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Oscillatory behavior of effective viscosity versus density for gases in the transition regime

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Oscillations of effective viscosity with density have been observed for helium, argon, nitrogen, and carbon dioxide in the transition regime. The experiments were performed with a torsion pendulum viscometer which has unequal distances between the moving surface and the two fixed surfaces. In general, two predominant oscillations were detected for each gas using Fourier analysis. From the mean positions of these two peaks we have calculated a molecular diameter using a simple relation between the wave lengths in density and the effective molecular cross section. Our values are in reasonable agreement with those measured by other methods for the four gases investigated.

In the study of transport phenomena in dilute gases one can distinguish three different regimes according to the ratio between the molecular mean free path, l, and a characteristic macroscopic dimension of the system, L. In the continuous regime ($l \ll L$) one can usually write down a local relation between a given flux and the gradient of the quantity which gives rise to such flux. Consider, for example, a dilute gas in which there exists a velocity flow in, say, the x direction. The momentum flux in the x direction across a surface perpendicular to the z direction, P_{zx}, is given by

$$P_{xz} = -\eta \frac{\partial u_x}{\partial z} , \tag{1}$$

where η is the coefficient of shear viscosity and u_x is the flow velocity.

In the transition regime the system size is of the order of a few mean free paths ($L \gtrsim l$). In this case one might expect non-negligible variations of the gradient over distances of the order of l. This in turn means that particles crossing a given element of area might have had their last collision at a point where the gradient has a different value than it has there. In other words, particles transporting momentum across such an area would have "sampled" different values of the velocity gradient, over a distance of the order of the mean free path. It is clear that this nonlocal character is not compatible with Eq. (1).

In the present Brief Report we report further experimental evidence of an oscillatory behavior of the effective viscosity as a function of the Kundsen number in the transition regime.[1] This effect seems difficult to describe in the framework of models using Eq. (1). On the other hand, we do not at present know whether the more elaborate models describing the transition regime of gases[2-9] can explain this effect. These models have, however, yielded accurate results of several transport properties.[2-7] Therefore it could be expected that these same models may be appropriate in our case provided that the boundary conditions considered reflect the complexity of the interaction between the gas and the walls.

We limit ourselves here to reporting experimental results for helium, argon, nitrogen, and carbon dioxide gases in the transition regime, and to calculating the corresponding molecular diameters using a simple relation between the wavelength in density Δn of the above-mentioned oscillations, and the effective molecular cross section σ,

$$\sigma = \frac{1}{\left[\dfrac{4}{3\pi^3}\right]^{1/2} \Delta n\, L} , \tag{2}$$

where L is the distance between the moving surface and one of the fixed surfaces. This relation arises in a rather crude model of the viscosity of gases in the transition regime, elaborated in previous papers.[8,9] Although this model does not explain all of the features of our experimental results, as expected given its simplicity, the molecular diameters calculated using Eq. (2) are in reasonable agreement with those determined by other methods[10] for the four gases investigated (see Table I). On the other hand, according to Eq. (2), a system with two characteristic macroscopic lengths L_1 and L_2 should have two oscillations of viscosity with density corresponding to different wavelengths in density Δn_1 and Δn_2. Fourier analysis of the data then provides an *unbiased* procedure for detecting the presence of such oscillations and verifying the approximate validity of Eq. (2).

Our measurements have been performed using a torsion pendulum viscometer. This viscometer consists of a cylindrical moving shell with a surface-mass ratio of 52.5 cm^2/g, enclosed and concentric with a stainless-steel cylinder 90 cm long and 51 cm in diameter. Enclosed within the moving shell there is a fixed stainless-steel tube 80 cm long and 5.14 cm in diameter. The moving shell is suspended from a quartz fiber 39 cm long. The external and internal fixed surfaces are separated from the moving shell by distances $L_1 = (18.60 \pm 0.10)$ and $L_2 = (3.98 \pm 0.10)$ cm, respectively. The period of the pendulum is measured by electronically timing ten periods using a laser beam reflected from a small mirror attached to the fiber.

The timing was started when the amplitude of the deflection was 45°. The period of the pendulum was large enough (~11 sec) to avoid turbulence. The range of pressures used for each gas was from 1 to 20 μm of Hg. The working temperature was (293.0 ± 0.2) K.

We calculated the ratio of the stress P_{zx} to the difference Δu between the velocities of the moving and fixed surfaces using the relation

$$\frac{P_{zx}}{\Delta u}=-\frac{4\pi M}{A}\frac{(T_i^2-T_0^2)^{1/2}}{T_iT_0}, \tag{3}$$

where T_0 is the period of the pendulum without gas friction and T_i is the period for a given gas pressure; M and A are the mass and the area of the moving surface, respectively.

The values of $P_{zx}/\Delta u$ as a function of pressure (μm Hg) have been calculated from the measured pendulum periods using Eq. (3) and taking for T_0 the minimum period measured in each experiment. We have proceeded in this way in order to minimize the effects on $P_{zx}/\Delta u$ due to changes in state of the quartz fiber from one exper-

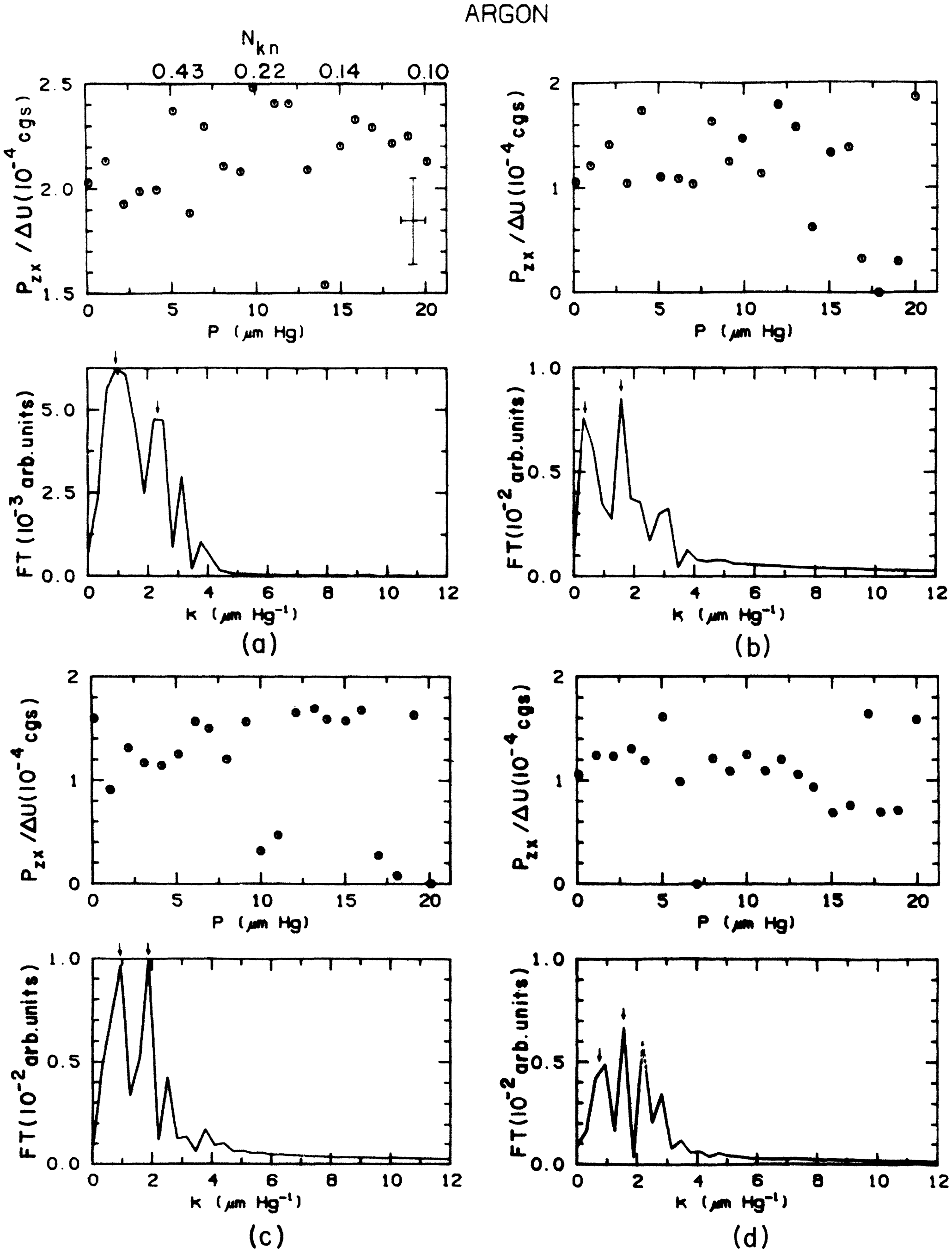

FIG. 1. $P_{zx}/\Delta u$ (in cgs units) for argon at (293.0 ± 0.2) K as a function of pressure for four experiments denoted (a), (b), (c), and (d), of 20 points each. A Knudsen-number (N_{kn}) scale is given above each figure. The error bars indicate the mean of the interexperimental errors at each pressure. Below each $P_{zx}/\Delta u$ plot is shown the corresponding Fourier-transform power spectrum (in arbitrary units). The small arrows indicate the "predominant" peaks chosen for the analysis.

iment to another. In Figs. 1 and 2, we show experimental results of argon and carbon dioxide gases, respectively. Four experiments, denoted (a), (b), (c), and (d) on these figures, are analyzed for each gas. The experimental results of helium and nitrogen show the same general features as those appearing in Figs. 1 and 2 (see Table I).

Shown below each of the plots of $P_{zx}/\Delta u$ versus pressure given in Figs. 1 and 2 is the corresponding Fourier-transform power spectrum. To obtain these, a straight line, with slope corresponding to the drift of $P_{zx}/\Delta u$ as a function of pressure, and with a mean value of zero over the interval 0–20 μm Hg, was subtracted from the $P_{zx}/\Delta u$ curve before performing the Fourier transform. This was done in order to remove the appearance of a spurious peak at low k. These Fourier transforms generally have more than the two peaks predicted by Eq. (2) and we have therefore had to choose two predominant peaks taking into account all of the four experiments for each gas. The peaks chosen are indicated with small arrows on the figures. In order to better separate the noise from our experiments we first analyzed each experiment using Fourier-transform analysis and then calculated the mean values and standard deviations of the wavelengths in density associated with each predominant peak, rather

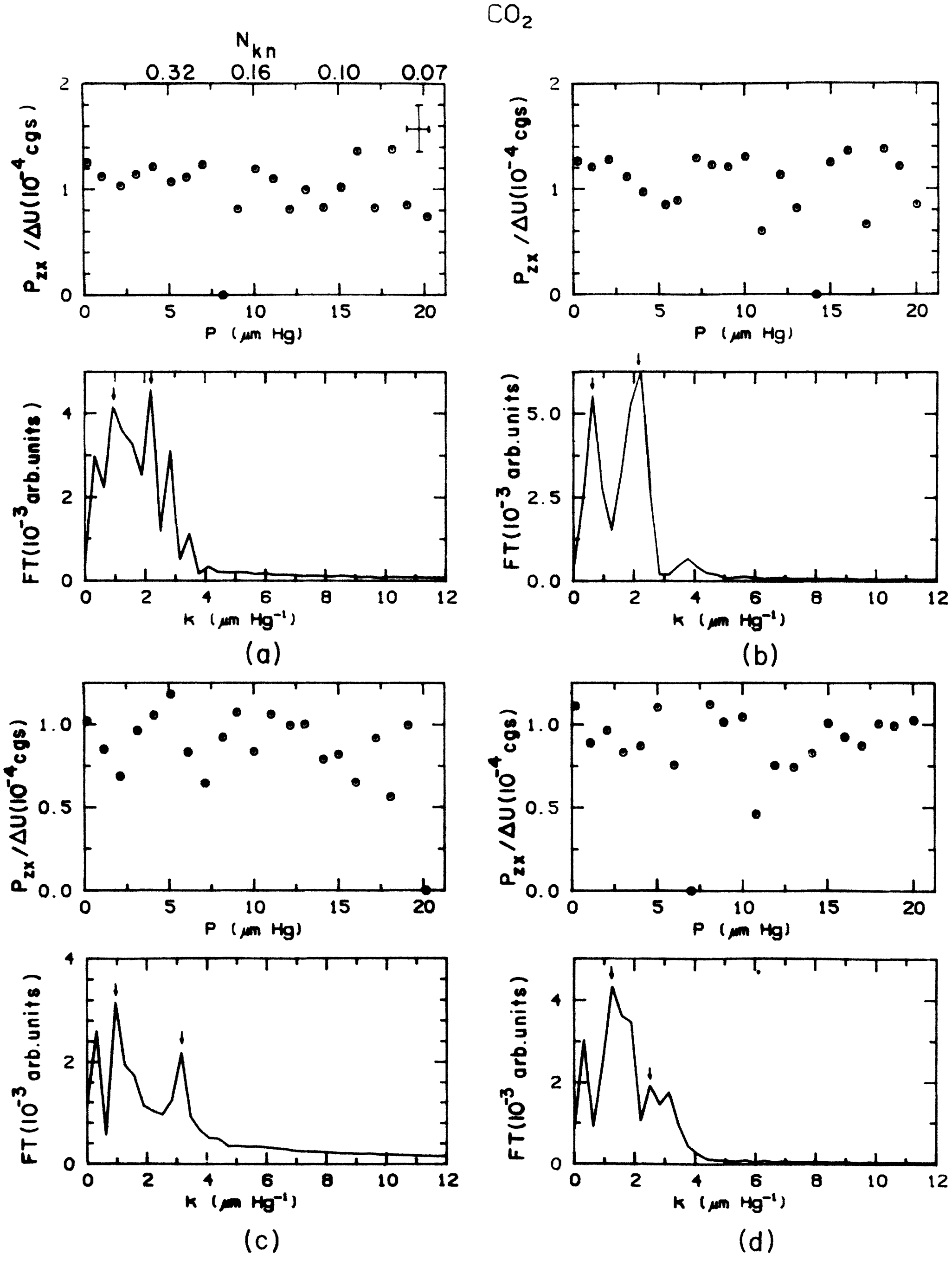

FIG. 2. $P_{zx}/\Delta u$ (in cgs units) for carbon dioxide at (293.0±0.2) K as a function of pressure for four experiments denoted (a), (b), (c), and (d), of 20 points each. The other features of the figure are the same as those of Fig. 1.

TABLE I. Wavelengths in density Δn and pressure Δp, molecular cross sections σ, and diameters d for He, Ar, N_2, and CO_2 at $T=(293.0\pm0.2)$ K, values corresponding to the two different system lengths L, are shown. Diameters obtained by viscosity in the continuous regime (Visc. cont.), van der Waals excluded volume (van der Waals), and heat conductivity (Heat cond.) measurements are included for comparison purposes (Ref. 10).

	L	Δn	Δp	σ	d	d (other methods) (10^{-8} cm)		
	(cm)	(10^{+14} cm^{-3})	(μm Hg)	(10^{-16} cm^{2})	(10^{-8} cm)	Visc. cont.	van der Waals	Heat cond.
He	18.6	1.64±0.39	4.99±1.18	15.7±3.7	2.24±0.51	1.90	2.65	2.30
	3.98	5.21±1.43	15.8±4.3	23.2±6.9	2.72±0.75			
Ar	18.6	1.10±0.15	3.33±0.47	23.6±3.3	2.74±0.37	2.88	2.94	2.86
	3.98	2.66±0.91	8.66±2.74	42.4±15.0	3.67±1.17			
N_2	18.6	1.07±0.17	3.24±0.53	24.3±3.9	2.78±0.44	3.15	3.15	3.53
	3.98	2.47±0.48	7.50±1.44	49.0±9.5	3.94±0.76			
CO_2	18.6	0.84±0.12	2.55±0.35	30.8±4.4	3.13±0.45	3.34	3.23	3.40
	3.98	2.33±0.60	7.08±1.82	52.0±13.4	4.06±1.05			

than doing a Fourier analysis of the mean value of $P_{zx}/\Delta u$ versus pressure for each gas. Nevertheless, we do give mean horizontal and vertical error bars in the upper right of the first plot of $P_{zx}/\Delta u$ versus pressure for each gas. These errors correspond to the mean of the set of errors obtained by considering the four experimental points at each pressure.

In Table I, we show the mean wavelengths in density Δn and pressure Δp and their standard deviations for each of the two peaks that were ascribed to each gas. From these values we calculated the molecular cross sections using Eq. (2) for each gas. The corresponding molecular diameters, $d \simeq (\sigma/\pi)^{1/2}$, are also given in Table I. For comparison we also give the molecular diameters that have been determined by other methods.[10] Our results can be seen to be in reasonable agreement with those obtained by the other methods. Differences are to be expected because these values correspond to different effective cross sections which are defined in terms of the different transport processes involved in each case. Perhaps more puzzling is the difference between the results corresponding to the two difference peaks. The molecular diameter determined by the smaller k value is always larger than that determined by the larger one. There are two factors that might account for this. (i) In our simple model we have not accounted for the effect of the curvature of the surfaces. These curvatures differ significantly and may affect the precise relationships between the obtained k values, the values of L, and the molecular diameters. (ii) The position of the peak with the smallest k value is more difficult to ascertain and, moreover, the finite size of the range of pressures investigated implies an uncertainty in the spectrum which becomes larger as k becomes smaller.

In summary we conclude that the observation of the peaks in the Fourier-transform power spectra and the agreement of the predicted molecular diameters with values obtained by other methods lends support to the previous observation of an oscillatory behavior of the effective viscosity of various gases as a function of density in the transition regime.

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