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Author
Russo, R.E.

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Response to 'Comment on "Evidence for phase-explosion and generation of large particles during high power nanosecond laser ablation of silicon"'
[Appl. Phys. Lett. 76, 783 (2000)]

J.H. Yoo, S.H. Jeong[a)], X.L. Mao, R. Greif[b)], and R.E. Russo[c)]

Lawrence Berkeley National Laboratory, University of California, Berkeley, CA 94720

The experimental results reported in [1] are briefly summarized as: i) 3-4 fold nonlinear increase in crater depth across the threshold irradiance of ~22 GW/cm^2, ii) crater depths up to 20 μm per single pulse ablation, iii) local peak and valleys at the bottom of the craters for irradiances above the threshold, iv) resolidified droplets scattered around the crater, v) ejection of particulates or droplets of approximately 10-30 μm in diameter, and vi) delay of mass ejection by about 300-400 ns from the beginning of the ablation pulse[2]. The sample was pure silicon of about 1 mm thickness. Each crater was formed by a single laser pulse in air. For laser irradiances above the threshold, the observed crater characteristics were attributed to a phase explosion (or explosive boiling) of a superheated liquid layer in conjunction with induced-transparency of the silicon near the thermodynamic critical temperature.

[a)] Kwangju Institute of Science and Technology, Kwangju 500-712, Republic of Korea
[b)] Department of Mechanical Engineering
[c)] To whom the correspondence should be addressed

The main concern in Craciun's comment, as noted in the last paragraph, is the higher threshold irradiance for explosive boiling determined in [1] than that reported in other studies. Craciun indicates that in those studies, the emission of droplets is been also attributed to volume or explosive boiling and was reported for lower laser irradiances. When comparing the experimental results reported in those studies to Reference [1], careful attention should be paid to what was actually observed in each experiment and exactly what phenomena the term 'volume or explosive boiling' represents. The experimental observations described as a result of 'volume boiling' are not consistent among these studies, leaving open the definition of volume boiling and evidence to determine it. The differences between the crater characteristics in [1] and those from the studies referenced in Craciun's comment will be discussed in detail to show that even though these studies use the same term 'volume or explosive boiling' the relevant processes in consideration are quite different.

Craciun emphasizes the generation of droplets in the referenced studies to support his argument that volume boiling would be probable under much lower irradiances than the value reported in [1]. The ablation process in [3] referred to 'volume boiling' largely because of a rough morphology on the crater bottom, as indicated in the comment. Because volume boiling was observed at 0.4 GW/cm^2 in this reference, Craciun questions that the threshold irradiance of ~22 GW/cm^2 in [1] is too high. If crater surface morphology was the only consideration, Craciun's argument would be reasonable. However, there are many other characteristics of the craters in [1] different from those in [3]. The *estimated* thickness of the *liquid layer* in [3] is only on the order of 1 μm while the *measured* depth of the *crater itself* in [1] ranges from 10-20 μm, implying that the thickness of the liquid layer would be even greater. Furthermore, in Ref. [3], a Ge sample was ablated with 10 pulses while in [1] only single pulse ablation was employed on silicon. Even though Craciun attributes the mechanism responsible for the craters in [3] to be volume boiling, this volume boiling mechanism can not be the same as the 'explosive boiling process' described in [1]. If the mechanisms were assumed to be the same, it would be impossible to explain the formation of deep craters in [1] with the shallow liquid layer observed in [3]. Similarly, in [4] the ejection of droplets is not specifically attributed to volume boiling, and the original article gave no explanation for the mechanism of droplet generation. Furthermore, the experiment was performed in ultra-high vacuum and the size of microparticles was not measured. Therefore, ejection of droplets at lower irradiance described in [4] is not a meaningful comparison in identifying the threshold for volume boiling. The data in [5] were reported for a Ni sample and the ablation depth per pulse was only about 0.05-0.1 μm, which is two orders of magnitude smaller than that in [1].

The studies in [6,7,9,10] were done on thin films of various materials. For thin films the substrate could serve as a thermal insulator influencing heat flow. If a thermal insulator is underneath the film, less heat will be lost to the substrate and thus it is possible to maintain the superheated liquid layer at reduced laser irradiance. Such an effect would have a large influence on the irradiance threshold for volume boiling.

The common observation in these studies was the emission of droplets which is the parameter Craciun uses to define the threshold for volume boiling. The authors contend that droplet emission alone is not enough evidence for phase explosion. The phase explosion phenomena described in [1] is not a mere observation of droplets alone, but instead represents collective phenomena including all of the features mentioned above. The actual processes for droplet formation in each of the studies are likely different, regardless that they were all called volume boiling.

Craciun's comment about numerical simulation on page 2 is a general statement applicable to any computer modeling of laser ablation. In [1] and [2], the assumptions and theoretical considerations employed in the model were clearly explained with the literature sources for thermal and optical properties. It is true that there are discrepancies between the calculated and measured crater depths below the threshold irradiance, and the source of these discrepancies is unclear. The thermal and optical properties used in the model and the inaccuracy of the model itself to precisely represent complex laser ablation processes all require further investigation. The numerical results do show that thermal evaporation and explosive boiling in conjunction with induced-transparency phenomena are probable processes to describe the measured crater depths below and above the threshold irradiance, respectively. It was also emphasized that the trends and order of magnitude of the calculated maximum crater depths below and above the threshold irradiance agreed with the measured data. The comments about the discrepancies in the calculated results below the threshold only point out general imperfections of a theoretical model rather than problems in the specific model employed in [1].

Craciun claimed that because explosive boiling was observed on 0.1 μm thin films that the critical radius is likely to be smaller than that value. The authors disagree and consider that the critical radius could be greater than the film thickness. It is possible that the vapor bubbles reach a size equal to the film thickness before the bubbles grow to a critical radius. An estimation of the critical radius of vapor bubbles using the theoretical expression provided in [1] and reference therein would to provide better insight to this issue.

In summary, the studies referenced by Craciun do not show the important characteristic observed in [1] such as the nonlinear increase of the crater depth by 3-4 fold across the threshold irradiance, craters of 20 μm deep per single pulse ablation, emission of particulates up to 10-30 μm in diameter, and mass ejection delay of about 300-400 ns from the beginning of ablation pulse. The references only show the production of droplets. Therefore, it seems inappropriate to directly compare the laser irradiances among different ablation processes to determine the threshold for volume boiling. The question should be focused on the real mechanisms responsible for the production of droplets at different ablation conditions rather than on the usage of the term 'volume or explosive boiling'.

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