

Optical waveguiding using thermal gradients across homogeneous liquids in microfluidic channels

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This letter describes the design and operation of a liquid-core liquid-cladding (L^2) optical waveguide composed of a thermal gradient across a compositionally homogeneous liquid flowing in a microfluidic channel at low Reynolds number. Two streams of liquid at a higher temperature (the cladding) sandwich a stream of liquid at a lower temperature (the core). This temperature difference results in a contrast in refractive index across the width of the channel that is sufficient to guide light. The use of a single homogeneous liquid in this L^2 system simplifies recycling, and facilitates closed-loop operation. Furthermore, with radiative and inline heating of the liquids, it should be possible to reconfigure this optical system with considerable flexibility. © 2006 American Institute of Physics. [DOI: 10.1063/1.2170435]

This letter describes the design and operation of an optical waveguide in which the difference in refractive index between the core and cladding regions arises from a thermal gradient across a homogeneous liquid flowing in a microfluidic channel. The change in refractive index across the channel reflects the temperature profile within the channel; this structure in refractive index forms the core and cladding of a liquid-core, liquid-cladding (L^2) waveguide. The ability to generate transient optical interfaces by local heating of flowing fluids presents a new approach to reconfigurable micro-optical systems.

There are a number of prior demonstrations of adaptable optical components containing fluid and/or deformable optical elements. These include elastomeric lenses and gratings,¹ liquid-core solid-cladding (LS) waveguides,^{2–6} liquid-core liquid-cladding waveguides,⁷ all-liquid lenses,⁸ reconfigurable microfluidic light sources,^{9,10} microfluidic lasers,^{11,12} and other optical components.^{13–15} We have been especially interested in L^2 waveguides. These optofluidic devices are waveguides in which the core and cladding regions are composed of two liquids with different composition (and refractive index) flowing in a microchannel at low Reynolds number (and thus laminarily, without turbulence) at constant temperature. A difference between L^2 waveguides and conventional solid and LS waveguides is that it is practical to reconfigure the properties and functions of the L^2 systems in real time by adjusting rates of flow and composition of liquids. The laminar flow of the liquids in a microchannel also ensures the smoothness of the optical interfaces, and makes their fabrication particularly simple.⁷

Here, we describe a type of L^2 waveguide in which the core and cladding liquids are the same liquid, but introduced into the device at different temperatures. Thermally defined optical structures are potentially more versatile than ones defined by chemically distinct components for several reasons: (i) It is easy to generate temperature gradients perpendicular to the direction of flow as well as parallel to it (the latter configuration is not easily accomplished with chemical gra-

dients). (ii) The use of a single common liquid simplifies recycling, and facilitates long-term continuous operation with a limited supply of liquid. (iii) Since heat can be supplied radiatively (e.g., with in-line heating elements), thermally based devices have the potential to adopt more flexible geometries or configurations than optofluidic systems that rely solely on mass transfer to form optical structures.

Thermally based L^2 waveguides also have a prominent disadvantage: Since thermal diffusion is more rapid than molecular diffusion, it is more difficult to maintain the difference in refractive index required for guiding of light with thermal gradients than with chemical gradients. This unfavorable characteristic can be circumvented (at least in part) by maintaining a high rate of flow of liquids in the microchannel; a short transit time of liquid through the channel minimizes the extent of diffusion across the width of the channel. This letter describes optical waveguides having gradients in temperature and refractive index perpendicular to the long axis of the channel.

In principle, any material can be used to make thermally defined optical structures, provided there is a large enough thermo-optical (TO) coefficient; that is, change in refractive index with change in temperature, or dn/dT . To establish a stable thermal gradient, it is also necessary to maintain the system away from the thermal equilibrium by balanced use of flows, heat sources, and sinks. Microfluidic systems are ideal for this application, because the heat can be readily supplied by injecting hot liquid and dissipated by radiation or contact with a cooled element during flow down a channel. Recent biological studies have already used thermal gradients in microfluidic channels.¹⁶

As temperature and refractive index of a liquid are almost always inversely related [Fig. 1(b)], we achieved light-guiding by constructing a device with a central cold (21 °C) stream of liquid sandwiched between two adjacent hot ≥ 37 °C streams in a single channel. We control the initial difference in temperature and the rate of flow of the liquid, to control the lateral diffusion of heat across the liquid during its transit of the device. We can, therefore, control the contrast in refractive index ($\Delta n = n_{\text{core}} - n_{\text{cladding}}$) across and along the channel.

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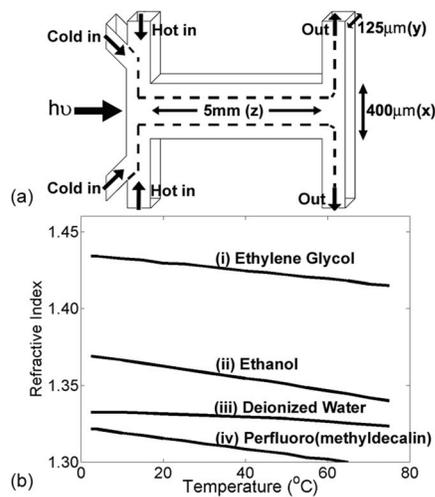


FIG. 1. (a) Schematic diagram of the microfluidic channel used for thermally generated optical waveguides. The length, width, and height of the channel are 5 mm, 400 μm , and 125 μm , respectively. The dotted line shows the boundary between hot and cold liquids under laminar flow conditions. (b) Graph of refractive indices measured using benchtop Bausch and Lomb refractometer as a function of temperature for: (i) ethylene glycol, (ii) ethanol, (iii) deionized water, and (iv) perfluoro(methyldecalin). The average slopes or TO coefficients ($dn_D/dT, K^{-1}$) are -2.6×10^{-4} (ethylene glycol), -4.0×10^{-4} (ethanol), -1.2×10^{-4} (water), and -3.4×10^{-4} [perfluoro(methyldecalin)], respectively.

We demonstrate the concept of optical waveguiding in thermally generated L^2 waveguides using several homogeneous liquids displaying a range of TO coefficients, including water, ethylene glycol, ethanol, and perfluoro(methyldecalin). We examine the temperature differential and rates of flow required to establish sufficient contrast in refractive index to guide light.

Figure 1(a) shows the design of the microfluidic device we used to evaluate the performance and characteristics of the thermal liquid waveguide. We fabricated the microfluidic channel in PDMS using standard procedures.¹⁷ The width (x), height (y), and length (z) of the channel were 400 μm , 125 μm , and 5 mm, respectively. The liquid with higher temperature was pumped into the two cladding inlets, while the liquid with lower temperature was pumped into the core inlets. The flow was pressure-driven and maintained with syringe pumps. Light from a laser diode (635 nm) or a quartz halogen lamp was coupled into the waveguide using a single-mode optical fiber [nominal numerical aperture, (NA)=0.11]. We imaged the light exiting the waveguide on a charge coupled device (CCD) camera through a microscope eyepiece (10 \times) and objective (10 \times). The liquid core was injected at 21 $^{\circ}\text{C}$ (room temperature) and the liquid claddings were preheated within a water bath and injected at temperatures between 30 $^{\circ}\text{C}$ and 80 $^{\circ}\text{C}$. Figure 1(b) shows a plot of refractive indices as a function of temperature for various liquids, measured with a benchtop Bausch and Lomb refractometer. The TO coefficient (K^{-1}) of each liquid can be extracted from these plots: $dn_D/dT = -1.2 \times 10^{-4}$ (water), -2.6×10^{-4} (ethylene glycol), -3.4×10^{-4} [perfluoro(methyldecalin)], and -4.0×10^{-4} (ethanol). The trend in these values is consistent with what is expected from reported thermal expansion coefficients (α, K^{-1}): 2.0×10^{-4} (water), and 6.5×10^{-4} (ethylene glycol), and 11×10^{-4} (ethanol).¹⁸ Thus, as expected, the TO coefficient correlates with α .

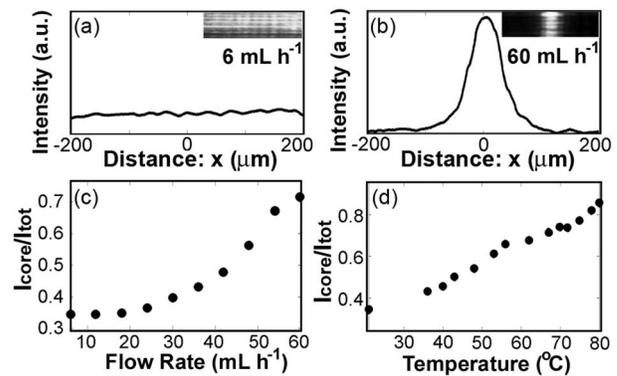


FIG. 2. (a) and (b) Plots of average intensity (arbitrary units, a.u.) of a digital micrograph taken of the light output of the water L^2 waveguide at the outlet of the microfluidic channel viewed through the transparent window for nonwaveguiding and waveguiding under various rates of flow and temperature settings. The insets show the digital micrographs used as the sources for the plots (data were smoothed prior to integration). In each case, the core was water at 21 $^{\circ}\text{C}$ and the cladding was water heated to 80 $^{\circ}\text{C}$. Total rates of flow were 6 mL h^{-1} (a) and 60 mL h^{-1} (b), respectively. (c) Plot of intensity ratio ($I_{\text{core}}/I_{\text{total}}$) at the light output as a function of total flow rate ($T_{\text{core}}=21^{\circ}\text{C}, T_{\text{cladding}}=72^{\circ}\text{C}$). (d) Plot of intensity ratio ($I_{\text{core}}/I_{\text{total}}$) as a function of temperature in the cladding region (total rate of flow=60 $\text{mL h}^{-1}, T_{\text{core}}=21^{\circ}\text{C}$). The error bars in both (c) and (d) were smaller than the size of the symbols.

In order to determine the conditions required for optical waveguiding with thermal gradients, we measured the output of liquid from the waveguide shown in Fig. 1(a) at various input temperatures and rates of flow. All of the liquids in Fig. 1(b) provided an adequate change in refractive index to produce waveguiding when $\Delta T \geq 16^{\circ}\text{C}$ (at the highest linear flow rate we used -0.4 m s^{-1}). We focus on water because it is the most relevant liquid biologically, and to demonstrate that even liquids with relatively modest values of α can be readily used to form thermally generated optical waveguides.

We evaluated the performance of the waveguide by examining the intensity profiles across the channel at the output of the waveguide. The intensity values, ranging from 0 (black) to 255 (white), were taken from horizontal line scans of the output of the waveguide with the CCD camera. Figures 2(a) and 2(b) show plots of the average intensity from left to right across the channel at two different total rates of flow (6 mL h^{-1} and 60 mL h^{-1}). The ratio of flow rates of the core and cladding streams was 1:2. In this case, the cladding streams were at 72 $^{\circ}\text{C}$ and the core was at 21 $^{\circ}\text{C}$ at their respective inlets. The inset images show optical micrographs for the output of the waveguide. The image in Fig. 2(a) shows the light output when the rate of flow does *not* meet the requirement for waveguiding. The image in Fig. 2(b) shows the light output observed when the rate of flow is sufficient to maintain the thermal gradient for the whole length of the channel, and thus to meet the requirement for waveguiding.

Figure 2(c) shows the ratio of the intensity of the core to the total intensity of the channel ($I_{\text{core}}/I_{\text{total}}$) as a function of total flow rate. Higher total rates of flow reduced lateral thermal diffusion, and maintained a steeper gradient in temperature and sharper contrast in refractive index across the width of the channel than did lower rates of flow. Thus, the efficiency of light confinement with the core liquid improved with the increase in the rate of flow. Figure 2(d) shows a plot of ($I_{\text{core}}/I_{\text{total}}$) as a function of inlet temperature at constant

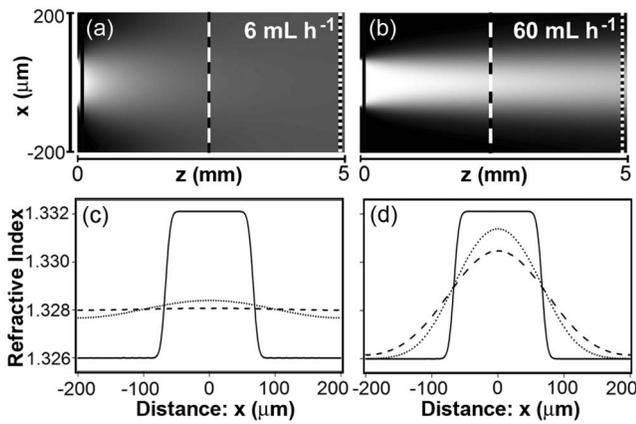


FIG. 3. Simulated two-dimensional (xz) distributions of refractive index in a 5 mm long waveguide formed by water at total rates of flow of 6 mL h^{-1} (a) and 60 mL h^{-1} (b), and $T_{\text{core}}=21 \text{ }^{\circ}\text{C}$ and $T_{\text{cladding}}=72 \text{ }^{\circ}\text{C}$. Plot of the refractive index as a function of distance from the center of the waveguide in the transverse (x) direction for three positions along a waveguide (in z direction)—i.e., the beginning (solid line), the middle (dashed line), and the end (dotted line) of the channel at total rates of flow of 6 mL h^{-1} (c) and 60 mL h^{-1} (d). The residence times are 0.12 s in (a) and 0.012 s in (b), respectively.

total rate of flow 60 mL h^{-1} . The inlet temperatures of water were: Core, $21 \text{ }^{\circ}\text{C}$; and cladding, between $21 \text{ }^{\circ}\text{C}$ and $80 \text{ }^{\circ}\text{C}$. A larger temperature difference resulted in a greater contrast in refractive index across the width of the channel, and thus a higher NA of the waveguide ($NA \propto \sqrt{\Delta n}$). The numerical aperture sets the theoretical maximum on the power of light that can be confined in the core region. With the temperature range used in the experiments, there was still an appreciable amount of light observable in the cladding regions, because the relatively low NA of our waveguides ($\lesssim 0.11$ with water at $21 \text{ }^{\circ}\text{C}$ in the core region and water at $72 \text{ }^{\circ}\text{C}$ in the cladding region) prevented complete coupling of light from the solid fiber to the input of the L^2 waveguide. The results we obtained for other liquids were similar to those for water. The minimum difference in temperature, and rates of flow, required for waveguiding were highest for water, followed (in order of their thermal expansion coefficients) by ethylene glycol, perfluoro(methyldecalin), and ethanol.

Figures 3(a) and 3(b) show the calculated profiles of refractive index due to thermal diffusion for water along the longitudinal axis (z direction) of the channel, and the profiles of temperature and refractive index across the width (x direction) of the channel at the entrance, middle, and exit of the waveguide. The temperature is calculated according to the heat conduction equation:

$$\frac{\partial T(r,t)}{\partial t} + v \nabla T(r,t) = \kappa \nabla^2 T(r,t), \quad (1)$$

where $\kappa = k/\rho C_p$ is the thermal diffusivity (and has units of $\text{m}^2 \text{ s}^{-1}$), v is the average velocity of the fluid moving down the channel (m s^{-1}), k is the thermal conductivity ($\text{W m}^{-1} \text{ K}^{-1}$), ρ is the density (kg m^{-3}), and C_p is the specific heat ($\text{J kg}^{-1} \text{ K}^{-1}$). As the thermal diffusivity of water ($\sim 1.5 \times 10^{-7} \text{ m}^2 \text{ s}^{-1}$) is higher than its mass diffusivity ($\sim 2.7 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$) by two orders of magnitude in the temperature range of interest ($21 \text{ }^{\circ}\text{C}$ – $80 \text{ }^{\circ}\text{C}$),¹⁹ diffusive broadening of the interface between the core and the cladding fluids

occurred in a shorter distance (or over a shorter residence time in the channel) than it did for waveguides composed of two miscible liquids with different composition. This broadening limited the maximum transit time and length of our waveguide to about 30 ms and 1.2 cm for flow velocity of 0.4 m s^{-1} for $400 \text{ } \mu\text{m}$ wide channels.

Thermally generated optical waveguides have several attractive features: (i) Rapid dissipation of heat in liquid systems should lead to rapid switching times when reconfiguring thermal optical structures (compared to systems that rely on mass diffusion). (ii) Any single liquid providing sufficient thermal expansion should be compatible with this system. (iii) The chemical homogeneity of the input liquids means no separation of components is required to recycle the fluids; such systems could thus be easily operated in closed loops. (iv) The properties of the waveguide are reconfigurable. By changing the total rate of flow of the liquid into the microfluidic channel, the pattern of hot and cold liquids, and the temperature difference between the core and cladding regions, we can fine-tune the contrast in refractive index across the waveguide.

These principles should also be applicable to the designs of other optical structures such as gradient index lenses, L^2 splitters or switches,⁷ and systems operating with in-line radiative heating elements.

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- ¹J. L. Wilbur, R. J. Jackman, G. M. Whitesides, E. Chang, L. Lee, and M. Prentiss, *Chem. Mater.* **8**, 1380 (1996).
- ²A. Datta, I.-Y. Eom, A. Dhar, P. Kuban, R. Manor, I. Ahmad, S. Gangopadhyay, T. Dallas, M. Holtz, H. Temkin, and P. K. Dasgupta, *IEEE Sens. J.* **3**, 788 (2003).
- ³P. Dress and H. Franke, *Appl. Phys. B: Lasers Opt.* **63**, 12 (1996).
- ⁴A. Hanning, J. Westberg, and J. Roeraade, *Electrophoresis* **21**, 3290 (2000).
- ⁵P. Mach, M. Dolinski, K. W. Baldwin, J. A. Rogers, C. Kerbage, R. S. Windeler, and B. J. Eggleton, *Appl. Phys. Lett.* **80**, 4294 (2002).
- ⁶O. J. A. Schueller, X.-M. Zhao, G. M. Whitesides, S. P. Smith, and M. Prentiss, *Adv. Mater. (Weinheim, Ger.)* **11**, 37 (1999).
- ⁷D. B. Wolfe, R. S. Conroy, P. Garstecki, B. T. Mayers, M. A. Fischbach, K. E. Paul, M. Prentiss, and G. M. Whitesides, *Proc. Natl. Acad. Sci. U.S.A.* **101**, 12434 (2004).
- ⁸S. Kuiper and B. H. W. Hendriks, *Appl. Phys. Lett.* **85**, 1128 (2004).
- ⁹B. T. Mayers, D. V. Vezenov, V. I. Vullev, and G. M. Whitesides, *Anal. Chem.* **77**, 1310 (2005).
- ¹⁰D. V. Vezenov, B. T. Mayers, D. B. Wolfe, and G. M. Whitesides, *Appl. Phys. Lett.* **86**, 041104/1 (2005).
- ¹¹S. Balslev and A. Kristensen, *Opt. Express* **13**, 344 (2005).
- ¹²Y. Cheng, K. Sugioka, and K. Midorikawa, *Opt. Lett.* **29**, 2007 (2004).
- ¹³G. Schilling, *Science* **299**, 1650 (2003).
- ¹⁴C. Kerbage and B. J. Eggleton, *Appl. Phys. Lett.* **82**, 1338 (2003).
- ¹⁵F. Cattaneo, P. Mach, J. Hsieh, T. Krupenkin, S. Yang, and J. A. Rogers, *Mater. Res. Soc. Symp. Proc.* **741**, 9 (2003).
- ¹⁶E. M. Lucchetta, J. H. Lee, L. A. Fu, N. H. Patel, and R. F. Ismagilov, *Nature (London)* **434**, 1134 (2005).
- ¹⁷J. C. McDonald and G. M. Whitesides, *Acc. Chem. Res.* **35**, 491 (2002).
- ¹⁸D. R. Lide, *CRC Handbook of Chemistry and Physics*, 83rd ed. (CRC Press, Boca Raton, FL, 2002).
- ¹⁹E. H. Abramson, J. M. Brown, and L. J. Slutsky, *J. Chem. Phys.* **115**, 10461 (2001).