Towards the optimization of dual-frequency driven sonochemistry: a four dimensional parameter scan of a single gas bubble accelerated by GPUs

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One of the main success stories of modern chemistry is the use of high frequency and high intensity ultrasound on a liquid domain to increase the chemical yield of various reactions. This phenomenon is called sonochemistry in which the key phenomenon is acoustic cavitation and the collapse of the emerging bubbles. According to many experimental observation, the chemical yield can be further increased by the use of two different frequencies during the irradiation. Due to the large involved parameter space, however, even in case of a single spherical bubble, a clear theoretical understanding of such a synergetic effect is still missing in the literature [1].

Our strategy is to employ a bottom-top approach; that is, investigate a model as simple as possible but perform a large dimensional and fine parameter scan. Later, the model complexity can be gradually increased until a suitable explanation for the synergy of dual-frequency driving is found. As an initial step, the main aim of the present this study is to present numerical simulations obtained by solving the Keller—Miksis equation well known in sonochemistry that is a simple second order ordinary differential equation describing the dynamics of a single spherical gas bubble [2].

The investigated four dimensional parameters space involves the amplitudes of the dual-frequency driving P_{A1} and P_{A2} varied between 0 and 2 bar; and the corresponding frequencies ω_{R1} and ω_{R2} spanned in the range 0.1 and 10, and normalized by the linear undamped eigenfrequency of the system ω_0. Even with a moderate resolution of 100 values of each parameters applied here (the effect of phase angle is neglected and the equilibrium bubble size is $R_E = 10$ μm), one hundred million transient initial value problems have to be solved. The initial condition is the equilibrium state of the unexcited system at each parameter combination. In order to obtain results within reasonable time, the high computational capacities of professional GPUs were exploited (2 Nvidia Tesla K20m). The overall computations took approximately one week. The integration algorithm was the adaptive Runge–Kutta–Cash–Karp method.

After the initial transient (1024 number of collapses), 64 additional collapses were simulated and their properties saved (maximum and minimum bubble radii, collapse times and maximum bubble wall velocities). A collapse is defined as the evolution of the bubble radius from a local maximum to a subsequent local minimum. The above quantities provide a good flexibility to describe the strength of a bubble collapse mandatory for efficient application. The data of the 64 number of collapses allows to make a coarse statistical investigation as well. In addition, the total time of the 64 collapses were also recorded, which makes it possible to determine the number of the strong collapses (a suitable threshold is required) in a unit time. In the present paper, we focus only in the examination of the **relative maximum bubble radius** $(R_{MAX} - R_E)/R_E$ as a measure of the collapse strength used also by many researchers [3, 4].

The building block of the investigation is the production of bi-parametric contour plots of the relative maximum bubble radii in the plane of the relative frequencies at fixed pressure amplitudes. Out of the $100 \times 100 = 10000$ plots, only one is presented in the left hand side of **Fig. 1** at $P_{A1} = 0.6\ \mathrm{bar}$ and at $P_{A2} = 1.2\ \mathrm{bar}$. It is clear that high maximum bubble radius can be achieved only at low frequencies (red-yellow domain). This low frequency region is known as the giant response region. During the evaluation of all the diagrams, no synergetic effect has been found. The only possible optimization strategy (in terms of the collapse strength) is the distribution of power between the ultrasonic transducers. If they are equal, the energy efficiency can be increased approximately by a factor two.

Another important factor in sonochemistry is the number of the strong collapses in a unit time. As an example, in the right hand side of **Fig. 1.**, the number of the strong collapses are plotted as a function of the excitation frequencies at fixed pressure amplitudes. Strong collapse here means that the relative maximum bubble radius is greater than two, which is a common threshold in the literature [5]. It is clear that very frequent strong collapses can be achieved near the resonance frequencies (read stripes near $\omega_{R1,2} = 10^0$). The detailed investigation of such collapse frequencies revealed that in case of nearly equal pressure amplitudes (optimal for energy efficiency), the number of the strong collapses dropped by a factor of two (compared to a single frequency driving) due to the canceling effect of the combined driving pressure signal. The main consequence is that the evolution of **the maximum bubble radius cannot explain the high increase** (sometimes 300% [5]) **of the efficacy of sonochemistry** compared to single-frequency driving. These results put the main outcome of some previous studies [3, 4] into question. Consequently, other explanation(s) need to be found and investigated in more details. Some possible candidates: other measures for the strength of the collapse and/or the number of strong collapse in a unit time may show different trends; dual frequency may stabilize the spherical shape of the bubbles; or the efficiency increase may occur inherently due to the collective dynamics of bubbles in a cluster.

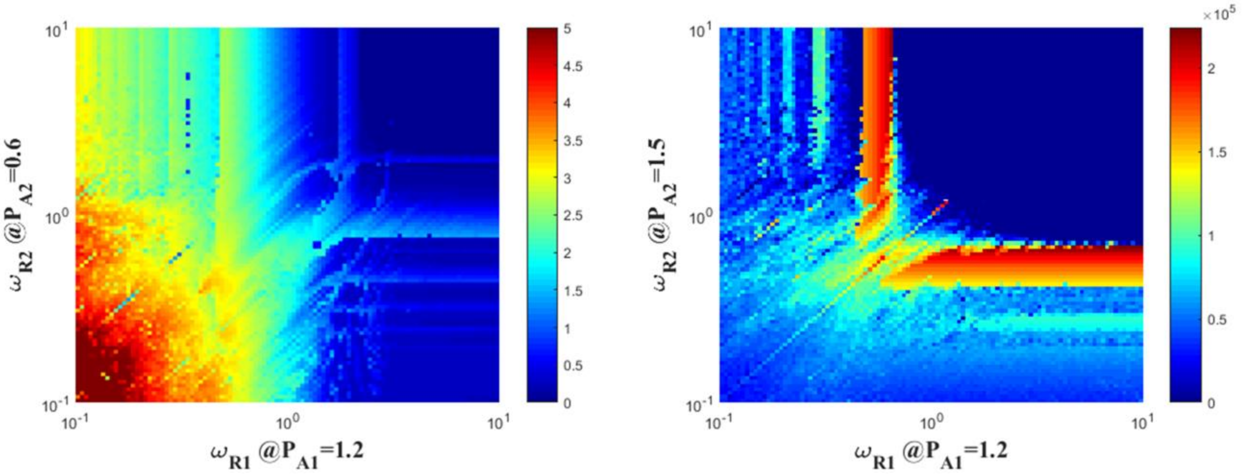

Figure 1. Left panel: Maximum bubble radii as a function of the relative frequencies ω_{R1} and ω_{R2} at pressure amplitudes $P_{A1} = 0.6\ \mathrm{bar}$ and $P_{A2} = 0.8\ \mathrm{bar}$. Right: The number of the strong collapses $(R_{MAX} - R_E)/R_E > 2$ as a function the relative frequencies ω_{R1} and ω_{R2} at pressure amplitudes $P_{A1} = 1.5\ \mathrm{bar}$ and $P_{A2} = 1.2\ \mathrm{bar}$.

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