

VII. MOLECULAR BEAMS*

Prof. J. R. Zacharias
 Prof. J. G. King
 Prof. C. L. Searle

Dr. R. F. C. Vessot
 H. H. Brown, Jr.
 R. Golub

E. L. Kamen
 F. J. O'Brien
 R. Weiss

A. SEARCH FOR A SMALL CHARGE CARRIED BY MOLECULES

The suggestion by Lyttleton and Bondi (1) that hydrogen atoms might carry a charge of approximately 2×10^{-18} of an elementary charge has stimulated a number of investigators to undertake experiments designed to measure this charge. At least two methods have been used; atomic beam deflection experiments (2, 3), and the method of Piccard and Kessler (4). The results published thus far are listed in Table VII-1. In this paper

Table VII-1. Upper limit on the charge carried by various molecules.

Molecule	Charge (elementary charge/molecule)	Experimental Method
CO ₂	$\leq 2 \times 10^{-19}$	Piccard-Kessler (ref. 4)
CsI	$\leq 4 \times 10^{-13}$	Molecular beam (ref. 2)
A	$< 8 \times 10^{-20}$	Piccard-Kessler (ref. 5)
N ₂	$< 12 \times 10^{-20}$	Piccard-Kessler (ref. 5)
CsF	$\leq 2 \times 10^{-14}$	Molecular beam (ref. 3)
KF	$\leq 1 \times 10^{-13}$	Molecular beam (ref. 3)
n	$< 6 \times 10^{-12}$	Molecular beam (ref. 6)

preliminary results of a Piccard-Kessler experiment are reported in which hydrogen molecules (and also helium atoms) exhibit a charge that is approximately 40 times less than that required by the hypothesis of Lyttleton and Bondi.

In our experiment the gas whose charge is to be measured is allowed to escape from an electrically insulated metal container connected to an electrometer. While the gas is escaping, a current that can be measured will flow to the container if the gas is charged. The charge on each atom can then be computed, although it is essential to distinguish charge carried by each of the hypothetically charged atoms from charge carried by ions, electrons, and charged dust particles. By passing the escaping gas through a de-ionizer

* This work was supported in part by Purchase Order DDL B-00306 with Lincoln Laboratory, a center for research operated by Massachusetts Institute of Technology with the joint support of the U.S. Army, Navy, and Air Force under Air Force Contract AF 19(604)-5200.

(VII. MOLECULAR BEAMS)

consisting of a coaxial capacitor charged by a battery within the container, ions and electrons, because of their relatively high mobilities, can be swept from the gas stream, while atoms carrying the hypothetical charge of 10^{-18} of an electron charge will be unaffected. Dust particles, which range in size from molecules to visible grains, may be sufficiently massive to pass through the de-ionizer, but their effect is likely to be both erratic and cumulative.

By recording the pressure and the output of the electrometer, both as a function of time, on a two-channel recorder, it is not only possible to measure the total charge carried by the gas, but also to see that the potential-versus-time curve has the expected shape for the flow of a uniformly charged gas. The course to be followed by the experimenter is plain. If the atoms seem to be charged, the computed charge per atom must be independent of all parameters, such as de-ionizer potential and polarity, rate of gas efflux, pressure, bias potential of the container with respect to ground, and, of course, details of apparatus construction and gas handling. It is to be expected that the charge per atom will be different for different gases. On the other hand, if the atoms appear uncharged, it is important to make certain that the electrometer is sensitive during the gas efflux and that mechanisms that might cancel a genuine effect are eliminated. Such cancellation is most likely to occur at the end of the outlet pipe from the container.

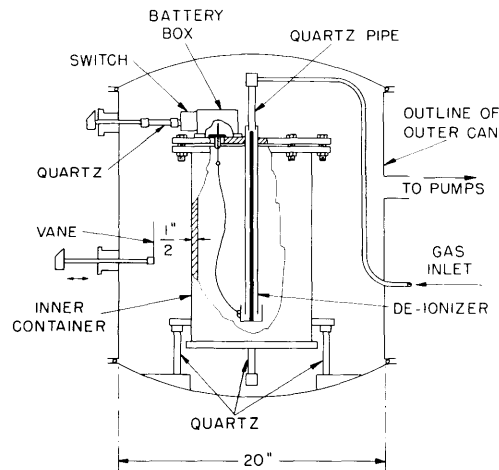


Fig. VII-1. Scale drawing of the apparatus.

Figure VII-1 is a scale drawing of the apparatus. The outer can serves both as a vacuum envelope and as an electrical shield for the container (11.2 liters) which stands on three quartz supports and is filled with gas through a quartz pipe. This quartz pipe is metalized at its ends so that it can be soft-soldered into place. Although it was designed to stand a pressure of 1000 psi, the container was usually operated at a pressure

(VII. MOLECULAR BEAMS)

of 265 psi. The outer can was pumped to 10^{-6} mm Hg to prevent electrical leakage and to reduce undesirable thermal effects. The central electrode of the de-ionizer is insulated by Teflon spacers and maintained at ± 90 volts, or zero volt with respect to ground, as determined by the setting of a switch in the battery box that is visible on top of the container. The switch can be controlled from outside by the insulated shaft. The approximate contact potential to ground of the container can be determined by a movable vane. The gas that is to be investigated is admitted to the container through a porous bronze filter with 5- μ pores, an electrostatic precipitator, and needle valves for controlling the flow. A pressure transducer generates a signal proportional to the pressure in the container which can be recorded on one channel of a two-channel Sanborn recorder. Filling the container rapidly resulted invariably in the appearance of erratic voltages when the gas was allowed to escape. This is believed to be due to the stirring up of a cloud of dust within the container because pumping it down and then filling it slowly always restored normal performance. All of the runs reported here were made with filling times of one-half hour, or more, which appeared to give reproducible results.

The gas escapes from the container through an external regulator that maintains constant pressure on a needle valve adjusted to give the desired rate of flow. It was thus possible to obtain a nearly linear drop in pressure in the container with respect to time. It was found that the pressure-versus-time curve departed from linearity because of temperature changes within the needle valve during the flow of gas. The pressure transducer was calibrated, and the behavior of the pressure-versus-time recording when the gas flow was suddenly stopped was examined closely. It was concluded that the pressure recordings are accurate and can be used as reliable indicators of the amount of gas in the container.

Potential changes of the container with respect to the outer can are measured with a GL5674 electrometer whose output is amplified by a chopper amplifier and recorded on the other channel of the Sanborn recorder. Potential differences of 10^{-4} volt, or currents of approximately 10^3 electrons per second, can be detected. The time constant is 60 ± 5 sec which agrees with the time constant predicted from independent measurements of the electrometer grid resistance and capacitance, 10^{12} ohms and 60 μf , respectively. The ground return of the electrometer is connected through a low-impedance voltage supply that makes it possible to place the container at different potentials with respect to the outer can. When the potential of the container was +0.01 volt, moving the vane was found to have negligible effect. Calibrating signals may be introduced at the ground end of the electrometer grid resistor. Since the time constant of the electrometer was determined by the input circuit, the rms noise voltage should be $(kT/C)^{1/2}$, which is approximately 10 μv , so that the electrometer performance is within a factor of 10 of that which might ultimately be obtained. Frequent calibrations of the electrometer were made with signals close to the minimum detectable signal

(VII. MOLECULAR BEAMS)

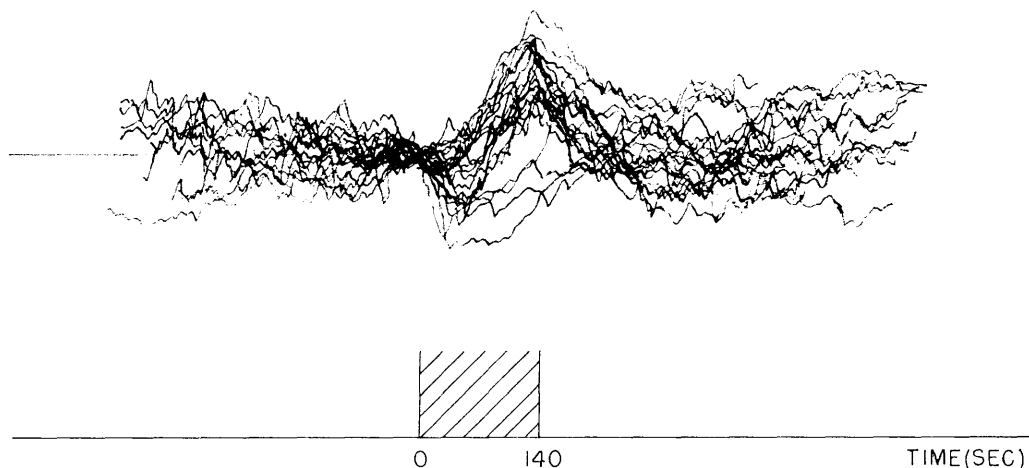


Fig. VII-2. Superimposed electrometer recordings of 21 hydrogen runs.

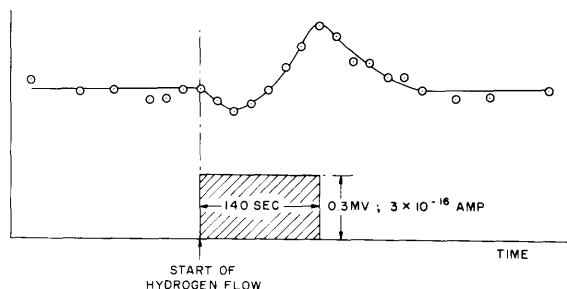


Fig. VII-3. Average curve constructed from the data of Fig. VII-2.

introduced at the ground end of the electrometer grid resistor. No consistent dependence of the electrometer sensitivity on the polarity of the calibrated signal, the amount of gas in the container, the polarity of the de-ionizer or any other parameter was found. Calibrations were also made while the gas was flowing from the container, and the sensitivity was found to be normal.

A charged gas leaving the container would induce charges on its inner wall and thus cause a current to flow in the electrometer grid resistor. To make certain that the electrometer was sensitive to the flow of such charges, a small probe was put in the container and connected by a shielded lead to a voltage source that was made to vary linearly with time; from the estimated capacitance between the probe and the container, the rate of voltage rise, and the size of the electrometer response, the value of the electrometer sensitivity could be determined and it was found to agree well with the value obtained by direct calibration.

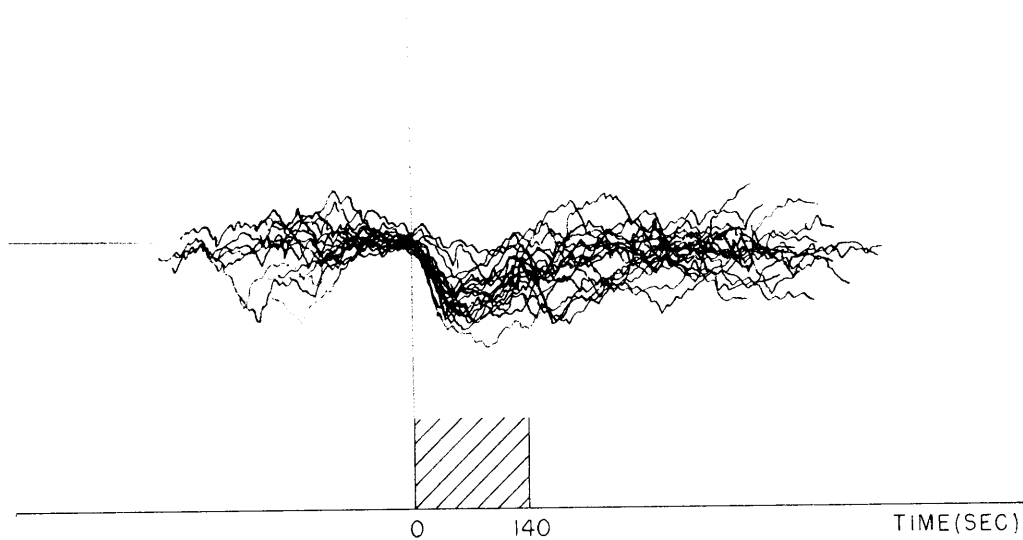


Fig. VII-4. Superimposed electrometer recordings of 19 helium runs.

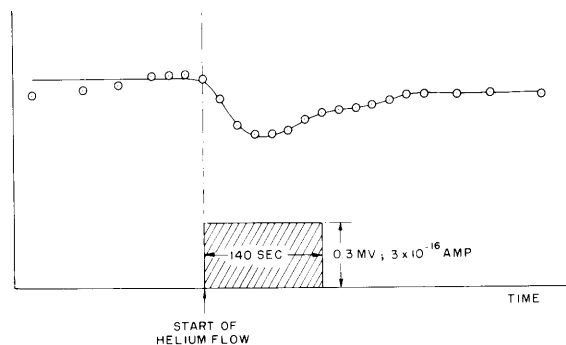


Fig. VII-5. Average curve constructed from the data of Fig. VII-4.

The 21 hydrogen and 19 helium runs at 265 psi and 140-sec efflux time (selected only because these parameters were the same) yielded electrometer recordings that are superimposed in Figs. VII-2 and VII-4, normalized at the start of flow. Average curves that were constructed for hydrogen and helium are plotted in Figs. VII-3 and VII-5. The shaded rectangle, 0.3 mv high and 140 sec long, represents 2.7×10^5 elementary charges, which is the amount of charge that would be observed if each of the 5.4×10^{20} molecules carried 5×10^{24} of an elementary charge. The apparent charge per molecule, determined from the average curves, is $2.5 \pm 1.5 \times 10^{-20}$ elementary charge/molecule for hydrogen, and $-4 \pm 2 \times 10^{-20}$ elementary charge/molecule for helium. The charge on hydrogen appears to be negative; the charge on helium, to be positive.

None of the recordings looks like the computed curve, which is plotted in Fig. VII-6,

(VII. MOLECULAR BEAMS)

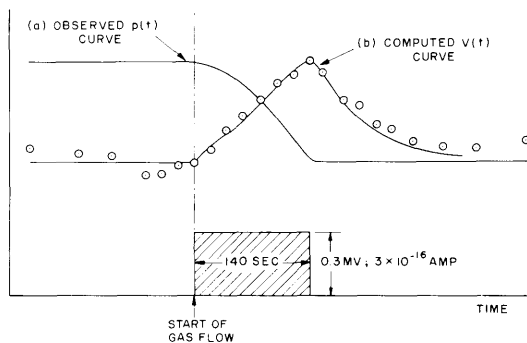


Fig. VII-6. Curve obtained by subtracting the curve of Fig. VII-5 from the curve of Fig. VII-3.

together with the pressure curve from which it was derived. The hydrogen curve goes negative and then positive, and the helium curve has a negative maximum that is too early by 30 sec. If it is assumed that the helium effect is spurious and that whatever produces it is likewise present in the hydrogen runs, the helium curve may be subtracted from the hydrogen curve. This procedure yields a curve (represented by circles in Fig. VII-6) that is in remarkable agreement with the computed curve. If this agreement is taken to be significant, we obtain a value of $7 \pm 2.5 \times 10^{-20}$ elementary charge/molecule for hydrogen, and $0 \pm 2 \times 10^{-20}$ elementary charge/molecule for helium.

The electrometer deflections recorded are still not understood; but the nature of these results, as well as the results of numerous side experiments, eliminates a few of the possible causes for the deflection, such as capacitative effects, piezoelectricity, and thermoelectricity. New and improved apparatus, use of other gases, including deuterium, and wider variations of parameters will provide the answer. In any case, the charge that we found for the hydrogen molecule, by interpreting our experiment in the obvious way, is far less than that required for the hydrogen atom by the hypothesis of Lyttleton and Bondi. A molecular-beam experiment is under way with cesium, but the difficulties of achieving sufficient sensitivity with hydrogen (and later atomic hydrogen) are considerable.

J. G. King

References

1. R. A. Lyttleton and H. Bondi, Proc. Roy. Soc. (London) A252, 313 (1959).
2. V. W. Hughes, Phys. Rev. 105, 170 (1957).
3. J. C. Zorn, G. E. Chamberlain, and V. W. Hughes, Bull. Am. Phys. Soc. Ser. II 5, 36 (1960).
4. A. Piccard and E. Kessler, Arch. Sci. Phys. et Nat. 7, 340 (1925).
5. A. M. Hillas and T. E. Cranshaw, Nature 184, 892 (1959).
6. I. Shapiro and V. Estulin, Soviet Phys. - JETP 3, 626 (1957).