New findings on plasma and cavity generation in aqueous media with fs and ns laser pulses, and their applications in cell surgery

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We investigated the transition from low- to high-density plasmas in femtosecond and nanosecond optical breakdown at large NA both experimentally and by numerical modeling. Experiments were performed in water but the results on plasma formation are largely transferable to breakdown in solid dielectrics.

Femtosecond breakdown: At threshold, cavitation bubbles with a maximum size smaller than the optical diffraction limited are produced (R = 200 nm for NA = 0.8), and the conversion rate of laser energy into bubble energy is only 0.0002%. The bubble formation threshold corresponds to a peak temperature in the focal volume of only 150°C as revealed by our model of thermoelastically induced bubble formation. We found excellent agreement between theoretically predicted bubble sizes and experimental results. For $T \geq 300$°C, a phase explosion occurs and the conversion rate of laser energy into bubble energy increases gradually until it reaches 1% at $E/E_{th} \approx 2$ and 10% at $E/E_{th} \approx 7$. For $E/E_{th} = 7$ (E = 165 nJ), we observed a luminescent plasma – a phenomenon never been reported for fs breakdown in bulk media at lower NAs. Luminescence indicates a large free-electron density and efficient laser plasma coupling. The corresponding plasma energy density deduced from the plasma volume and absorption is 24 kJ/cm^{-3}, the pressure inferred from the size of the bubble reached after plasma expansion is 13.5 kbar. These results are in striking contrast to recent claims (Juodkazis et al., PRL 96, 2006) that multimegabar pressures are achieved during fs breakdown in solid bulk media.

Nanosecond breakdown: Ns breakdown in commonly believed to be associated with bright plasma luminescence. However, we found that for VIS and UV laser pulses ns-breakdown is a two-step process. In the first step, a non-luminescent low-density plasma is formed the expansion of which creates minute bubbles (R = 500 nm – 10 µm, depending on pulse energy). Electron-hole recombination limits the free-electron density to values $\leq 10^{20}$ cm^{-3}, and the conversion efficiency of laser energy into bubble energy is, at threshold, as small as 0.00003%. For energies 10-30× above the bubble formation threshold (for 532 nm and 355 nm, respectively), plasma suddenly assumes a much larger size (1600× the focal volume), bright luminescence is observed, and large bubbles ($R \geq$ 200 µm) are produced. This plasma inflation is attributed to a thermal ionization runaway that overcomes recombination and results in high plasma densities reaching full ionization. Plasma energy density is limited by energy transport out of the absorbing region by UV radiation and/or ejection of fast electrons. When the laser energy is further increased, the plasma grows along thin streaks into the cone angle. The bright and strongly scattering streaks, which resemble Lichtenberg figures, are the origin of the diffuse plasma luminescence observed in a much larger volume. The interplay of multiphoton ionization and avalanche ionization usually considered in plasma models can only account for the non-luminescent low-density plasmas if recombination is also taken into account. A correct description of the formation of the luminescent high-density plasmas requires consideration of thermal ionization. We established a rate equation model including thermal ionization that predicts the two steps of ns breakdown in excellent agreement with our experimental findings.

Practical consequences: The discovery that minute plasma-mediated effects can be produced not only with femtosecond pulses but also with VIS and IR nanosecond pulses is of particular importance for cost-effective cell surgery and nanomorphing of solid dielectrics.