then excite more slow-moving atoms (through their Lorentz tail) than velocity-selected atoms because so few have the required velocity.

Figure IV-3 shows results for Na^* ($3p_{1/2}$ and $3p_{3/2}$) + Rb $\rightarrow$ Na + Rb^* ($5p_{3/2}$). In each case we find that $Q(v)$ varies as $1/v$ within experimental error. The $p_{3/2}$ data were taken with both linear and circularly polarized light in order to study the influence of level crossings which can increase the observed Na fluorescence slightly. Our data in which the Rb^* is in the $5p_{1/2}$ state display similar velocity dependence. This is consistent with the behavior of cross sections for quenching of metastable Hg^* (3p) by collisions with molecular targets.[2]

Work is now in progress to extend these measurements to higher velocities by both reducing background and increasing the cell temperature. We are also preparing to study the system Na^*+Ba, in which kinetic energy must be added to make the transfer; here we hope to study the velocity dependence in the threshold region.

References

1. W. D. Phillips and D. E. Pritchard, Phys. Rev. Letters 33, 1254 (1974).
2. H. F. Krause, S. Datz, and S. G. Johnson, J. Chem. Phys. 58, 367 (1973).