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Ultrasonic Wave Mixing for Nonlinear Ultrasonics in a Microfluidic Capillary

Andriejus Demčenko
Division of Biomedical Engineering
University of Glasgow
Glasgow, UK
andriejus.demcenko@glasgow.ac.uk

Christian Witte
Division of Biomedical Engineering
University of Glasgow
Glasgow, UK
christian.witte@glasgow.ac.uk

Julien Reboud
Division of Biomedical Engineering
University of Glasgow
Glasgow, UK
julien.reboud@glasgow.ac.uk

Jonathan M. Cooper
Division of Biomedical Engineering
University of Glasgow
Glasgow, UK
jon.cooper@glasgow.ac.uk

Abstract—In this study we show that nonlinear ultrasonics, based on nonlinear wave mixing, can enable the measurement of the interactions between acoustic waves and a microliter liquid sample in a glass capillary microchannel. This has the potential to deliver new techniques with a high sensitivity to acoustic material properties.

Index Terms—microfluidics, nonlinear ultrasonics, wave mixing

I. Introduction

The interest in probing the properties of small liquid sample volumes (on the order of a microliter) has given rise to a range of ultrasonic techniques, which can be divided into linear [1], [2] and nonlinear [3], [4] methods. In the linear regime, samples are characterized by measuring the wave's velocity, attenuation [2] and superposition [1] while harmonic generation [4] and parametric wave mixing [3] have been used in the nonlinear regime. The nonlinear waves depend not only on the linear material properties, but also on the nonlinear properties, opening up the potential to higher measurement sensitivity, as well as enabling the characterisation of the nonlinear properties of samples.

Liquid biosamples are often only available in small volumes (usually in order of micro to nanoliters) to minimise the discomfort and risk of the sampling procedures (e.g. blood fingerprick in point-of-care diagnostic strategies), or organism size (e.g. scorpion venom). The measurement of acoustic parameters in such samples is currently challenging not only in the linear regime, but more especially in the nonlinear regime, due to the limited wave propagation path and diffraction effects. However, information about sample properties (e.g. mechanical characteristic) can add to the understanding of disease progression (cancer) or the differentiation process of stem cells in tissue engineering applications [5].

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In this work, a nonlinear ultrasonic measurement technique was used to sense fluid mixtures in a microfluidic glass capillary. We used parametric wave mixing due to its multiple advantages over the harmonic generation techniques (including controlled wave interaction volume, frequency separation, lower power [6]).

II. Microfluidic measurement setup and its operation

Fig. 1 shows the measurement setup for the characterisation of liquid samples using on nonlinear wave mixing. The setup consisted of a lithium niobate wafer (128° Y cut, diameter 10.16 cm, thickness 1 mm) with two opposing single-phase unidirectional transducers (SPUDT) of different frequencies (9.8 MHz and 11.7 MHz) to generate surface acoustic waves (SAWs). A glass capillary channel (20 mm × 5 mm × 0.4 mm) was placed in the middle of the SPUDTs. The propagating SAWs were coupled into the capillary via mode conversion through a coupling liquid. Burst signals of 5 µs (modulated by a Hamming window) with a digital delay were used to excite the initial counter propagating SAWs with the wavevectors

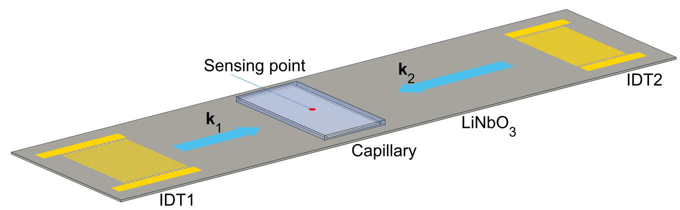

Fig. 1. Experimental setup for the sensing nonlinear ultrasonic response from viscous fluids in the capillary channel coupled to the lithium niobate ($LiNbO_3$) waveguide.

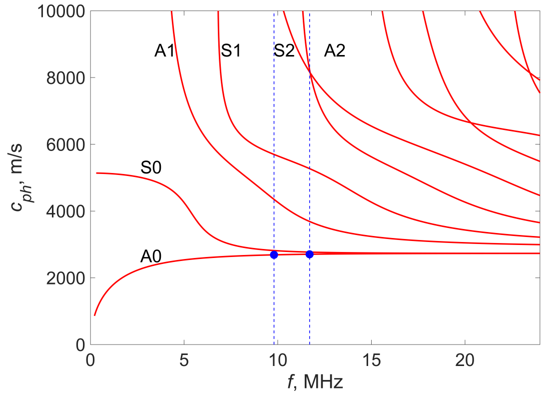

Fig. 2. Calculated guided wave modes in the capillary wall where two blue points denote the initial dominant wave modes at the 9.8 MHz and 11.7 MHz frequencies.

$\mathbf{k}_1$ and $\mathbf{k}_2$. Leaky guided waves interacted in a test liquid (water and glycerol mixtures) in the capillary channel, and generated nonlinear waves of combined (sum and difference) frequencies. The initial and sum frequency waves were detected by a needle hydrophone (diameter 0.5 mm) from the top side of the capillary channel via coupling liquid.

Fig. 2 shows dispersion curves of symmetric (S) and antisymmetric (A) guided waves in the unloaded capillary wall of 0.4 mm when longitudinal and transverse wave velocities are 6150 m/s and 2920 m/s, respectively; volumetric mass density is 2592 kg/m^3. Due to the SAW coupling to the capillary, the antisymmetric guided wave modes are most significant (A0 and A1). We focused on the analysis of the the fundamental A0 mode which is dominant in this configuration.

In fluids, classical plane wave interactions are possible when the interacting waves propagate in a line. However, these interactions occur also at different wave intersection angles [7]. When there is an acoustic loading on the waveguide, in our case the capillary filled with a fluid, the guided wave leaks energy into the fluid at critical angles θ_1 and θ_2, forming longitudinal leaky waves, see Fig. 3. These two waves intersect at the angle $\theta = \theta_1 + \theta_2$, which plays a crucial role in the nonlinear wave interactions. The interaction angle θ varies in the range 67° (water) – 90° (glycerol). This shows that the nonlinear interactions occur not only at different interaction angles in various mixtures, but the generated nonlinear wave propagates also in different directions. It has to be noted that an

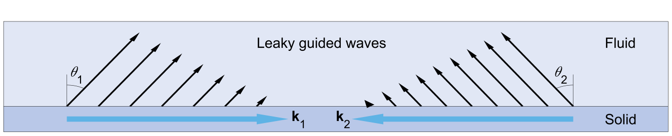

Fig. 3. Leaky guided waves in the fluid generated by guided waves propagating in a wall of the capillary.

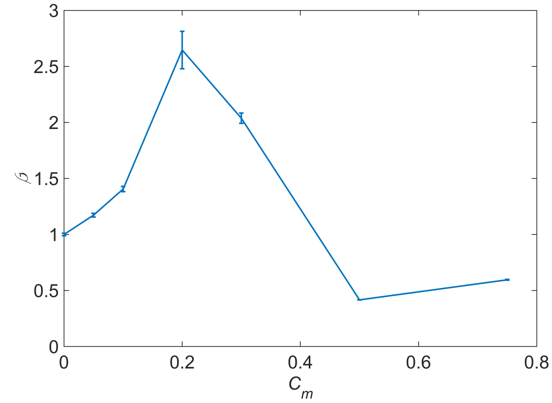

Fig. 4. Relative nonlinear ultrasonic response "beta" from a range of glycerol-water mixtures, where error bars represent the standard deviation over 3 independent measurements for each point. The measurements were normalised to water (glycerol 0%)

influence of the higher order mode (e.g. A1) should also be considered in the analysis of the nonlinear wave interactions.

III. Measurement results

Fig. 5 shows typical filtered nonlinear wave signals when the glycerol-water mixture concentration is 0 and 0.75 (C_m in % wt). Although the signal suffered from a significant absorption in the glycerol-water mixture, both of them show excellent signal to noise ratios. Fig. 4 depicts experimental results of the nonlinear ultrasonic response of the glycerol-water mixtures. The nonlinear response is presented as a relative parameter $\beta = a_3/(a_1 a_2)$, where a_i is the peak-to-peak amplitude of the initial waves 1 and 2, and the 3 denotes the nonlinear wave parameter.These results show that the complex nonlinear ultrasonic response from the glycerol-water mixture in

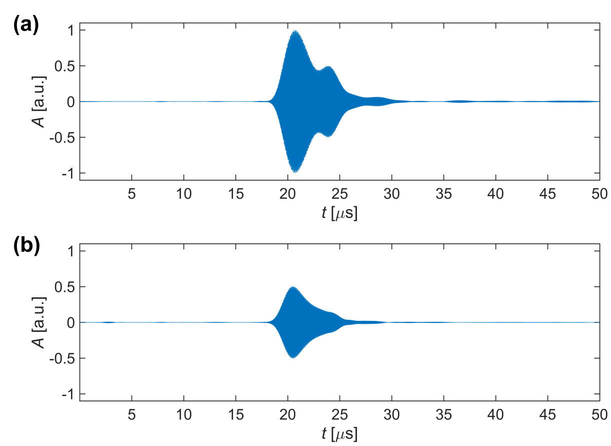

Fig. 5. Nonlinear ultrasonic signals when the capillary was filled with water (a), and glycerol-water mixture of concentration 0.75, (b). Both amplitudes of the signals were normalized by a maximum amplitude of the reference signal (water).

the capillary reached a maximum at the concentration $C_m = 0.2$. After this point, β decreased until it reached a minimum at $C_m = 0.5$. At the higher concentration, $C_m = 0.75$, a small increase was observed in β.

The complex behaviour of the nonlinear parameter could be explained by a number of factors:

- Parameters such as nonlinearity, wave velocity and viscosity increased with increasing glycerol content in the mixture [8], [9].
- Due to the changes in the wave velocity in the glycerol-water mixtures, the interaction angle θ changed for different concentrations, leading to the nonlinear wave propagating in different directions, potentially decreasing the signal obtained at the microphone.
- The resonances of the ultrasonic signals in the capillary varied with the different acoustic properties of the mixtures.

IV. Conclusions

Non-classical nonlinear wave interactions were used to probe the acoustic response of liquids in a microfluidic capillary. The experimental results showed a complex nonlinear ultrasonic response from the glycerol-water mixtures.

Our experiments demonstrated that nonlinear wave mixing can be utilized in microfluidics for sensing of nonlinear properties in various fluids of small volumes. However more detailed studies are required to characterise the angular dependence and how the geometry of the microchannels can impact on the measurements.

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