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An atmospheric-pressure,
high-aspect-ratio, cold micro-plasma

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An atmospheric pressure nonequilibrium Ar micro-plasma generated inside a micro-tube with plasma radius of 3 μm and length of 2.7 cm is reported. The electron density of the plasma plume estimated from the broadening of the Ar emission line reaches as high as $3 \times 10^{16} \text{ cm}^{-3}$. The electron temperature obtained from CR model is 1.5 eV while the gas temperature of the plasma estimated from the N_2 rotational spectrum is close to room temperature. The sheath thickness of the plasma could be close to the radius of the plasma. The ignition voltages of the plasma increase one order when the radius of the dielectric tube is decreased from 1 mm to 3 μm .

Non-equilibrium low-temperature plasmas sustained at atmospheric pressure, which are very promising for a range of advanced applications from health care and medicine to materials science and nanotechnology^{1–6}. Upon reduction to micrometer dimensions, plasmas not only show unique physical properties (e.g., extreme thermal non-equilibrium), but also enable an unprecedented access to a largely unexplored region of the plasma phase parameter space⁷. Present-day micro-plasmas are confined to micro-cavities of the typical dimensions in the ten-to-hundreds micrometers and the aspect (length-to-depth/width) ratios in the 1 : 1 to 20 : 1 ranges^{7–14}. The electron/ion density of such micro-plasmas is typically $\sim 10^{11}–10^{15} \text{ cm}^{-3}$ ^{7–14}. These and other interesting properties make micro-plasmas warranted for applications in chemical analysis¹⁵, materials synthesis and processing¹⁶, nanotechnology^{17,18}, micro- and nanofabrication¹⁹, and several other areas.

As the discharge size is reduced, it becomes more difficult to generate the plasma and maintain its charge neutrality ($n_e = n_i$), where n_e and n_i are the electron and ion densities, respectively. Moreover, the number of electrons and ions within the Debye sphere

$$N_D = 4/3 \pi n_e \lambda_D^3 \quad (1)$$

should remain large as the Debye length $\lambda_D = (\epsilon_0 k_B T_e / n_e e^2)^{1/2}$ becomes smaller and smaller. Here, T_e is the electron temperature, k_B is Boltzmann's constant, e is the electron charge, and ϵ_0 is the dielectric constant of vacuum. When the discharge size is reduced, the electron density should increase accordingly to minimize λ_D compared to the plasma dimension. Otherwise, the charge separation area (plasma sheath) may span across the whole discharge area thus leading to the charge neutrality loss. However, increasing the electron density in micrometer gaps further represents a major challenge for atmospheric-pressure discharges, mainly because of the extremely high losses on the discharge confining surfaces leading to imbalanced species production-loss and unstable discharge behavior.

This challenge is resolved in this work by confining micro-plasmas within a channel of 3 μm radius within a thin glass tube and adopting an advanced micro-fluidic approach to enable a slip-flow regime²⁰, because the common continuous gas flow regime (used in nearly all plasma discharges within tubes) fails in so narrow gaps where the Knudsen numbers are very low (~ 0.02). By using easily accessible AC ignition voltages of up to 60 kV, we demonstrate a stable plasma plume with a radius of 3 μm , length of ~ 2.7 cm (maximum aspect ratio of ~ 9000) sustained inside the thin glass tube carrying an Ar flow (Fig. 1). The plasma is highly-non-equilibrium because the electron density n_e reaches as high as $3 \times 10^{16} \text{ cm}^{-3}$, while the electron temperature is ~ 1.5 eV and the gas temperature of the plasma remains at room temperature.

Furthermore, the Debye length λ_D of the plasma plume is ~ 50 nm and the number of electrons and ions within the Debye sphere is $\sim 15–20$. On the one hand, the sheath of the micro-plasma reported here is probably more like high-voltage sheath (the plasma is generated between the high voltage electrode and the surrounding air which serves as the ground), the actual sheath thickness could be tens of Debye lengths²¹. Thus the dimension of the

Figure 1 | (a) The schematic of the experimental setup. (b) The photograph of the Ar plasma plume.

sheath of the micro-plasma could be close to the radius of the plasma. In other words, the charge neutrality of the plasma may fail. The sheath-bulk balance of the plasma may be disrupted with the creation of a structure of sheath-only plasma.

Results

The device is sketched in Fig. 1(a). When alternating voltage is applied and Ar gas is fed into the tubes, a plasma plume in the glass tube is generated, as shown in Fig. 1(b). The length of the plasma plume could reach ~ 2.7 cm.

The discharge current-voltage waveforms are shown in Fig. 2. One current pulse in each half cycle of applied voltage is observed, as shown in Fig. 2(a). But the amplitude and the onset time of the discharge current are not constant. The peak value of the discharge current pulse is $0.1 \sim 0.25$ A. Figure 2(b) shows a single discharge current pulse. The full width at half maximum of the current pulse is about 30 ns. Importantly, the power needed to sustain the plasma plume was only about 1.8 W. However, the peak current density and the power density of the plasma plume were as high as $\sim 3.5 \sim 8.8 \times 10^5 \text{ A}\cdot\text{cm}^{-2}$ and $2.3 \times 10^6 \text{ W}\cdot\text{cm}^{-3}$, respectively.

Moreover, even though the dissipated power density is quite high ($\sim 2.3 \times 10^6 \text{ W}\cdot\text{cm}^{-3}$), the gas temperature of the Ar plasma plume is close to room temperature. To confirm this, we estimate the rota-

tional gas temperature by comparing the simulated spectra of the $C^3\Pi_u-B^3\Pi_g$ ($\Delta v = 0$) band transition of N_2 (trace amount of N_2 from the surrounding air due to diffusion) with the experimental recorded spectra. The result clearly shows that the simulated spectra at $T_{\text{rot}} = 350$ K give a good fit to the experimental spectra. Therefore, the gas temperature of the plasma plume is close to 350 K. However, because the Ar metastables $\text{Ar}(4s)$ can easily transfer energy to the ground state N_2 molecules to form the excited state N_2 ($C^3\Pi_u$) at high rotational levels, resulting in a relatively broad distribution of rotational N_2 temperatures, the above gas temperature determined by the T_{rot} of N_2 is overestimated. On the other hand, the temperature of the glass tube measured by a thermometer shows it is at room temperature. One of the reasons that the plasma can stay close to room temperature even with the deposited power density on the order of $\text{MW}\cdot\text{cm}^{-3}$ is because of the very large surface-to-volume ratio of the plasma, which enhances heat exchange between the gas and the environment thereby reducing the gas temperature.

The electron density n_e of the plasma is measured through the emission line broadening. It is about $3 \times 10^{16} \text{ cm}^{-3}$. The Stark broadening of the Ar atom emission line at 696.5 nm is used to determine the electron density of the plasma plume. When the electron density of the plasma is higher than 10^{15} cm^{-3} , the Stark effect of Ar lines broadening plays an important role and can be used for the purpose

Figure 2 | The discharge current and applied voltage waveforms of the plasma plume. (a) Multiple current pulses. (b) Single discharge current pulse.

of electron density diagnostics^{22,23}. Besides the Stark broadening, the spectral line emitted from the plasma can be broadened by other mechanisms, and the total broadening of the line profile includes contributions of all these effects. In this experiment, the relevant sources of broadening include Stark, Doppler, and the van der Waals effects, in addition to the broadening induced by the instrument, have been rigorously included. The natural and resonance broadening are negligible for the high density plasmas of this work.

The profiles arising from these mechanisms can be accurately approximated by Gaussian (Doppler and instrumental broadening) or Lorentzian (Van der Waals and Stark broadening) forms, with their combination representing a Voigt profile. $\Delta\lambda_L$ and $\Delta\lambda_G$ are the broadening widths (full width at half maximum) of the Lorentzian and Gaussian profiles, respectively, given by $\Delta\lambda_L = \Delta\lambda_S + \Delta\lambda_V$ and $\Delta\lambda_G^2 = \Delta\lambda_D^2 + \Delta\lambda_i^2$, where $\Delta\lambda_S$, $\Delta\lambda_V$, $\Delta\lambda_D$, and $\Delta\lambda_i$ refer to the Stark, van der Waals, Doppler and instrument broadening widths, respectively. The part of the line broadening corresponding only to the Stark broadening can be obtained by separating (deconvolution) the Stark broadening from the total broadened profile, with the knowledge of other broadening components. The instrumental broadening width $\Delta\lambda_i$ is determined by using a low-pressure Hg lamp. The instrumental line profile at 691.5 nm is fitted by a Gaussian profile, and the $\Delta\lambda_i$ is obtained to be 0.088 nm, as shown in Fig. 3(a).

The Doppler broadening is determined from the emitter temperature T_g ²²

$$\Delta\lambda_D(\text{nm}) = 7.16 \times 10^{-7} \lambda_0 (T_g/M)^{1/2} \quad (2)$$

where the gas temperature T_g is given in Kelvin, M is the mass of the emitters expressed in atomic mass units and λ_0 is the wavelength of the line in nm. Under the conditions of this work ($T_g = 350$ K, $\lambda_0 = 696.5$ nm, and $M = 40$), $\Delta\lambda_D$ is estimated to be 1.4×10^{-3} nm.

The van der Waals broadening²²

$$\Delta\lambda_V = K_i (T_g/\mu)^{3/10} N \quad (3)$$

is a consequence of a dipolar interaction between the emitter and the neutral perturbers, where μ is the reduced mass of the emitter-perturber pair, N is the neutral gas density, K_i is a constant that depends on the spectral line and the emitter polarizability¹⁴. The value of $\Delta\lambda_V$ is calculated to be 2.36×10^{-2} nm.

The Stark broadening width

$$\Delta\lambda_s = 2w(T_e)n_e [1 + 1.75 \times 10^{-4} n_e^{1/4} \alpha(T_e) \times (1 - 0.068 n_e^{1/6} T_e^{-1/2})] \times 10^{-16} \quad (4)$$

is expressed in nm and is a complex function of the electron density n_e (cm^{-3}) and temperature T_e (K)²³, where $\alpha(T_e)$ is the static ion-

broadening parameter, and $w(T_e)$ is electron impact half-width. Figure 3(b) shows the relationship between $\Delta\lambda_s$ and n_e for $T_e = 10000$ K and $T_e = 40000$ K, which clearly suggests that the value of $\Delta\lambda_s$ strongly depends on n_e while is weakly affected by the electron temperature. The Stark broadening width is separated from the Voigt profile by deconvolution, which is calculated to be 0.038 nm, as shown in Fig. 3(c). Therefore, the electron densities for $T_e = 10000$ K and $T_e = 40000$ K are $3.4 \times 10^{16} \text{ cm}^{-3}$ and $2.1 \times 10^{16} \text{ cm}^{-3}$, respectively.

The accuracy of the electron density measurements from the Stark broadening depends on the values of the electron temperature, which was estimated to be ~ 1.5 eV using a simple collisional-radiative (CR) model from Ar excited levels²⁴. The intensity of Ar emission lines can be obtained from the emission spectrum, as shown in Fig. 4. Our calculations relied on the emission lines and transition probabilities²⁵, cross-section data for electron collision excitation from the ground state²⁶, as well as the electron-impact excitation cross-sections from the metastable levels to the four $4p$ levels^{27,28}. The calculated electron temperature of 1.5 eV yields the electron density of the plasma plume of approximately $3 \times 10^{16} \text{ cm}^{-3}$.

Discussion

We now highlight and discuss several unique features of the plasma plume. First, for the electron temperature of just above 1 eV and near-room-temperature gas, this is the highest electron density of room temperature non-equilibrium plasmas ever been reported. The electron density of our plasma plume is orders of magnitude higher than previously reported^{7–11}.

Second, we also studied the relationship between the inner radius and the plasma ignition voltages. It was found that the ignition voltages increase steeper and steeper as the inner radius of the tube is reduced; this trend is particularly strong when the inner radius becomes less than 5 μm as shown in Fig. 5. This may be due to the higher relative importance of plasma sheath when the plasma volume decreases while the inner radius becomes smaller. It would be expected that even more interesting phenomena could be observed if the radius of the plasma is further reduced. We would like to pointed out that Jogi et al. recently also found that the sustaining voltage of a helium plasma jet increase from about 6 kV to 14 kV when the diameter of the plasma jet is decreased from 500 μm to 100 μm ²⁹.

Third, the aspect ratio of our micro-discharge can reach $\sim 9000:1$ which is ~ 450 times higher than the reported maximum aspect ratios of $\sim 20:1$.

Finally, from the application point of view, this high density non-equilibrium atmospheric pressure plasma may be used to novel applications such as micro-fluid control, chemical analysis, radiation sources, plasma medicine, as well as inner surface modification or surface coating of micro-tubes.

Figure 3 | (a) Gaussian fitting of the Hg 691.5 nm line profile recorded using the optical system with the grating of 1200 groove/mm and slit width of 50 μm . (b) The relationship between the Stark broadening width and the electron density for $T_e = 10,000$ K and $T_e = 40,000$ K. (c) Voigt fitting of the Ar 696.5 nm line profile recorded using the optical system with the grating of 1200 groove/mm and the slit width of 50 μm .

Figure 4 | The emission spectrum of the Ar plasma plume in the range 690–850 nm. The grating and slit width of the spectrometer are 1200 groove/mm and 100 μm , respectively.

In conclusion, a room-temperature Ar plasma plume with a radius of 3 μm and a length of 2.7 cm, driven by kHz AC power supply, is reported. The electron density of the plasma plume, calculated from the Ar emission line broadening, can reach as high as $\sim 3 \times 10^{16} \text{ cm}^{-3}$. The electron temperature of the plasma determined by the collisional-radiative (CR) model for Ar excited levels is about 1.5 eV. Importantly, even though the input power density for the plasma plume is quite high ($\sim 2.3 \text{ MW} \cdot \text{cm}^{-3}$), the gas temperature stays close to room-temperature. This is due to the large surface-to-volume ratio of the plasmas under extremely non-equilibrium conditions. The discharge ignition voltage increases to 40 kV as the inner radius of tube decreases to 3 μm , which is ten times higher compared to the plasma plumes with the radius in the mm range. The plasma sheath may span across the whole discharge area thus leading to the charge neutrality loss.

Methods

Figure 1(a) shows the schematic of experiment setup. The high-voltage (HV) electrode is made of stainless steel needle (radius of the needle tip is about 50 μm) and is inserted into a quartz tube with the inner radius of 150 μm . A glass tube with the inner radius of 3 μm and the outer radius of 12 μm is connected to the quartz tube. The joint between the two tubes is sealed to prevent gas leakage. The distance between the

Figure 5 | The ignition voltage of the Ar plasma plume vs. the inner radius of the glass tube.

tip of the needle and the left end of the glass tube is about 1 mm. Ar is used as working gas with a flow rate of $3.6 \times 10^{-4} \text{ ml/min}$. Alternating voltage (AC voltage V_{p-p} : 42 kV, frequency: 10.5 kHz, fixed throughout the paper) is used to drive the plasma and Ar gas is used as working gas. The applied voltage and currents are measured by a P6015 Tektronix high-voltage probe and a current probe (Pearson 6585), respectively. The distance between the tip of the needle and the current probe is fixed at about 10 mm. The voltage and current waveforms are recorded by a Tektronix DPO3034 broadband digital oscilloscope. The power density of the plasma plume is calculated according to the measured current and voltage waveforms. The optical emission spectra of the plasma are measured by a Princeton Instruments Acton SpectraHub 2500i spectrometer. The grating and the slit width of the spectrometer are set at 3600 groove/mm and 50 μm , respectively. The rotational gas temperature of the plasma is estimated by comparing the simulated spectra of the $C^2\Pi_u-B^2\Pi_g$ ($\Delta v = 0$) band transition of N_2 (trace amount of N_2 from the surrounding air due to diffusion) with the experimental recorded spectra. The electron density n_e of the plasma is measured through the Stark broadening of the Ar atom emission line at 696.5 nm. As we know that, besides the Stark broadening, the spectral line emitted from the plasma can be broadened by other mechanisms, and the total broadening of the line profile includes contributions of all these effects. In this paper, the relevant sources of broadening include Stark, Doppler, and the van der Waals effects, in addition to the broadening induced by the instrument, have been rigorously included. The natural and resonance broadening are negligible for the high density plasmas of this work. The obtained electron density of the plasma is about $3 \times 10^{16} \text{ cm}^{-3}$. The electron temperature is estimated by using a simple collisional-radiative (CR) model from Ar excited levels. The model assumes that the population of four of the $4p$ levels of Ar is dominated by electron excitation kinetics, and spontaneous radiative decay is the main process of de-excitation from the excited energy levels. Here the $2p_2$, $2p_6$, $2p_8$, and $2p_9$ levels (in Paschen notation) are chosen. The intensity of Ar emission lines can be obtained from the emission spectrum, as shown in Fig. 4.

- Shashurin, A., Keidar, M., Bronnikov, S., Jurjus, A. R. & Stepp, A. M. Living tissue under treatment of cold plasma atmospheric jet. *Appl. Phys. Lett.* **93**, 181501 (2008).
- Lu, X., Naidis, V. G., Laroussi, M. & Ostrikov, K. Guided ionization waves: Theory and experiments. *Phys. Rep.* **540**, 123 (2014).
- Kolb, F. J. et al. Cold atmospheric pressure air plasma jet for medical application. *Appl. Phys. Lett.* **92**, 241501 (2008).
- Xian, Y. et al. From short pulses to short breaks exotic plasma bullets via residual electron control. *Sci. Rep.* **3**, 1599 (2013).
- Keidar, M. et al. Cold atmospheric in cancer therapy. *Phys. Plasmas* **20**, 057101 (2013).
- Douat, C., Bauville, G., Fleury, M., Laroussi, M. & Puech, V. Dynamics of colliding microplasma jets. *Plasma Sources Sci. T.* **21**, 034010 (2012).
- Kushner, M. Modeling of microdischarge devices: Pyramidal structures. *J. Appl. Phys.* **95**, 846 (2004).
- Park, S., Eden, J., Chen, J. & Liu, C. Microdischarge devices with 10 or 30 μm square silicon cathode cavities: pd scaling and production of the XeO excimer. *Appl. Phys. Lett.* **85**, 4869 (2004).
- Boeuf, J., Pitchford, L. & Schoenbach, K. Predicted properties of microhollow cathode discharges in xenon. *Appl. Phys. Lett.* **86**, 071501 (2005).
- Schoenbach, K., Verhappen, R., Tessnow, T., Peterkin, P. & Byszewski, W. Microhollow cathode discharges. *Appl. Phys. Lett.* **68**, 13 (1996).
- Schoenbach, K., El-Habachi, A., Shi, W. & Ciocca, M. High-pressure hollow cathode discharges. *Plasma Sources Sci. T.* **6**, 468 (1997).
- Becker, K., Schoenbach, K. & Eden, J. Microplasmas and applications. *J. Phys. D* **39**, R55 (2006).
- Eden, G. J. & Park, J. S. New opportunities for plasma science in nonequilibrium, low-temperature plasmas confined to microcavities: There's plenty of room at the bottom. *Phys. Plasmas* **13**, 057101 (2006).
- Wagner, A., Mariotti, D., Yurchenko, K. & Das, T. Experimental study of a planar atmospheric-pressure plasma operating in the microplasma regime. *Phys. Rev. E* **80**, 065401 (2009).
- Janasek, D., Franzk, J. & Manz, A. Scaling and the design of miniaturized chemical-analysis systems. *Nature* **442**, 374 (2006).
- Tai, K., Houlihan, J. T., Eden, G. J. & Dillon, J. S. Integration of microplasma with transmission electron microscopy: Real-time observation of gold sputtering and island formation. *Sci. Rep.* **3**, 1325 (2013).
- Ostrikov, K., Neyts, C. E. & Meyyappan, M. Plasma nanoscience: from nano-solids in plasmas to nano-plasmas in solids. *Adv. Phys.* **62**, 113 (2013).
- Wheeler, R. A. Putting electrowetting to work. *Science* **322**, 539 (2008).
- Harrison, D. et al. Micromachining a miniaturized capillary electrophoresis-based chemical analysis system on a chip. *Science* **261**, 895–897 (1993).
- Zhang, W., Meng, G. & Wei, X. A review on slip models for gas microflows. *Microfluid. Nanofluid.* **13**, 845 (2012).
- Lieberman, M. & Lichtenberg, A. *Principles of plasma discharges and materials processing* [164–166] (John Wiley & Sons, New York, 1994).
- Torres, J. et al. A stark broadening method to determine simultaneously the electron temperature and density in high-pressure microwave plasmas. *J. Phys. D: Appl. Phys.* **40**, 5929 (2007).

23. Konjevic, R. & Konjevic, N. On the use of non-hydrogenic spectral line profiles for electron density diagnostics of inductively coupled plasmas. *Spectrochim Acta B*, **52**, 2077–2084 (1997).
24. Mariotti, D., Shimizu, Y., Sasaki, T. & Koshizaki, N. Method to determine argon metastable number density and plasma electron temperature from spectral emission originating from four 4p argon levels. *Appl. Phys. Lett.* **89**, 201502 (2006).
25. Kramida, A., Ralchenko, Yu. & Reader, J. and NIST ASD Team, <http://physics.nist.gov/PhysRefData>, Atomic spectra database – Version 5 (2014) Date of access: 22/09/2014.
26. Yanguas-Gil, A., Cotrino, J. & Alves, L. L. An update of argon inelastic cross sections for plasma discharges. *J. Phys. D: Appl. Phys.* **38**, 1588–1598 (2005).
27. Bartschat, K. & Zeman, V. Electron-impact excitation from the (3p⁵4s) metastable states of argon. *Phys. Rev. A*, **59**, R2552 (1999).
28. Dasgupta, A., Blaha, M. & Giuliani, L. J. Electron-impact excitation from the ground and the metastable levels of Ar I. *Phys. Rev. A*, **61**, 012703 (1999).
29. Jogi, I., Talviste, R., Raud, J., Piip, K. & Paris P. The influence of the tube diameter on the properties of an atmospheric pressure He micro-plasma jet. *J. Phys. D: Appl. Phys.* **47**, 415202 (2014).

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Author contributions

X.L. and Y.P. initiated the idea to generate the high density plasma in micro-tubes. X.L. and S.W. wrote the main manuscript text, S.W. and J.G. prepared figures 1–5. All authors reviewed the manuscript.

Additional information

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