Supporting Information: Non-photochemical laser-induced nucleation of supercooled glacial acetic acid

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1. Near-infrared absorbance of acetic acid

The near-infrared absorption of glacial acetic acid (GAA) was measured using a spectrometer (Perkin–Elmer, Lambda 900). The heat capacity (C_p = 1.930 J g^{-1} K^{-1}) and density (ρ_L = 1.080 g cm^{-3}) of GAA at –9 °C were estimated by fitting and extrapolation of data from the literature.[1, 2] For a single pulse at a peak power of 40 MW cm^{-2}, the energy absorbed by acetic acid in the illuminated volume (0.046 cm^3) corresponds to a temperature rise of 6 mK; at 900 MW cm^{-2}, the rise is 0.14 K. We assume that convection and conduction are sufficiently fast to allow redistribution of heat throughout the entire volume of the sample (2 cm^3). At a peak power of 40 MW cm^{-2}, the energy absorbed by acetic acid would be 0.6 mJ pulse^{-1}, corresponding to a temperature rise of 14 mK for 100 pulses.

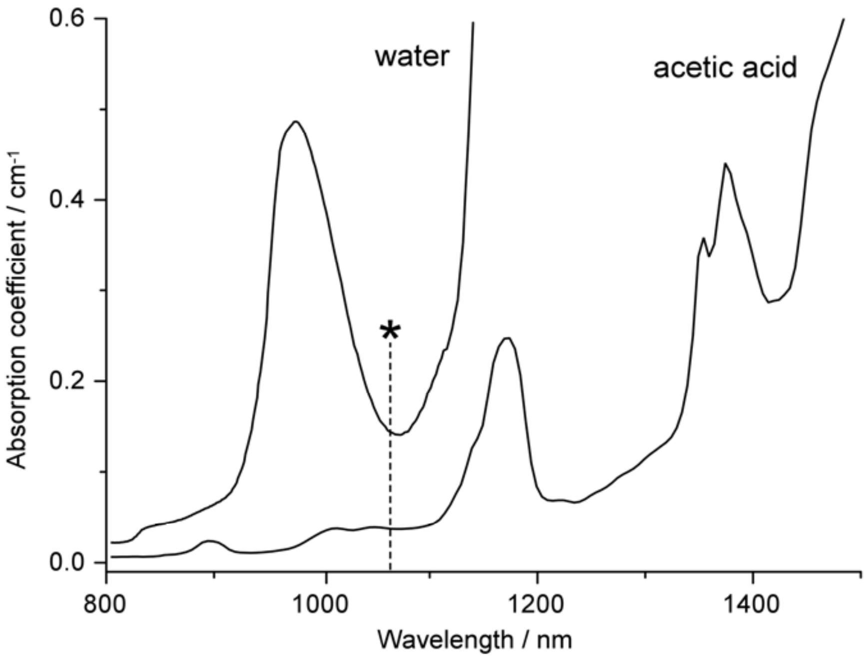

Figure S1. Plot of the near-infrared absorption coefficient of water and acetic acid. The absorption coefficients at 1064 nm, marked by the arrow, are 0.144 cm^{-1} (water, Kou et al.[3]) and 0.0370 cm^{-1} (acetic acid, present work).

2. Refractive indices of acetic acid

The refractive indices of GAA at 1064 nm and –9 °C are not readily available, therefore we estimated them by extrapolation from available data in the literature. For the liquid, we extrapolate measurements of El-Kashef to find n_L = 1.3614 (at 1064 nm, –9 °C).[4] For the solid, we use the Lorenz–Lorentz relation to estimate n_S from n_L at a specific frequency and temperature, using the densities of the solid (ρ_S = 1.274 g cm^{-3} by interpolation from literature data[1]) and the liquid (ρ_L = 1.080 g cm^{-3}).[5] We calculate n_S = 1.4357 at 1064 nm, which is in very good agreement with our calculations for a crystalline cluster consisting of 27 unit cells using the semi-empirical PM6–DH+ method (n = 1.4385).[6-8]

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