

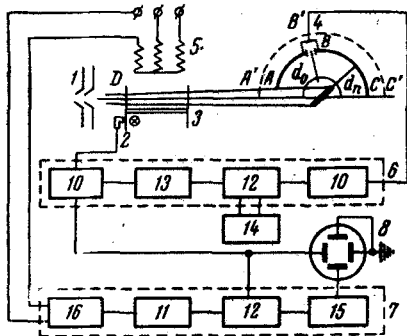
# STUDY OF ENERGY TRANSFER DURING THE COLLISION OF A MOLECULAR STREAM WITH A SURFACE

V. B. Leonas

Zhurnal Prikladnoi Mekhaniki i Tekhnicheskoi Fiziki, No. 2, pp. 84-86, 1965

A method of direct study of the energy transfer between a molecular stream and the surface of a solid body is described. The energy loss of particles (Ar, CO<sub>2</sub>) colliding with metal surfaces (Cu, Fe, Ta) was measured at various temperatures. The capture of the molecules by these surfaces was observed, and the corresponding adsorption times were measured.

Direct study of the effects of the interaction between molecules and a solid surface was made possible by the use of the molecular-beam method [1]. In its application to the study of energy transfer, the method consists in generating a collimated beam of atoms (or molecules), shooting these at the surface under investigation, and measuring the change in particle energy associated with the collision. Clearly, one can determine the energy loss if, knowing the speeds of the incident particles, one can measure the speeds of the reflected particles. Disregarding internal degrees of freedom, one can determine the total energy of the reflected particles from their speeds, and thus calculate the energy loss associated with the collision. As the molecular-beam method is based on direct measurements of energy transfer, which makes the interpretation of the data much simpler, it is possible, in principle, to control more thoroughly the state of the investigated surface (operation at high vacuum and temperature, considerable reduction in measuring time; see, for example [2]).



The experimental setup used in this work was described in [3]. Here we shall describe only the measuring circuit and the elements directly connected with the measurements. In the figure, which represents a block diagram of the measuring elements, the notation is as follows: 1) source of molecular

beam, 2) time-datum sensor, 3) mechanical molecule-velocity chopper, 4) movable ionization-type beam receiver, 5) stator winding of chopper, 6) electronic measuring circuit, 7) electronic circuit for stabilizing chopper frequency, 8) oscilloscope for control of chopper frequency, 9) investigated material, in the form of a plate, 10) tuned amplifier, 11) power amplifier, 12) synchronous detector, 13) phase shifter, 14) recorder, 15) master oscillator, 16) relay, A), B), C) different positions of receiver during measurements. The plate (20 × 2 × 0.5 mm) was held in a movable holder and could be turned about its axis, which coincided with the axis of rotation of the receiver. The plate could be heated to temperatures of the order of a thousand degrees by an electron beam from an oxide cathode located behind it. Temperature was measured either by a pyrometer or by a thermocouple.

The chopper was operated at a number of rigorously controlled frequencies, which made it possible to vary the particle velocities of the monokinetic beam stepwise from 1100 to 1520 m/sec. A more detailed description of the chopper and the recording system can be found in [4, 5].

We shall now discuss the method of velocity measurement. Apart from the method based on the chopper, the velocity of the particles can also be measured by the time-of-flight method. Thus, by measuring the time of flight over the distances DA, DC, DB, DB' (see figure), one can determine the speeds of the incident and reflected particles, as well as their adsorption time at the surface. In fact, the difference between the times of flight over DA and DC is equal to the time of flight over the known distance AC (the speed calculated by this method should be the same as that determined by the chopper frequency, as indeed was the case in our experiments). In an analogous manner, the time difference between DB and DB' is the time of flight over the known distance BB', which determines the speed of the reflected particles.

If the measured time of flight over AB is not equal to the time calculated from the known speeds, the difference is the adsorption time of the molecules at the surface. In the absence of surface adsorption of molecules, the measurement of the time of flight over AB yields the velocity loss associated with the collision of the particles with the surface (accommodation coefficient). In the case of adsorption, the accommodation is, clearly, perfect. The time of flight was determined from the time lag of the ion-current pulses with respect to synchronous pulses of photocurrent, which served as time datum. For practical convenience, sequences of pulses were transformed into sinusoidal voltages, and the time lag was measured as a phase shift.

Time (phase shift) was measured by the null method, in which a reference phase shift (a figure) was generated by a special phase shifter and was fed into one input channel of the synchronous detector, which served as a null indicator. By using an inertia-type indicator (synchronous detector), it was possible to preserve high sensitivity even in the presence of fluctuating noise.

The use of the phase method with the transformation of a pulse sequence into sinusoidal voltage requires that the ion-current pulses generated by clusters of incident and reflected particles be constant in form. In the case of a uniform distribution of the speeds of the reflected particles, the pulse form is not distorted. In the other extreme case – Maxwellian distribution of velocities – one can calculate the additional phase shift.

We shall now discuss the main results obtained by shooting beams of Ar and CO<sub>2</sub> at technical-grade surfaces of Cu, Fe, and Ta. Measurements of the time of flight of the reflected particles showed that for all possible surface-gas combinations the measured speed corresponds to the mean thermal speed at the temperature of the surface. Thus, for a surface temperature of 300° K, the speed calculated from the time of flight was 430 m/sec for the Ar beam and 410 m/sec for the CO<sub>2</sub> beam. When the temperature was raised to 1100° K, the speeds were 820 and 780 m/sec, respectively. As the reflected speeds corresponding to different incident speeds differed by no more than 10% (the order of the experimental error), the above values represent the arithmetic means of reflected speeds corresponding to different incident speeds.

In order to be able to draw conclusions as to the energy transfer from speed measurement data, one must know the speed distribution of the reflected particles. Taking into account the fact that the incident beam is monokinetic, one can assume that the measured speed represents the mean speed of the reflected beam. An oblique corroboration is provided by the fact that the speed of the reflected particles was found to be independent of the angle of emission and that the total reflected particle flux (sum of the ion currents of beams reflected in all directions) was equal to the total incident particle flux (ion current of incident beam).

Thus, if one interprets the speed measurements in terms of an accommodation coefficient, one may assume that perfect thermal accommodation takes place. The results allow one to conclude that perfect thermal accommodation also took place at higher temperatures.

Comparing this result with the results of [6], obtained by the wire method, it is clear that in the latter case perfect accommodation was practically never observed. The cause of this discrepancy could be the considerably different vacuum conditions in the two sets of experiments.

In our second series of experiments the time of flight over DB was compared with the time calculated from the measured values of the incident and reflected particles. It was found that the former was significantly longer than the latter. This can be explained only by the adsorption of the molecules at the surface. Thus, the comparison allows one to conclude that the molecules are captured by the surface and to find (taking into account the effect of pulse distortion) the retardation time of the molecules on the surface.

This retardation time, which is also expressed as the arithmetic mean over particles with different initial speeds, is, in the case of Ar, 31 μsec on Cu, 41 μsec on Fe, and 49 μsec on Ta. In the case of CO<sub>2</sub>, these times were 36, 43, and 46 μsec, respectively. The measurements showed little effect of temperature on retardation time. Thus, at a temperature of 1100° K the times for Ar were 23 (Cu), 34 (Fe), and 44 μsec (Ta), and for CO<sub>2</sub> 23, 35, and 44 μsec, respectively. Using the measured retardation times and a known relation [6], one can find the adsorption energy. This was found to be 1.8 eV and practically independent of temperature.

In connection with the measurement of retardation time, it should be noted that the present method is applicable only if this time is shorter than the pulse period. Otherwise the retardation time would be indeterminate to a multiple of the period. This possibility should, however, be rejected, since the measured retardation time would then depend on chopper frequency, which was not observed in the experiments.

The author wishes to thank N. V. Kamyshev for his part in this research.

## REFERENCES

1. N. F. Ramsay, *Molecular Beams* [Russian translation], IL, 1960.
2. U. A. Arifov, *Interaction of Atomic Particles with Metal Surfaces* [in Russian], izd-vo AN UzSSR, Tashkent, 1961.
3. V. B. Leonas, "Study of the interaction of a molecular beam with a wall," *PMTF*, no. 6, p. 39, 1962.
4. V. B. Leonas and V. K. Rubtsov, "A chopper for the study of molecular speeds," *Pribery i tekhn. eksperim.*, no. 2, p. 115, 1960.

5. V. B. Leonas, "Detection of beams of neutral molecules," *Pribory i tekhn. eksperim.*, no. 3, p. 127, 1962.
6. J. H. De Boer, *The Dynamical Character of Adsorption* [Russian translation], IL, 1962.

20 May 1963

Moscow