Thermal bending of nanojets: Molecular dynamics simulations of an asymmetrically heated nozzle

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(Received 8 July 2008; accepted 1 September 2008; published online 25 September 2008)

A method for deflecting high velocity liquid nanojets through nonuniform asymmetric heating of a cylindrical nozzle is described using molecular dynamics simulations of liquid propane. The deflection originates mainly from a strong, highly nonisotropic evaporation flux from the liquid near the exit, resulting in overall cooling and a force that bends the nanojet in the lower temperature direction. Also discussed is the formation of a self-convergent virtual nozzle in the injector-confined liquid with an exit radius smaller than that of the injector, portraying the temperature and density profiles in the heated liquid and resulting in a nanojet of reduced dimensions. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2988282]

Fundamental and applied research interests pertaining to the nature of fluids and flow at highly reduced dimensions and potential technological applications of such systems in the printing of miniaturized circuitry, nanoscale surface patterning, nanofluidics, and biomedical applications motivated recent efforts directed at understanding the properties of liquid jets of nanometer-scale dimensions [that is, nanojets (NJs)] and at developing methods for their generation and manipulation.[1–6] Early molecular dynamics (MD) simulations[1] have demonstrated the formation of liquid (propane) NJs, generated by the application of high back-pressures and convergent nozzles. To alleviate certain difficulties pertaining to the fabrication of nozzles of limiting miniaturized dimensions, we introduced most recently[6] the concept of a "virtual NJ nozzle," where NJs are generated via the flow of a fluid (propane) through a heated (straight) cylindrical injector nozzle; see the profiles of the density and temperature distributions in Fig. 1 and an atomistic view [Fig. 2(a)] obtained from MD simulations[7] of a NJ with a 2 nm radius, generated by a heated cylindrical nozzle (length $L=30$ nm, inner radius $R=6$ nm, wall temperature $T_W=500$ K, driving back-pressure of 220 MPa, and the temperature and velocity of the liquid entering the heated nozzle section being 150 K and 250 m/s, respectively), with a propagation velocity of 700 m/s and a breakup length $L_b>500$ nm (L_b is the jet length before breakup into droplets).

Here we demonstrate through the use of MD simulations[7] that employment of asymmetrical (nonuniform) heating of the injector walls allows bending (deflection) of the NJ, and we show that the "vectorization" of the NJ originates from nonisotropic strong evaporation from the emanating heated fluid in the vicinity of the exit from the injector, which cools and self-narrows the NJ, as well as bends its propagation direction. Previous work about the thermal bending[4,5] of jets employed asymmetric heating of the nozzle and attributed[4] the bending to temperature dependencies of the fluid's properties (surface tension and viscosity).

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Recent MD and hydrodynamic simulations have illustrated that by heating the nozzle walls one can create a temperature gradient between the walls and the central core of the liquid in the injector.[6] The temperature profile in the liquid varies also along the axial direction (z), and this is reflected in the temperature and density profiles, which exhibit (inside the injector, $z\leqslant 0$) a (self) converging shape toward the exit [located at $z=0$, see contours in Figs. 1(a) and 1(b) and profiles in Figs. 1(c) and 1(d)]; density contours with $\rho \geqslant 0.4\times 10^3$ kg/m^3 correspond to a liquid, and those with a lower density represent surrounding vapor molecules. The self-convergence of the liquid flow inside the cylindrical (straight) injector increases significantly the flow velocity in the convergent region, in the same way that a geometrically

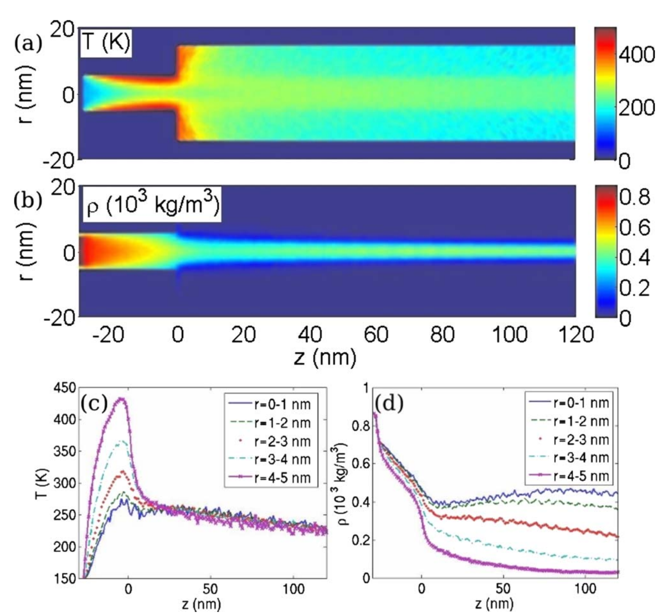

FIG. 1. (Color online) Steady state temperature and density contours of a liquid propane NJ injected by a uniformly heated nozzle, $T_W=500$ K, with $R=6$ nm; contours obtained from averages in annular rings of 1 nm width and thickness. Temperature and density scales corresponding to different shades are given on the right. Averaged (c) temperature and (d) density profiles. Different line types correspond to different annular shells, spanning radial ranges as indicated.

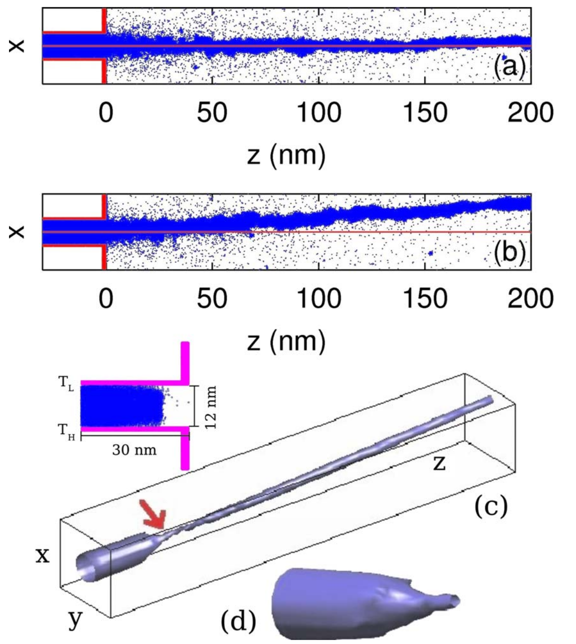

FIG. 2. (Color online) Atomic configurations of steady state flow of a NJ generated by (a) a uniformly heated nozzle (see also Fig. 1) and (b) a deflected NJ generated by the same nozzle but heated nonuniformly. Inset: two-dimensional schematic of the injector nozzle. (c) Density contour surface at $\rho=0.4\times10^3$ kg/m^3 of the deflected NJ; box dimensions: $30\times30\times190$ nm^3. (d) Enlargement of the contour surface in (c) focusing on the part inside the nozzle and demonstrating the formation of an asymmetric VN.

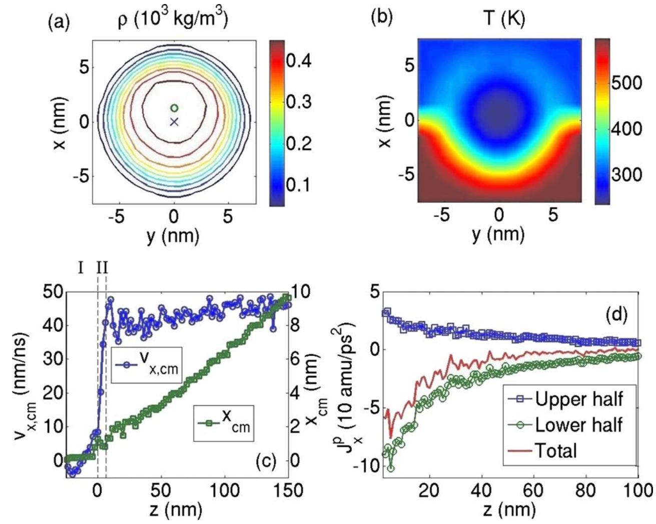

FIG. 3. (Color online) Properties of the deflected NJ described in Figs. 2(b) and 2(c). (a) Contours of the density on the transverse (xy) exit plane of the nozzle (at $z=0$). The density unit is 10^3 kg/m^3. The geometrical center of the nozzle and the center of gravity of the confined fluid are denoted by an "x" and a circle, respectively. (b) Temperature contours at the nozzle exit. The density and temperature scales are on the right of the figures. (c) x-component of the center-of-mass (c.m.) velocity, $v_{x,\mathrm{c.m.}}$ (circles, blue online, left axis), and the x-component of the c.m. of the NJ, $x_{\mathrm{c.m.}}$ (squares, green online, right axis). Region I is inside the nozzle (nozzle exit at $z=0$), and region II is the 4 nm interval where an abrupt increase in $v_{x,\mathrm{c.m.}}$ occurs. (d) x-component of the momentum flux per unit length (nm), $j^p{}_x$, resulting from nonisotropic molecular evaporation from the heated fluid; upward $x>0$ (squares), downward $x<0$ (circles), and the total (net) flux depicted by a solid line (red online).

designed convergent nozzle operates (Bernoulli effect), but without the complications brought about by fabrication difficulties of a material convergent nozzle of highly miniaturized dimensions.

The exit of the heated liquid from the nozzle is accompanied by massive evaporation that occurs mostly in the vicinity of the nozzle exit [Fig. 2(a)]. This results in a NJ that is of lower temperature and reduced diameter compared to that of the injector (see Figs. 1 and 2). At a distance of about 60 nm away from the nozzle, the radius of the NJ decreases to $\approx$2 nm, which is about a third of the radius of the nozzle (located at $z=0$). As a result of the evaporation process, the propagating NJ undergoes "self-narrowing" [see Figs. 1(b) and 1(d)]. In particular, in Fig. 1(d) we observe large dispersion of the molecular densities as the distance ($z>0$) from the exit increases, with only the two inner shells [$R_{\mathrm{sh}}=0-1$ nm and $R_{\mathrm{sh}}=1-2$ nm in Fig. 1(d)] having liquid densities, while the outer shells correspond to vaporlike shells; note also the densification outside the nozzle exhibited for the two innermost liquid shells [Fig. 1(d)].

A steady state flow configuration of a NJ generated by uniform (symmetrical) heating ($T_W=500$ K) of a nozzle and propagating (with a velocity $v=700$ m/s) along the axial (z) direction is shown in Fig. 2(a). For comparison we show in Figs. 2(b)–2(d) the results for a NJ generated by nonuniform (asymmetrical) heating of a nozzle[8] (with the lower π sector maintained at $T_H=600$ K and the upper sector kept at $T_L=300$ K, see the schematic in the inset in Fig. 2), with propagation velocities (past the exit) $v_z=700$ m/s, $v_x=45$ m/s, and $v_y=0$ [Fig. 3(c)], resulting in a $\theta=4°$ deflection of the NJ from the z axis in the upward (lower temperature, $x>0$) direction [see the atomistic flow configuration in Fig. 2(b) and the corresponding contour surface for $\rho=0.4\times10^3$ kg/m^3 shown in Fig. 2(c), with the nozzle exit marked by an arrow]. The nozzle parameters for both cases displayed in Fig. 2 are as those given for the nozzle in Fig. 1. Varying T_H from 500 to 700 K (with $T_L=300$ K), the deflection angle changes in the range of $3.1°<\theta<4.95°$.

Underlying the NJ deflection is the shape change of the virtual convergent nozzle inside the heated nozzle and the massive highly nonisotropic evaporation of the outer layers of the ejected NJ near the exit, with the latter effect playing a dominant role (see below). The shape change of the virtual nozzle (VN) inside the injector is displayed in Fig. 2(d), which is a focused view of the liquid density 0.4×10^3 kg/m^3 contour shown in Fig. 2(c). The density contours shown in Fig. 3(a), recorded (for steady state flow) at the exit plane of the injector for the nonuniform heating case [see also the corresponding temperature contours in the same region in Fig. 3(b)] further illustrate the small asymmetry of the VN; note the small upward shift of the center of mass of the fluid (marked by a circle), which is displaced in the direction ($x>0$) of the lower temperature in the VN with respect to the geometrical center (marked by a cross) of the injecting nozzle. This displacement of the fluid center of mass ($x_{\mathrm{c.m.}}$) is also seen in Fig. 3(c); see the data marked as squares in interval I, corresponding to the interior of the injector, showing $\Delta x_{\mathrm{c.m.}}\approx0.5$ nm near $z=0$). The deflection of the flow in the VN is further evidenced by the development of a $v_{x,\mathrm{c.m.}}=9.8$ m/s component of the liquid center of mass flow velocity. Together with $v_z=700$ m/s (and $v_{y,\mathrm{c.m.}}=0$), we obtain a small internal deflection angle of 0.8°, which is too small to account for the observed deflection of the propagating NJ ($\theta=4°$).

The understanding of the thermal deflection phenomenon emerges from inspection of the transverse velocity $v_{x,\mathrm{c.m.}}$ in Fig. 3(c), which shows a dramatic increase in the 4 nm interval (marked II) near the exit, changing from $v_{x,\mathrm{c.m.}}=9.8$ m/s at the exit to $v_{x,\mathrm{c.m.}} \geqslant 40$ m/s at $z=4$ nm, with a subsequent slow increase afterward and with saturation ($v_{x,\mathrm{c.m.}} \approx 45$ m/s) at $z \approx 80$ nm; the above sharp velocity change is reflected also in the abrupt increase in the slope of the $x_{\mathrm{c.m.}}$ displacement at the exit of the nozzle. The origins of the above sharp transverse velocity increase are revealed by the records of the transverse momentum flux per unit length ($j^p{}_x$), exhibiting a negative value of the total (net) flux for $z \leqslant 80$ nm [see the solid line in Fig. 3(d)], as a result of a larger evaporative flux from the lower (hotter) part of the emanating fluid [compare circles and squares in Fig. 3(d)]. The conservation of momentum in the transverse plane dictates that the evaporative momentum imbalance will be offset by an increased velocity of the liquid core of the NJ in the opposite (i.e., $x>0$) direction (which may be expressed in terms of an effective force[9]), resulting in the bending of the propagation direction of the NJ in the lower temperature direction (upward), as observed in Figs. 2(b) and 2(c).

In summary, we have demonstrated here via MD simulations a method for deflecting high velocity liquid NJs through nonuniform asymmetric heating of a nozzle. The deflection originates from a highly nonisotropic strong molecular evaporation flux from the liquid near the exit, resulting in an effective force that bends the NJ in the direction of the lower temperature. Such vectorization of NJs may allow directional control and targeting of nanoscale jet flows, and it is expected to find uses in basic research and technological applications.

This work was supported by the NSF, AFOSR, and DOE. Computations were done at the Georgia Tech Center for Computational Materials Science.

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[7]The methodology of our MD simulations, where Newton's equations of motion for propane molecules interacting between themselves and with the atoms of a Pt nozzle are integrated on the computer (with an integration time step of 5×10^{-15} s), is the same as that given in Refs. 1 and 6. In the reservoir, before the entrance to the heated nozzle, the temperature of the "stored" liquid propane is 150 K, which is well below the boiling point (230 K) of propane. The liquid is pushed from the reservoir by applying a constant back-pressure, and when it passes through the heated (wall temperature T_W) cylindrical nozzle, it heats up and a temperature gradient is formed. For the choice of the heated nozzle length ($L=30$ nm in the present simulations), see Ref. 6.

[8]Steady state virtual NJ flow characteristics and stable operation of the deflection mode require sustained temperature gradients in the fluid inside the injector, particularly in the plane normal to the propagation direction (z). In the z direction, the Peclet number (expressing the relative effectiveness of convective and diffusive heat transport, $\mathrm{Pe}=VL/\kappa$, where V, L, and κ are the characteristic velocity, length, and thermal diffusivity, $\kappa=100$ nm^2/ns for the propane liquid) is large, $\mathrm{Pe}\approx200$ (with $L=30$ nm and $V_z=700$ m/s), reflecting the dominance of convection. However, in the perpendicular plane, the characteristic velocities are very small, corresponding to an exceedingly small Pe. The small transverse Peclet number, together with the small Reynolds number for our system ($\mathrm{Re}\approx30$, i.e., nonturbulent flow), supports the formation of stable gradients and results in the steady state temperature profiles observed in our simulations.

[9]The change in the total momentum flux (per unit length) caused by the nonuniform evaporation in the first 6 ps, the time it takes the exiting heated jet to traverse a 4 nm distance from the exit [i.e., region II in Fig. 3(c), with an exit velocity of 700 m/s], is about 70 amu/ps^2, and from the simulations we determine also that the mass per unit length (nm) enclosed by a cylindrical surface of about 6–7 nm diameter is $\approx2\times10^4$ amu/nm, yielding an acceleration of about 4.0×10^3 nm/ns^2. The corresponding force (per unit length) is $\approx$20 pN/nm (acting on the liquid core of the NJ for about 6 ps in the $x>0$, upward, direction), estimated by using the mass distribution of the NJ after it leaves the strong evaporation region (i.e., at the end of region II) where the mass distribution is about 3500 amu/nm (the corresponding radius of the liquid core of the NJ is about 2–3 nm).