How pinning and contact angle hysteresis governs quasi-static liquid drop transfer

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S1. Details of surface fabrication

Table S1. Technical details on the fabrication of surfaces used in this study. Except Silicon and OTS, all surfaces were fabricated with the techniques similar to one described in Ref.32.

Surface Name	Method	Material and fabrication details
Silicon	N/A	100mm-orientationsiliconwafers(Silicon Valley Microelectronics, Inc.)
PMMA(1)	Spin coat	Solution: 2wt% of PMMA (Aldrich Mw~120000) in tolueneSpin coater speed: 1000 RPMSpin time: 60s
PMMA(2)	Spin coat	Solution:0.2wt% of PMMA (Aldrich Mw~120000) in tolueneSpin coater speed:2000 RPMSpin time:Spin coater speed:2000 RPMSpin time:
PMMA(3)	Spin coat	Solution: 1wt% of PMMA (Aldrich Mw~120000) in toluene Spin coater speed: 2000 RPM Spin time: 60s
Blend (1)	Spin coat	Solution:1wt%PMMA (AldrichMw~120,000)andPolystyrene(AldrichMw~35,000)mixed in toluenesolution, ratio of PMMA:PS=15:1.Spin coater speed:1500 RPMSpin time:60s
Blend (2)	Spin coat	Solution: 1wt% PMMA (Aldrich Mw~120,000) and Polystyrene (Aldrich Mw~35,000) mixed in toluene solution , ratio of PMMA: PS = 6:1. Spin coater speed: 1500 RPM Spin time: 60s
PS (1)	Heat press	Material: Polystyrene (Aldrich Mw~35,000) Plate: Two slices of spin coated Teflon AF surface Temperature: 185°C Time: 2 mins. Pressure: 645 Pa After heat press, surface was placed into 20 °C distilled water immediately. Cleaned by FC-75 again, after detached from the plate.

PEMA	Spin coat	Solution: 1wt% solution of Poly (ethyl methacrylate), (Aldrich Mw~515,000) in toluene		
		Spin coater speed: 2000 RPM Spin time: 60s		
PS (2)	Spin coat	Solution: 1wt% solution of Polystyrene, (Aldrich Mw~35,000) in toluene. Spin coater speed: 2000 RPM Spin time: 60s		
OTS	N/A	Microscope slides wafers were soaked in 95% OTS solution (30 mins) and cleaned with ethanol		
Teflon AF	Spin coat	Solution: Teflon AF (DuPont Teflon AF 601s2-100-6)diluted with FC-75 (3M) in the ratio of 1:5Spin coater speed: 1500 RPMSpin time: 60s		
РР	Material: Polypropylene (Aldrich Mw~12,000) Plate: Two slices of spin coated Teflon AF surface Temperature: 200°C Time: 2 mins. Pressure: 645 Pa			

S2. Evidence for vanishing Regime II when at least one of the surfaces has $\vartheta_r > 90^\circ$

Here, we show the process of 2.0µl water transfer from surface ① to surface ② (Case SA) and from surface ③ to surface ② (Case SB), simulated using the theoretical model. The receding contact angle of surface ①, ② and ③ are 95.5°, 95° and 94.5°, respectively. The evolutions of contact radius and contact angle are shown in Fig.s S2 and S3, respectively. For Case SA, the pinning of contact line is found on the acceptor surface. For case SB, the receding contact angle of donor surfaces is only decreased very slightly, from 95.5° to 94.5°, but the contact line pinning is observed only on the donor surface. Therefore, Case SB is in Regime I, and Case SA is in Regime III (Fig. S4), which is evidence for the vanishing width of Regime II when the receding angles exceed 90°.



Fig. S1. Evolution of contact radius and contact angle of the liquid transfer from surface (1) to surface (2).



Fig. S2. Evolution of contact radius and contact angle of the liquid transfer from surface ③ to surface ②.



Fig. S3. Locations of Cases SA and SB in the transfer regime Map.

S.3 Effect of surface tension

dθ

Two liquid bridges which have the same H (distance between two surfaces: 0.8 mm), volume (2. 2µl) and contact angle (lower: 60° Upper: 100°), but different surface tensions (72mN/m and 63mN/m) are simulated. Their profiles are shown in Fig. S5(a). It can be seen that the shape of the two bridges are exactly the same despite their different surface tensions. The only difference between the two bridges is the value of ΔP (Laplace pressure difference), which is found to be 43.1N/m² for the liquid with surface tension of 72 mN/m, but 37.7 N/m² for the liquid with surface tension of 63 mN/m.

The observation can be understood by examining the equations (EQ(S1) - (S3))that are used to solve the profile of liquid bridge:

$$\frac{dX}{dS} = \cos\theta \tag{S1}$$

$$\frac{dZ}{dS} = \sin\theta \tag{S2}$$

$$\frac{d\theta}{dS} = \frac{\Delta P}{\gamma} - \frac{\sin\theta}{X} \tag{S3}$$

where X and Z are the horizontal and vertical coordinates of the axisymmetric liquid bridge, S is the arc length measured from the contact point of the liquid with the lower surface, and θ is the angle between the local tangent of the liquid surface and the horizontal axis; γ is the surface tension of the liquid. These equations are solved with boundary conditions on the two solid surfaces, either presented as contact radius or as contact angle. The Laplace pressure ΔP is not unknown a priori and is part of the solution. It can be seen that the surface tension γ only appears in the EQ (S3) together with ΔP . Therefore, if one defines a normalized Laplace

pressure
$$\Delta P' = \frac{\Delta P}{\gamma}$$
, then EQ (S3) can be re-written as
 $\frac{d\theta}{dS} = \Delta P' - \frac{\sin\theta}{X}$ (S4)

EQs (S1), (S2) and (S4), together with boundary conditions on the surfaces, completely determine the liquid profile and the value of $\triangle P'$, while the surface tension does not play a role in the solution. In other words, ΔP accommodates the change of surface tension. Therefore, the transfer regime Map of quasi-static liquid transfer (Fig. 3 in the main text) is not affected by the surface tension of the liquid. We also simulated two more liquid bridges, which have the same contact radius as the boundary conditions (upper: 1.2 mm, lower: 1.0 mm), but different surfaces tension (72mN/m and 63mN/m). Again the profiles of the two liquid bridges are identical, as shown in Fig. S5(b).



Fig. S4. (a) Profiles of liquid bridges have the same H, volume, lower and upper contact angles but different surface tension. (b) Profiles of liquid bridges have the same H, volume, lower and upper contact radius but different surface tension.

S.4 Effect of *R_{min}* on the position of the liquid bridge neck

Associated with the change of R_{min} on the donor and/or acceptor surfaces, the neck i.e. the narrowest point of the liquid bridge also changes its location. Specifically, as $(\vartheta_r)_{don}$ increases or as $(\vartheta_r)_{acc}$ decreases, the liquid becomes more attracted to the acceptor surface, R_{min} on the acceptor surface increases and/or R_{min} on the donor surface decreases. Consequently, the neck of the liquid bridge is located closer to the donor surface. Fig. S5 shows the experimental data for $H^* = H_N$ /*H* as a function of $(\vartheta_r)_{don}$, where H_N is the distance between the neck of the liquid bridge and the donor surface, when the bridge breaks. The corresponding transfer ratios are also shown in Fig. S5. It can be seen that, as expected, the height of the neck location decreases with the increase of $(\vartheta_r)_{don}$. Since the liquid bridge breaks at the neck, with the decrease in H^* , the transfer ratio should increase. When $(\vartheta_r)_{don}$ is larger than 90°, H^* reduces to zero, the liquid bridge breaks at the donor surface and a complete transfer ($\alpha = 100\%$) can be observed.



Fig. S5. H^* of experimental cases 1 to 10 as a function of $(\vartheta_r)_{don}$, and the corresponding transfer ratio.

S.5 Details of the nine more experiments to find *m* and *n* used in the regression analysis

Table S2. Details of the nine more experimental results used in the regression analysis to find m and n coefficients in EQ.(2). All experiments follow the same procedures as described in the main text.

Liquid	Name of acceptor surface and its ϑ_r	Name of donor surface and its ϑ_r	Transfer ratio (%)
Glycerol	PEMA, ϑ _r :63.1°	Silicon, $\vartheta_r:34.5^\circ$	2.5
Glycerol	PEMA, ϑ _r :63.1º	PMMA(1), ϑ _r :58.1°	6
Glycerol	PEMA, ϑ _r :63.1°	PS(1), ϑ _r :61.2°	18
Glycerol	PEMA, ϑ _r :63.1°	OTS, ϑ _r :85.9°	98.5
Glycerol	PEMA, ϑ _r :63.1°	Teflon AF, ϑ _r : 107.3°	100
Ethylene glycol	PEMA, ϑ _r :53.1º	PS(1), ϑ _r : 51.1º	17
Ethylene glycol	PEMA, ϑ _r :53.1º	OTS, ϑ _r : 68.1º	96
Ethylene glycol	PEMA, ϑ _r :53.1º	Teflon AF, ϑ _r : 96.2°	99.9
Silicon oil	PEMA, ϑ _r : ∼4º	Teflon AF, ϑ_r : 50.2°	89

S.6 The evolutions of the adhesion force, contact angle and contact line of Case A and B

Fig. S6 (a) and (b) show the evolutions of the contact angle on both surfaces in cases A and B. The evolutions of the adhesion force are also shown in the plots. It can be seen that, for case A, F_{max} happens within the period of contact line pinning on the acceptor surface, but after the contact line pinning on the donor surface. Therefore, based on the discussion in Section 3.4.1, the governing parameters for F_{max} in case A are $(\vartheta_a)_{acc}$ and $(\vartheta_r)_{don}$. However, for case B, which has a much larger $(\vartheta_a)_{don}$, F_{max} happens at after the contact line pinning on the acceptor surface and the end of the contact line pinning on the donor surface ; hence F_{max} in case B is governed by $(\vartheta_r)_{acc}$, $(\vartheta_a)_{don}$ and $(\vartheta_r)_{don}$, i.e., $(\vartheta_a)_{don}$ still plays a role at this large $(\vartheta_r)_{don}$.

The different $(\vartheta_a)_{don}$ in cases A and B also lead to different F_{max} values. From Fig. S6(a) and (b), it can be seen that in both cases the adhesion force experiences a rapid increase during the period of contact line pinning on both surfaces (*a*1 to *a*2 in case A, *b*1 to *b*2 in case B). Due to the very small *CAH* (2°) of the donor surface in case A, the contact line pinning on the donor surface (*a*1 to *a*2 in Fig. S6(a)) is much shorter compared to that of case B (*b*1 to *b*2 in Fig. S6(b)). This has limited the increase in F_{max} and results in smaller F_{max} in case A compared with F_{max} in case B.



Fig. S6. (a) Evolutions of contact angle and adhesion force of cases A. (b) Evolutions of contact angle and adhesion force of cases B. F_{max}