PHYSICAL SCIENCES

Molecular beam brightening by shock-wave suppression

Yair Segev,* Natan Bibelnik, Nitzan Akerman, Yuval Shagam, Alon Luski, Michael Karpov, Julia Narevicius, Edvardas Narevicius*

Supersonic beams are a prevalent source of cold molecules used in the study of chemical reactions, atom interferometry, gas-surface interactions, precision spectroscopy, molecular cooling, and more. The triumph of this method emanates from the high densities produced in relation to other methods; however, beam density remains fundamentally limited by interference with shock waves reflected from collimating surfaces. We show experimentally that this shock interaction can be reduced or even eliminated by cryocooling the interacting surface. An increase of nearly an order of magnitude in beam density was measured at the lowest surface temperature, with no further fundamental limitation reached. Visualization of the shock waves by plasma discharge and reproduction with direct simulation Monte Carlo calculations both indicate that the suppression of the shock structure is partially caused by lowering the momentum flux of reflected particles and significantly enhanced by the adsorption of particles to the surface. We observe that the scaling of beam density with source pressure is recovered, paving the way to order-of-magnitude brighter, cold molecular beams.

INTRODUCTION

Atomic and molecular beams have enabled groundbreaking experiments in physics and chemistry for almost a century. Pioneering applications of these methods, such as the Stern-Gerlach experiment (1), and later the maser (2), used effusive beams. These beams were inherently limited to low densities and relatively high temperatures, as particles would leave the source in free-molecular flow. Supersonic nozzles (3) introduced markedly denser ("brighter") beams of significantly colder particles, leading to major advances in various fields such as reaction studies (4), nanomaterials (5), and atom interferometry (6). More recently, supersonic beams have been applied as sources for decelerators (7, 8) used for novel methods of molecular cooling (9), in the study of quantum phenomena in cold collisions (10-12), and for molecular interferometry (13).

The success of supersonic beams stemmed largely from the high densities they offered; however, continuous (CW) flow experiments often remained limited by maintainable background pressure because they required high pumping speeds (14). Advances in pulsed nozzles have effectively removed these restrictions (15), enabling the use of source pressures up to three orders of magnitude higher than with a CW beam. However, beam brightness seems only to have reached a new limit, induced by interference of the beam with necessary collimating diaphragms known as skimmers (16). Sufficiently high densities lead to "clogging," an interaction of the flow through the skimmer with particles reflected from the skimmer walls that manifests as shock waves (17) and limits the beam flux (18). Thus, to date, common practice with pulsed beams remains to limit the beam brightness by either restricting source pressure, distancing the nozzle from the skimmer, or both (16).

Here, we demonstrate that the shock-wave interaction with the skimmer and the associated clogging can be significantly reduced or even eliminated by cryocooling the skimmer. We trace the elimination of clogging to two effects, each of which lowers the momentum flux reflected from the surface. Cooling the surface reduces the thermal velocity 2017 © The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. Distributed under a Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC).

of reflected particles and decreases their mean free path into the undisturbed beam. Still lower temperatures lead to adsorption of the impacting particles to the skimmer. We observe that the suppression of shock waves by reduction of reflected momentum is possible because the shock propagation in a rarefied medium is governed by direct collisions between the beam and the reflected particles. This mechanism is in stark contrast with the well-studied propagation of shocks through a supersonic fluid in the continuum regime. We measure a resulting increase of a factor of 8 in beam flux, with no further fundamental limitation reached in our experiment.

Formation of shocks in supersonic beams

Supersonic beams are generated by releasing gas through a nozzle into a vacuum. Flow emanating from the stagnant source into the lowpressure volume expands due to collisions between flow particles, leading to narrowing of the velocity spread from the source's thermal distribution to a low-temperature, high-velocity beam. At any point in the early expansion region, the flow conditions-velocity magnitude, density, and temperature-are connected through the isentropic relations to the local Mach number and the stagnation conditions (19). The free-jet shock structures present in most continuous beams vanish at the sufficiently low background pressures commonly obtained with low duty-cycle pulsed valves (14), and the isentropic expansion continues until the distance between collisions, as measured in the laboratory frame, is non-negligible. At this point, typically occurring at distances of hundreds of nozzle throat diameters, the flow gradually "freezes" (20)-the temperature gradient eases while the density continues to decrease as the square of the distance from the nozzle. For most applications, it is advantageous to conduct experiments as early as possible in this rarefied region, making use of the maximum available density. However, supersonic expansion creates a broad plume of gas, so the axial part of the beam must first be "skimmed" from the plume to produce a collimated jet in the test chamber.

Typical skimmers are straight or flared (21) cones, or two-dimensional slits (22), with the common intention of minimizing the interaction of the centerline region of the beam with particles reflected from either the inner surface into the skimmer or the outer surface into the oncoming plume. If the beam is sufficiently bright, then both reflection directions will manifest as shock waves (17)—abrupt disruptions in the supersonic flow that introduce entropy and heat from the walls. In the interior of the skimmer, the

Department of Chemical Physics, Weizmann Institute of Science, Rehovot 7610001, Israel.

^{*}Corresponding author. Email: yair.segev@weizmann.ac.il (Y.S.); edn@weizmann. ac.il (E.N.)



Fig. 1. Skimmer clogging due to shock-wave interference. A cold supersonic plume is generated by expansion from a pulsed valve into a vacuum. A skimmer transmits the axial portion of the plume as a collimated beam into the detection chamber. With high beam densities, shock waves are reflected from the skimmer lip, interfering with the transmitted beam. In imaging experiments only, a discharge pulse from a high-voltage grid induces plasma glow in the denser portions of the flow, revealing the shock waves. A large split skimmer is used here for the observation of the internal shock structure.

shock waves from different points on the lip may combine to clog the skimmer, as demonstrated in Fig. 1. These shocks slow and heat the beam before a second expansion process follows downstream. The resulting jet has different terminal properties (23), because the stagnation temperature of the beam is changed by the energy introduced from the surfaces. Common practice is to avoid this "skimmer interference" by restricting the density at the skimmer entrance, either by increasing the distance between the nozzle and the skimmer or by decreasing the source pressure. In this manner, skimmer interference is often the limiting factor in creating bright, cold beams.

To reduce the effect of skimmer interference without restricting the density, we propose to cool the skimmer wall. The use of a cooled skimmer was previously tested with a CW beam at the Arnold Engineering Development Complex (24-27); however, reaching sufficient source pressures for skimmer interference with a continuous source required enormous pumping speeds in excess of 500,000 liter/s and quickly led to filling of the skimmer orifice with frozen particles (24).

RESULTS AND DISCUSSION

The experimental setup consists of a source chamber, as shown in Fig. 1, and a detection chamber. The beam source is a pulsed Even-Lavie valve (28, 29) with variable stagnation pressure P_0 and temperature. The



Fig. 2. Measured beam densities of different species for varying values of skimmer temperature T_w . (A to D) Cooling the skimmer initially increases the peak density until a limit is achieved at the unclogging temperature. Time is measured from the trigger to the valve. The skimmer is conical. The source conditions are set to achieve clogging for a hot skimmer. Additional parameters are listed in Materials and Methods. a.u., arbitrary units.

skimmer separating the chambers is held at a temperature of $T_{\rm w}$ at a variable distance $r_{\rm ns}$ from the nozzle. A residual gas analyzer is used to measure the density of the beam in the detection chamber.

We define a case as "unclogged" if an increase in source pressure by a factor of k leads to a similar increase in peak density and "clogged" if the increase is significantly smaller than k. Measurements of clogged beams at varying T_{w} , as shown in Fig. 2, reveal a sensitivity of the density to the skimmer temperature for different gases from constant source conditions. At high surface temperatures, the measured beams exhibit a sharp peak, followed by a dip and a long "tail" lasting tens of milliseconds. As the

temperature of the skimmer is lowered, the first peak initially increases, whereas the tail gradually diminishes, with increasing sensitivity at lower temperatures. Below a certain temperature, the density profile stops changing. Concurrently, we observe that the peak density becomes sensitive to source pressure, so the skimmer is now effectively unclogged. This temperature, termed the unclogging temperature $T_{\rm u}$, varies with species—it is approximately 60 K for krypton, 40 K for argon and molecular oxygen, and 20 K for neon.

The variation in beam density corresponds to a change in the flow structure through and around the skimmer. To view the flow phenomena



Fig. 3. Visualization of the density field using a pulsed discharge. (A to I) Shock waves enveloping the skimmer recline and eventually vanish as the skimmer temperature is lowered. The internal shock structure exhibits similar phenomena. For each case, the species is indicated in the lower left corner, and skimmer temperature is indicated in the lower right corner. The source conditions are set to achieve clogging for a hot skimmer. Additional parameters are listed in Materials and Methods.



Fig. 4. External shock-wave angle as a function of skimmer temperature. The shock angle, defined on the right over an image with a helium beam, initially decreases gradually with the temperature. Near the unclogging temperature, the sensitivity increases until the shock becomes parallel to the wall and vanishes. Dashed lines serve as guides to the eye. The angle is defined relative to the line of maximum intensity within the shock. The source conditions are set to achieve clogging for a hot skimmer. Additional parameters are listed in Materials and Methods.

responsible for this, we add a pulsed-discharge grid in front of the valve. The plasma glow from the denser portions of the flow reveals the oblique shock waves that envelop the skimmer (Fig. 3). At high temperatures (Fig. 3, A and B), the angle between the surface and the shock is far larger than the corresponding shock angle in flows of similar Mach number but lower Knudsen number. This indicates that the disturbance to the beam propagates directly by collisions with reflected particles, even at distances comparable with the skimmer dimensions. This effect is a result of the large mean free path of particles reflected from the surface into a rarefied flow, in contrast with the typical shock waves of the continuum regime, which propagate by collisions between particles of the medium. At lower surface temperatures, the shock angle significantly decreases (Fig. 3, D and E), approaching the characteristic acute angles expected at high Mach numbers. Near or below T_{uv} the shock is almost parallel to the skimmer and may even vanish entirely (Fig. 3, G and H). In this regime, condensation of frozen beam particles gradually appears on the skimmer over time.

The variation of the external shock angle with skimmer temperature (Fig. 4) shows a general trend similar to that of the density profile, with increasing sensitivity as the temperature decreases to $T_{\rm u}$. This correlation derives from a near mirroring of the external shock that occurs in the flow within the skimmer. Using a "split" skimmer, we observe oblique shock waves emanating from the lip and intersecting at the centerline (Fig. 3C), the flow structure identified with skimmer clogging (17). As we lower the skimmer temperature, the high-density region behind the shock fronts becomes less intense and more elongated, and the shocks recline (Fig. 3F). This structure eventually vanishes below $T_{\rm w}$ (Fig. 3I).

Further insight into the mechanism of skimmer interference and its dependence on temperature emerges from numerical simulation of the flow using the direct simulation Monte Carlo (DSMC) method. We use the DS2V code (30) with unsteady sampling, species-dependent parameters from Bird (31), and beam parameters estimated from analytical and semiempirical expressions for different stagnation conditions (19, 32). Reflections from the skimmer surface are defined as fully diffuse at temperature $T_{\rm w}$, initially assuming no adsorption. The calculated flow exhibits shock waves for modified Knudsen numbers (*33*) of the order of unity and less. Figure 5 presents such cases for a krypton beam impacting "hot" (Fig. 5A) and "cold" (Fig. 5B) conical skimmers. With decreasing skimmer temperature, the shock waves recline, and the internal shock structure elongates, qualitatively reproducing the experimentally observed trend. In the simulation, the mechanism behind this trend can only derive from the change in the thermal velocity of reflected particles, with lower temperature leading to lower mean free path of these particles and more acute shock angles.

The transmitted beam in the simulated skimmers presents a relatively dense "bullet," followed by a hot, sparser "cloud" of expanding gas. The bullet corresponds to the portion of the beam that passes the skimmer entrance before the shock structure has completely formed, whereas the cloud corresponds to the reexpansion of a jet behind a strong shock. The intensity and length of the bullet increases as T_w decreases, whereas the total flux emitted later from the cloud decreases. Thus, for a pulsed beam sampled in time at a constant point downstream of the shock structure, as in Fig. 5C, the density qualitatively matches the peaks and tails measured experimentally and presented in Fig. 2.

The duration of the transmitted portion of the beam peak is limited by the shock formation time, $t_s \approx D_s/2v_t$, where v_t is the thermal velocity of the gas arriving from a skimmer lip held at temperature T_w to the centerline. This expression holds as long as the mean free path of reflected particles and the skimmer radius are of the same order of magnitude. The measured rise times of the transmitted peaks roughly match the corresponding values of t_s , decreasing with lower T_w and lighter species, as evident in Fig. 2. The skimmer transmits higher peak densities when t_s is a sufficiently large fraction of the full-pulse rise time. Because t_s scales with the square root of molecular mass, the shock formation times at the lowest temperatures available in the current experiment are sufficiently long to observe the onset of unclogging in beams of neon, argon, molecular oxygen, and krypton, but unclogging a helium beam requires even lower temperatures. Note that an additional consequence of a finite clogging time is that, when skimmer clogging occurs, only the



Fig. 5. DSMC calculations of a krypton beam impacting a conical skimmer. The density field is presented in logarithmic scale for cases of a hot ($T_w = 300$ K) (**A**) and a cold ($T_w = 30$ K) (**B**) skimmer. The transmitted beam consists of a dense bullet followed by a sparse cloud corresponding to the peaks and tails in experimentally measured beams. Changes in the shock structures with skimmer temperature qualitatively match the experimental observations above the unclogging temperature. (**C**) The centerline density 30 mm downstream of the skimmer entrance also exhibits a beam intensification trend at lower temperatures, but only the addition of significant adsorption to the surfaces enables complete unclogging. Additional parameters are listed in Materials and Methods.



Fig. 6. Peak measured density for a neon beam transmitted through hot ($T_w = 300$ K) and cold ($T_w = 13$ K) conical skimmers with varying beam density. The density at the skimmer entrance is controlled either by varying the source pressure (**A**) at a constant nozzle-skimmer distance ($r_{ns} = 45$ mm) or by varying this distance (**B**) at a constant stagnation pressure ($P_0 = 40$ atm). Increasing the density by either method eventually clogs the hot skimmer, whereas the cold skimmer transmits a continuously increasing peak density with no observed limit. The source temperature is 300 K. Dotted lines serve as guides to the eye.

faster portion of a pulsed beam is transmitted without interference. This effect is detrimental for applications where a lower mean beam velocity is beneficial, such as decelerators.

The mechanism of decreased thermal velocity alone fails to predict the existence of a complete unclogging temperature because the transmitted pulse duration is far longer than t_s at the values of T_u relevant for each species. Furthermore, the rapid reduction in shock angle as T_w approaches T_w followed by vanishing of the shock, significantly exceeds the numerically predicted effect. It appears that the onset of significant adsorption enhances the reduction in reflected momentum and explains the gradual appearance of condensation on the skimmer at these temperatures. Adding sufficient adsorption fractions to the surfaces in the DSMC calculations also reproduces unclogged skimmers (Fig. 5C). We conclude that, at sufficiently low temperatures, skimmer clogging is relieved by condensation of many or most of the impacting particles.

To demonstrate that beam transmission with a cold skimmer is no longer limited by density, we present the peak measured density with increasing source pressure (Fig. 6A) and with decreasing nozzle-skimmer distance (Fig. 6B). At either large distances or low source pressures, a room temperature skimmer and a skimmer cooled below T_u identically transmit the beam. However, larger densities lead to the clogging of the hot skimmer, causing a leveling off of the density. In contrast, the peak transmitted density with a cold skimmer continues to rise without any observed limit. Our measurements have found an increase of a factor of 8 with neon from a maximum source pressure of 85 atm and a minimum r_{ns} of 28 mm. We expect that significantly higher densities can yet be achieved with higher source pressures.

Summary and outlook

We have demonstrated the successful suppression of the shock waves that frequently interfere with molecular beam experiments. By cryocooling the collimating surfaces, the flux of reflected momentum causing this interference was reduced, resulting in the revival of density scaling with source pressure. An increase in density by a factor of 8 was measured, with no new fundamental limit reached. The removal of this shockwave interference lifts the long-standing limitation on the brightness of molecular beams.

Boosting the beam brightness by an order of magnitude is beneficial for a wide variety of experiments. Higher densities can provide increased

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sensitivity in atom and molecular interferometry. In the case of bimolecular collisions, the collision rates will be increased by two orders of magnitude, assisting in the study of slow processes that proceed by tunneling. Finally, attaining higher beam density may bring new and general molecular cooling methods based on collisions, such as sympathetic or evaporative cooling, within reach.

MATERIALS AND METHODS

Experimental setup

The beam source was an Even-Lavie valve (28, 29) with a nozzle diameter of 0.2 mm. The skimmer was attached to a 10 K cryostat, and its temperature was monitored with a diode and maintained with a heating resistor. In some of the experiments, the valve was attached to a second cryostat to vary the stagnation temperature, whereas, in others, it was attached to a manipulator, controlling the nozzle-skimmer distance. The skimmer, made of oxygen-free high-thermal conductivity copper, is a cone with a tapered wall—2 mm thick near the base for sufficient heat conduction, and sharpening toward the tip. The entry diameter of this skimmer is 4 mm. A residual gas analyzer was used to measure the density of the beam 215 mm downstream of the skimmer entrance in the detection chamber. Additional data regarding the setup and skimmer are available in the Supplementary Materials.

Numerical calculations

For rarefied flow calculations, we used the DS2V code (30) with unsteady sampling, species-dependent parameters from Bird (31), and beam parameters estimated from analytical and semiempirical expressions for different stagnation conditions (19, 32). Reflections from the skimmer surface were defined as fully diffuse at temperature T_w . The fraction of impacting particles adsorbed to the surface was set to 0 or 1, as noted.

Additional data for figures

The parameters of the experiment were set to achieve clogging when the skimmer was at room temperature. For the measurements presented in Fig. 2, the source temperature was 300 K, and the source pressure and nozzle-skimmer distance in each panel were as follows: 15 atm, 45 mm (Fig. 2A); 45 atm, 28 mm (Fig. 2B); 80 atm, 25 mm (Fig. 2C); 60 atm,

28 mm (Fig. 2D). For the measurements presented in Figs. 3 and 4, the source temperature was 300 K, the nozzle-skimmer distance was 45 mm, and the source pressure for each species was 25 atm (Ne), 45 atm (Kr), and 35 atm (Ar).

For the calculations presented in Fig. 5, the free-stream flow temperature was 0.1 K, the particle density was 5×10^{15} cm⁻³, and the velocity was 385 m/s. For Fig. 5 (A and B), the elapsed time was 90 µs from the time of beam impact at the skimmer tip. For Fig. 5C, the total pulse duration was 70 µs.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/ content/full/3/3/e1602258/DC1

fig. S1. Experimental setup for measuring the effect of skimmer temperature on the transmitted beam.

fig. S2. Schematics of the conical skimmer used in the quantitative experiments.

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