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Detection of modulated molecular beams with electron bombardment detectors*

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Abstract. By means of refinements in the modulated molecular beam technique the signal-to-noise ratio can be greatly improved, and differential cross-sections, for collision of molecules of the same species, can be measured. This was accomplished by combining beam modulation and phase sensitive detection with very sharp turning on the front end of the lock-in-amplifier and long integration times on the output. In addition, the signal-to-noise ratio of the Ar-Ar system as a function of integration time was investigated using two different types of electron bombardment detectors an Aberth ion-source and a quadrupole mass filter. With an integration time of 40 min the estimated upper limit to the signal-to-noise ratio is 1500 to 1 for the Aberth ion-source. Using quadrupole mass filter with an integration time of 60 min the estimated upper limit to the signal-to-noise ratio is 5×10^4 to 1. For chemical kinetics studies this ratio may be two orders of magnitude higher.

Keywords. Molecular beams; neutral beam detection; molecular beam scattering; collision cross-sections.

1. Introduction

The molecular beam technique is a powerful tool for investigating intermolecular forces between atoms and molecules. Indeed, the differential scattering cross-section (DSCS) shows a very sharp sensitivity to the intermolecular potential (Mueller and Marchi 1963 and Luoma and Mueller 1965). The problem of measuring the DSCS is somewhat formidable as the beam intensities are a factor of 10^2 to 10^5 lower than in total cross-section measurements.

For systems in which the scattering beam molecules are made up of the same gas, the problem is further complicated because it now require detection of a signal which is submerged in a background several orders of magnitude greater. In addition to these obstacles, measurement of an absolute DSCS as opposed to a relative DSCS requires a knowledge of the scattering particle density as well as scattering path length. For this apparatus these two factors can be determined according to the work of Landorf and Mueller (1966). Knaur (1933) and Zabel (1933) studied scattering as a function of angle for systems in which the incident beam molecules and the scattering gas molecules are the same. In both these studies a primary beam was allowed to enter a scattering chamber filled with the target gas. The background in these experiments is quite complicated and these early measurements probably have large systematic errors. The technique of beam modulation and phase sensitive

*Measurements were carried out at the Purdue University, Lafayette, Indiana, USA.

detection (Brackman and Fite 1961) provides a partial solution. Although differential scattering cross-sections have been measured recently by several authors who used crossed molecular beams, the scattering systems have mostly been limited to cases where the primary beam molecules differ from the secondary beam molecules, and to cases where electron bombardment detection is not required.

2. Apparatus and experimental technique

The apparatus, a good part of the detection system and the measuring technique has been previously described in the literature (Landorf and Mueller 1966 and Mueller *et al* 1971). Here we will describe in brief the essential parts of the set-up. The primary beam is modulated by a 41 cps tuning fork chopper and its reference output is utilized to synchronize a PAR HR 8 lock-in-amplifier.

The modulated beam is crossed at right angles by a secondary beam, and the scattered beam is ionized by the Aberth detector in one case, and the quadrupole mass filter in the other. The resultant ion beam is then focussed and accelerated into an electron multiplier.

The output from a twelve stage Ag-Mg electron multiplier (Dumount Model SPM-DL-100) is fed into a preamplifier which consists of four cascaded White transistor active band pass filters (White Instruments, Inc. Austin, Texas). These have the effect of allowing very sharp tuning on the input, or front end of the lock-in-amplifier. The output of the lock-in-amplifier is then recorded and averaged over

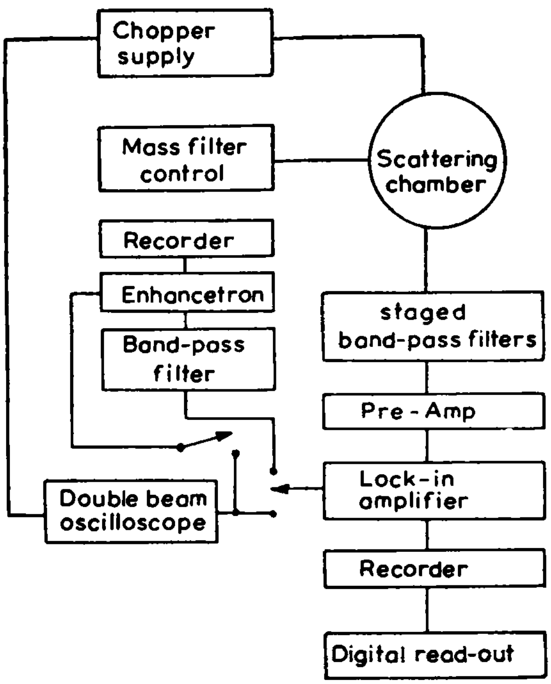

Figure 1. Block diagram of the electronics. For the Aberth gun, the mass spectrometer control is replaced by Kepco and Fluke voltage supplies. The enhancetron is connected to the HR-8 Signal monitor.

Table 1. Estimated signal-to-noise ratios for a standard* argon beam

Signal-to-noise ratio	Conditions	Comments
	(a) *Aberth Detector*	
Not detectable	No White filters	
10/1	3 White filters $\tau = 3$ sec	Main beam
32/1	4 White filters $\tau = 3$ sec	Main beam
1500/1	4 White filters $\tau = 40$ min	Scattered beam
	(b) *Quadrupole mass filter*	
600/1	3 White filters $\tau = 3$ sec	Main beam
2000/1	3 White filters $\tau = 3$ min	Scattered beam
20000/1	3 White filters $\tau = 30$ min	Scattered beam
$5 \times 10^4/1$	3 White filters $\tau = 60$ min	Scattered beam

*A standard beam has a flux of 5×10^{16} molecules/Sr. sec. Total beam flux is $1{\cdot}8 \times 10^{13}$ molecules/sec. Maximum flux into the detector was 2×10^{12} molecules/sec.

long integration times by means of a mechanical recorder. A block diagram of the electronics is shown in figure 1.

The major part of the noise in these experiments was due to the bursting of Aberth ion-source. This ion-source noise (Minturn *et al* 1960) which contain a large component of very short duration (μs) high intensity bursts is to be eliminated before lock-in. Four cascade White transistor active filters consisting of two type 256 and two 254 band-pass filters serves as the preamplifier. Each is especially made for 41Hz and has a band width 1·5%. The four stages were used in a type 256-254-254-256 geometry and the stages were isolated and shielded from each other to prevent feedback and ringing.

A four-cycle technique was used to measure the cross-sections. Measurements were made using both the Aberth detector and the quadropole. With the Aberth detector, emission currents were in the range of 100–300 mA. The emission current with the quadrupole mass filter was 1·1 mA. The primary beam pressure was 200μ. The pressure in the scattering chamber was $3{\cdot}2 \times 10^{-6}$ torr.

In addition to the sharp front-end tuning, it was necessary to have long integration times on the output in order to average out the noise bursts. The filter time constant of the lock-in-amplifier is limited to 100 sec with capacitance filtering. However, by using a disc integrator with digital output, the time constant of the lock-in amplifier could be extended indefinitely. The integrator is a model 224 CZ, which is available from Disc Instruments, Santa Ana, California. The recorder used in conjunction with the disc integrater was a Leeds-Northrup Spectromax Type W, Model S, Indicating Recorder. Table 1 shows the improvement in the signal-to-noise ratio (S/N) as longer integration times and more filters are used.

3. Comparison of electron bombardment and mass spectrometric detection

The replacement of the Aberth gun by an EAI quadrupole caused an improvement of about 35 in the signal-to-noise ratio, even though the background is of the same species as the beam. We believe that its function is largely to eliminate source noise. Probably much of the noise in the Aberth gun was from positive ion bursts from the anode which were eliminated by mass selection. There is a better signal-to-noise ratio when the beam and background are of different species, but experiments with He-Ar system have not revealed any really dramatic differences.

The character of the noise did not appear to be inversely proportional to the square root of the integration time. In fact, for integration times of one-half to two hours it appeared to vary inversely with the period of integration. The noise does not have a gaussian distribution but appears to have some cyclic characteristics.

This noise was also in phase with the primary beam signal and appeared to come from bouncing of the primary beam off the walls of the vacuum envelope. This has subsequently been confirmed by the use of an enhancetron to obtain the averaged wave form of the background, and by more direct expedient of removing separately the primary and secondary beams. Apparently, the tuning was sharp enough to reduce the noise from the residual background gas below the noise level arising from the primary beam. Further noise reduction will depend on effective means of preventing the primary beam from entering the ionization region after first pass rather than by general reduction of the background.

In the enhancetron experiments, the signal from the lock-in monitor was sent through an additional tuned stage before entering the time-averaging system. The reduction in signal-to-noise is so striking that this technique may be superior to the use of lock-in amplification.

We also examined the noise off the primary beam and off the mass peak of the primary and secondary beams. This kind of noise will be important in measurements of beam kinetics. The signal-to-noise ratio was at least two orders of magnitude higher for this kind of experiment. The noise was highly cyclic in nature.

4. Comparison with other experiments

Determinations of signal-to-noise ratios are given in table 1. All the experiments with quadrupole were done with three filters and a bandwidth of 0·3 Hz. The background was generally 3 to 4×10^{-6} mm and of the same species as the beam. The only direct comparison is with a series of single beam experiments with small backgrounds, Bennewitz and Wedemeyer (1963) has reported a signal-to-noise ratio of 750 for argon with a time constant of 1·25 sec. This is slightly better than our ratio of 600 with a time constant of 3 sec. The effect of the sharpened tuning has eliminated most of the background noise even though the background pressure is at least two orders of magnitude higher in our experiments.

The use of long integration times gives an improvement of 60 over that of Bennewitz and it is only by the use of long integration times that the lock-in system makes a substantial improvement. With a time constant of one second the lock-in contributes almost nothing to the reduction of noise because the response time of the tuned circuit is roughly the same.

In these experiments, most of the noise came from the primary beam. Signal-to-noise ratios obtained from reducing primary beam intensities to the lowest detectable level are apt to be over-optimistic since the noise level is also reduced.

The system which was described in this article has been used for the measurement

Table 2. The argon differential scattering cross-section

Centre of mass angle (Degrees)	Differential cross-sections (C. M. system, A^2)
1·83	$2{\cdot}316 \times 10^4$
3·00	$1{\cdot}768 \times 10^4$
4·00	$5{\cdot}69 \times 10^3$
5·00	$2{\cdot}60 \times 10^3$

of rare gas differential scattering cross-sections (Penta *et al* 1967). These experiments must be conducted in the presence of an intense primary beam. Under these circumstances, signal-to-noise ratios on the scattered beam are the only meaningful quantities.

5. Data

Table 2 gives data on the differential cross-section of argon using the quadrupole mass filter. These data were collected over a 7 hr period. More extensive data have been published elsewhere (Mueller *et al* 1971) on this and other rare gas systems.

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