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SIMULATION OF AXISYMMETRIC FREE SURFACE VISCOUS FLOW AROUND  
A NON-SPHERICAL BUBBLE IN THE SONOLUMINESCENCE REGIME

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## 1 Introduction

Sonoluminescence, the phenomenon by which sound is turned into light was first documented in the 1930's by German scientists studying the catalysing effect of sound waves on chemical reactions. Using ultrasound transducers invented during the First World War, H.Frenzel and H.Schultes of the university of Köln reported in 1934 light emission from clouds of cavitating bubbles generated in their solution. Their first interpretation of the phenomenon is now called *triboluminescence*. They suggested electric surface charges could be generated during the strong oscillations of collapsing bubbles. Electric sparks would then be the cause of light emission. Several other researchers studied this problem and looked for possible light emission mechanisms. It is currently believed that high temperatures are reached in the bubbles due to compression during the collapse. This would then explain both the catalysing effect of ultrasound and the measured spectra corresponding to temperatures up to 3000 °K.

However, due to the complexity of the dynamics of these large clouds of cavitating bubbles it was difficult to extract more information from the experiments and a theoretical model was difficult to find. In 1988, F. Gaitan and L. Crum managed to trap a single sonoluminescing bubble at the antinode of a stationary ultrasound wave [8]. Their discovery renewed the interest in the study of the mechanisms of sonoluminescence because the phenomenon was very stable allowing the different parameters of single-bubble sonoluminescence to be studied in detail.

The apparatus for single bubble sonoluminescence is very simple. Usually two piezo-electric transducers are glued diametrically opposite one another on a spherical glass sphere (around 10 cm in diameter). A driving current of around 25 kHz frequency (depending on the resonant frequency of the container) generates a standing sound wave with an amplitude of around 1 atmosphere. A bubble can be levitated at the antinode of the sound wave. The force counteracting gravity is called the Bjerknes force and is an average of the driving pressure gradient over one acoustic cycle. When the pressure falls, the bubble expands and the force due to the pressure gradient ( $-v\nabla P$  where  $v$  is the volume of the bubble) is greater than during the collapse phase, giving rise to a net force over one cycle. For certain values of the bubble radius and the amplitude of the acoustic forcing, this force counterbalances gravity and the bubble is *on average* stationary.

Not all trapped bubbles sonoluminesce however and the phenomenon appears to be very sensitive to a variety of parameters. One important discovery made by Gaitan and Crum was the need for partially degassed water. This could appear surprising at first. One would think that a bubble in degassed water would tend to dissolve and ultimately disappear, whereas single-bubble sonoluminescence has been observed to last for several minutes even hours. The beginning of an explanation is given by the phenomenon of *rectified diffusion*. When the bubble expands the pressure inside decreases and air dissolved in water flows in. The reverse occurs during contraction, but the surface is smaller giving rise to a positive net mass inflow. This can compensate for the loss due to classical diffusion. A number of theoretical studies of the parameter range for stability of this process have been conducted [15, 4, 3] and this theory accounts satisfactorily for the experimental observations.

The light intensity itself appears very sensitive to temperature [2], and gas composition [12, 13].

Cooling the liquid from 20°C to 1°C increases the light intensity by a factor of 200, at which point the light spot is clearly visible even with ambient light.

Another striking feature of the light emission is the brevity of the emitted pulses. The light emission is indeed not continuous and occurs once during each acoustic cycle with a phase shift. Trying to characterize the light emission mechanism S. Putterman and B. Barber measured the duration of the pulses to be less than 50 picoseconds [1]. They had to use the fastest photo-multipliers at their disposal and this duration is only an upper bound. This is obviously extremely short (one millionth of the acoustic period) and emphasizes the extremely high concentration of energy which occurs.

The spectrum of light emission reveals another strange feature [11]. The maximum of intensity appears to be beyond ultraviolet which suggests that very high temperatures are reached (50,000°K and higher). These available spectral measurements were made using quartz containers, but due to the cutoff wavelength of water, no data regarding the exact location of the maximum of intensity are available yet. This, together with the brevity of the light pulses suggests that the hypothesis of compressional heating retained as the mechanism for multiple-bubble sonoluminescence does not hold for single-bubble sonoluminescence. This hypothesis leads to temperatures of around 5000°K and pulses duration of 20 nanoseconds (500 times too long).

Lead by these considerations, H.P. Greenspan and A. Nadim [9] and C. Wu and P.H. Roberts [25] proposed an explanation based on a shock-wave hypothesis. As the bubble collapses, the bubble walls eventually reach supersonic speed, launching a spherical shock wave in the gas. It is known from Guderley's self-similarity solution ([24], pp. 196-198) that in the ideal case the Mach number tends to infinity as the radius of the spherical shock wave tends to zero. Obviously in the real case the stability of the shock wave will limit this growth but this theory accounts well for the high temperatures and brevity of the light emission.

This theory relies however on the hypothesis of spherical symmetry. Following the arguments given by A. Prosperetti [23], there are strong reasons to believe that the bubble can not remain spherical during the collapse phase. Due to the asymmetry induced by gravity, the bubble will actually oscillate slightly around its equilibrium position along the vertical axis. During the collapse phase momentum conservation implies the formation of a high velocity jet oriented in the direction of motion. This is analogous to what happens for cavitation bubbles in the neighborhood of a solid wall. Due to the asymmetry induced by the solid wall, a high-velocity jet concentrating the momentum of the collapsing (and moving) bubble forms and eventually hits the wall. This is thought to be the explanation for cavitation damage.

The shockwave hypothesis may still be right however, but probably depends on the shape history of the bubble and the resulting shape of the shock wave [7]. If the bubble is still spherical enough, the shockwave will remain spherical and stable for long enough for the gas to reach high temperatures. On the other hand, if the bubble is already deformed, the hypothesis of a spherical shock wave does not hold anymore. As an alternative explanation A. Prosperetti proposes the jet impact to be responsible for the light emission.

The phenomenon of single-bubble sonoluminescence is thus largely unexplained. The mechanism of light emission itself is unknown and the motion and shape of the bubble when it reaches its minimum radius (thought to be less than one micron) is very difficult to measure experimentally due to its brevity and small size.

These considerations lead us to consider the feasibility of a numerical simulation of a bubble in the sonoluminescence regime. All the numerical models developed to date are based on one-dimensional formulations and we are not aware of any two or three-dimensional Navier-Stokes simulations. Ideally a numerical model would give information on the shape of the emitted shock wave or on the formation of a high velocity jet.

## 2 Numerical method

All the results presented here are computed using a free-surface version of the code described in [17, 21]. The pressure in the bubble is considered to be uniform and is given by an appropriate equation of state. In all the calculations presented we assume an isothermal pressure law ( $p = p_0 V/V_0$ , where  $V$  is the bubble volume). In the liquid phase, the full incompressible axisymmetric Navier-Stokes equations are solved using a projection method. The velocity and pressure fields are discretized on a staggered, fixed, Cartesian and regular mesh (figure 1). The code is first order in time and second-order in space. The

Poisson-like pressure equation is solved using multigrid-accelerated Gauss-Seidel relaxations [22].

The interface is discretized using a set of marker points linked by cubic splines. This description allows the precise knowledge of the position and curvature of the interface, necessary to include the surface tension terms.

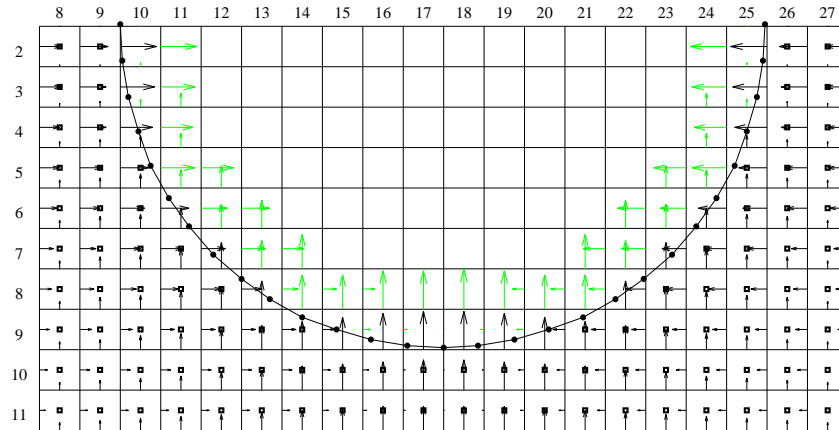


Figure 1: *Mesh and discretization of the velocity and pressure fields. The marker points as well as the connecting cubic splines are represented. The light arrows are extrapolated values of the velocity field. The squares indicate the location of the pressure discretization when the pressure equation is solved.*

As in all free-surface codes [10, 5, 18, 14], the most delicate point is the treatment of free-surface boundary conditions. The pressure on the interface on the fluid side is given by

$$p = p_i + \sigma \kappa + \mu \mathbf{n} \cdot \mathbf{D} \cdot \mathbf{n}, \quad (1)$$

where  $p_i$  is the pressure in the bubble,  $\sigma$  the surface tension coefficient,  $\kappa$  the curvature,  $\mu$  the dynamic viscosity,  $\mathbf{n}$  the normal to the interface and  $\mathbf{D}$  the deviatoric part of the stress tensor. Using the irregular stars scheme described in [5] and the splines description of the interface, one can impose this condition during the relaxation of the pressure equation.

As we use a fixed grid to solve the Navier-Stokes equations, we need to extrapolate the velocity field far enough inside the bubble to get the velocity values necessary for the marker points advection and for the resolution of the Navier-Stokes equations on the fluid boundary (figure 1). Moreover, this must be done while fulfilling the zero tangential stress interface boundary condition

$$\mathbf{t} \cdot \mathbf{D} \cdot \mathbf{n} = 0, \quad (2)$$

where  $\mathbf{t}$  is the tangent to the interface. This is quite difficult to achieve but fitting two-dimensional polynomials through the points with the constraint given by (2) gives satisfactory results [17].

The marker points are advected using a simple bilinear interpolation scheme and a redistribution is done at every time step to insure a uniform distribution as the bubble deforms. As underlined in [21] the computational time needed by the marker part of the algorithm scales as  $1/n$  where  $n$  is the number of grid points along one dimension and is then negligible for reasonable domain sizes ( $64 \times 64$  and up). All the calculations presented were done on a 200 MHz IBM compatible PC running Linux. The achieved typical speed is approximately 30,000 grid points/second.

### 3 Bubble dynamics and stability

As we have seen, sonoluminescence is certainly closely linked to cavitation even if the length scales involved are very different (millimeters for cavitation, micrometers for sonoluminescence). Interest in cavitation dates back to the First World War with cavitation around ship or submarines propellers. Rayleigh solved in 1917 the problem of the collapse of an empty spherical cavity in a large mass of liquid and showed the radius  $R(t)$  to obey the relation

$$R\ddot{R} + \frac{3}{2}(\dot{R})^2 = \frac{p(R) - p_\infty}{\rho}, \quad (3)$$

where  $\rho$  is the liquid density,  $p_\infty$  is the pressure in the liquid far from the bubble and  $p(R)$  is the pressure in the liquid at the bubble boundary. Plesset extended this relation to include viscous effects and surface tension [20]

$$R\ddot{R} + \frac{3}{2}(\dot{R})^2 = \frac{1}{\rho} \left( p_i - p_\infty - \frac{2\sigma}{R} - \frac{4\mu}{R} \dot{R} \right), \quad (4)$$

where the pressure in the gas at the bubble wall  $p_i$  may be a function of time as well as the pressure at infinity  $p_\infty$ . This equation is often referred to as the Rayleigh-Plesset equation. The choice of an appropriate equation of state for the gas inside the bubble is necessary to close the system. As a first approximation, a polytropic law of the form  $p_i = p_0(R/R_0)^{3\kappa}$  with  $\kappa$  constant is convenient ( $\kappa = 1$  for an isothermal gas,  $\kappa = \gamma$  if adiabatic). The Rayleigh-Plesset equation has been shown experimentally to give a good description of the behavior of a sonoluminescing bubble. Several researchers made laser scattering measurements of the evolution of the radius of the bubble with time which are well fitted by Rayleigh-Plesset equations where the pressure at infinity and the equilibrium radius  $R_0 = 2\sigma/(p_i - p_\infty)$  are adjustable parameters [2]. An example of the solution of the Rayleigh-Plesset equation for parameters typical of sonoluminescence is given figure 2. As we see the behavior of the bubble is quite complex. The

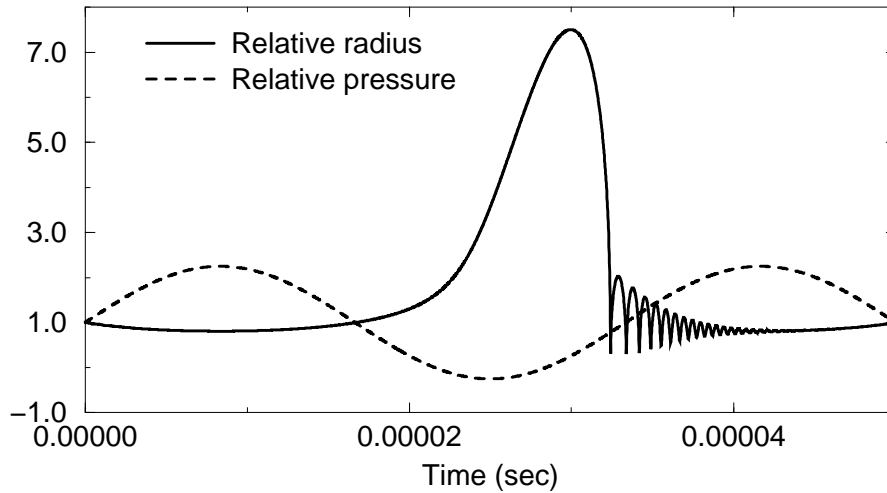


Figure 2: *Solution of the Rayleigh-Plesset equation for parameters typical of sonoluminescence. Equilibrium radius: 2.5  $\mu\text{m}$ , pressure amplitude: 1.25 atmospheres, frequency: 30 kHz.*

first growth of the bubble is followed by a violent collapse and several rebounds before the next expansion. Experimentally, the light is shown to be emitted during the first and most violent collapse.

Using the Rayleigh-Plesset equation one can investigate the linear stability of non-spherical bubbles. Using spherical harmonics the bubble shape can be described as

$$S(r, \theta, \phi, t) = r - R(t) - \sum_{n,m} a_{nm} Y_n^m(\theta, \phi), \quad (5)$$

where  $S(r, \theta, \phi, t) = 0$  is the equation of the bubble surface,  $R(t)$  is the instantaneous average radius and  $a_{nm}(t)$  is the amplitude of the spherical harmonic component [20]. Plesset & Mitchell were able to solve the equation for the  $a_{nm}$  in the case of a fixed pressure difference  $p_i - p_\infty$ . While it appears that the perturbations tend to a finite value in the case of an expansion of the bubble (due to the stabilizing effect of the divergence of the streamlines), the contraction is shown to be unconditionally unstable for all modes. In this case, the perturbation  $a_n$  is shown to be proportional to  $R^{-1/4} \exp(\pm icn^{1/2} \int R^{-5/2} dt)$ , where  $c$  is a constant. The linear theory thus predicts a growth proportional to  $R^{-1/4}$  with a superposed oscillation of increasing frequency.

We should note however that this theory neglected both the viscosity and the surface tension which have both a stabilizing effect. This is true however only for small scales and will act mainly on short wavelengths (high  $n$ ).

This unconditional instability of a contracting bubble is a major problem for numerical simulations. The growth in  $R^{-1/4}$  will not only affect any initial deviation from sphericity but also all the numerical errors introduced at each time step. While the initial perturbations are part of the physics of the problem, one does not want the numerical errors to become dominant.

## 4 Results

### 4.1 Rayleigh-Plesset oscillations of a bubble in an acoustic field

A first test relevant to sonoluminescence is the radial oscillation of a spherical bubble far from any boundaries. In this case the spherically symmetric Rayleigh-Plesset equation gives the evolution of the radius with time. An axisymmetric simulation should reproduce a similar behavior of the radius. Figure 3 illustrates such a simulation. The viscosity of the liquid is that of water and the surface tension coefficient is that for water/air.

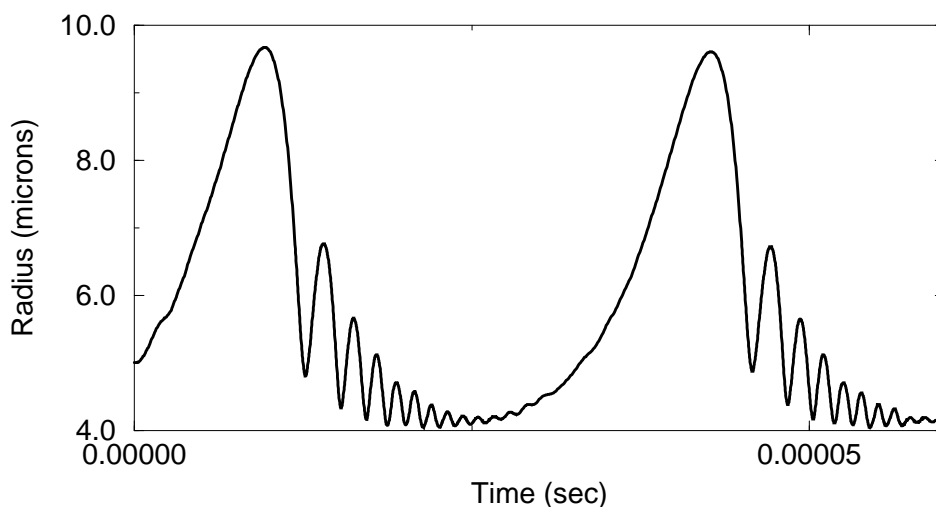


Figure 3: *Evolution of the radius with time for oscillations of an axisymmetric bubble in an acoustic field. The equilibrium radius is 5 microns, the amplitude of the acoustic forcing is 0.5 atmosphere, frequency is 30 kHz.*

We must note that the pressure forcing is only 0.5 atmosphere. In typical sonoluminescence conditions it should be close to one atmosphere, leading to much higher compression ratios as in figure 2. We have not implemented yet the adaptative grid refinement necessary to deal with radii varying from 100 to 1 microns as happens in sonoluminescence conditions.

### 4.2 Collapse of an empty cavity (cavitation bubble) near a solid wall

As seen above, we are interested in bubble deformations and particularly jet formation in the case of non-symmetrical flows. A case of particular interest is the collapse of a cavity near a wall, as happens during cavitation phenomena. Beautiful high-speed films of this phenomenon have been realized by W. Lauterborn [16, 6], using laser-generated vapor bubbles in water. M.S. Plesset & R.P. Chapman showed that the experimental results were well fitted by their boundary-integral calculations of a collapsing bubble [19] (neglecting both viscosity and surface tension).

Figure 4 shows the results of such a collapse and looks very similar to those obtained by M.S. Plesset & R.B. Chapman [19]. A detailed comparison is not possible however, the simulation by Plesset & Chapman being a boundary integral calculation neglecting both viscosity and surface tension. The velocity after jet formation reaches 130 m/s, which is in agreement with their results.

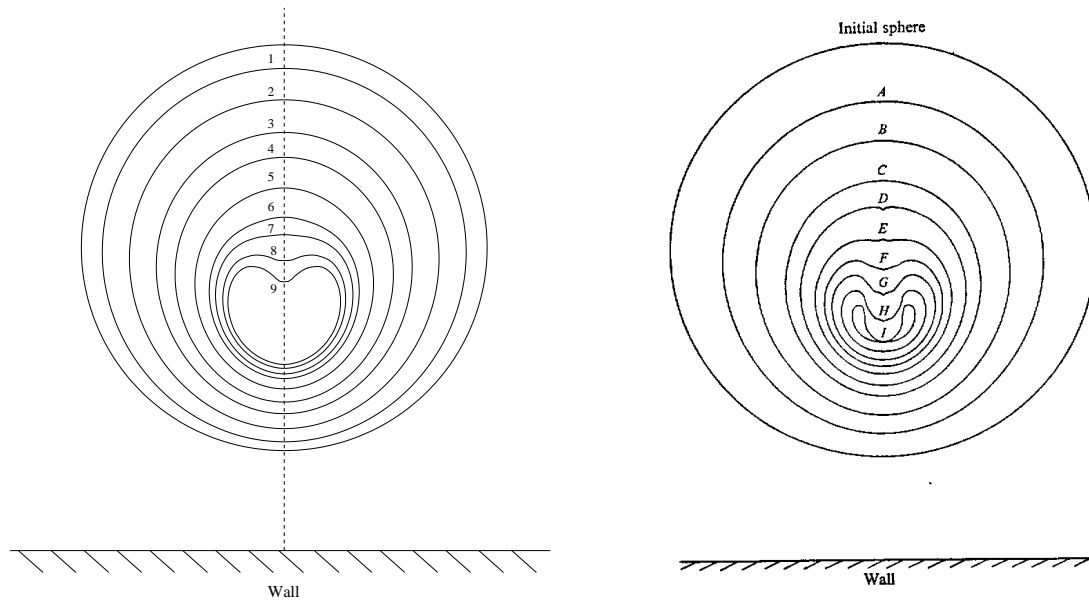


Figure 4: left: Collapse of an empty cavity near a wall. The interface markers are represented for different times. The bubble is initially spherical, the closest point being half a radius from the wall. The initial bubble diameter is 0.33 millimeters, the surrounding fluid is water and the surface tension coefficient is water/air:  $0.07 \text{ kg.s}^{-2}$ . The times represented are as follows (in  $\mu\text{s}$ ). 1: 7.55, 2: 10.59, 3: 12.59, 4: 13.51, 5: 14.28, 6: 14.71, 7: 14.88, 8: 15.03, 9: 15.11. right: Boundary-integral simulation of the collapse by M.S. Plesset & R.B. Chapman.

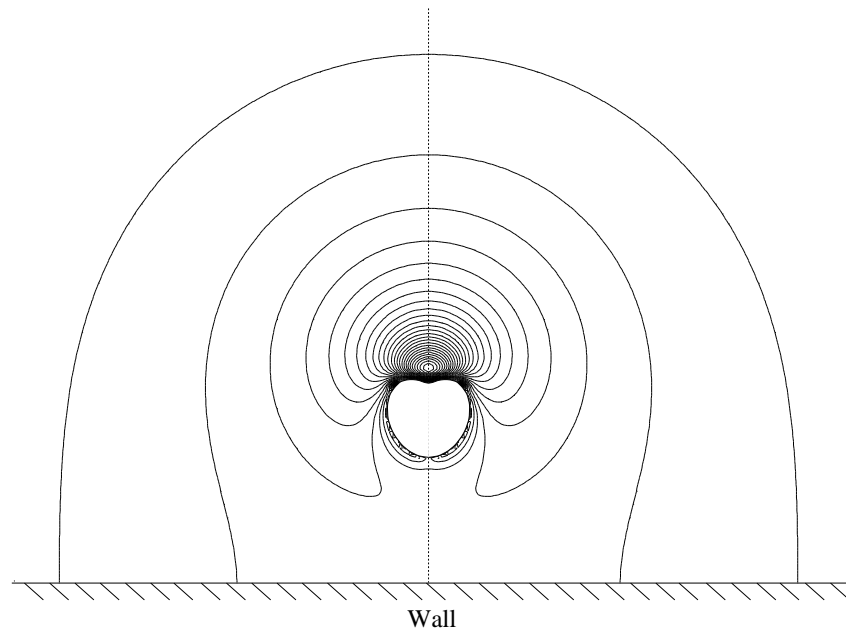


Figure 5: Pressure isolines soon after the beginning of jet formation. The high overpressure and pressure gradient at the origin of the jet are clearly visible.

## 5 Conclusion

In this paper, we have underlined the potential of full Navier-Stokes numerical simulations for the investigation of bubble dynamics problems. The case of sonoluminescence and cavitation are particularly relevant. Numerical simulations can potentially give useful information where experiments are difficult to perform. The analogy between sonoluminescence and cavitation, through the theory of jet formation of A. Prosperetti, can lead to interesting links between these phenomena. However, the unconditional physical instability of collapsing bubbles is a difficult challenge for numerical computations.

First results have been presented, including radial oscillations of a spherical bubble and jet formation for a cavitation bubble (with realistic physical parameters). These results are only preliminary and the simulation code requires further development but is already usable for the investigation of cavitation process. As far as sonoluminescence is concerned, the large spread of time and space scales make the simulation difficult and requires the use of adaptative grid refinement schemes. However, if the simulations can show that with realistic parameters, a jet really appears early enough during the compression phase, this will trigger interest for new theories trying to explain sonoluminescence.

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