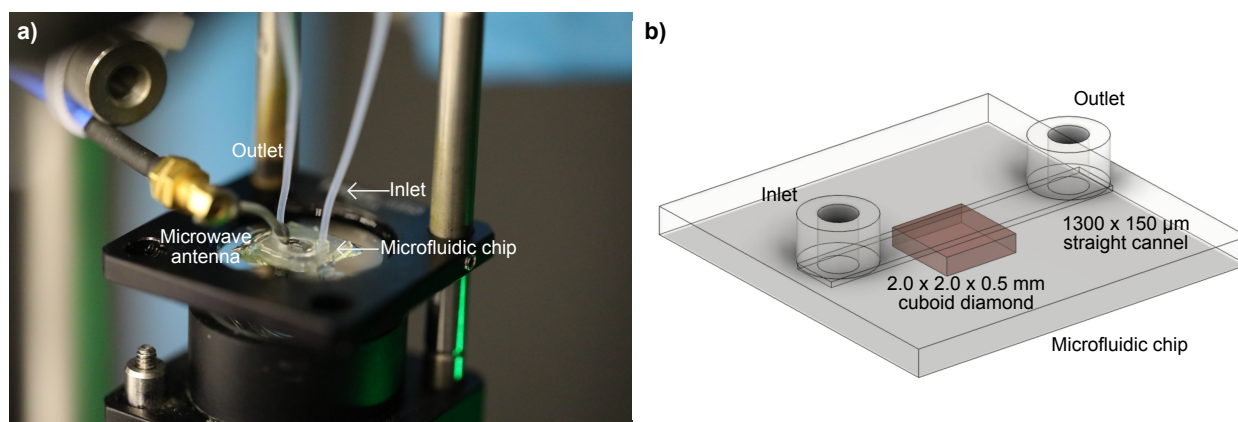# S Supplementary information

S.1 Second example chip design

Fig. S1: Second microfluidic chip design. a) Photograph of the second microfuidic chip design inside a NV center experiment. **b)** CAD image of the second chip design which incorporates a 2.0 x 2.0 x 0.5 mm cubiod-shaped diamond and a 1,300 x 150 µm straight example channel. The inlet and outlet ports in this design are on top of the channel to allow easy optical access through the diamond by, *e.g.*, solid-immersion lenses (SILs) or microscopy objectives.

The second microfluidic chip design fits (see Fig. S1) standard 2.0 x 2.0 x 0.5 mm NV center-doped cuboid diamonds, demonstrating that the microfluidic quantum sensing platform can easily be adapted to different diamond shapes and sizes. Furthermore, in this design, the inlet and outlet ports for the microfluidics are placed on top of the chip to facilitate optical access for NV center excitation and read-out from the bottom through the diamond. This geometry has the drawback that a total internal reflection geometry is more difficult to realize but allows for a more rigid chip design where no cut-out for the excitation pathway is needed (compare main text Fig. 3a). We envision applications using confocal microscopy or other nanoscale experiments, where photodegradation of biological or chemical samples is less of a concern. In this example, the microfluidic channel is 2,000 x 1,300 x 150 µm, which can be fully adapted to more complicated structures. A technical drawing of the microfluidic chip is depicted in Fig. S2c).

S.2 Technical drawings

Fig. S2 depicts simplified technical drawings of several example microfluidic chips that have been used for the microfluidic quantum sensing platform. The microfluidic chip designs are attached in the Supplementary Data as .STL files.
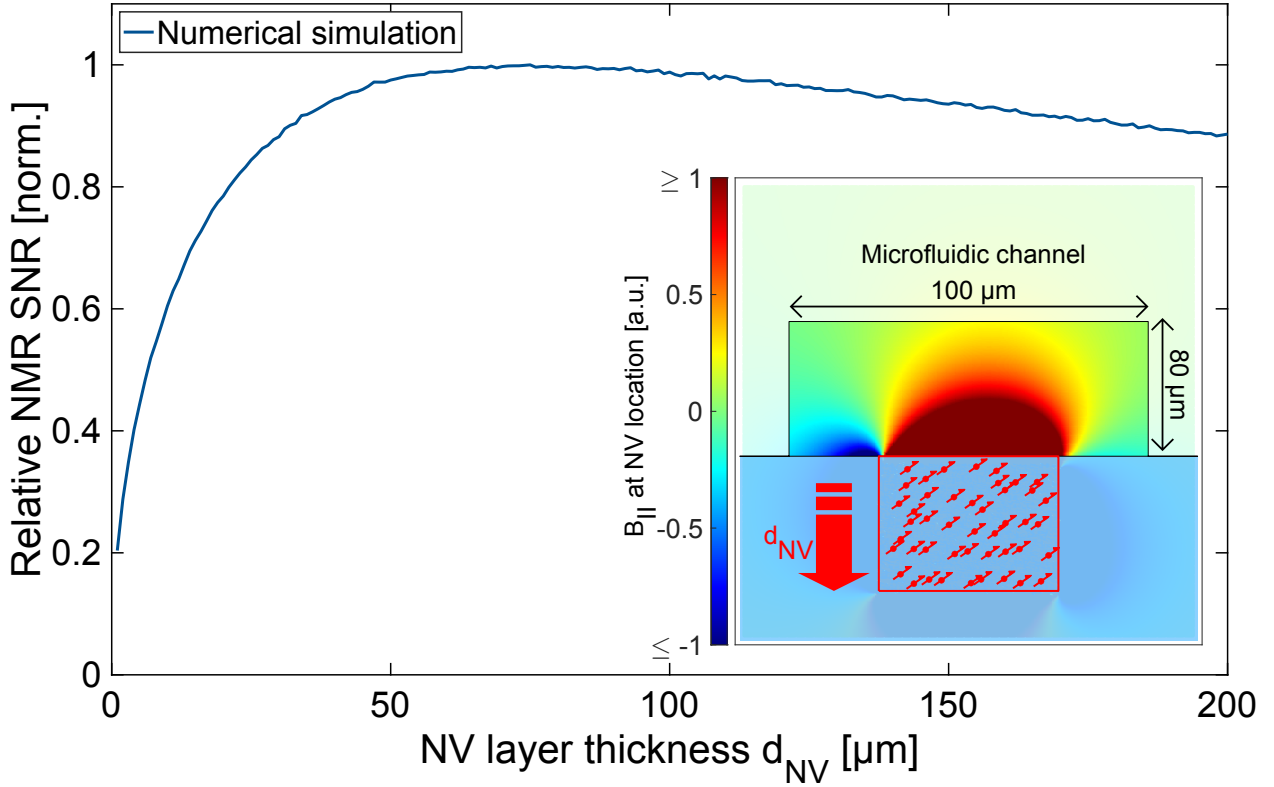
S.3 Optimization of the NV layer thickness for NV-NMR within our microfluidic chips

The microscale NV-NMR signal has a strong volume and geometry dependence [17]. In order to choose the NV-layer thickness for the highest sensitivity, we follow the approach according to Bruckmaier *et al.* [24]. Using Monte Carlo simulation, we estimate the NMR signal strength at the NV center position (red arrows, see Fig. S3), caused by the dipolar magnetic fields of nuclear sample spins (protons). These spins are confined in our 1,000 x 100 x 80 µm microfluidic channel (see Fig. 3a). The cylindrical NV center volume, *i.e.*, the sensor volume, is 45 µm in diameter (typical value in microscale NV-NMR experiments, see Fig. 3d) and has a variable height (d_{NV}) form the surface of the diamond. For each sample spin position, the dipolar field within the detection volume is projected along the NV orientation, (*i.e.*, the magnetic bias field B_0 direction).
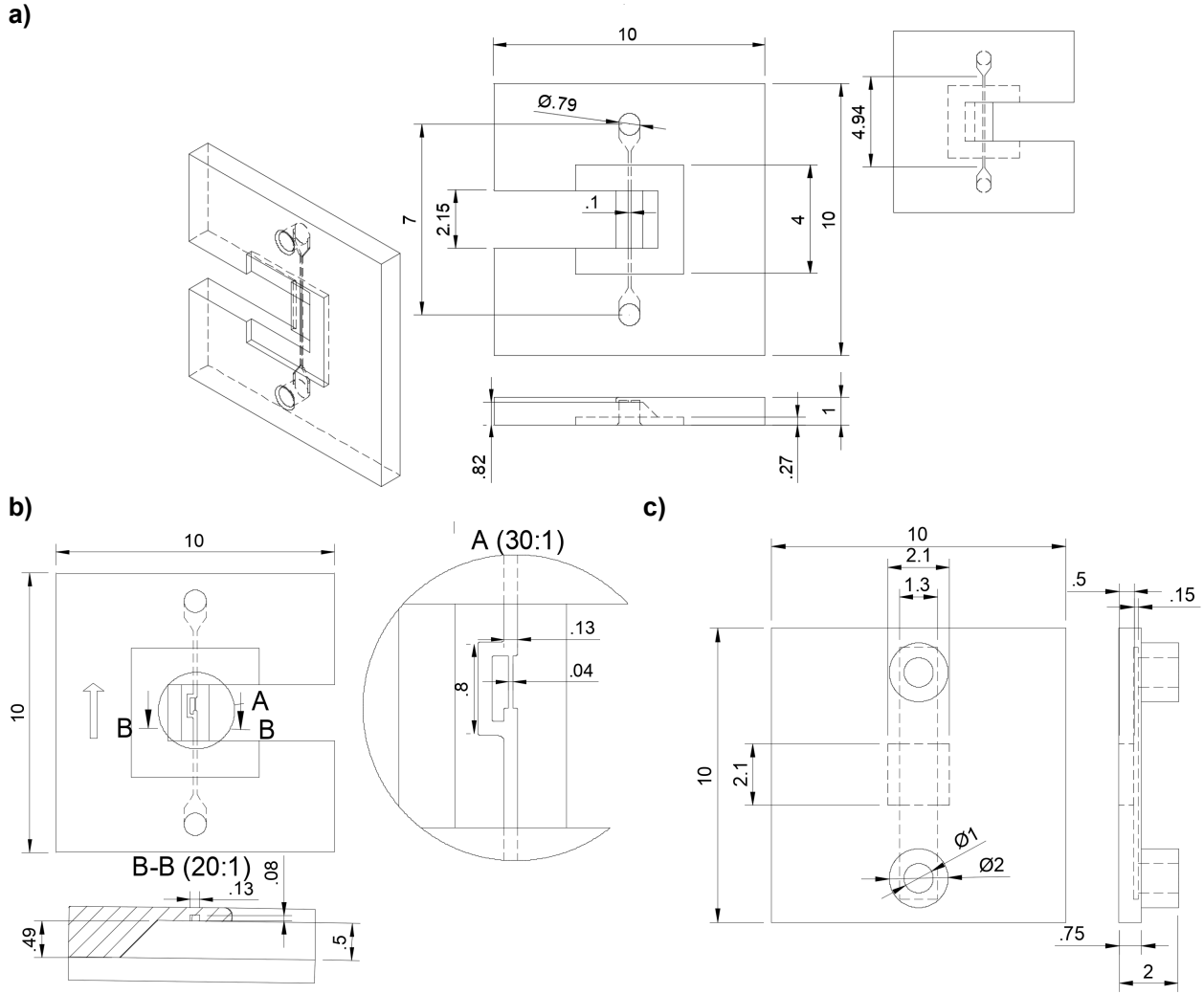
Fig. S2: Technical drawings of example microfluidic chips. a) Simplified technical drawing of the base microfluidic chip with a 100 x 80 µm straight channel (see main text Fig. 3a), **b)** of a modified microfluidic chip with a fluidic bypass channel (see main text Fig. 3b), and **c)** of the second microfluidic chip design shown in SI Section S1 (see Fig. S1).

Each point was averaged 10,000 times for this simulation with 40 randomly selected NV center positions within the volume and 32,000 randomly selected sample spins within the microfluidic channel for each average. While the average signal size of each of our NV center within the ensemble decreases with the depth d_{NV}, the sensitivity (relative NMR SNR) reaches a maximum at $\sim$ 50 - 70 µm due to the increasing number of NV centers contributing to the measurement (see Fig. S3). For that reason, we grew a 50 µm thick NV-layer for our microscale NV-NMR experiments within the channel, yielding the highest sensitivity for our microfluidic chip dimensions.

Fig. S3: Optimization of the NV layer thickness to the microfluidic dimensions for microscale NV-NMR.
Inset: Visualization of the NMR signal origin for a microscale NV-NMR experiment with a cylindrical NV center (red arrows) volume, *i.e.*, sensor volume, of 45 µm diameter and variable height (d_{NV}) at the surface of the diamond. The sample spins are contained in a 1,000 x 100 x 80 µm microfluidic channel. For each sample spin position, the dipolar field within the detection volume is projected along the magnetic bias field B_0 and averaged. Red areas depict sample spin positions with overall positive signal contribution, and the blue regions sample spin positions with overall negative signal contribution, respectively. **Graph:** Normalized signal $B_{II}\sqrt{d_{NV}}$, *i.e.* the relative sensitivity of the NV-NMR experiment, against the thickness of NV ensemble layer d_{NV} according to a numerical simulation using the Monte Carlo method.