Supplementary Information

Optical-resonance-assisted generation of super-monodisperse microdroplets and microbeads with nanometer precision

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1 Encoding

To encode data into droplet sizes, one needs to map a string of characters uniquely to a set of \( m \) different unordered droplet sizes. The sizes of droplets must not be too similar, so that the spectral peaks of whispering gallery modes (WGMs) are distinguishable. This limits the minimal step size which, together with the droplet size range, gives the total number of possible diameters for each droplet \( D \). Using \( m \) droplets, the number of distinct combinations is \( N = \binom{D}{m} \), allowing to encode \( \log_2 (N) \) bits of information. In order to encode information, represented by a binary integer number between 1 and \( N \), one generally needs to define a way to count all \( N \) possible sets of three diameters, which defines a discrete bijective mapping \( f \) between two sets of integers:

\[ f : \{1, 2, \ldots, N\} \rightarrow \{\{I_1, \ldots, I_m\} : I_i \in \{1, \ldots, D\}, I_i \neq I_j, \text{ for } i \neq j\}. \] (1)

To demonstrate this principle we encode the surname of Jožef Stefan, a 6-character string "STEFAN" into three droplet sizes. Using 5-bit encoding, this string is a sequence of 30 bits. In a minimalist approach, we use the last 5 bits of the standard ASCII 7-bit encoding (in matlab, the numerical representation of a character \texttt{char} is calculated by \texttt{mod(double(char), 2^5)}). This enables the use of characters, indexed in standard 7-bit ASCII by 64 through 95, corresponding to all english capital letters and 6 additional characters. For our simplified encoding we use \( m = 3 \) droplets with \( D = 3071 \) distinct diameters, which translates to 32 bits of storage. Therefore, the step size is 4 nm and the range of used diameters is \( 24.096 \mu m - 36.380 \mu m \).

Our task is to store 30 bits of information to a set of three 12-bit integer numbers \( I_i \), mapped to the three diameters \( d_i \) by

\[ d_i[\mu m] = 20 \mu m + \frac{20 \mu m}{5000} I_i, \] (2)

for \( i \in \{1, 2, 3\} \) denoting three droplets. To understand the way we stored information, we look at the data in bit-form. The first 2 bits in a binary representation of the droplet size \( I_i \) encode the position of the next 10 bits in an array of 30 bits that represent our original information, the initial string. The three droplets’ sizes \( I_i \) in binary start with 01, 10, 11, ensuring not too similar diameters and therefore distinct WGM barcodes.
In our case, each diameter stores exactly two 5-bit characters, although the encoding allows for the information of a single character to be shared between more droplets. This ensures that the method exploits all available bits independently of the character encoding, number of droplet sizes $D$ and string length. Using this simplified encoding, decoding the information from droplet sizes is also straightforward. After reading the diameters from WGMs, we order them by size, use the inverse of the mapping in Eq. 2, read the last 10 bits of each droplet size $I_i$, and stack them into a 30-bit sequence. This, if read in 5-bit encoding, decodes the original string.

2 Supplementary Figures

Supplementary Figure 1: a) Combined fluorescence and brightfield image of three representative monodispersed oil droplets with diameter of 22.75 µm. b) Corresponding WGM spectra of 8 monodispersed droplets. c) Spectral overlap of two polymerised microbeads with diameter of 21 µm. d) Corresponding SEM image of a 21 µm microbead.
Supplementary Figure 2: a) Polymerized microbeads of different diameters. Last image shows fluorescent signal of polymerised bead. Scalebar for first four images corresponds to 10 µm and 20 µm for the last two images. b) Representative WGM spectra for a 9 µm diameter microbead (left) and a 39 µm microbead (right), respectively.

Supplementary Figure 3: Long term stability of a 34 µm diameter polymerized microbead. Spectra are measured 4 and 300 days after polymerization.
Supplementary Figure 4: a) Fluorescence image of doped droplet at the tip in a microfluidic device. Fluorescently doped fluid is being constantly pushed though the microcapillary by a syringe pump. b) Brightfield image of growing droplet at tip. c) Time evolution of the WGM spectrum for a large droplet growing at the tip of a microfluidic device. d) Time lapse of microfluidic droplet generation. Arrows indicate the flow direction of outer (red) fluid and inner (green) fluid. The outer flow is initiated when the droplet reaches it target size to release it from the tip of the microcapillary. Inset: Continuous generation of smaller droplets by using a smaller capillary.