

### III. ATOMIC RESONANCE AND SCATTERING\*

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#### RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

We are concerned with the properties of electrons and nuclei as manifested by their interactions in simple atoms, and with the interactions among atoms, molecules, and the radiation field. Our experiments generally utilize atomic-beam methods, including both radiofrequency and collision spectroscopy.

##### 1. Spin Exchange Scattering

We have observed the spin-dependence of the differential cross section for various alkali-molecule systems. Both elastic and inelastic processes have been found. In a related project, we are attempting to create a molecular beam of Van der Waals molecules with a view to studying their magnetic resonance spectrum.

##### 2. Hydrogen Maser Studies

Instrumentation has been completed and data taking has been initiated in an experiment to determine the effect of atomic binding on the magnetic moment of the electron. The measurements are made in a hydrogen maser that has been double-moded to operate simultaneously on hydrogen and deuterium.

##### 3. Interactions in the Excited State

As a natural development of our scattering and maser experiments, we have initiated work on the interactions of atoms in excited states. Many new and interesting experiments appear now to be feasible, because of recent developments in atomic beams and lasers. We describe in the present report plans for one such experiment: measurement of the energy dependence of an alkali fine-structure transfer cross section.

D. Kleppner

#### A. ALKALI-MOLECULE SCATTERING – PRELIMINARY RESULTS

We have completed preliminary measurements of the spin-dependent differential cross sections for scattering of potassium by the molecules  $N_2$ , NO, and  $O_2$ . As shown in

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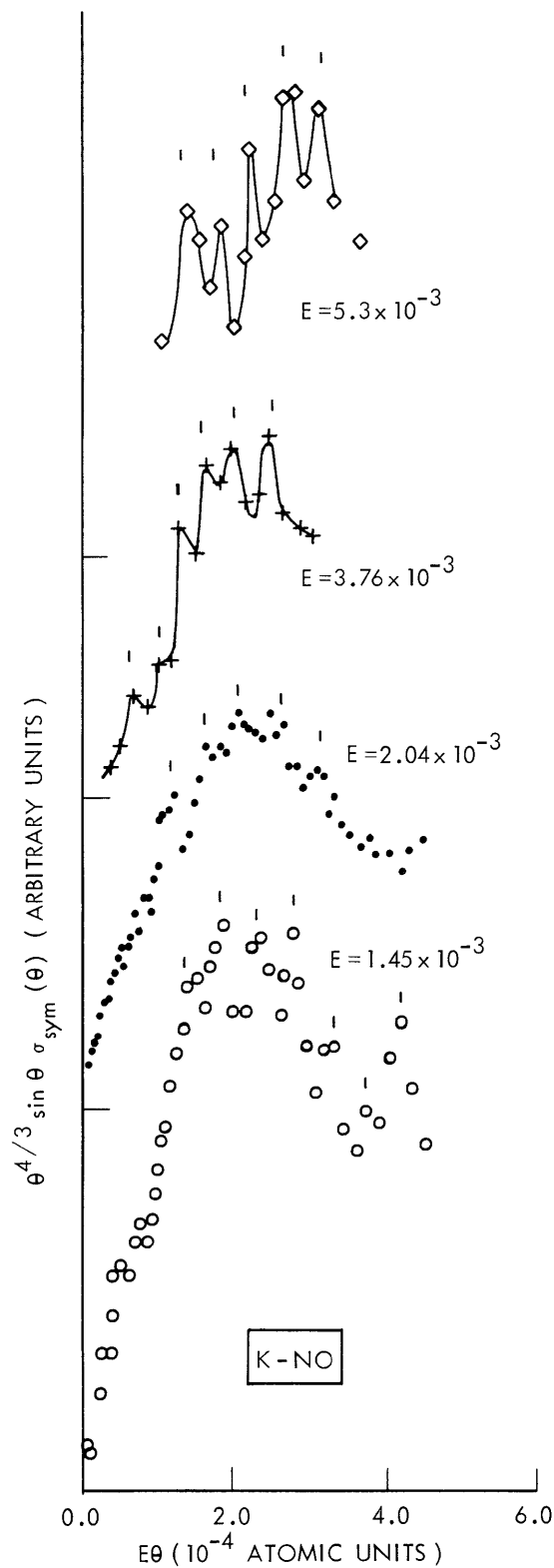


Fig. III-1.

$\sigma_{\text{sum}}$  vs  $E\theta$ : results for K-NO. Vertical bars over the cross sections indicate our assignment of the maxima of the oscillations. The zero of the cross sections taken at the three highest energies are indicated by horizontal lines.  $\sigma_{\text{sum}}$  has been multiplied by  $\theta^{4/3} \sin \theta$  to suppress unwanted angular dependence.

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Fig. III-1, there is striking structure in the sum differential cross section (the sum is over final spin state) at small angles. The rapid oscillations occur in all three systems that have been studied. By studying these systems at several energies and comparing the change of the period of the oscillation with energy, we have obtained insight into their origin.

If the oscillation results from the interference between two classically allowed paths (i. e., if classical collisions with impact parameter  $b_1$  or  $b_2$  both cause the same deflection), then the period of oscillation is

$$\Delta\theta = \frac{\lambda}{d}, \tag{1}$$

where  $d$  is the distance between the interfering impact parameters, and  $\lambda$  is the de Broglie wavelength

$$\lambda = h/p = h/mv. \tag{2}$$

Since  $d$  generally depends only weakly on the energy and angle, we would expect that

$$d = \frac{\lambda}{\Delta\theta} = \frac{h}{m} (v\Delta\theta)^{-1} \tag{3}$$

should not change with energy. The right-hand side of Eq. 3 is tabulated in Table III-1, and it is clear that it is constant within error for the systems K-O<sub>2</sub> and K-N<sub>2</sub>. This

Table III-1. Small-angle interface structure.

System	Energy (a. u. $\times 10^{-3}$ )	n	$\Delta\theta$ (mrad)	d [a <sub>0</sub> ]	$\Delta(E\theta)$ (a. u. $\times 10^{-6}$ )
K-N <sub>2</sub>	1.43	4	29.5	22.8 $\pm$ .8	43
	2.91	3	20.5	23.0 $\pm$ 1.2	60
K-O <sub>2</sub>	1.99	2	28.5	19.3 $\pm$ 1.0	57
	3.07	4	22.5	19.9 $\pm$ 0.6	69
	4.49	3	18.5	19.0 $\pm$ 0.8	83
K-NO	1.45	5	31.7	21	46 $\pm$ 1
	2.04	4	23.5	24	48 $\pm$ 1.3
	2.73	2	17.3	28	47 $\pm$ 2
	3.76	5	12.8	32	48 $\pm$ 1
	6.43	4	7.3	43	47 $\pm$ 1.3

Note. n is the number of complete cycles averaged to find  $\Delta\theta$ .

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means that the oscillatory structure in the K-N<sub>2</sub> and K-O<sub>2</sub> systems is most probably due to interference processes arising in purely elastic scattering. (It is tempting to say that the K-N<sub>2</sub> interaction has a larger range than the K-O<sub>2</sub> interaction. This conclusion is not warranted, however, because the K-N<sub>2</sub> structure was observed in the angular range  $10^{-4} < E\theta < 3 \times 10^{-4}$  (atomic units), whereas the K-O<sub>2</sub> structure was observed over the range  $2.5 \times 10^{-4} < E\theta < 5 \times 10^{-4}$ .)

It is apparent that the interference structure in the data for K-NO does not yield a consistent result for d. This strongly suggests that the oscillatory structure for this interaction does not result from an elastic interference process, but rather from some inelastic or quasi-elastic process such as level crossing. (The K-NO molecule is complicated, and energy levels with different quantum numbers may become degenerate, or degenerate save for the effect of some very small coupling, at certain internuclear separations.) Because level crossings occur at well-defined internuclear separations, they should lead to features that occur at constant values of  $E\theta$ , provided that the impact approximation to the collision is valid. A series of evenly spaced level crossings could lead to the periodic behavior observed in the data. Evidence in support of this thesis is that the values of  $\Delta(E\theta)$  stay remarkably constant as the energy is changed (see Table III-1).

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### B. EFFECT OF BINDING ON THE MAGNETIC MOMENT OF THE ELECTRON

We have undertaken a precise measurement of the effect of atomic binding on the magnetic moment of the electron. Recent calculations<sup>1, 2</sup> yield the following result for the g-factor of an electron bound in the 1S state of hydrogen:

$$g_{1S} = g_S \left( 1 - \frac{1}{3} \alpha^2 + \frac{1}{4\pi} \alpha^3 + \frac{1}{2} \alpha^2 \frac{m}{M} \right).$$

In order to verify the mass-dependent term, we are comparing the electron g-factor in hydrogen and deuterium. To order  $\alpha^2 m/M$  the ratio is predicted to be

$$\frac{g_{1S}^{(H)}}{g_{1S}^{(D)}} = 1 + \frac{1}{4} \alpha^2 \frac{m}{M} = 1 + 7.25 \times 10^{-9}.$$

Two recent measurements<sup>3, 4</sup> of this quantity have achieved an accuracy of 1 part in  $10^9$ , but disagree by several times their estimated error. Our goal is to achieve an accuracy of 1 part in  $10^{10}$ .

#### 1. Method

The experiment consists in observing with a hydrogen maser the two transitions indicated in Fig. III-2. The frequencies are given by the Breit-Rabi formula which,

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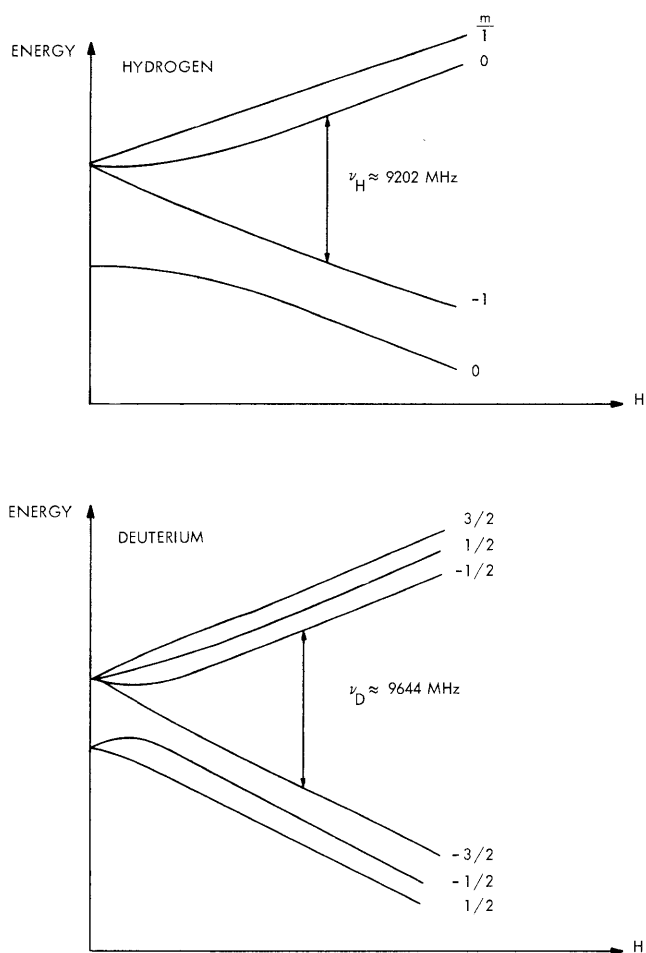


Fig. III-2. Energy levels and observed transitions for hydrogen and deuterium in the  $^2S_{1/2}$  ground state.

for purposes of numerical inversion, is conveniently written in the following fashion:

$$\nu_H = \left\{ \frac{(1-1/R_H)}{2} \left[ (1-1/x_H) + \left( 1 + \frac{1}{x_H^2} \right)^{1/2} \right] + \frac{1}{R_H} \right\} f_H$$

where

$$f_H = \frac{g_J(H) \mu_o H}{h}$$

$$x_H = \frac{(1-1/R_H)f_H}{\Delta\nu_H}$$

$$R_H = g_J(H)/g_p(H)$$

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$$\nu_D = \left\{ \frac{(1-1/R_D)}{2} \left[ (1-1/x_D) + \left( 1 + \frac{2}{3x_D} - \frac{1}{x_D^2} \right)^{1/2} \right] + \frac{1}{R_D} \right\} f_D,$$

with  $f_D = g_J(D) \mu_o H/h$ ,  $x_D = (1-1/R_D) f_D/\Delta\nu_D$ , and  $R_D = g_J(D)/g_d(D)$ . The g-factor ratio is found from  $f_H$  and  $f_D$ :

$$\frac{g_J(H)}{g_J(D)} = \frac{f_H}{f_D}.$$

The experiment is performed with a hydrogen maser operating at X-band frequencies in the 3.5 kG field of a permanent magnet. By using a double-mode cavity, we are able to operate the maser simultaneously on hydrogen and deuterium. The maser operates in a pulsed fashion in which the hydrogen and deuterium moments are stimulated by an

applied  $90^\circ$  pulse and then allowed to radiate freely. The frequencies are determined by measuring the period of a fixed number of cycles. Great care is taken to ensure that the hydrogen and deuterium signals are compared over identical periods so that fluctuations in the field have little effect on the ratio.

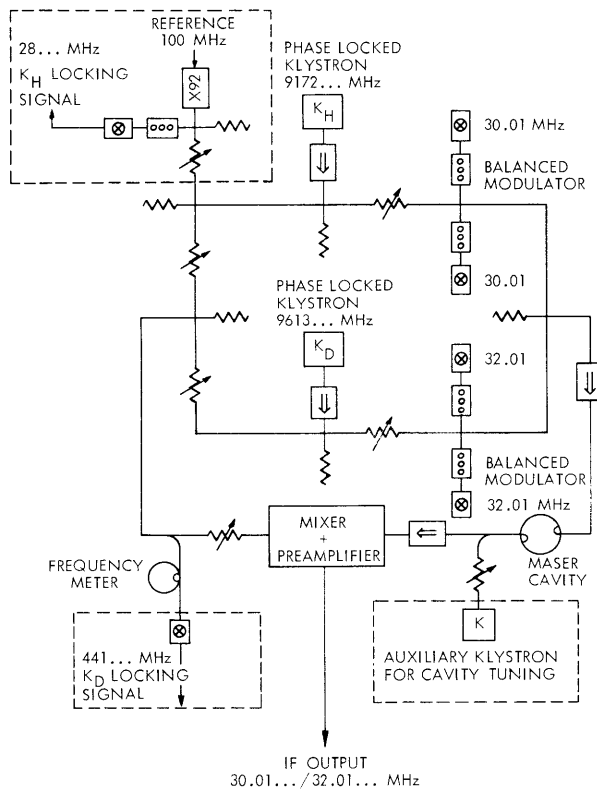


Fig. III-3. Signal-generating system.

## 2. Electronic Instrumentation

The microwave generation system is shown in Fig. III-3. Two phase-locked klystrons provide the stimulating power for the hydrogen and deuterium signal. The klystrons are offset from resonance by 30.01 MHz (hydrogen) and 32.01 MHz (deuterium); sidebands are added to obtain the resonance frequencies. The klystrons also provide local-oscillator power.

Accurate reference frequencies are derived by standard techniques from a stable crystal oscillator which is continuously monitored by a VLF comparator against the broadcast time standard from the National Bureau of Standards.

The detection and counting systems are shown schematically in Fig. III-4. The

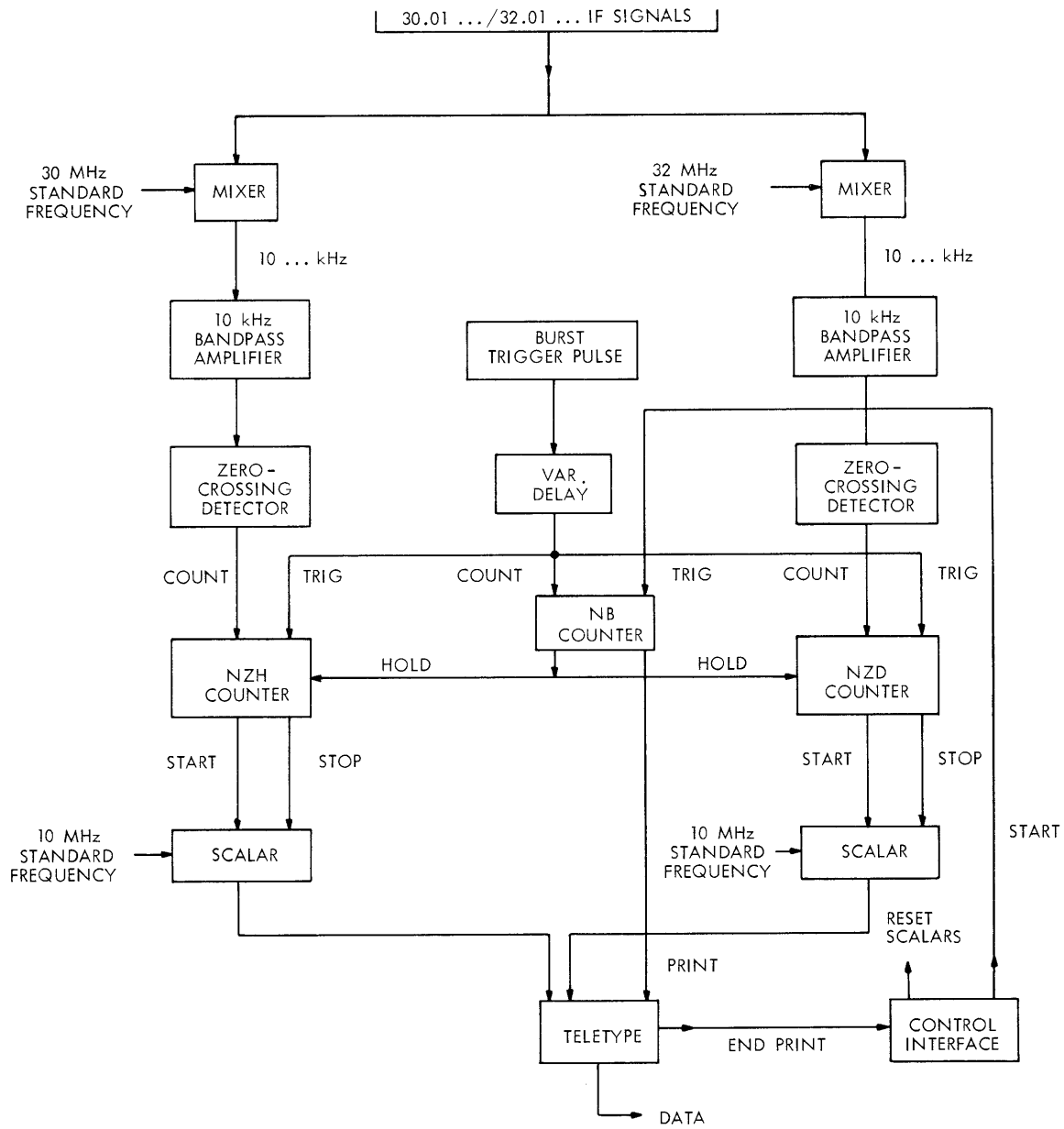


Fig. III-4. Detection and data accumulation systems.

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signals are separately processed in identical channels. Each is heterodyned to 10 kHz and then filtered by a bandpass amplifier. The average frequency for a short burst of decaying radiation is determined by measuring the period of a given number of cycles. The signal is passed through a sensitive zero-crossing detector, the output of which drives special counters providing indicator pulses at the beginning and end of a predetermined number of cycles, NZ. These pulses in turn are used to gate a scalar that counts a reference frequency. The final scalar count measures the average period of the radiation during a burst, and hence its frequency. The process is initiated by a "burst" trigger pulse, and the counting cycle is repeated a preselected number of times, NB, after which the data are read by a teletype and recorded on paper tape for subsequent computer analysis.

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### C. MOLECULAR-BEAM STUDIES OF VAN DER WAALS MOLECULES

We are attempting molecular-beam resonance studies of an unusual species of molecules, the "Van der Waals molecules," in which Van der Waals forces create bound-state molecules for atoms that have no chemical affinity.

Rare-gas dimers provide the prototype Van der Waals molecule. Although these have been formed in abundance in molecular beams,<sup>1,2</sup> they do not lend themselves to magnetic-resonance studies. There is a large class of molecules, however, for which one of the partners is an alkali. Such molecules are attractive, because of their interesting spectra and on account of several experimental advantages, including ease of detection and deflection, and (we hope) production. Among species for which a sizeable number of bound states can be predicted on the basis of accurate potential data are alkali-Hg, alkali-rare gas, and triplet-state alkali-alkali. Also, there are some possible alkali-molecule systems.

The magnetic-resonance spectra of these molecules can yield precise values for interactions that at present are matters for conjecture: for instance, the radial



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dependence of the spin rotation constant, the effects of binding on hyperfine coupling, and various spin-spin interactions.

The optical emission spectra of Van der Waals molecules from high-pressure sources have been recognized for some time.<sup>3</sup> Bouchiat and her colleagues have found direct evidence for alkali-rare gas molecules in optical pumping experiments.<sup>4</sup> Here the formation of bound states leads to various relaxation phenomena from which a number of parameters of the molecule can be deduced. Thus far, however, these molecules have not been examined by resonance methods.

It is a straightforward matter to calculate the equilibrium fraction of Van der Waals molecules. Except for interatomic potentials which support only one or a few bound states, the number of bound states is not particularly sensitive to the shape of the potential. In fact, one can estimate the number of bound vibrational states,  $n_v$ , to an accuracy of perhaps 20% simply by fitting the potential minimum with a parabola and counting the number of levels below the dissociation threshold. In this way it can be shown that

$$n_v \cong \text{const. } \sqrt{\beta},$$

where the constant depends on the analytical form of the potential (for a 6-12 potential it has the value  $1/6\sqrt{2}$ ), and  $\beta$  is a reduced parameter that characterizes the potential:

$$\beta = \mu \epsilon r_m^2 / \hbar^2.$$

Here  $\mu$  is the reduced mass,  $\epsilon$  is the well depth, and  $r_m$  is the position of the potential minimum. To the same approximation, the number of bound rotational states in the lowest vibrational state is

$$N_o = 2\sqrt{\beta}.$$

An exact expression for the number of bound states in a 6-10 potential has been derived by Mahan and Lapp.<sup>5</sup> Their method has been used to compile the results shown in Table III-2 where we give the potential parameter for 3 alkali Van der Waals molecules, together with the total number of bound states,  $Q$ , and the fractional association,  $X = n(\text{molecules})/n(\text{atoms})$ , calculated at 100-Torr pressure with equal densities of the two atomic species.  $X$  has been evaluated in the high-temperature approximation, i. e., with the Boltzmann factor taken as 1. (If positive-energy quasi-bound states are included, the total number of bound states is increased by approximately  $2/3$ .<sup>6</sup> Since most of these states decay in a time that is short compared with the flight time in a molecular beam, we omit them in our estimate.)

Since  $X$  is proportional to the pressure, it is obvious that the equilibrium population is unfavorable at a typical molecular-beam source pressure of 1 Torr. This naturally

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Table III-2. Parameters of some Van der Waals molecules.

Species	G (°K)	$r_m$ (Å)	Q	X
Rb-Kr	115	4.4	$4 \times 10^4$	$1.2 \times 10^{-3}$
K-Cs (triplet)	350	6.5	$7.2 \times 10^5$	$3.3 \times 10^{-2}$
Na-Hg	620	4.7	$2.8 \times 10^5$	$2.2 \times 10^{-2}$

suggests the use of a jet beam operating at high source pressure. Furthermore, the cooling that accompanies the beam expansion offers the possibility of enhancing the fractional association.

A jet source differs from a conventional atomic-beam effusive source by operating in the high-pressure limit where the mean-free path is short compared with the source aperture. The emerging gas undergoes adiabatic expansion during which the translational temperature falls and the available energy, plus the energy of expansion, goes into the translational energy of the beam. If the temperature of the source is  $T_s$  and the translational velocity of the beam is  $u$ , then for a single atomic species of mass  $m$ , we have

$$\frac{1}{2} mu^2 = \frac{3}{2} kT_s + kT_s.$$

From this it follows that

$$u = \sqrt{5/3} a_s,$$

where  $a_s$  is the most probable source velocity.

This description predicts a monochromatic beam. In practice this limit can be approached closely: velocity spreads corresponding to an internal beam temperature of a few degrees are possible. Jet sources are often characterized by the Mach number,  $M$ , the ratio of velocity of the beam to the velocity of sound in the beam. If  $\Delta v$  is the full width at half-maximum of the velocity distribution, then for an atomic beam the Mach number is given approximately by

$$M = 1.82 \frac{u}{\Delta v}.$$

This expression is accurate in the limit  $M \gg 1$ . The internal temperature of an atomic beam is related to the oven temperature by

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$$T_b = \frac{7.4}{M^2} T_o.$$

In many cases determination of the Mach number is limited by velocity resolution of the apparatus. A Mach number of 20 can be readily obtained, corresponding to a reduction of the temperature by a factor of 50, say, from 800°K to 16°K. It is apparent that the translational temperature of a jet beam cannot have too much to do with the degree of association – at such low temperatures one would expect the ultimate in association, a solid. The expansion is too rapid for association or condensation to maintain equilibrium. Nevertheless, there can be a considerable enhancement of the association fraction over that in the source and several groups have used this method to produce intense beams of alkali singlet dimers.<sup>7-9</sup> In particular, Gordon and Hershbach<sup>9</sup> have produced dimer beams with a molar concentration of 30%, a concentration 30 times greater than the source equilibrium value. Since little of the reaction energy appeared in the transitional mode, they concluded that the molecules were in highly excited vibrational states. There are indications that the molecules are rotationally cooled, which is reasonable in view of the relative ease with which rotational cooling occurs.

Although it is difficult to predict accurately the rate of production of alkali-Van der Waals molecules in a beam, these considerations indicate that it may indeed be feasible to produce a reasonably robust beam. The molar fraction is most likely still to be low, however, which means that there may be a problem of separating the molecules from the atomic background. Conventional deflection techniques can be of assistance here: since all species from a jet source move at approximately the same velocity, the more massive molecules will be deflected less than the free atoms. (This is not true for alkali triplet dimers.) The more reliable method for identifying the molecules is by their magnetic resonance spectrum. Fortunately, estimates of the spin rotation constants for the alkali-rare gas molecules are available,<sup>4</sup> and this may offer assistance in the search for these molecules.

Our initial efforts are to produce K-Ar. The experiment is being undertaken with the spin-exchange scattering apparatus that has been demoted temporarily to a molecular-beam apparatus. We have modified the source by adding a gas inlet to introduce pressurized Argon to the oven and we have also added a C-magnet and an RF loop in the scattering chamber. Some preliminary work on a pure potassium jet beam with a nozzle diameter of 0.0015 in. and a hot skimmer situated 50 nozzle diameters downstream gave evidence of skimmer-jet interaction. The situation improves considerably when the hot skimmer is removed, and a beam of Mach number 9 has been formed in which the singlet dimer concentration is 25%. A new liquid-nitrogen-cooled oven housing has been completed, and a rotating cold skimmer is under construction. We hope this will eliminate the skimmer-jet interaction problem.

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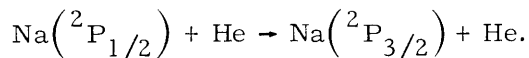
#### D. MERGING ATOMIC BEAMS

We have investigated the feasibility of observing atomic scattering processes with variable but well-defined relative energy through the use of merging atomic beams. Briefly, if the two species approach with identical velocities,  $v$ , then the relative energy is

$$K_r = 2\mu v^2 \sin^2(\theta/2),$$

where  $\mu$  is the reduced mass, and  $\theta$  is the angle subtended by the beams. With conventional atomic beams the large thermal velocity spread prevents this method from being practical. The low internal temperature in jet beams, which was described in Section III-B, allows good energy resolution, however, down to very small values of  $K_r$ .

As a prototype experiment we have considered measuring the energy dependence of the total cross section for the following process.



In this process an excited Na atom undergoes a fine-structure transition, because of a collision with helium before it radiates to the ground state. The process has been studied extensively by Krause<sup>1</sup> and others, in gas cell experiments in which one of the D lines is excited and fluorescence from the other D line is studied as a function of rare-gas pressure. Such experiments measure  $\bar{Q}$ , the cross section averaged over all collision energies. Recent theoretical work by Reid<sup>2</sup> on  $Q(E)$  reveals that the cross section has a rich energy dependence that is sensitive to details of the interaction between Na(3P) and He. The maximum of the cross section occurs at an energy of  $\sim 60^\circ\text{K}$ , and there is

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structure attributable to orbiting collisions with a width of  $\sim 15^\circ\text{K}$ . None of this is observable in room-temperature experiments involving thermal-energy spread of  $300^\circ\text{K}$ .

In the projected experiment a jet beam of Na merges with a jet beam of He in an interaction region which is illuminated with a pulsed dye laser tuned to the lower D line. The yield of photons of the upper D line during the fluorescent decay is studied as a function of the interaction energy. The temperature of the helium source is adjusted to produce a beam velocity equal to that of the sodium velocity.

A numerical analysis has been made of the relative energy distribution,  $f(E)$ , as a function of scattering angle. The beam velocity distributions were assumed

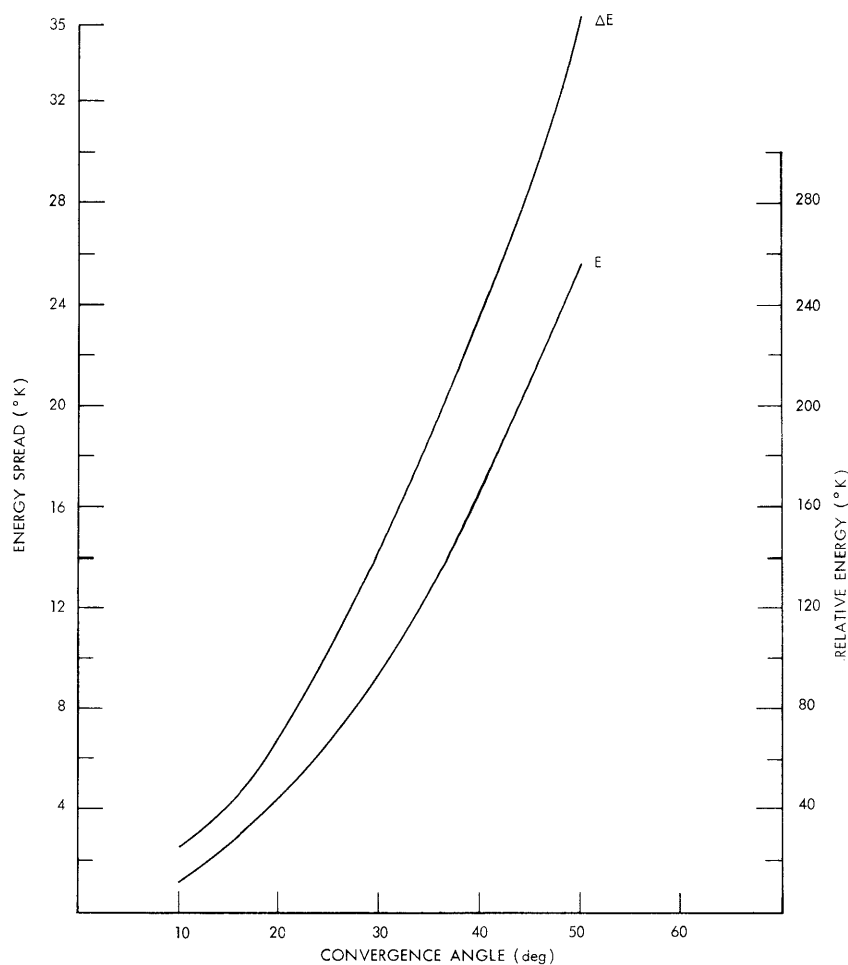


Fig. III-5. Energy distribution for merging beams of Na and He with  $u = 1.32 \times 10^5$  cm/s,  $u/\Delta v = 10$ , and a convergence angle of  $25^\circ$ .

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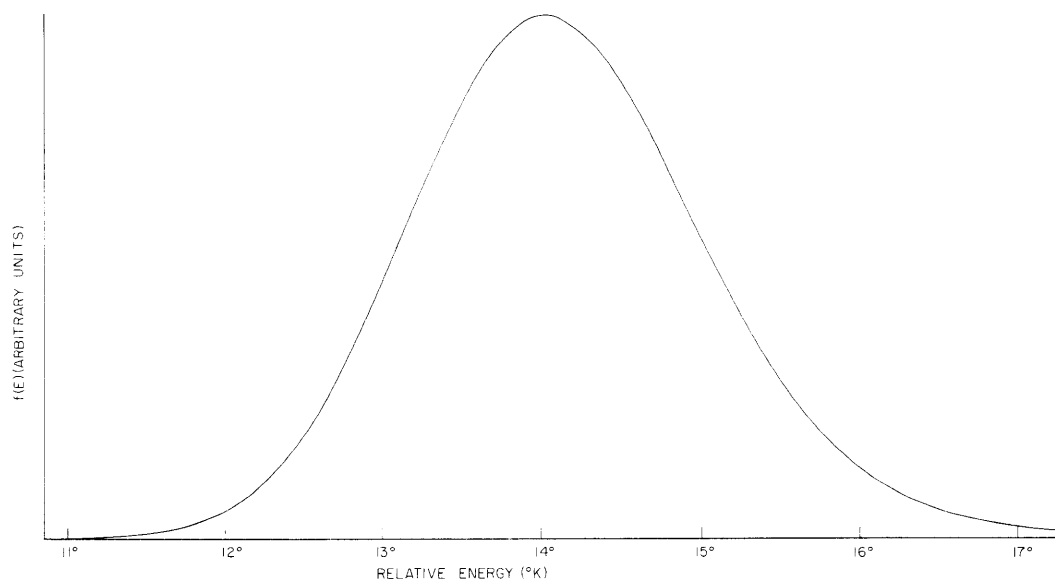


Fig. III-6. Mean energy and energy spread (full width at half-maximum) vs convergence angle.

to be Maxwellian distributions with a full width at half-height of  $\Delta v$ , superimposed on a high translational velocity,  $u$ . Figure III-5 shows a typical result for  $f(E)$  for the case  $u/\Delta v = 10$ . The shape of the curve is practically unchanged, except at low scattering angles where the curve becomes skewed and the energy resolution decreases. Figure III-6 gives the mean energy and energy spread for various angles.

The results of this analysis indicate that this merging beam technique is capable of giving excellent energy resolution, and is potentially a valuable tool for a variety of collision studies.

D. Kleppner, C. E. Wieman

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#### E. METASTABLE HELIUM SOURCE

We are developing a metastable helium source with the aim of measuring differential scattering cross sections for helium in the metastable  $1s2s$  configuration. The traditional method of producing metastable helium is by electron bombardment of an atomic beam. In order to obtain a higher flux of metastable helium than is possible with an effusive source, we are using a hollow-cathode discharge (HCD) as a source. This type

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of discharge has been investigated by Lidsky, et al.,<sup>1</sup> and more recently by Delcroix, et al.<sup>2</sup> The advantages of the HCD for our purposes are: (i) it can operate in the "jet" mode, thereby giving an atomic beam of greater intensity and narrower energy spread than that produced by an effusive source; (ii) the high degree of ionization known to exist in the plasma produced by the HCD gives promise of a large population of metastables (Delcroix and his co-workers have calculated a theoretical fraction of 10% metastables at the end of the cathode of an argon HCD<sup>3</sup>); and (iii) the fast expansion of the jet into a low-pressure region will help minimize loss of metastables through collisions with background gas and other atoms.

We have constructed a simple HCD source in order to measure its production characteristics. Time-of-flight spectroscopy will be used to study the velocity distribution of the beam, with a continuous-strip channel electron multiplier detecting the metastable atoms. At the present time, we are in the debugging stage. In a related experiment, the possibility of using nuclear photographic emulsions to detect metastable atoms is being investigated.

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#### F. MULTIPLE-RESOLUTION VELOCITY SELECTOR

We have constructed a slotted-disc velocity selector for use in atomic beams. The selector has no thick discs, and consequently it has several velocity passbands.<sup>1</sup> In applications where there is no uncertainty about which passband is in use (for example, where the width of the velocity distributions is reduced by state selectors, other velocity selectors, or by supersonic jetting action) the existence of multiple passbands is advantageous because the resolution of the selector can be changed simply by changing the selector speed so that a different passband is used.

Figure III-7 is a rolled-out representation of a typical thin disc velocity selector whose discs are positioned at integral multiples of some unit distance (similar to Kinsey's method<sup>1</sup> of disc positioning, but with no thick disc). It is clear that there is a regular array of straight-line paths through the selector, each with a different pitch. Characterizing the paths by the integer index  $m$  (see Fig. III-7), we can show that the central velocity of the  $m^{\text{th}}$  passband is

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$$V_m = \frac{NfL}{a + mn},$$

where  $N$  is the number of teeth per disc,  $f$  is the rotational frequency (in Hz), taken positive when the  $m = 0$  passband is in use, and  $a$ , the asymmetry parameter, is measured in units of the tooth spacing. The remaining variables are defined in Fig. III-7. The corresponding resolution (full width at half-maximum) is very nearly

$$R_m = \frac{\eta}{|a + mn|},$$

where  $\eta$  is the fractional open space (the ratio of tooth width to tooth spacing;  $\ell_1/(2\bar{\ell})$  in Hostettler and Bernstein's<sup>2</sup> notation).

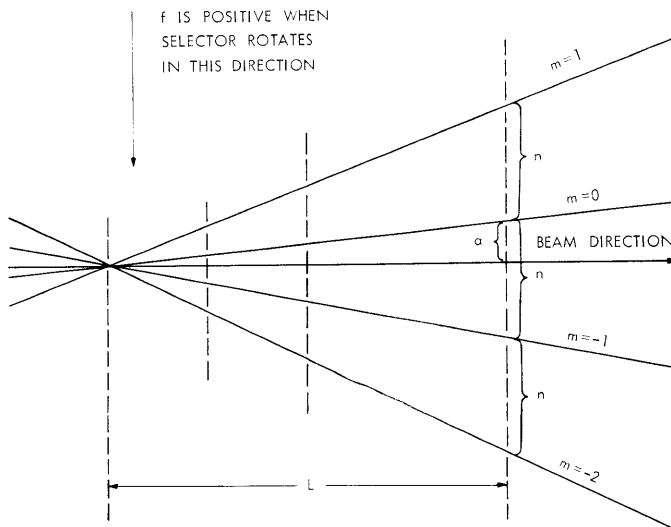


Fig. III-7.

Schematic representation of slit orientation and possible trajectories for thin disc velocity selector.

Proper choice of  $a$  and  $n$  can yield quite different selectors. For example, if  $a = 0$  it is possible to align the apparatus visually with the selector in place. More variety is achieved by choosing  $a \neq 0$ ; for example, a selector with resolutions of 16%, 8%, 4%, and 3.2% results from the choice  $a = 2$ ,  $n = 9$ , and  $\eta = 0.48$ . In any application, it is necessary to make measurements of the same (preferably sharply peaked) velocity distribution in order to determine  $a$  exactly. This eliminates any velocity error caused by misalignment of the selector.<sup>3</sup>

We use a simple passive phase-shift network which facilitates changes in selector speed by providing a 90° phase shift over a 20:1 frequency range.<sup>4</sup>

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References

1. J. L. Kinsey, Rev. Sci. Instr. 37, 61 (1966).
2. S. U. Hostettler and R. B. Bernstein, Rev. Sci. Instr. 31, 872 (1960).
3. A. E. Grosser, Rev. Sci. Instr. 38, 257 (1967). Grosser's design utilizes thick discs to eliminate all but the  $m = 0$  and  $m = -1$  sidebands.
4. "A Phased Single-Sideband Exciter," in The Radio Amateur's Handbook (American Radio Relay League, Newington, Conn., 1954), p. 298. (Any handbook 1955  $\pm$  3 A.D. should contain a description of this exciter. If the capacitors in the exciter are increased by some factor, the operating frequency range will be lowered by this factor.)

