



Microfluidic Memory and Control Devices

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This results in a lower volume. The difference between the two phases lies in the phonon dynamics. Although the phonons in δ Pu are fairly harmonic, our calculation reveals substantial instability in several modes of ϵ Pu. This anharmonicity results in a much higher phonon entropy, which in turn stabilizes the ϵ -Pu phase at high temperatures.

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Microfluidic Memory and Control Devices

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We demonstrate microscopic fluidic control and memory elements through the use of an aqueous viscoelastic polymer solution as a working fluid. By exploiting the fluid's non-Newtonian rheological properties, we were able to demonstrate both a flux stabilizer and a bistable flip-flop memory. These circuit elements are analogous to their solid-state electronic counterparts and could be used as components of control systems for integrated microfluidic devices. Such miniaturized fluidic circuits are insensitive to electromagnetic interference and may also find medical applications for implanted drug-delivery devices.

Fluidics, the construction of fluidic devices similar to electronic circuits, was an active field of research technology in the 1960s and early 1970s. Fluidic circuits were an alternative to vacuum tubes and solid-state electronics (1, 2), and advanced fluidic devices such as oscillators, flip-flops, amplifiers, and logic gates were built and integrated into sophisticated control systems (1, 2). Fluidic circuits ultimately lost the competition with semiconductor electronics in part because they could not be easily miniaturized, placing severe limits on their size and ultimate complexity. Miniaturization was not feasible because the devices are based on nonlinear inertial flow phenomena at high Reynolds numbers, Re (3). When the size is reduced, achieving sufficiently large Re to generate these nonlinearities requires increasingly high

driving pressures and becomes impractical at the submillimeter scale.

With the advent of miniaturized plumbing and other microfluidic technologies, there is an increased need for logic and control systems to operate such devices without the use of external electronics or interfaces. Although there are some examples showing how complexity and nonlinearity can be introduced into microfluidic devices through the use of multiphase flows (4) or chemically responsive materials (5), few logic, memory, or control elements have been demonstrated. Here, we show that major nonlinear flow effects can be achieved in microfluidic devices with a single fluid and no moving parts. We demonstrate two microscale nonlinear fluidic devices that can operate at arbitrarily low Re : a flux stabilizer, which is analogous to an electronic constant-current source, and a bistable flip-flop, which is analogous to a digital flip-flop memory.

The nonlinearity required for the operation of these devices derives from the use of a working fluid with nonlinear mechanical properties: a viscoelastic polymer solution

(6). The elastic properties of solutions of flexible polymers can lead to many interesting nonlinear phenomena, including purely elastic flow instabilities (7) and elastic turbulence (8). The strength of nonlinear elastic effects depends on the Weissenberg number (5) $Wi = \lambda \nabla V$, where ∇V is the rate of deformation in the flow and λ is the polymer relaxation time. These elastic effects are essentially independent of Re and should not diminish when the device is miniaturized (6), but such transitions have not previously been observed in microscopic flows. In this work, we found substantial transitions in microchannels even when using polymer solutions with such low viscosities and short relaxation times that viscoelastic effects would be difficult to measure in macroscopic experiments.

The flux stabilizer that we constructed is shown in Fig. 1. It is a nonlinear fluidic resistor, where the volumetric flux rate, Q , through a channel of a special shape depends on the applied pressure in a nonlinear fashion. The channel is a chain of halves of broad ellipsoidal rings interconnected by narrow bottlenecks (9), thus forcing the working fluid through a sequence of expansions and contractions (10). When a fluid element passes through a contraction, it becomes extended along the flow direction, as do the polymer molecules within it. If the rate of extension, $\dot{\epsilon}$, is sufficiently large compared to $1/\lambda$, the polymer molecules can unravel (11). Their contribution to the flow resistance grows quickly and nonlinearly, and the apparent viscosity of the polymer solution may increase by a few orders of magnitude (12). Transition to this nonlinear flow regime is independent of Re and should occur when Wi is greater than 0.5 (6, 11, 12).

The dependence of Q on the pressure drop per segment, ΔP (13), is shown in Fig. 2. Ini-

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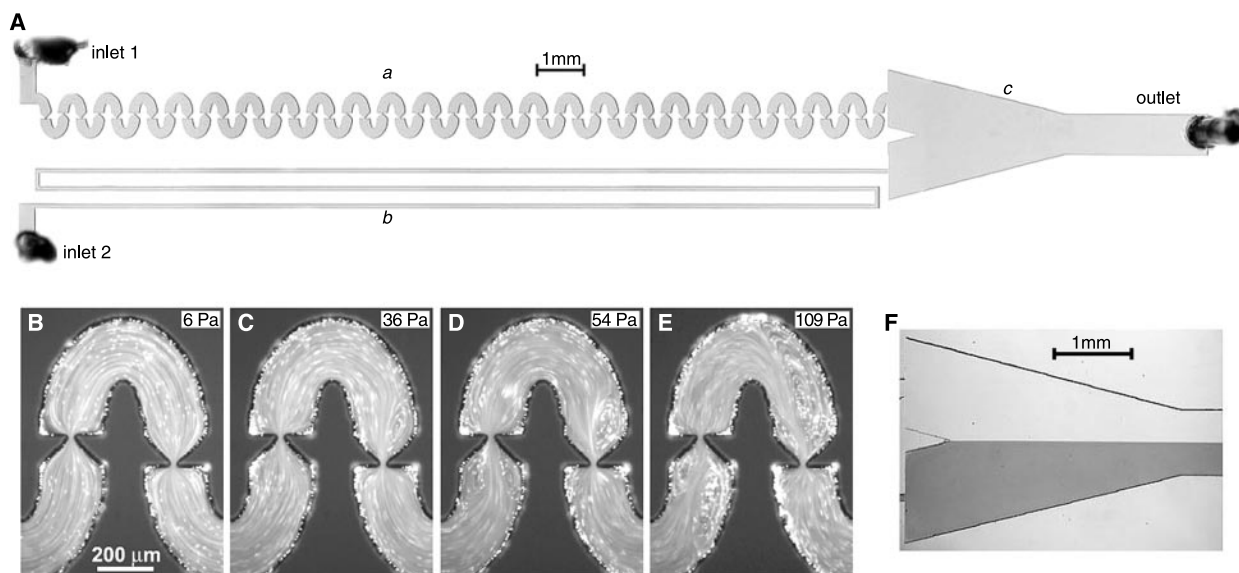


Fig. 1. (A) An image of the nonlinear resistor. The microfluidic setup is planar and has a uniform thickness of $h = 100 \mu\text{m}$. Nonlinear channel a is a chain of 46 identical segments (the adjacent segments are mirror images of each other). A segment is a half of an ellipsoidal ring with inner half axes of 185 and $65 \mu\text{m}$ and outer half axes of 410 and $290 \mu\text{m}$. The bottleneck contractions between the segments have $d \approx 30 \mu\text{m}$. The rectilinear channel b has a width of $90 \mu\text{m}$ and a total length of about 48 mm . Channel a is the functional nonlinear device, whereas

auxiliary channel b and the comparator region, c, serve to make a differential in situ measurement. (B to E) Streak lines of typical flow patterns in nonlinear channel a at different pressure drops per segment. A small volume fraction of $1.1\text{-}\mu\text{m}$ fluorescent beads was added for the flow visualization. (F) Flow in comparator region c when a compensated measurement of the flow rate is performed. The concentrated suspension of beads emerging from channel b makes a clear straight boundary with the polymer solution in the middle of the region.

tially the flux grows linearly with the pressure, but when Q reaches about 15 nl s^{-1} a flow transition occurs. It is manifested in a slower growth of the flux and in a sharp increase of the differential flow resistance dP/dQ . If we imagine a planar radial flow in front of the contractions, the maximal rate of extension can be estimated as $\dot{\epsilon} \approx Q/(2hd^2)$. Here, $h = 100 \mu\text{m}$ is the thickness of the channel, and $d \sim 30 \mu\text{m}$ is the width of the contraction. At the transition, $\dot{\epsilon}$ is about 75 s^{-1} , which gives an estimate of $Wi \approx 1$. Re , which is equal to $Q/(h\nu)$ (where $\nu = 0.026 \text{ cm}^2 \text{ s}^{-1}$ is the kinematic viscosity), has a value of about 0.06 , and thus inertial effects do not contribute to the transition.

The nonlinearity in the resistance of the channel is striking and, to our knowledge, does not have any counterpart in the fluidic devices constructed for high Re flows. In the nonlinear regime ($Q > 18 \text{ nl s}^{-1}$), the differential resistance dP/dQ is always at least 20 times higher than that in the linear regime. When the pressure is reduced by a factor of 10, from 220 to 22 Pa per segment, the flux only decreases by 38% . Similarly, when ΔP is doubled from 50 to 100 Pa , the flux increases by only 7% and dP/dQ in this plateau region is about 35 times higher than at low flow rates. Thus, the nonlinear resistor can indeed be considered as a constant current control device that stabilizes the flux at $Q = 18.7 \pm 0.6 \text{ nl s}^{-1}$ for $\Delta P = 50 - 100 \text{ Pa}$ and $Q = 20 \pm 5 \text{ nl s}^{-1}$ for $\Delta P = 22 - 220 \text{ Pa}$.

The flow patterns in the channel change significantly with the flow rate. The linear regime is a laminar forward flow (Fig. 1B). Above

the transition, recirculating vortices develop near the bottleneck entrances (Fig. 1C). As the pressure increases, they fluctuate and grow in size, ultimately becoming three dimensional (Fig. 1, D and E). Similar vortices and nonlinear growth of flow resistance at low Re were previously found in macroscopic channels that had contractions using polymer solutions with substantially higher viscosities and longer relaxation times (6, 14–16). However, the observed growth was not as dramatic as in Fig. 2, and no distinct plateaus were reported.

The second set of nonlinear microfluidic devices is shown in Fig. 3. They are the flip-flop memory: a switchable bistable system with two metastable flow states. For the flip-flop in Fig. 3A, the two flow states generate different fluxes through the two outlets, whereas for the flip-flop in Fig. 3B they create differential pressures. Both devices have the same active element, shown in the insets in Fig. 3, A and B. It is a “crossroad,” where the liquid is fed from two inlets and from where it is evacuated to two outlets. The two channels leading to the outlets are identical, whereas the feeding channels have distinct geometries. The channel originating at inlet 1 is relatively broad, and the major part of the flow resistance from inlet 1 to either outlet (Fig. 3A) is due to the contraction at the exit from the crossroad (Fig. 3A, inset), where polymer molecules are stretched and large elastic stresses are created at high Wi . In contrast, the polymer solution fed from inlet 2 passes through a contraction and an extensional flow region before it enters the crossroad. Thus, the polymer

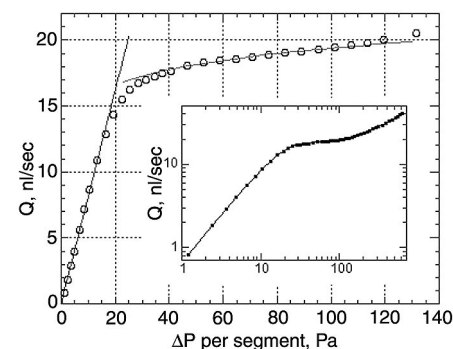


Fig. 2. Flow rate, Q , through the nonlinear resistor as a function of the pressure drop per segment, ΔP . A straight line and a power law curve, $Q \sim \Delta P^{0.095}$, are drawn in the linear and the plateau regions, respectively, to guide the eye. (Inset) Dependence of Q on ΔP in a broader range in logarithmic scale.

molecules entering the crossroad are already extended, and less additional energy and pressure drop are required to squeeze this “pre-stretched” polymer solution through the exit contraction.

Typical flow patterns at large Wi in the flip-flop from Fig. 3A are shown in Fig. 3, C and D. These are the two metastable states, which occur at the same pressures at the inlets and the outlets. The outlet pressures are also always equal, so the system is truly symmetric with respect to the outlets. One can see a notable separation between the streams: Almost all the liquid coming from a given inlet flows to a

Fig. 3. (A) A snapshot of a microfluidic flip-flop, where the flow rate through the two outlets changes between two metastable flow states. The inlet 2 channel has a width of $90\text{ }\mu\text{m}$ and a $30\text{-}\mu\text{m}$ contraction at the "crossroad" entrance, whereas the two contractions at the crossroad exits are $37\text{ }\mu\text{m}$ wide. The auxiliary inlets, comparator (comp). 1 and comp. 2, and wide comparator regions near the outlets serve the purpose of measuring the rate of flow from the crossroad to the outlets by the same method as for the nonlinear resistor (13). (Inset) Enlarged image of the active crossroad region. (B) A snapshot of a flip-flop with modulation of pressure between the metastable states. The channels leading from the crossroad exits to the outlets are $90\text{ }\mu\text{m}$ wide and 15 mm long. The pressure is measured locally, near the crossroad exits, with the aid of two auxiliary inlets, comp. 1 and comp. 2. They are connected to the outlet channels by high-resistance narrow ducts, $5\text{ }\mu\text{m}$ by $35\text{ }\mu\text{m}$ by $450\text{ }\mu\text{m}$, seen in the upper inset and do not disturb the main flow. The pressure at comp. 1 and comp. 2 is tuned hydrostatically until flow through the ducts is stopped, which gives a precision of about 1 Pa in the local pressure measurements. (C and D) Streak lines of flow

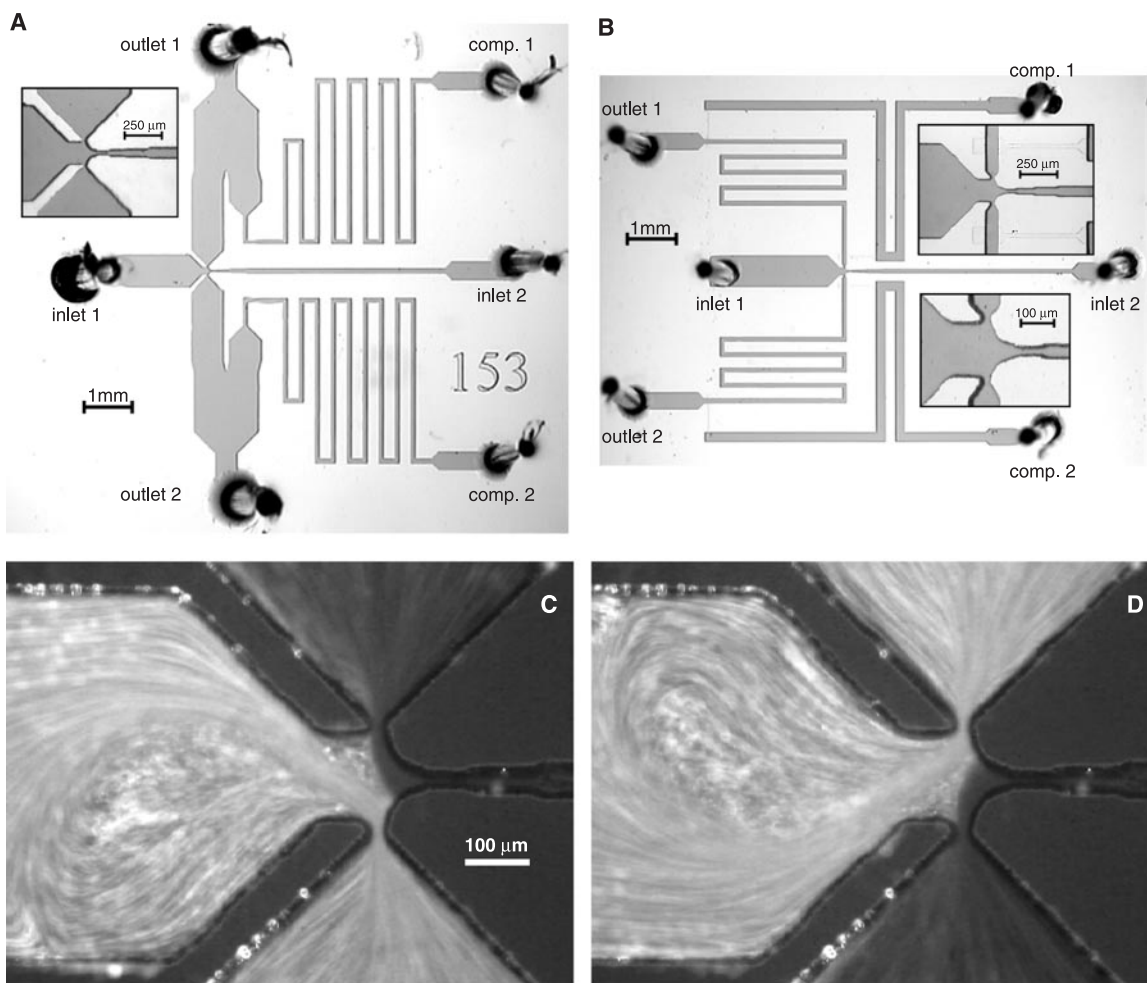
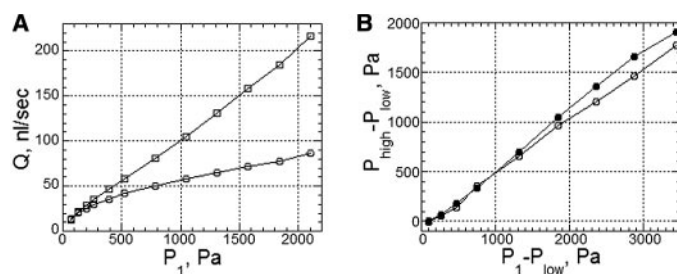


Fig. 4. (A) Flow rate, Q , through the outlets in the flip-flop from Fig. 3A as a function of the pressure difference between inlet 1 and the outlets. The pressure applied to inlet 2 was typically about eight times higher, and it was tuned for the best performance. The upper curve (squares) shows Q averaged between the two outlets in the "high" state, when the liquid coming to the outlet is mainly from inlet 2 (the outlet 1 in Fig. 3A and the outlet 2 in Fig. 3B). The lower curve (circles) shows the average flow rate in the "low" state. (B) Modulation of the pressure between the "high" and "low" states for the two exits of the flip-flop in Fig. 3B (open and solid circles) plotted as a function of the pressure difference between inlet 1 and the exit in the "low" state. An exit is "high" when flux from inlet 2 is directed to it.



single outlet. The streak-line patterns in Fig. 3, C and D, look very much like mirror images of each other. They feature large recirculating vortices in front of the crossroad exit contractions in the stream originating from inlet 1.

The plot in Fig. 4A shows that the two metastable flow states illustrated in Fig. 3, C

and D, differ by Q through the two crossroad exits. The curves for the "high" and the "low" state of an outlet start to diverge at a Q of about 20 nl s^{-1} , corresponding to $Wi \approx 1.3$. Consistent with the above argument, the outlet where the stream from inlet 2 is directed has a higher flux rate. At the highest driving pressure tested,

in the active region in the two metastable states. To make the injected liquids distinguishable, we added dye to the solution fed to inlet 2. Otherwise, the polymer solutions coming from the two inlets are identical, with the same small volume fraction of suspended fluorescent beads.

the difference between flux rates in the "high" and "low" states was a factor of 2.5. The highest flow rate was about 200 nl s^{-1} , so Re was always below unity and inertial effects were minor. No bistability was observed with Newtonian fluids in the microchannel, in which case flux from the inlets was always evenly split between the two outlets.

The two states of the flip-flop are stable to small perturbations and persist as long as the flow continues. A transition can occur, however, when a sufficiently large perturbation is applied. A fast and reliable way to switch between the states is by briefly blocking one of the outlets (and forcing all the flux to go through the other), for example by applying a pulse of a high pressure to it, which can be as short as 30 ms at high flow rates. Independent of the initial state of the flip-flop, a pressure pulse applied to outlet 1 brings it to the flow state in Fig. 3D (outlet 1, low; outlet 2, high), and a pulse applied to outlet 2 results in the state shown in Fig. 3C. Therefore, the flow originating from inlet 2 keeps its trajectory, whereas the flow from the inlet 1 is

diverted to the outlet, which opens after the high-pressure pulse passes. At high flow rates, the time of transition between the two states (which is a measure of the storage device time delay) is around 100 ms. The stability of the narrow stream from inlet 2 is because of the large extensional stresses, resembling the phenomenon of an "open siphon" (6, 14), well known in polymer solutions.

The difference in fluxes between the two states can be converted to a difference in pressures by putting fluidic resistors (long, narrow channels) between the active element exits and the outlets, Fig. 3B. The pressure modulation between the two states is shown in Fig. 4B as a function of the total maximal pressure difference. The pressure modulation is more than 50% at highest ΔP , reaching 1800 Pa.

The transition to the functional nonlinear regime in these devices is determined by $Wi = \lambda \nabla V$, whereas the driving pressure and pressure modulation are proportional to η/λ . Therefore, the characteristic pressures should remain constant when the devices are scaled down to dimensions of a few tens of micrometers. We verified that by building functional flip-flops with four-times-smaller channel depths and active element areas. Further, by using a lower molecular weight polymer sample with smaller λ , we can increase the characteristic fluxes and pressure modulations while the response time is reduced. The use of smaller polymer molecules should also allow further miniaturization of the fluidic devices. So, we believe that a reduction in size of the devices described in this report by a factor of at least 10 is quite feasible.

We have observed purely elastic flow transitions and elasticity-generated flow patterns in dilute aqueous polymer solutions in microchannels of sub-100- μm width and demonstrated that those nonlinear flow phenomena can be used to construct functional microfluidic memory and control devices, which can work at arbitrarily low Re and are insensitive to electromagnetic noise. The pressure modulation achieved in the flip-flop is nearly sufficient to actuate other functional elements such as membrane valves (17), thus opening the possibility for fabrication of cascaded devices and multielement control circuits. If a biocompatible polymer solution is used, the flux stabilizer could find medical applications for implanted drug-delivery devices, for which a constant flux of the pharmaceutical into the patient is required (18).

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9. Microchannels were fabricated by molding an optically transparent silicon elastomer, polydimethylsiloxane (PDMS). Molds were prepared by spin coating a silicon wafer with a layer about 100 μm thick of an ultraviolet (UV)-curable epoxy (MicroChem SU8-50) and exposing it to UV light through a photomask, which had a resolution of 7.1 μm . The mold was used to make a 4-mm-thick cast of PDMS (RTV 615 by General Electric) with 100- μm -deep rectangular grooves on its surface. Finally, holes were punched through the elastomer for feeding the liquids, and the side with the grooves was bonded to a microscope coverslip by overnight baking in an 80°C oven. To fabricate the pressure sensors for the device in Fig. 3B, we performed two consecutive photolithography steps in mold fabrication; the first used a 5- μm -thick coating of SU8-2005, whereas the second was as described above.
10. A 250 parts per million by weight solution of a high molecular weight (M_w) polyacrylamide, $M_w = 1.8 \times 10^7$, from Polysciences (Warrington, PA) was used. The Newtonian solvent was an aqueous solution of 13% sucrose, 1% NaCl, and 0.1% Tween (Sigma) surfactant. The sucrose was added to bring the solution density to 1.055 g cm^{-3} and to ensure neutral buoyancy of suspended polystyrene beads, and salt was added to fix the ionic contents. The solvent viscosity, η_s , was $1.37 \times 10^{-3} \text{ Pa} \cdot \text{s}$ at the room temperature of 22°C. The solution viscosity was $2.78 \times 10^{-3} \text{ Pa} \cdot \text{s}$ at a shear rate of 150 s^{-1} , suggesting a dilute-to-semidilute polymer solution. The relaxation time, λ , was estimated as 13 ms from the relaxation-time measurements of a solution of the same polymer in a more viscous sugar syrup (8), with the assumption that λ scales linearly with η_s .
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13. The pressures were generated hydrostatically with the use of two long vertical rails with precise rulers and

sliding stages. Working liquids were kept in three 30-ml plastic syringes to minimize effects of surface tension and level variation during the experimental runs. The syringes were held upright, open to the atmosphere, and connected to the inlets and the outlet by plastic tubing with an internal diameter of 0.76 mm. The pressure drop in the tubing was estimated to be below 1% of the total. The two syringes feeding the inlets were attached to the sliding stages. The difference in liquid elevation between these two syringes and the outlet syringe was measured with a precision of about 0.1 mm, corresponding to 1 Pa in pressure. During the experiment, the polymer solution was driven from inlet 1 (Fig. 1A) through the nonlinear channel a at varying pressure P_p . Pure solvent seeded with 1- μm beads at a high concentration was pumped through channel b from inlet 2 at pressure P_s . The two streams merged in region c with a sharp and laminar separation line (Fig. 1D). P_s was always tuned to bring the separation line to the same position at the center of channel c. After the dependence of P_s on P_p was found, polymer solution was pumped again through channel a at varying Q with the use of a syringe pump. Again, P_s was tuned to put the separation line at the center, and a smooth curve connecting Q and P_s was obtained. Thus, we could calculate the dependence of Q on P_p with an estimated relative error of about 0.5% without any direct measurements of the flow velocity. The error is mostly due to uncertainty in the tuning of P_s .

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A Mesoscale Iron Enrichment in the Western Subarctic Pacific Induces a Large Centric Diatom Bloom

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We have performed an in situ test of the iron limitation hypothesis in the subarctic North Pacific Ocean. A single enrichment of dissolved iron caused a large increase in phytoplankton standing stock and decreases in macronutrients and dissolved carbon dioxide. The dominant phytoplankton species shifted after the iron addition from pennate diatoms to a centric diatom, *Chaetoceros debilis*, that showed a very high growth rate, 2.6 doublings per day. We conclude that the bioavailability of iron regulates the magnitude of the phytoplankton biomass and the key phytoplankton species that determine the biogeochemical sensitivity to iron supply of high-nitrate, low-chlorophyll waters.

Low iron bioavailability is thought to be one of the causes of high-nitrate and low-chlorophyll (HNLC) ocean waters (1, 2) and may play a

major role in linking global climate change and ocean productivity (3, 4). Recent mesoscale iron enrichment experiments conducted in the equa-