Validation of Earlier Work

I t has only now come to my attention that Computing in Science & Engineering published an article by Leo Kadanoff in its March/April 2004 issue that is critical of the experimental work on sonoluminescence (SL) that came out of my group. Kadanoff questions the accuracy of the data we published in 1992, but subsequent experiments have, in fact, confirmed our groundbreaking work.

The rebirth of interest in SL, a phenomenon studied since the 1930s, was driven in large part by the appearance of a 1991 paper that showed that the flash widths of light emitted by imploding bubbles could be under 100 picoseconds (ps). Later work claimed that for bubbles formed from well-degassed water, these flash widths could be smaller than 50 ps. Kadanoff says that “this short width opened the door to novel mechanisms for explaining the emitted light’s total intensity. Later developments suggested that the short pulse width was a misstep by the experimentalists” and “led simulators and theorists astray.” Furthermore, “a host of incorrect speculations and mechanisms ran through the field, intended to explain the ‘observed’ behavior” and “continued unhindered until an experiment by Gompf and colleagues showed that the pulse width was much longer than previously believed.”

B. Gompf’s measurements revealed flash widths of 60 ps, with an uncertainty of 10 ps for SL in strongly degassed water. Surely the theory of SL hasn’t progressed to the point where experimental discrepancies on the order of a standard deviation can be blamed for sending theorists astray? But we can say more: our application of Gompf’s excellent method to SL from bubbles formed from well-degassed water found flash widths ranging down to less than 40 ps. Figure 46 of reference 9 in Kadanoff’s article actually shows a theoretical line going through these “very short widths for the emitted pulse of light.” Is Kadanoff implying that this 2002 work, by his student and his postdoc, constitutes yet more theory led “quite astray”?

In 1994, R.A. Hiller and colleagues showed that SL is sensitive to the gas concentration of the fluid from which a bubble is formed. Later research found that higher gas levels in water led to longer flash widths: at 150 Torr air in water, the flash width becomes approximately 100 ps, or roughly twice as long as for strongly degassed air in water. Gompf and colleagues cited us for qualitatively warning in our earliest papers that the flash width might be increasing with gas concentration as the water ages. Thus, Kadanoff is wrong when he says that “in contrast to the excellent work in SL in the post 1997 period, this misstep in the measurement of the flash width led simulators and theorists astray. Post 1997 work has, in fact, confirmed the validity of our earlier flash width experiments on SL.

In saying that simulations “never led to any advances,” Kadanoff’s negativity causes him to miss the impending opportunity presented by current SL research. Soon SL will provide an experimental test bed for molecular, even plasma, dynamic simulations that contain sufficient particles for a complete description of a natural phenomenon. As the acoustic frequency is increased, the size of a bubble decreases; research has demonstrated that at 1 MHz there are only $5 \times 10^7$ atoms inside. Molecular dynamics simulations have already addressed Kadanoff’s concern that neglect of heat conduction would lead to higher bubble temperatures. Experimentally, the situation is different; SL from helium has the highest spectral temperature despite it having the highest value of thermal conductivity as compared to other noble gases. Molecular dynamics simulations suggest an explanation; when heat bath boundary conditions are applied to collapsing bubbles, there is greater energy focusing inside the bubble. These simulations also find that the mean free path becomes comparable to the distance over which energy density varies, thus...
indicating the shortcomings of hydrodynamic approaches.

Kadanoff cites the work of C.C. Wu and P.H. Roberts at the University of California, Los Angeles, as an example of one of the “failures” of the “earliest computer simulations” of SL.12 Their letter to the editor (which follows this one) explains why Kadanoff’s criticism is unfounded. Ultimately, Kadanoff’s commentary would have had the appearance of balance if he had said who he meant by “they” when he mentions that “experimentalists made the major discoveries and started a program of work” and “by 1997 they set out the values of some of the most important parameters.” In addition to establishing short flash widths and the noble gas doping effect,7 the UCLA effort laid out the bubble radii used in all future calculations13 as well as the amazing clock-like synchronicity of SL.

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References


Shocks in Sonoluminescence

Sonoluminescence (SL) is the light emitted when a bubble of gas trapped in a liquid is violently compressed by strong acoustic forcing (amplitude $P'$. We recently discovered that in a previous issue of Cise,1 Leo Kadanoff told readers our simulation2 of SL was one of several “wrong ones.” We thank the editors for this belated opportunity to defend ourselves.

Our 1993/1994 publications2,3 provided the first self-consistent SL calculations, with the dynamics of the gas and surrounding water being solved simultaneously. It was unknown why, when, and for how long the light would be emitted each acoustic cycle, because light emission depends sensitively on gas temperature $T(r,t)$ and density $\rho(r,t)$, the determination of which was our primary objective. Our secondary aim was to estimate bubble luminosity $L(t)$ from $T$ and $\rho$. Our results are qualitatively describable through a “threshold” $P'_0$, where for $P' < P'_0$, light production is negligible, but as $P'$ increases through $P'_0$, implosion-generated shocks become progressively stronger and more numerous, and light emission increases up to bubble destruction at $P' \sim 1.5$ atmospheres (atm).

We concentrated on air bubbles before it was realized that the bubble is a chemical separator (except for H$_2$ bubbles4), so the monatomic case $\gamma = 5/3$ (He, Ar, Xe) is more generally applicable to SL than $\gamma = 7/5$.

V.Q. Vuong and A.J. Szeri integrated the Navier-Stokes (NS) equations with nonzero viscosity $\mu$ and conductivity $k$.5 They found no shocks for $\gamma = 5/3$ and $P' = 1.3$ atm, whereas we obtained strong shocks for $\gamma = 7/5$ and the same $P'$ from the Euler equations ($\mu = k = 0$). Kadanoff erroneously concludes that the finite $\mu$ and $k$ prevented a 5/3 shock, and would, if included, have suppressed the 7/5 shock, too. The true explanation is different: an increase in $\gamma$ means increased sound speed $c$. Implosion-generated compressions must travel nearer to $r = 0$ before steepening into shocks. $P'_0$ is thus larger: in fact, $P'_0(5/3) > 1.3$ atm, but $P'_0(7/5) < 1.3$ atm. Using finite $\mu$ and $k$, Vuong and Szeri had already confirmed strong shocks for $\gamma = 7/5$, $P' = 1.3$ atm.

Kadanoff’s belief in shock suppression through diffusion is contradicted by the following $\gamma = 5/3$ results, which show that finite diffusion promotes the formation of SL shocks. In Figure 1a and 1c, for example, the water temperature at the bubble surface $r = R(t)$ is held at 300 K, and in Figure 1c, $k$ is 90 percent reduced. The water chills the compressing gas only in a thin boundary layer; the shock is so weak that we needed 2,000 grid points to resolve it.
convincingly. For the full $k$, chilling penetrates deeper, and the concomitant reduction in $c$ engenders a strong shock (Figure 1a). Vuong and Szeri included heat conduction in the water, and their Figure 12 shows a strong increase in $T(R)$ at implosion. We approximated their $T(R)$, re-integrated, but found the shock only slightly weakened (Figure 1b).

NS equations are as valid as Euler equations in describing shocks. They’re equally incapable of realistically describing the solution at implosion ($t = t_i$) when $T$ and $p$ are momentarily infinite; molecular dynamics simulations cure this. Importantly, the infinitely brief $\delta$-function singularities at ($r$, $t$) = (0, $t_i$) don’t vitiate the Euler solution for other ($r$, $t$); energy and other physical properties are conserved across $t = t_i$. Also, “The major contribution to $L$ arises from the vicinity of $r = 0.07 \mu m$… It is worth noticing that the singularity that occurs at ($r$, $t$) = (0, $t_i$) makes virtually no contribution to $L(t)$.”

Kadanoff implies that the infinities invalidate our simulations, but L.D. Landau and E.M. Lifshitz include such a solution, recognizing its physical integrity.

The success of our work is evident from our calculated $L(t)$ and the total radiation $q$ per flash, obtained by evaluating ionization levels from Saha’s equation and computing the resulting bremsstrahlung, assuming the emitting region is transparent. We got an approximately correct $q$ (up to $\approx 5 \times 10^6$ eV) and explained the observed sensitivity of $q$ to $P'$. We predicted that the flash’s duration $\tau$ increases with $P'$, mainly through multiple shocks. Although our $\tau$ was about right (tens to hundreds of ps), shocks gave part of $q$ in sharp peaks of $L(t)$, but only a few ps wide. Perhaps the incapability of current experiments to resolve such peaks led to the speculation that shocks are irrelevant to SL. Subsequent investigators (adopting our Saha/bremsstrahlung approach) assumed the entire bubble to be in a nearly uniform adiabatic state, which is approximately true only for $P' \leq P_0'$. Mysteriously, their results are claimed to be as encouraging as ours.

Clearly, SL research still presents interesting challenges that readers might find engaging (provided they’re undeterred by august but ill-informed criticism)!

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References


Proof through Simulation?

Thank you for sending me the comments of Professors Roberts, Wu, and Putterman occasioned by my article. I was pleased to see these scholars invoke past perspectives on the subject of sonoluminescence. My article’s main point about sonoluminescence was that some of the previous simulations had dealt with an ill-posed problem and hence were not necessarily re-
liable. Professors Roberts and Wu carried this point somewhat further than I had, by asking whether any hydrodynamic simulation could capture the behavior of these cavitation phenomena.

The subject has moved forward since my article was written. New information about sonoluminescence can be found in, for example, D.J. Flannigan and K.S. Suslick’s “Plasma Formation and Temperature Measurement During Single-Bubble Cavitation” (Nature, vol. 434, 2005, pp. 52–55) and the commentary “Cavitation Heats Up” (Nature, vol. 434, 2005, pp. 33–34). I think we shall once more see that good experiments are more effective than simulations in changing people’s scientific opinions.

Sincerely yours,
Leo Kadanoff