

# Low-energy helium nozzle beam

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A high-intensity supersonic nozzle beam source is described, whose beam energy can be varied between 2 and 170 meV (helium). Experimental performance and time-of-flight measurements on cold helium beams are reported. Condensation in the free jet expansions of helium has been studied by mass spectrometric detection of  $\text{He}_2^+$ . A correlation between the maximum speed ratio and the onset of clustering is observed.

## INTRODUCTION

Atomic and molecular beam surface scattering has become a powerful tool in surface science. The rapid progress in the last decade is mainly based on the use of highly expanded nozzle beams with their high center-line intensity and narrow velocity distribution. In particular, the dynamics of inelastic gas-surface interaction is of increasing interest. In order to observe single phonon events, i.e., annihilation or creation processes, *light* particle beams of *low* energy should be preferred.<sup>1</sup> High monochromaticity and intensity are still prerequisites. A measure for the monochromaticity is the speed ratio  $S = \bar{v}(m/2k_B T_b)^{1/2}$ , where  $m$  is the mass,  $k_B$  the Boltzmann constant, and  $T_b$  the translational beam temperature, as measured by an observer moving with the mean flow velocity  $\bar{v}$  of the expanding gas. Thus, high monochromaticity is related to very low beam temperatures, in extreme cases as low as  $T_b = 10^{-3}$  K. Accordingly, even He cluster formation becomes probable; a limitation of the monochromaticity may be the consequence.

We have designed a nozzle source which can be used to produce helium beams in the energy range of 2 to 170 meV with speed ratios up to 180. Source design and time-of-flight measurements (TOF) with helium free jets (stagnation temperature  $T_0 = 13\text{--}300$  K) are reported. The limitation of the speed ratio due to the onset of condensation is discussed.

## I. EXPERIMENTAL

### A. Source design

Figure 1 shows a cross-sectional view of the source assembly which together with a differential pumping system is mounted to the time-of-flight machine described in Refs. 2 and 3. The source chamber (I) is evacuated by a  $1500\text{ s}^{-1}$  turbomolecular pump. The supersonic expansion is skimmed by two electroplated thin nickel skimmers<sup>4</sup> with 0.29 and 0.2-mm opening diameter, respectively, and double differentially pumped ( $150\text{ s}^{-1}$  turbomolecular pumps). The nozzle-first skimmer distance is 18 mm.

The nozzle (A) is a commercial electron microscope aperture disk made of platinum,<sup>5</sup> 2 mm in diameter and 0.25 mm thick. The aperture diameter (optical) is  $10\text{ }\mu\text{m}$ . Operation at high stagnation pressures imposes severe constraints to the mounting of the aperture disk at the end of the gas supply tube. The aperture disk is electron beam welded to a stainless-steel tube, 4 mm in diameter, 1.25-mm wall thickness, and 80-mm length (B). The design allows also fast temperature changes of the nozzle. The rear of the tube (B) is electron beam welded to a high-pressure tube (C), which can be attached to the gas supply system by means of a high-pressure fitting. The tube with the nozzle on its tip is plugged into a cylindrical copper block (D), outer diameter 12 mm, which is enclosed in a hard-soldered copper tube helix (E). For cooling, the helix can be supplied with liquid helium or liquid nitrogen. Between the coils of the helix a thermocoax heater element (F), specific resistance  $12.5\text{ }\Omega\text{m}^{-1}$ , is wound. The whole assembly is enclosed in a tight fitting gold-plated copper jacket (G). The evaporated helium (or nitrogen) gas is used for the cooling of the radiation shield (H), a chromium-plated thin-walled copper tube, which essentially reduces the radiation and convection to the cold elements. The temperature of the nozzle is stabilized to better than 1 K between 10 and 800 K by cooling and/or heating the copper block (D)

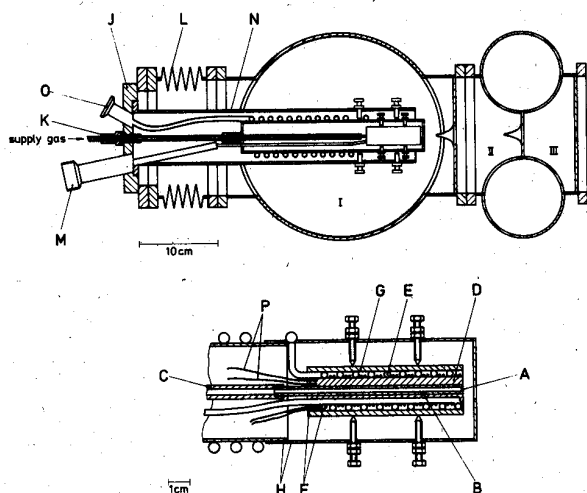


FIG. 1. Cross-sectional view of the source assembly mounted to the differential pumping unit. (A) nozzle, (B) nozzle tube, (C) high-pressure tube, (D) copper block, (E) copper tube helix for cooling, (F) thermocoax heater element, (G) copper jacket, (H) radiation shield, (J) end flange, (K) O-ring-type fitting, (L) alignment facility, (M) coolant feedthrough, (N) supporting tube, (O) waste gas feedthrough, (P) thermocouples.

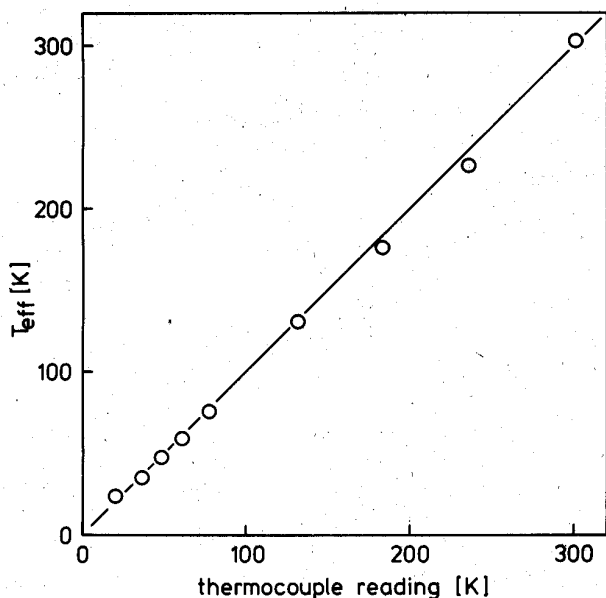


FIG. 2. Nozzle thermocouple calibration. Indicated thermocouple readings are plotted versus the effective stagnation temperature  $T_{\text{eff}}$  as determined from TOF analysis.

and monitored by an iron–constantan or gold/iron–chromel thermocouple.

The high pressure tube (C) is attached to a flange (J) of the vacuum system by means of a homemade O-ring type fitting (K). The end flange (J), which is mounted to the alignment facility (L), has also several feedthroughs for the thermocouples, electrical power, coolant liquid, and waste gas. The coolant feedthrough (M) enables the liquid–helium transfer from a standard storage container into the copper tube helix. A stainless-steel tube (N) for the radial support of the nozzle assembly is also attached to the end flange. This arrangement gives stable support of the nozzle tube while allowing axial movement due to thermal contraction or expansion of the different tubes upon cooling or heating.

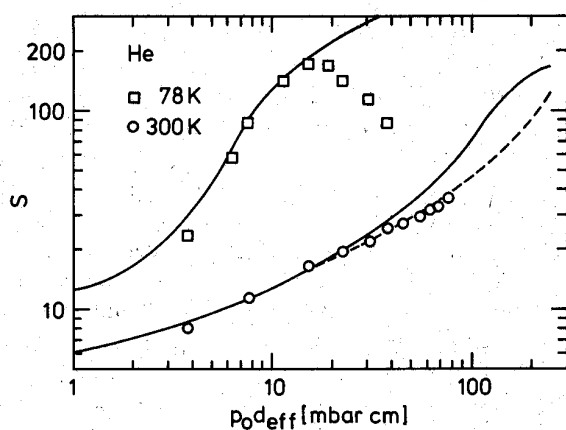


FIG. 3. Comparison between measured and calculated speed ratios  $S$  in helium expansions. The effective nozzle diameter is  $d_{\text{eff}} = 7.6 \mu\text{m}$ . The solid curves show the moments solution data of the Boltzmann equation given by Toennies and Winkelmann (Ref. 6). The dashed line shows the Monte-Carlo calculation of Chatwani (Ref. 7). Circles and squares are the data measured in the present experiment.

Great care has been taken to avoid nozzle clogging by impurities. Extra-high purity helium is used (99.9999 vol %). Also 10- and  $4\text{-}\mu\text{m}$  pore filters, cooled to liquid-nitrogen temperature, are mounted in the gas supply line.

## B. Working parameters

For a real gas the effective diameter of the nozzle throat is smaller than the optical diameter; a boundary layer is always present due to the viscosity of real gases. The effective diameter can be determined from total flow rate measurements; the value obtained here is  $d_{\text{eff}} = 7.6 \mu\text{m}$  for helium. No noticeable dependence of the effective diameter on the stagnation pressure was observed ( $5 \text{ bar} < p_0 < 30 \text{ bar}$ ,  $T_0 = 78$  and  $300 \text{ K}$ ).

The nozzle temperature was measured with great care. However, the temperature gradient along the nozzle tube, the radiative heating by surrounding walls, etc. may give rise to a difference between the measured nozzle temperature and the effective stagnation temperature which ultimately enters the beam parameters. In order to determine this difference, we compared the measured nozzle temperature with the effective stagnation temperature  $T_{\text{eff}}$  obtained from TOF analysis. The ideal expansion relation

$$\frac{m\bar{v}^2}{2} = \frac{\gamma}{(\gamma - 1)} k_B T_{\text{eff}}, \quad (1)$$

where  $m$  is the He mass and  $\gamma$  the ratio of specific heats, enables the evaluation of  $T_{\text{eff}}$  when the average velocity  $\bar{v}$  is known. Pseudorandom TOF measurement as described in detail in Refs. 2 and 3 is used (the flight path is increased here to  $790 \text{ mm}$ ). In Fig. 2,  $T_{\text{eff}}$  values as determined from TOF data via Eq. (1) are plotted versus the nozzle temperature obtained from thermocouple readings. It appears that within the experimental errors of the present experiment the thermocouple reading of the nozzle temperature coincides with the effective stagnation temperature.

## II. BEAM CHARACTERIZATION

The beam monochromaticity is the result of the large number of collisions in the hydrodynamic expansion of a gas: the larger the number of collisions, the higher the speed ratio  $S$ . Accordingly, high monochromaticity is obtained by using high values of  $p_0 d_{\text{eff}}$  and/or low stagnation temperatures. The latter increases the number of collisions because, at low relative velocities, the collision cross section of atoms increases. Due to quantum effects this increase is particularly pronounced for helium. In Fig. 3 speed ratio values are plotted versus  $p_0 d_{\text{eff}}$  at two stagnation temperatures ( $T_0 = 300$  and  $78 \text{ K}$ ). The data were obtained for He with the beam system described above. The full lines are theoretical dependencies due to Toennies and Winkelmann<sup>6</sup> who solved the Boltzmann equation by a moments solution. The dashed line corresponds to the Monte-Carlo solution obtained by Chatwani<sup>7</sup> by using the same collision cross sections as in Ref. 6. At  $300 \text{ K}$ , the data plots favor the Monte-Carlo solution, while the earlier data of Brusdeylins *et al.*<sup>8</sup> rather meet the moments solution. At  $78 \text{ K}$ , our data coincide with the earlier ones<sup>8</sup>: Below  $p_0 d_{\text{eff}} \approx 12 \text{ mbar cm}$  they follow the theoretical curve while above a marked deviation is obvious;

the stagnation conditions for which the speed ratio collapses. They were deduced from curves like those in Fig. 5. In all cases the stagnation conditions correspond to a dimer ion fraction of about  $2 \times 10^{-4}$  (at a background signal of about  $4 \times 10^{-5}$ ). This shows directly that the appearance of condensation limits the speed ratio increase. A cluster concentration corresponding to this dimer ion fraction appears to heat up the beam sufficiently to cause a breakdown of the speed ratio.

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<sup>9</sup>P. W. Stephens and J. G. King, *Phys. Rev. Lett.* **51**, 1538 (1983).

<sup>10</sup>The question of the existence of a bound van der Waals molecule  $\text{He}_2$  has long since been discussed. Recent results favor a very weak bound state with  $\epsilon = -4.3 \times 10^{-4}$  K (Ref. 11), respectively  $\epsilon = -8.3 \times 10^{-4}$  K (Ref. 12). However, no mass spectrometric evidence is found that the  $\text{He}_2^+$  ion originates from a helium dimer. Regardless of that, the  $\text{He}_2^+$  formed by fragmentation, can be used as an indicator for the abundant cluster species formed in the expansion.

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