

3. Atomic Resonance and Scattering

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3.1 Atomic Physics Program

Daniel Kleppner

Introduction

The Atomic Physics Group is concerned with problems of atomic structure and the interaction of atoms with static and dynamic fields. The experiments generally employ highly excited Rydberg atoms, and utilize techniques of laser excitation with detection by ionization in an applied electric field. (A recent article by members of the group in *Scientific American*¹ provides a general introduction to Rydberg atoms.) Interest in the past focused on the structure of Rydberg atoms in strong electric fields; more recently it has turned to the problem of structure in a strong magnetic field. In a somewhat different effort, a series of studies of the interaction of Rydberg atoms with blackbody radiation was carried out during the past year. These have led to a new avenue of research in quantum electronics: the interaction of atoms with single modes of the radiation field. An experimental demonstration of the suppression of blackbody absorption by cavity cutoff effects was successfully executed. This phenomenon, which had never previously been observed (nor, to our knowledge, contemplated), provides evidence that experiments on the most fundamental aspects of the interaction of an atom with the vacuum radiation field are now feasible.

Other work during the year includes a study of electron correlation effects in an alkaline earth Rydberg atom by microwave spectroscopy: in the development of techniques for transferring a population of Rydberg atoms to high angular momentum states and progress in the development of a low-temperature source of photoexcited hydrogen atoms.

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3.2 Atoms in Strong Magnetic Fields

Magnetic fields normally do little more than perturb atoms. However, the ratio of the magnetic interaction to the Coulomb interaction scales as n^6 , where n is the principal quantum number. Values of n of 50 or more are achievable, and in such cases the magnetic interaction can be dominant. Atoms under these conditions bear little similarity to normal atoms. Many of their properties are still a mystery.

Our discovery of an approximate symmetry for hydrogen in a strong magnetic field² has helped to stimulate new theoretical interest in the problem: more than ten papers were published on the subject in 1981. Our next experimental goal is to carry out high-resolution spectroscopy. Use of a cw laser should increase the resolution by a factor of over 100, and allow us to study details of avoided level crossings and linewidths which may provide strong clues to the underlying regularities that characterize this simple but elusive system.

3.3 Structural Studies - Nonadiabatic Effects in Calcium

A highly excited electron can be perturbed by the ionic core of a Rydberg atom. The major effect of the core is to produce a polarization potential which varies as $1/r^4$. However, the polarization potential can be affected by the energy of the Rydberg electron. Physically, the core may not be able to distort fast enough to follow the Rydberg electron; such behavior is described as nonadiabatic.

Our early efforts to identify nonadiabatic behavior in alkali metal atoms were not fruitful,³ but Ganesh Vaidyanathan recognized that the effects should be much larger in alkaline earth atoms. Together with Dr. Philip Shorer of the Smithsonian Astrophysical Observatory he developed methods to calculate nonadiabatic effects in calcium.⁴ The effects are so large that their neglect could lead to serious errors in the interpretation of calcium spectra. Mr. Vaidyanathan has carried out a series of studies of Rydberg states of calcium by microwave spectroscopy which confirms the accuracy of his theory.⁵

3.4 Rydberg Atoms and Radiation

Rydberg atoms interact strongly with radiation fields due to their enormous size. The interaction scales as n^4 compared to normal atoms. As a result, interactions which are often unobservable can have dramatic effects. In particular, blackbody radiation at room temperature can cause appreciable redistribution of the atoms into nearby energy levels, and even photoionize the atoms.

We have carried out a series of studies on Rydberg atoms and radiation intended to confirm our theoretical understanding, as well as our experimental ability to observe the radiative effects with clarity and precision. These include studies of spontaneous lifetimes in sodium,⁶ radiative transfer by blackbody radiation,⁷ and photoionization by blackbody radiation.⁸ These studies are all relevant to

the use of Rydberg atoms for detecting infrared radiation.

3.5 Inhibited Spontaneous Emission

Our work on Rydberg atoms and blackbody radiation has opened the way to a novel and potentially important new line of research: the quantum electrodynamics of atoms in a cavity.

Spontaneous emission by a free atom is usually regarded as a basic property of matter, but the spontaneous emission rate can be altered by a cavity, either enhanced or inhibited. The possibility of experimentally studying such effects is compelling because of the basic nature of the process; spontaneous emission is the fundamental irreversible process in nature - it is the fundamental source of noise.

The new experimental possibilities have been described in a Phys. Rev. Letter.⁹ We have carried out a related preliminary experiment, the "turning off" of blackbody absorption by radiative cutoff between two parallel planes.¹⁰ This first experimental success appears to open the way to a large number of studies on atoms and radiation, including novel studies of coherence. One has the opportunity to study a single atom strongly coupled to cavity by the vacuum field. The system can be regarded as a pair of coupled oscillators, one atomic, one man-made.

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3.6 Rotationally Inelastic Collisions

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Introduction

The most frequent type of inelastic collision between molecules is the rotationally inelastic (RI) collision. Before detailed understanding of more violent atom-molecule collision processes (e.g., vibrationally inelastic or reactive) may be obtained, it is necessary to understand RI collisions first. Large RI collision cross sections indicate that these collisions must occur predominantly at relatively large atom-molecule separations where there is reason to expect a similarity of atom-molecule potentials for different atom-molecule systems - hence there is some *a priori* reason to hope for some universality in these collisions. These considerations motivated us to begin a detailed experimental study of such collisions roughly five years ago.

During the past few years we have developed a general picture of rotationally inelastic (RI) collisions between atoms and molecules involving both theoretical and experimental advances. This picture appears to be able to account for a substantial majority of the existing experimental results for total RI cross sections, and for many of the theoretical calculations as well. There are two basic components of this picture: one is a simple classical calculation showing that the cross sections for transfer from the j^{th} rotational quantum level to the 0^{th} quantum level, $\sigma_{j \rightarrow 0}$, decrease as a power of j ; the other is the energy-corrected sudden (ECS) approximation of DePristo and Rabitz which expresses the cross section between arbitrary initial and final quantum levels in terms of the subject $\sigma_{j \rightarrow 0}$. These ideas were initially developed to fit the large arrays of rate-constant matrices (a rate constant is a thermal average of the cross section); their outstanding success at this has made them useful as a means of quantitatively accounting for RI processes in a variety of physical situations (gas lasers, freely expanding gases, and the atmosphere of Venus are three examples). We derived simple analytic expressions for these fitting laws³ and have shown that under many circumstances they are equivalent to two simpler energy-based fitting laws which we discovered empirically several years earlier.^{1,2} Recently we have shown that our picture of RI collisions also explains the measured

velocity dependence of RI cross sections - this demonstration of predictive power beyond the area of its origination (thermally averaged data) is striking evidence for the underlying validity of our general picture of RI collisions.

3.7 Two Basic Ideas

1. Impulsive classical approximation yields Power Law Dependence.⁷ The key result is that

$$\sigma_{j \rightarrow 0}(v) = Av^{-\alpha}[j(j+1)]^{-1-\alpha}, \quad (1)$$

where v is the relative velocity. If the anisotropy of the atom-molecule potential decreases as $c_n R^{-n}$ at large atom-molecule separation, R , then A may be predicted and $\alpha = 1/(n-1)$. Since $n = 4$ to 6 for typical atom-molecular systems, the overall exponent of the j -dependence is predicted to be around 1.25 , in accord with observations;⁴ A can be predicted with $\sim 20\%$ error for the heavier rare-gas targets.⁷

2. The energy-corrected sudden approximation. This mathematical expression gives $\sigma_{j_i \rightarrow j_f}(v)$ in terms of $\sigma_{j \rightarrow 0}(v)$.

$$\sigma_{j_i \rightarrow j_f}(v) = (2j_f + 1) \sum_j (2j + 1) \begin{pmatrix} j_i & j_f & j \\ 0 & 0 & 0 \end{pmatrix}^2 \left[1 + (\ell j_{>}/v)^2 \right]^{-2} \sigma_{j \rightarrow 0}(v), \quad (2)$$

where $\begin{pmatrix} j_i & j_f & j \\ 0 & 0 & 0 \end{pmatrix}$ is a standard 3- j angular momentum coupling coefficient, $j_{>}$ is the greater of j_i and j_f , and ℓ is a characteristic length parameter which is found from a fit to data (calculated or measured).

The energy-corrected sudden approximation was developed by DePristo and Rabitz. It was partly motivated by our data - a fact reflected by its application to our $\text{Na}_2^* \text{-Xe}$ data by Augustin et al., about a year before in their publication of their general theory.

3.8 Fitting Laws

The combination of the two basic ideas mentioned above leads to a fitting law - an expression which may be used to fit a large mass of $\sigma_{j_i \rightarrow j_f}$ data by using three free parameters (A , α , and ℓ). (It is possible to predict A and α if the potential is known, but not ℓ). We have demonstrated this fitting law's ability to fit data (typically ~ 80 thermally averaged rate constants with $\sim 7\%$ average error) within $\sim 10\%$ over an enormous range of j_i and j_f in collisions of Na_2^* with 8 targets⁴ and I_2^* with Xe.⁶ Example of fits to Na_2^* colliding with He and Xe. The similarity of the slope of the data for $j_i = 4$ in the two systems reflects the near equality of α for the two systems, the data for $j_i = 100$ are relatively lower in Xe than He, a reflection of the fact that the lower average velocity of the Xe collides accentuates the reduction caused by the bracketed expression in the ECS scaling law (Eq.2).

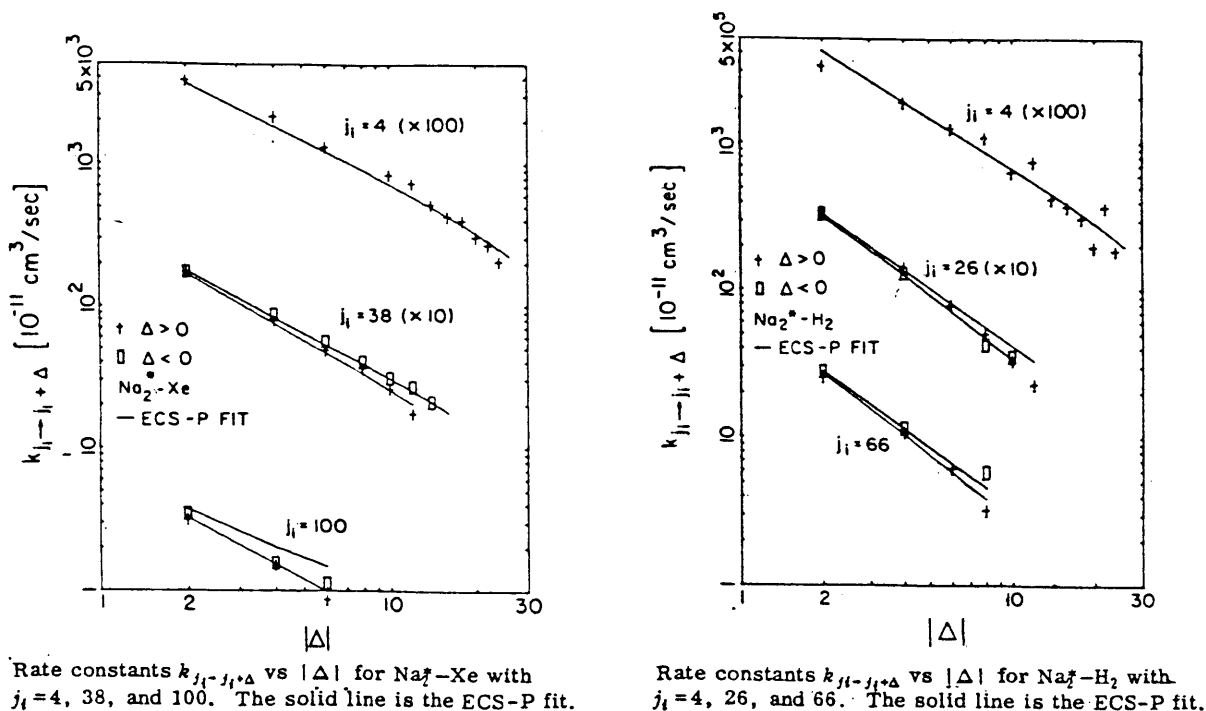


Figure 3-1.

3.9 Velocity Dependence

The velocity dependence of RI collision cross sections $\sigma_{j_i \rightarrow j_f}(v)$ has recently been determined in the system $\text{Li}_2^* - \text{Xe}^8$ for a variety of j_i, j_f pairs. (The experimental technique makes use of the Doppler shift to excite a velocity-selected group of molecules; it permits the velocity dependence to be determined from measurements made in a gas cell.⁵) Measurements in which $j_f = j_i - 2$, are shown below (error bars) for $j_i = 8, 22, \text{ and } 42$. There is a surprisingly strong decrease of the cross section at high ratios of j/v , as well as a maximum in the cross section $\sigma_{8 \rightarrow 6}$ at intermediate velocities. Both of these features are reproduced by the solid curves which are the predictions of Eqs. 1 and 2 (A and t were adjusted for best fit; we took $n = 6$).

3.10 Doppler Probe of Energy Transfer in Adiabatically Expanding Gas Scientific Objective

The objective of this project is to develop and apply new techniques for studying energy-transfer collisions at low relative velocities based on use of Doppler velocity-selection techniques applied to adiabatically expanding gas mixtures.

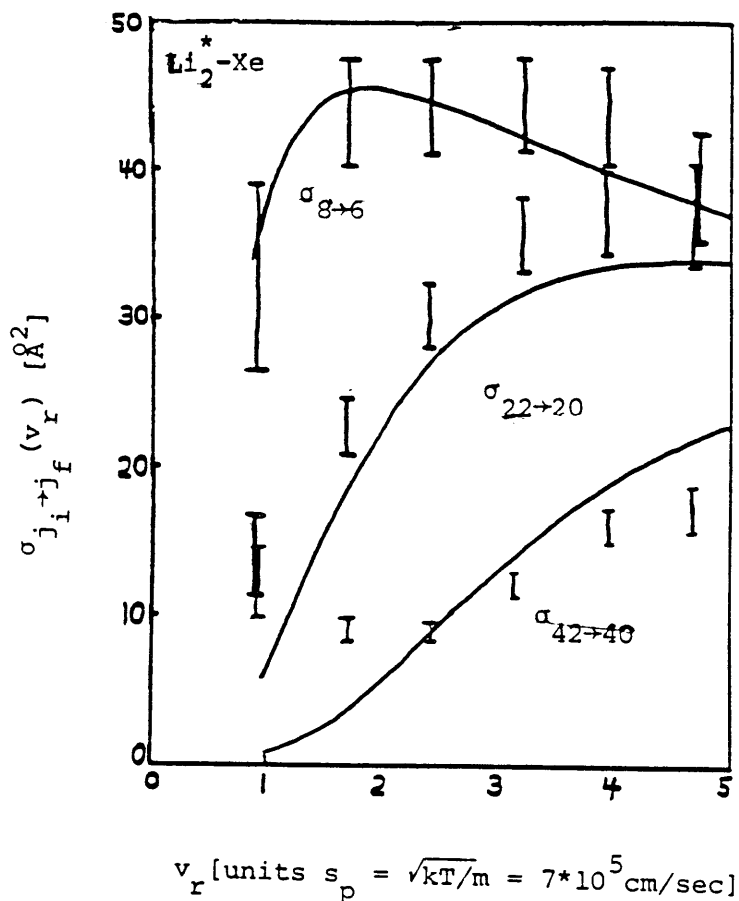


Figure 3-2.

Introduction

We propose to study energy-transfer processes in molecule-atom systems at very low temperatures and in situations of excess internal energy (i.e., when vibrational and rotational temperatures are "hotter" than the kinetic environment). A new technique will be developed for this purpose which combines the recently developed velocity selection by Doppler shift (VSDS) technique with existing technology for producing high-pressure seeded molecular beams. The essential idea is that the seeded molecular beam - basically an adiabatically expanding mixture of the molecule-atom system which is to be studied - will be probed at the point in its expansion where it has the desired density and kinetic temperature. By utilizing VSDS, the desired energy-transfer processes can be studied over a range of relative velocities characteristic of this temperature. This technique also permits study of the details of the expanding molecular beam, but such studies are incidental to the work proposed here.

3.11 Scientific Background

a. Seeded Molecular Beams

Over the past decade several research groups (SAF77, LBM79), including our own (ALP77), have developed high-pressure (~100 atmosphere) molecular-beam sources of rare gases seeded with molecular impurities. These sources generally have on-axis intensities in excess of 10^{20} atoms per steradian per second, and are therefore the first two component molecular-beam sources to live up to the original proposal of Kantrowitz and Grey that such sources would produce beams of adiabatically expanding (and cooling) gas sufficiently dense to prevent incursion by the hotter background gas. This region is termed a "zone of silence," and temperatures well below 1°K have been observed in them (HAB77, LMB79).

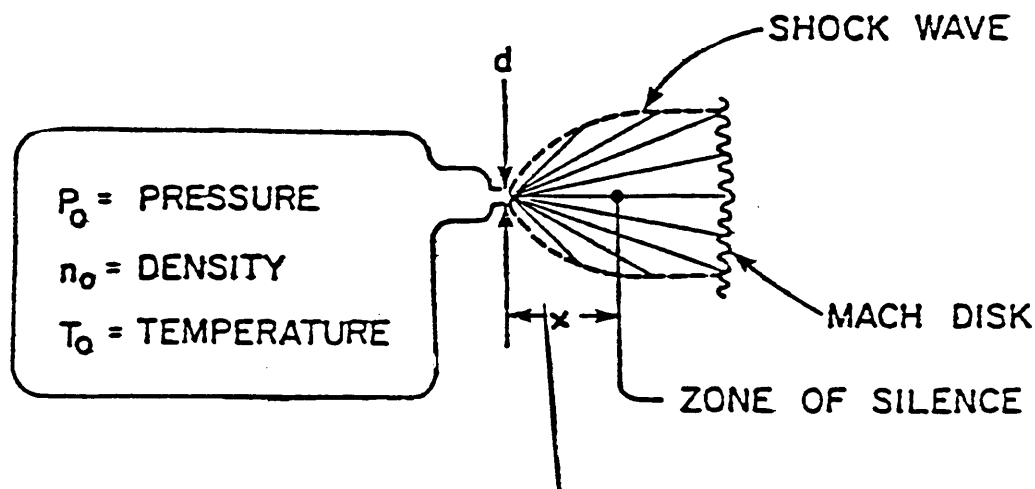


Figure 3-3.

Thermodynamic conditions in such a molecular beam are easy to calculate because the flow is a simple adiabatic radial expansion away from the nozzle. If the source pictured above has on-line intensity I_0 , then the density at a point downstream is (for a carrier gas of mass m_c)

$$n = I_0 / (\bar{v}^2),$$

where $v = \sqrt{5k_B T_0 / m_c}$ is the average speed of the beam (k_B is Boltzmann's constant, and T_0 is the nozzle temperature). The temperature follows from the laws of adiabatic expansion:

$$T(D) = T_o \left(\frac{6x^2}{d^2} \right)^{-2/3}$$

within $\sim +30\%$, depending on the exact configuration of the flow near the nozzle (of diameter d). The local temperature determines the relative velocity of atoms and molecules in the usual way, so that for cross section Q the collision rate will be

$$\Gamma = n v_{rel} Q$$

$$\approx 10^6 d^{2/3} \text{ (c.g.s.)},$$

where the numbers are for molecular-beam systems like those in our laboratory (with $Q = 100 \text{ \AA}^2$).

The preceding discussion of molecular beams holds only for a limited distance downstream. At best, the adiabatic expansion will terminate at the Mach disk, located at

$$x_M \sim d(P_o/P_{\text{background}})^{1/2},$$

because the momentum of the beam is spread over too large an area to resist the pressure of the background gas. For carrier atoms other than He, appreciable numbers of dimers form if kT decreases to less than $\sim 1/8$ of the dimer binding energy, and the heat of dimerization prevents further cooling of the expansion. This effect limits T to $\sim 2^\circ\text{K}$ for Ne and to $\sim 25^\circ\text{K}$ for Xe. Finally, for seeded beams the internal temperatures of the seeded molecules may decouple from the translational temperature of the expansion [termed "freeze out" (HAB77)] due to a decrease at low relative velocity of cross sections for the energy-transfer processes necessary to maintain equilibrium.

For the excited-state collisions considered here, the two most important features of the flow are the collision rate and temperature; the collision rate must be $\sim 5\%$ of the radiative rate (typically 10^6 - 10^8 sec^{-1}) for there to be sufficient energy transfer processes to measure without serious problems due to multiple collisions. The temperature determines the accessible velocity range. Our "laboratory-standard" seeded molecular beam (ALP77) has a throughput of $\sim 3 \times 10^{20}$ atoms/steradian per second, and can operate with any nozzle size above 30μ (where a pressure of ~ 2000 psi is required to achieve the desired throughput). The following figure summarizes the temperature and collision rate range accessible in such a machine. Note that $(T/T_o) * 10^3$ is close to the temperature in degrees (T_o is typically ~ 800 degrees). The figure shows the relationship of T and Γ at various downstream locations, for beams from nozzles of two different sizes. The limits to the useful range of downstream observation distances, $d < x < x_M$, are indicated.

Optically excited species have decay rates in the range 10^7 - 10^8 s^{-1} . For optimal study of their collisional properties, the collision rate must be roughly an order of magnitude smaller. It is evident

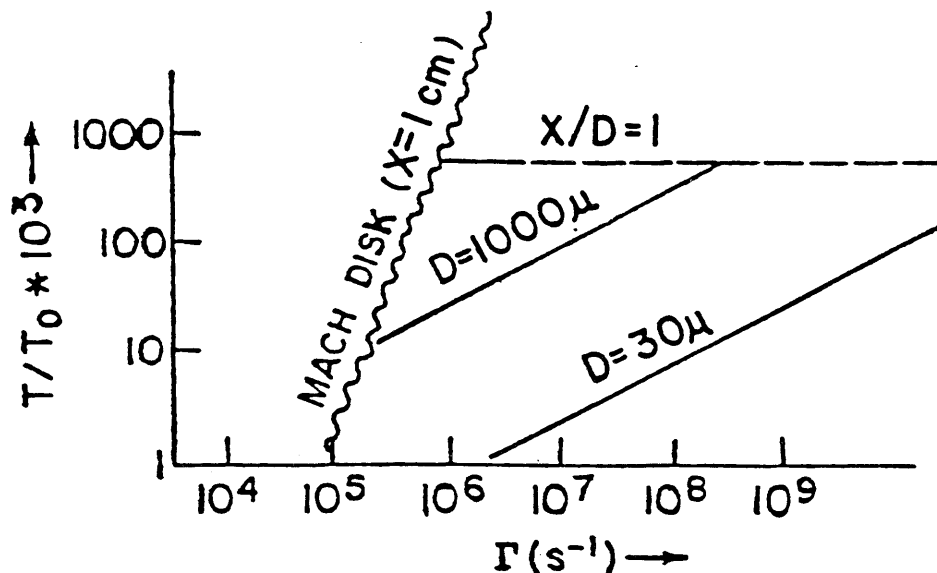


Figure 3-4.

from the figure that our new technique will permit probing the collisional properties of such species over a range of temperatures extending from room temperature down to a few degrees K.

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