Speed Ratios Greater than 1000 and Temperatures Less than 1 mK in a Pulsed He Beam

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Speed ratios $v/\Delta v$ greater than 1000 and temperatures less than 1 mK have been achieved for the first time in a gas expansion into vacuum. The pulsed-beam source used in these experiments had an open time of 150 μsec, a nozzle diameter of 0.125 mm, and was operated with pure He gas at 294 K and pressures between 34 and 136 atm. Speed distributions for the collimated He beam were measured by time of flight. It is possible that such temperatures may be low enough to probe the low-energy limiting behavior predicted by Wigner in 1948 for the single-collision quantum dynamics of atoms and molecules.

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Several recent experiments[1-6] and related calculations[7-9] have suggested that the microscopic collision dynamics of atomic and molecular systems may exhibit fundamentally interesting and unique features in the limit of extremely low kinetic energies. Not only are quantum resonances[10] of several different types generally expected to become prominent at very low energies, but as the kinetic energy approaches zero the dynamics must become dominated by the fundamental quantum threshold singularities predicted by Wigner in 1948.[11,12] According to the Wigner threshold laws for a two-channel system not subject to Coulomb forces at large separations, the cross section for collisional relaxation of a molecular excited state becomes infinite in the *s* wave, zero collision velocity limit, while the corresponding microscopic rate coefficient becomes a finite constant. Although the threshold laws are thought to be rigorous, very little is known at present about their range of validity for heavy-particle systems. Two recent calculations,[8,9] both motivated by questions concerning the interpretation of vibrational relaxation data for the I_2+He system,[1,2,4] showed the threshold regime for a model of that system to be limited to temperatures less than 1 mK. The implications for molecular dynamics and spectroscopy experiments will be very important if it proves to be possible to access the Wigner threshold regime experimentally, since the threshold laws provide a ubiquitous mechanism for relaxation of gas-phase molecules to arbitrarily low temperatures. The question is whether such low temperatures or kinetic energies are accessible in the gas phase. As a first step toward answering that question, we report here the results from a novel approach to the generation of ultracold He beams by means of extreme supersonic expansions. The purpose of these experiments is to learn whether submillikelvin translational temperatures in the gas phase can actually be achieved and measured in a nominally pure beam of He, which is the most favorable case. If so, this would offer hope for our performing similar experiments with dilute mixtures of a second species in He.

The behavior of ^{4}He in an extreme supersonic expansion is unusual because the weakly attractive ^{4}He-^{4}He interaction can support at most one vibrational state, which must lie very close in energy to that of the separated stationary atoms. The question of whether that state is real (bound) or virtual (unbound) has provided a longstanding challenge to computational quantum theory, but has never been answered experimentally. The best evidence currently available comes from the fitting of model potential forms to a wide variety of scattering and transport data. A recent and comprehensive analysis by Aziz, McCourt, and Wong[13] yielded a positive binding energy of 1.684 mK, while a previous Aziz potential[14] analyzed by Uang and Stwalley[15] gave a value of 0.83 mK. Feltgen *et al.*[16] independently arrived at a value of 0.46 mK by means of a similar analysis. If the $^4He_2$ binding energy is actually this small, then previous attempts to observe the dimer in a supersonic expansion have been doomed by the failure to achieve sufficiently low temperatures. By contrast, larger clusters of ^{4}He have been observed in relatively mild supersonic expansions from nozzles near the ^{4}He boiling temperature.[17,18]

Whether the dimer state in question is slightly bound or slightly unbound, its existence has a profound influence on the low-energy elastic-scattering cross section. Effective range theory[16] predicts a scattering length which goes to $\pm\infty$ in the limit of low kinetic energies, as the binding energy approaches zero from the positive and negative directions, respectively. In either case, a nearly zero binding energy implies a very large and constant scattering cross section in the limit of zero kinetic energy. The value calculated by Uang and Stwalley was 1.9×10^{-11} cm^2, or about 20000 times the room-temperature gas-kinetic cross section. This resonance enhancement of the cross section is responsible for very efficient continued cooling of the expansion at very low temperatures and densities,[19,20] and for the very large distances from the nozzle which are required for extreme ^{4}He expansions to reach terminal conditions.[21,22]

The use of supersonic gas expansions to produce narrow speed distributions and low internal temperatures is widespread today in both scattering and spectroscopy experiments. However, the degree of cooling which is obtainable by this method tends to scale with the product of the source pressure P_0 and the nozzle diameter d, implying practical constraints due to mechanical features of the apparatus, most notably the pumping speed of the vacuum system.[20,21] These constraints have so far limited almost all such experiments to temperatures of about 1 K or higher. Two important exceptions were reported by Campargue,[19] and Toennies and co-workers,[20] who achieved translational temperatures down to 6–8 mK in steady-state beams of He before encountering pumping-speed limits. Our approach has been to eliminate the pumping-speed constraints altogether by using a fast *pulsed* source.[23] Because the length of the gas pulse (about 30 cm) is less than the dimensions of the vacuum chamber into which the gas is expanding, the pumps play no role until the gas pulse reaches the opposite wall of the expansion chamber and scatters randomly.

The pulsed-beam source in these experiments was mounted in one side of a vacuum chamber 1.22 m in diameter and 2 m long, joined to a long tube which provides a path for time-of-flight (TOF) analysis of the beam. The background pressure in the expansion chamber, maintained by a single 50-cm-diam oil diffusion pump, was 2×10^{-6} Torr. A skimmer[24] having an orifice diameter of 3.0 mm was located on the opposite side of the chamber at a distance of 1.12 m from the nozzle in one set of experiments. In a second set of experiments to examine the effect of varying the nozzle-skimmer distance, the skimmer was mounted on a temporary internal partition at a distance of 0.44 m from the nozzle. In both cases the beam was chopped at a distance of 1.86 m from the nozzle, further collimated with a second skimmer, and detected with two fast ionization gauges[23] at distances of 15.8 and 915.0 cm from the chopper. Because of the very high beam intensities in these experiments, a conventional slotted-disk chopper causes interference due to scattering of atoms in the transmitted part of the beam by other atoms reflected from the chopper surface. To circumvent this problem, we used instead a chopper which is the optical complement of the slotted disk, i.e., a single tooth (actually a string in some experiments) which scatters only a thin slice from an otherwise uninterrupted gas pulse. The TOF distribution for the resulting "hole" in the beam is then just the inverted TOF spectrum of the atoms removed by the chopper.

The pulsed-beam source was of a new high-pressure design in which a steel "hammer" is accelerated in a solenoid field for about 1 ms before it collides impulsively with a spring-loaded stainless-steel needle, which forms a metal-to-metal seal with the Cu-Be alloy nozzle. The shape of the nozzle was determined by a rather delicate empirical compromise between a shape which withstands the large needle forces required to produce a vacuum-tight seal and a shape which permits the aspect ratio (length/diameter) in the throat to be reasonably small. The nozzle entrance has a converging conical section of about 100° included angle and 0.15-mm depth, which merges with a cylindrical throat of aspect ratio about unity, which in turn merges with a diverging exit cone of about 120° included angle, extending over the remainder of the 1.5-mm total nozzle length. The entire profile of the nozzle is mechanically polished to achieve smoothly rounded contours and an optical finish. The throat diameter at the waist, determined by means of an optical comparator, was 0.125 mm. Under typical operating conditions the pulse duration was about 150 μsec, and both intensity and TOF measurements indicated full nozzle conductance and steady-state flow for at least 120 μsec in the center of the pulse. The source was operated at 294 K, with He gas of stated purity 99.995% and normal isotopic abundance. Our experiments covered a range in pressure between 34 and 136 atm, and in P_0d between 320 and 1290 Torr cm. The repetition rate of the experiment was 1 Hz, and the pumpout time constant of the vacuum system was 0.2 sec.

In Fig. 1, parts (a) and (b) show the chopper signals, i.e., the "holes" in the beam, measured at the two distances x from the chopper, for $P_0d = 970$ Torr cm, the

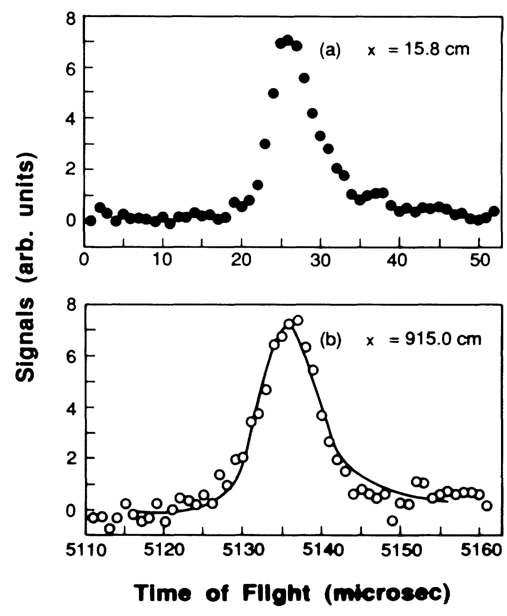

FIG. 1. TOF distributions measured at two distances x from the chopper. Both signals are actually negative with respect to those for the unchopped beam, as explained in the text, but have been inverted in these figures so as to show peaks in the conventional way. The solid line in part (b) is the best fit from convolving a Gaussian speed distribution with the signal shown in part (a).

conditions which resulted in the highest measured speed ratio in these experiments. Part (b) also show a curve obtained by convolution of the data from part (a) with a Gaussian speed distribution function $f(v) = C\exp[-(v-v_0)^2/a^2]$, in which a is adjusted to achieve a least-squares fit to the data in part (b). The weighted average of six such independent experiments and fits gives a parallel speed ratio $S = v_0/a = 1530 \pm 70$, and a parallel beam temperature[19-22] $T = ma^2/2k = 0.32 \pm 0.03$ mK. The Mach number[21,22] for this case is 1660 ± 80. The error estimates given are standard deviations from the fit statistics. It should be recognized that there are many sources of systematic error which can broaden the apparent TOF distribution and make the apparent value of S too small. Some of these, such as the finite detector spatial resolution (about 3 mm) and electronic rise time (< 1 μsec), we believe make negligible contributions. Others, however, such as skimmer or chopper perturbations of the speed distribution. we cannot estimate quantitatively. Although we have tried to minimize these effects, it is possible that S in a completely unperturbed beam would be significantly higher than these measurements indicate.

Brusdeylins *et al.*[20] found previously that deviations from a simple Gaussian speed distribution were observed for P_0d values around 100 Torr cm, which diminished for larger P_0d values. These deviations were attributed to nonuniform cooling in the expansion and could be fitted if they assumed a superposition of two distributions at different temperatures. We saw no evidence of such effects in our data, due possibly to the fact that small deviations from a single-temperature fit are not statistically significant, given the experimental signal-to-noise ratio and the extremely narrow speed distributions which are achieved. In view of the fact that an atomic beam is not an equilibrium state of matter, the values of S and T which we report should be interpreted as the best single-temperature representations of a speed distribution which might possibly be more complicated.

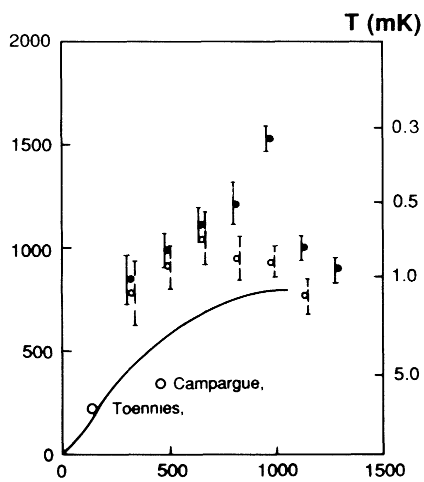

FIG. 2. Speed ratio S and parallel temperature T as functions of the P_0d product. The filled and open circles correspond to measurements with nozzle-skimmer distances of 1.12 and 0.44 m, respectively. For comparison, the largest S values reported by Campargue (Ref. 19) and Toennies and co-workers (Ref. 20) are also given, along with the theoretical predictions (solid line) of Toennies and Winkelmann (Ref. 20).

Figure 2 shows the dependence of S and T on P_0d for two nozzle-skimmer distances. Within experimental uncertainties, the results are independent of the nozzle-skimmer distance for small values of P_0d, indicating that the terminal expansion is reached without skimmer or background gas interference[21,22] under those conditions. At higher values of P_0d, S declines but reaches a higher limiting value for the larger nozzle-skimmer distance. Evidently the skimmer location is limiting the terminal speed distribution, implying that even higher speed ratios and lower beam temperatures might be obtained by making the expansion chamber larger. It seems likely that as P_0d increases and the transition from continuum to free particle flow[21,22] occurs at larger distances from the nozzle, a larger distance to the skimmer is required to avoid the reheating of the beam by small-angle scattering of the transmitted atoms by those reflected from the skimmer and bulkhead. Another possibility is that the maximum in S corresponds to the onset of clustering, which might release heat of condensation into the translational energy distribution. However, the formation of clusters requires *both* a sufficiently low beam temperature and a sufficiently high density for the occurrence of three-body collisions. In view of the fact that kT in these expansions barely reaches the range of the best available predictions for the $^4He_2$ binding energy, significant cluster formation seems unlikely.

Also shown in Fig. 2 are the values of S obtained by Campargue[19] and by Toennies and co-workers[20] at the upper limit of the P_0d ranges which they covered, and the results of the quantum calculation of Toennies and Winkelmann.[20] At the same P_0d values, our speed ratios are more than a factor of 2 higher than those obtained in the steady-state beam experiments, indicating that those results also are probably influenced by skimmer or background gas interference at the highest flow rates and background pressures. Interestingly, our values of S also exceed the theoretical values, probably as a result of inadequacies in the interaction potential assumed in the calculations, which must underestimate the scattering cross section at very low kinetic energies. As mentioned above, the scattering length for ^4He interactions is extremely sensitive to the absolute value of the dimer binding energy.

On the basis of these results, we conclude that the submillikelvin regime for gas-phase interactions is indeed accessible experimentally, at least within a nominally pure ^4He beam. Thus, there is reason to pursue the further question of whether similar temperatures may be achieved in dilute mixtures of a second species M with ^{4}He, which could provide a medium for the study of a wide variety of molecular relaxation processes in the extreme low-energy quantum limit.

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