Supporting Online Material
Materials and Methods

Fabrication of the Microfluidic Devices and Data Collection. We fabricated the microfluidic devices using standard photolithographic and soft-lithographic methods.(*S1*-*S5*) We created a master with photoresist on a silicon wafer, and then exposed the surface to a fluorinated silane.(*S6*) Polydimethylsiloxane (PDMS) poured onto the master and allowed to cure in a 60 °C oven formed the stamp that contained the channels. After being exposed to an oxygen plasma for 1 minute, the stamp was sealed irreversibly to a glass microscope slide that also had been exposed to the plasma for 6 minutes. We used a high-speed CCD video camera capable of recording up to 160,000 frames per second (Phantom V7, Vision Research) mounted on a microscope (Leica) to capture movies of the behavior of the system. We analyzed the movies using Matlab and software created in-house.

Controlling the Flow of Liquids in the System. We controlled the flows of both the aqueous phase and the hexadecane by using nitrogen gas to pressurize the reservoirs of the two liquids. An electronically controlled valve (Pneutronics OEM-EPC 15 PSI, Parker Life Sciences) also metered the pressure of the gas applied to the aqueous phase. This valve was powered by a power supply set to 24 V. House nitrogen supplied the backing pressure for the valve. A separate power supply, precise to 0.0005 V, controlled the degree to which the valve was open, and hence the pressure that it supplied to the reservoir of aqueous solution.

The vial that contained the aqueous solution was placed on a micropositioner; raising or lowering the reservoir provided a method of fine control of the pressure applied to the stream. The line that fed gas into the vial of hexadecane connected via a three-way valve to the outlet of the system. A second three-way valve controlled whether the outlet of the system connected to the pressure supplied by the nitrogen or to a waste container. By changing the positions of the two valves, we controlled whether the droplets moved forward or backwards through the system.

Bifurcation Cascade. We used an aqueous solution of pen ink (Waterman) as the dispersed phase. The lengths of the shorter and longer branches of the loop were 1.78 and 1.98 mm. Fig. S1a and S1b show schematic diagrams of the microfluidic network that we used for this experiment.

Reversibility. We used an aqueous solution of Meldola's Blue as the dispersed phase. The device used in the reversibility experiment is shown in fig. S2a and S2b. The channels were 80 μm tall and 100 μm wide. The lengths of the shorter and longer branches of the loop were 1.74 mm and 1.94 mm. Fig S2c is a schematic diagram of the valves that controlled the flow of the gas. The three-way valves closed off one of the paths and kept two open. When the valves were set in the configuration depicted in the figure, the nitrogen was applied to the outlet of the microfluidic device.

Encoding/Decoding. We used an aqueous pen-ink solution (Waterman) as the dispersed phase. The loops of the encoder/decoder devices are shown schematically in fig. 4a and 4b. The channels for the device used to process the analog signal were 72 μm tall and 100 μm wide. The loops in this device had shorter and longer branches that were 1.69 mm and 1.89 mm long. The channels for the device used to process the digital signal were 86 μm tall and 100 μm wide. The branches of the loops in this device were 1.61 mm and 1.41 mm long.

For the digital coding experiment, we set the pressure applied to the hexadecane at 3.26 psi. We set the backing pressure applied to the electronically controlled valve at 20.00 psi and the voltage used to power the device at 23.7 V. To make a 0 or a 1, we changed the control voltage by ± 1 mV, which corresponds to ± 0.003 psi, for ~1 second, and then returned the voltage to its baseline value.

SOM References

S1. D. C. Duffy, J. C. McDonald, O. J. A. Schueller, G. M. Whitesides. *Anal. Chem.* **70**, 4974 (1998).

S2. P. J. A. Kenis, R. F. Ismagilov, G. M. Whitesides. *Science* **285**, 83 (1999).

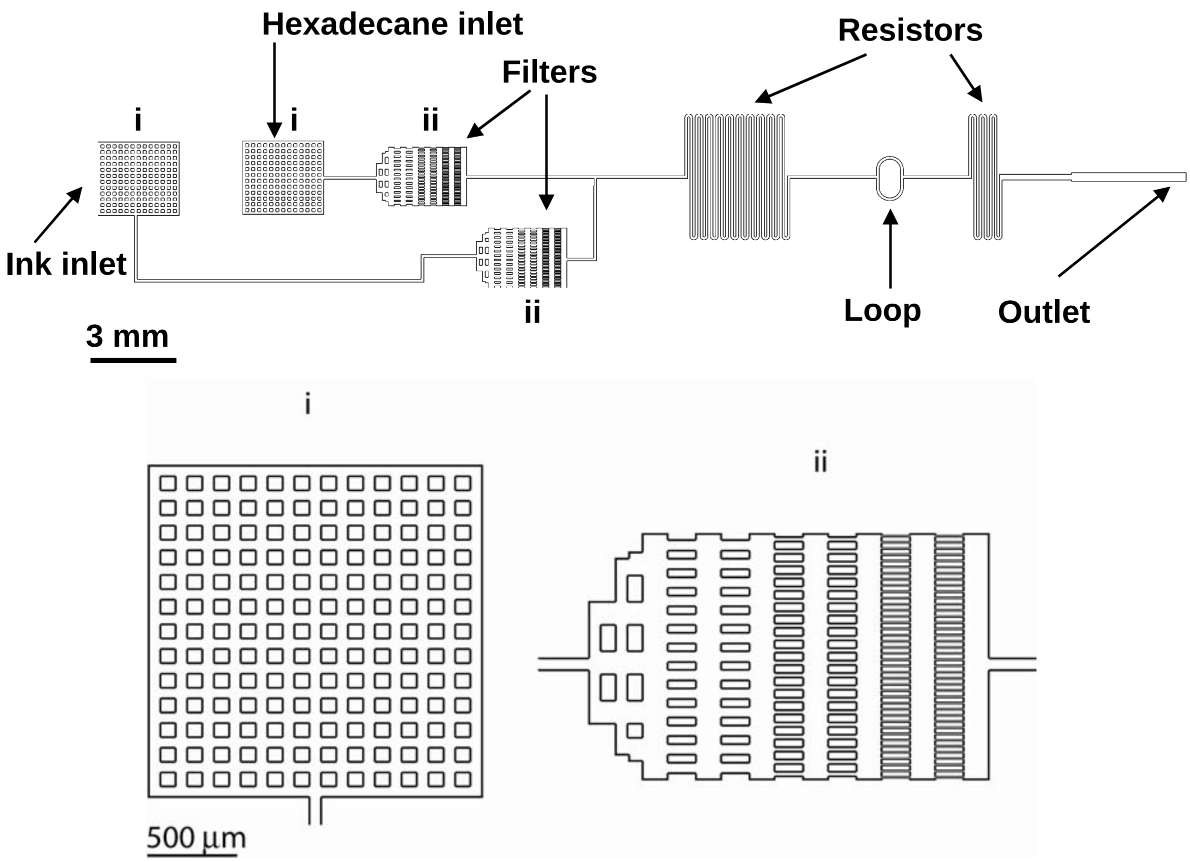
S3. J. C. McDonald *et al. Electrophoresis* **21**, 27 (2000).

S4. D. Qin, Y. N. Xia, G. M. Whitesides. *Adv. Mater.* **8**, 917 (1996).

S5. Y. Xia, G. M. Whitesides. *Angew. Chem. Int. Ed.* **37**, 551 (1998).

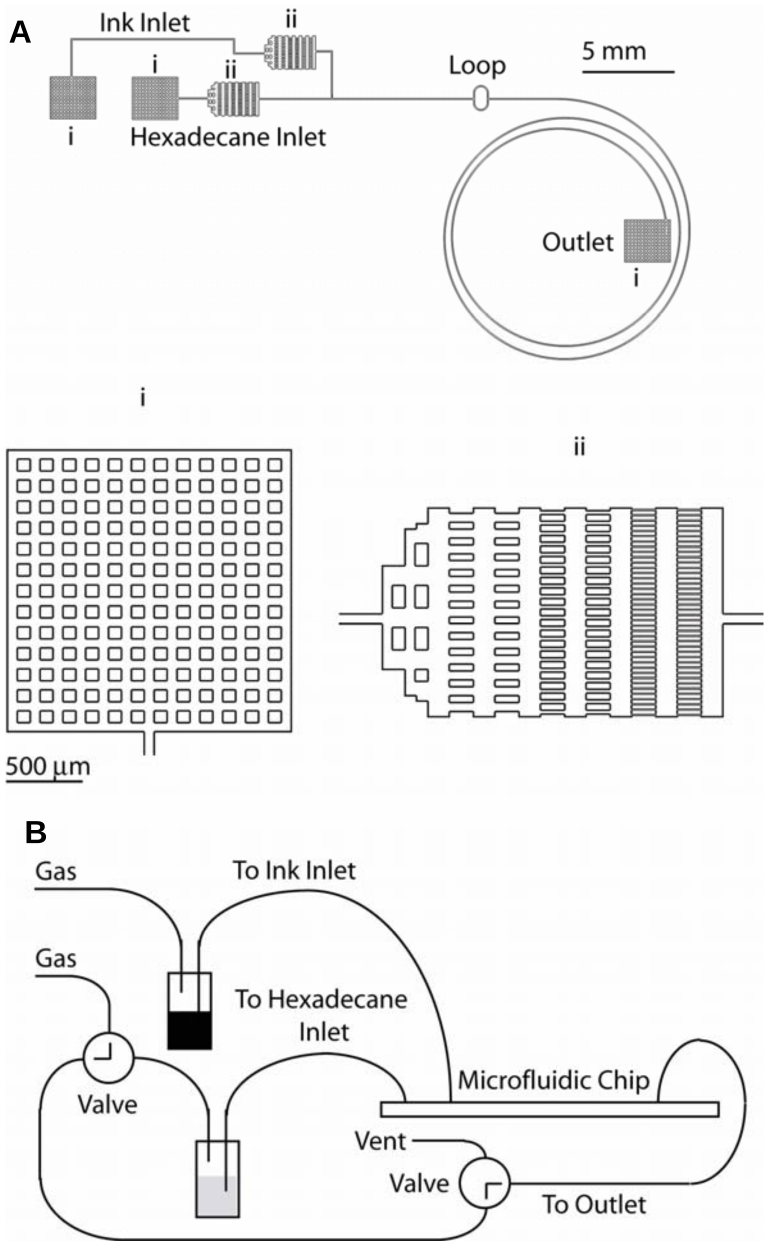
S6. M. K. Chaudhury, G. M. Whitesides. *Langmuir* **7**, 1013 (1991).

Figure S1



A schematic diagram of the system we used to obtain the bifurcation cascade. The insets i and ii are diagrams of filters that were used to trap particles that could have clogged the device. The square posts in the ink and hexadecane inlets provided more means to filter the liquids as they entered the device. In addition, the presence of the posts prevented the hexadecane from swelling shut the inlet. An optical micrograph of the loop in this device is shown in Fig. 1B.

Figure S2



A. A schematic diagram of a system that we used to demonstrate reversibility. The droplets were stored in the spiral as they move towards the outlet. Reversing the direction of flow through the system sent the droplets back through the loop towards the inlets. As in Fig. S1, the insets i and ii are diagrams of filters that were used to trap particles that could have clogged the device. The filter depicted in i also prevented the hexadecane from swelling shut the inlet. B. A diagram of the plumbing that controlled the direction of flow through the device. Both three-way valves kept one route closed and connected the other two. When the valves were set according to the configuration depicted in the figure, the nitrogen gas was applied to the outlet of the microfluidic chip.