Supporting Information

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SI Text

Data Acquisition and Analysis. ***Summary of data acquisition and instances of data rejection.*** We here summarize some features of our scalar image data acquisition. Further details can be found in ref. 1. The quantitative scalar field data we present in Figs. 2 and 3 are based on image sequences of 45 experiments comprising 45 applied electric fields (nominal electric fields from 333 to 1122 V/cm). These imaging experiments were strictly limited by trade-offs between camera sensitivity, frame rate, region of interest analyzed (subset of pixels used in CCD), and the amount of data that fit in the RAM of our data acquisition computer. We used a 16-bit, Peltier-cooled Photometrics CCD with on-chip gain (Tucson, AZ). We chose a frame rate of 390 Hz. The data sets each consisted of 1620 16-bit images with a CCD region of interest of only 3×512 pixels (to reduce required memory and increase data rate). One exception to this is the data of the lowest electric field (a stable case) for which we obtained only about 1000 images (and extended these data to match the duration of the other image series). The data of about half of the 12th electric field and all of the 13th electric field (at $E = 455$ and 466 V/cm) were likely corrupted by severe background illumination (we attribute this to unshielded lighting from the room). We therefore replaced the last approximately one-third of data record 12 with a copy of its first approximate one-third, and replaced all of the data record 13 with the new data record 12. In all, we replaced <3% of the data (about 1.3 of the 45 records) shown in Fig. 2.

Further details of power spectrum analysis. We performed the KPSS Test (2) to assess the stationarity of each of the experiments shown in Fig. 2. We used the kpsstest function in Matlab (Mathworks) and used parameters recommended by Kwiatkowski et al (2), including a 95% significance (alpha $= 0.05$) level. In 40 of the 45 experiments, the test showed a failure to reject the null hypothesis that the time series was trend stationary; suggesting the data were trend stationary. Four of the remaining data sets were spread apparently randomly in the low-electric field periodic regimes (at the 3rd, 14th, 16th, and 25th lowest Ra_e cases shown in Fig. 2). We attribute this to some slight fluctuation of the illumination intensity supplied by our microscope's mercury bulb. The remaining, single data series, which was suggested by the test to be nonstationary occurred within the first chaotic region in Fig. 2. However, the KPSS Test suggested the other six experiments in this same chaotic region were stationary.

Prior to performing the power spectra, we removed slight linear trends. These linear trends are common in fluorescence quantitation of electrokinetic microflows and attributable to various effects including variations in light intensity of the illumination (in our case, the mercury bulb of the microscope), effects of electrochemical reactions (at end channel reservoirs containing electrodes) on the tracer dye, and/or photobleaching of the dye. The typical linear trend in the data consisted of a variation of less than about $\pm 3\%$ of the measured mean value. The linear trend in 42 of 45 cases varied less than $\pm 7\%$ and all varied less than $\pm 11\%$.

Prior to computing power spectra on the data, we used a symmetric Hann window to minimize frequency leakage. Fig. 2 presents a map of 45 power spectra of the windowed time series up to the Nyquist folding frequency of 195 Hz.

Lastly, the data shown in Fig. 2 used a virtual point detector centered at $x/w = 2$. We integrated the intensity in a 3×3 region in this subimage as a virtual point detector for our power spectra. We also analyzed power spectra for temporal fluctuations of the scalar at following additional downstream locations: $x/w = 3.5$, 5.3, and 7.1. These power spectra maps were qualitatively very similar to those near $x/w = 2$, except for an expected attenuation of the high frequency power due to the dispersion associated with the combined effects of advection and molecular diffusion. This similarity included the harmonics of the first (low electric field) periodic regime, evidence of period doubling, the clear delination of two chaotic regions, and the harmonics of the second (high electric field) periodic regime.

1. Posner JD, Santiago JG (2006) Convective instability of electrokinetic flows in a cross-shaped microchannel. *J Fluid Mech* 555:1–42.
2. Kwiatkowski D, Phillips PCB, Schmidt P, Shin Y (1992) Testing the null hypothesis of stationarity against the alternative of a unit root. *J Ecol* 54:159–178.