Electronic Supplementary Information (ESI) to "Nanopore-mediated ultrashort laser-induced formation and erasure of volume nanogratings in glass"

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Table I. Optical and thermo-mechanical properties of fused silica.

Physical properties	Fused silica
Density $\rho[g/cm^3]$	2.2[a,c,n,p,q,t]
Refractive index ($\lambda = 800$ nm)	1.45[d,g]
Electron band gap $E_g[eV]$	9[d], 9.3[m], 7.2-7.8[c,f,j]
Kerr effect $10^{-16}[cm^2/W]$	3.54[d,g], 3.89[i]
Electron collision time $[fs]$	0.2-23.3: 0.5[v], 1.27[d], 10[g]
Electron recombination time $\tau_{rec}[ps]$	0.06[s], 0.15[i,g,r,d,m], 1-2[u,v], 30[c], > 100[u,v]
Photoionization rate w_{pi} $\lambda = 800$ nm $\sigma_6[\frac{m^9}{W^6 s}]$	Keldysh[g] 6-photon: $2 \cdot 10^{-65}$[d,m,g], $6 \cdot 10^{-63}$[e,l,g], $3.15 \cdot 10^{-67}$[s,g]
Heat capacity $C_i[J/(kgK)]$	772-790[a,t], 730-752[b,d,n], 1335-1440[x](1600-2400K), 1450[c,w] (1873K), 704-845[f], 1100[q]
Thermal conductivity $k_i[W/(mK)]$	1.31[t], 1.38-1.4[a,n], 1.4-2.514[b], 1.38-1.67[f], 1.4-3.0[h,w] (300-2500K)
Softening temperature $T_{melt}[K]$	2000-2006[b,q,e], 1873-1875[c,t], 1750[n], 1993[p]
Thermal diffusivity, $10^{-7}D[m^2/s]$	5.9
Shear modulus $G[GPa]$	31.2[b,w], 30-33.5[k] (200-1800K), 31-33[p] (300-1700K)

[a−f]References 1, 2, 3, 4, 5, 6. [g−l]References 7, 8, 9, 10, 11, 12. [m−t]References 13, 14, 15, 16, 17, 18, 19,20. [u−x]References 21, 22, 23, 24.

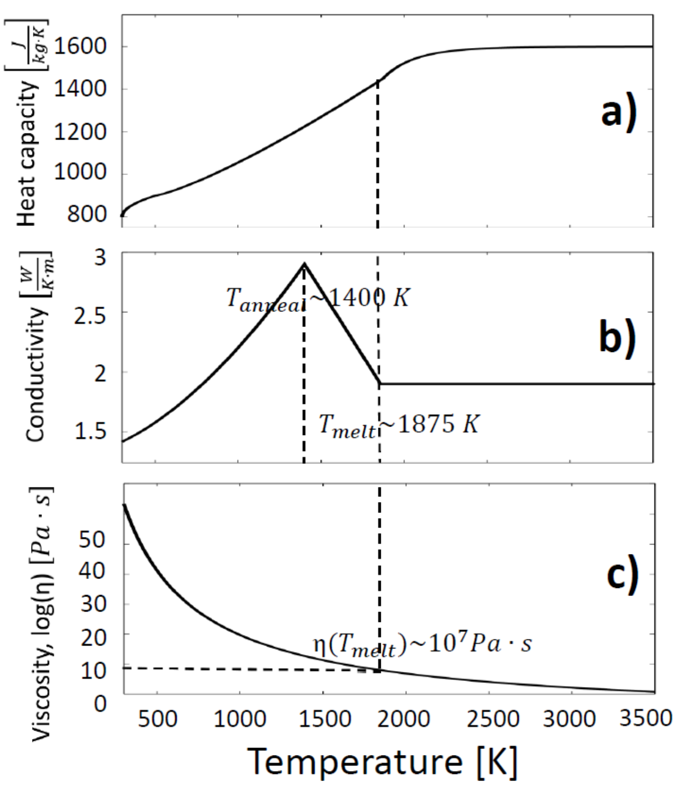

Figure 1. Heat capacity, thermal conductivity and viscosity temperature dependencies for fused silica glass.

APPENDIX: THERMO-ELASTIC PROPERTIES OF FUSED SILICA

The temperature dependence of ion heat capacity for fused silica is plotted in Fig. 1(a). The values are taken from recent experimental data [3, 25] approximated as

$$C_i = \begin{cases} 1.6 \cdot \left(1 + \sqrt{\frac{T_i - 300}{200}}/8\right) \frac{MJ}{m^3K}, 300K \le T_i < 500K \\ 1.8 \cdot \left[1 + \left(\frac{T_i - 500}{1375}\right)^{1.25} \frac{11}{18}\right] \frac{MJ}{m^3K}, 500K \le T_i < 1875K \\ [3.2 - 0.3 \left(\frac{1875}{T_i}\right)^{10}] \frac{MJ}{m^3K}, \quad T_i \ge 1875K, \end{cases} \tag{1}$$

consistent with the experimentally reported values given in Table I. In fact, the heat capacity C_i significantly increases with temperature (see Suppl. Info in Ref. [24]). For instance, if the value corresponding to the initial conditions is taken ($C_i \approx 800$ J/(kgK)) instead of the temperature dependent heat capacity given by (1), the lattice temperatures for the considered laser irradiation conditions are overestimated by more than 50%.

An adaptive fit is used to reproduce the temperature dependence of the fused silica thermal conductivity with slope discontinuities at the annealing temperature $T_{anneal} = 1400$ K and the melting temperature $T_{melt} = 1875$ K [8] as follows

$$k_i = \begin{cases} 1.3 + 1.6 \cdot \left(\frac{T_i}{1400}\right)^{1.7} \frac{W}{Km}, \quad 300K \le T_i < 1400K \\ 2.9 - \frac{T_i - 1400}{475} \frac{W}{Km}, \quad 1400K \le T_i < 1875K \\ 1.9 \frac{W}{Km}, \quad T_i \ge 1875K. \end{cases} \tag{2}$$

The temperature dependence is plotted in Fig. 1(b) and the experimentally reported values are summarized in Table I. The thermal conductivity k_i rises up to the annealing temperature. It then drops down to the melting point, according to the recent experimental measurements performed by using infrared thermography [8, 23]. This discontinuity significantly influences the temperature evolution in the multi-pulse accumulation regimes since the cooling time of the lattice is related to the diffusivity $D = k_i/\rho C_i$ as follows $\tau_{cool} = {w_0}^2/D$.

The corresponding temperature dependency of viscosity is described by the Vogel-Fulcher-Tammann (VTF) model, fitting the viscosity data for the intermediate temperatures over many orders of magnitude with high accuracy [26]

$$log[\eta(T_i)] = A + B/(T_i - T_V), \tag{3}$$

where $A = -7.925$ and $B = 31555.9$ K are constants, and $T_V = -142$ K is Vogel's temperature. For fused silica, the parameters are taken from Ref. [27]. The resulting viscosity curve shown in Fig. 1(c) fits well the experimental measurements [28–30] and is consistent with the other models [26]. The structural relaxation time is defined as $\tau_M = \eta/G$, where the shear bulk modulus is taken equal to $G = 31$ GPa, in agreement with a number of the experimental measurements performed for a wide range of temperatures [11, 16, 25].

[1] M. Lancry, F. Zimmerman, R. Desmarchelier, J. Tian, F. Brisset, S. Nolte, and B. Poumellec, Applied Physics B **122**, 1 (2016).
[2] Y. P. Meshcheryakov, M. V. Shugaev, T. Mattle, T. Lippert, and N. M. Bulgakova, Applied Physics A **113**, 521 (2013).
[3] D. Puerto, J. Siegel, W. Gawelda, M. Galvan-Sosa, L. Ehrentraut, J. Bonse, and J. Solis, JOSA B **27**, 1065 (2010).
[4] S. Najafi, A. S. Arabanian, and R. Massudi, Journal of Physics D: Applied Physics **49**, 255101 (2016).
[5] N. M. Bulgakova, R. Stoian, and A. Rosenfeld, Quantum Electronics **40**, 966 (2010).
[6] E. A. Romanova and A. I. Konyukhov, Optics and Spectroscopy **104**, 784 (2008).
[7] A. Couairon, L. Sudrie, M. Franco, B. Prade, and A. Mysyrowicz, Phys. Rev. B **71**, 125435 (2005).
[8] P. Combis, P. Cormont, L. Gallais, D. Hebert, L. Robin, and J.-L. Rullier, Applied Physics Letters **101**, 211908 (2012).
[9] P. Martin, S. Guizard, P. Daguzan, G. Petite, P. D'Oliveira, P. Meynadier, and M. Perdrix, Phys. Rev. B **55**, 5799 (1997).
[10] M. Sakakura, M. Terazima, Y. Shimotsuma, K. Miura, and K. Hirao, Las. Chem. **2010**, 1 (2010).
[11] R. L. Parc, C. Levelut, J. Pelous, V. Martinez, and B. Champagnon, Journal of Physics: Condensed Matter **18**, 7507 (2006).
[12] M. Lenzner, J. Krüger, S. Sartania, Z. Cheng, C. Spielmann, G. Mourou, W. Kautek, and F. Krausz, Phys. Rev. Lett. **80**, 4076 (1998).
[13] L. Hallo, A. Bourgeade, V. T. Tikhonchuk, C. Mezel, and J. Breil, Phys. Rev. B **76**, 024101 (2007).
[14] J.-x. Cai, X. Qu, H. Li, and G.-y. Jin (2015) pp. 96711C–96711C–6.
[15] M. Schmidt, F. Vollertsen, M. Geiger, K. Cvecek, I. Alexeev, I. Miyamoto, and M. Schmidt, Physics Procedia **5**, 495 (2010).

[16] M. Fukuhara and A. Sanpei, Japanese Journal of Applied Physics **33**, 2890 (1994).
[17] Y. Okamoto, I. Miyamoto, K. Cvecek, A. Okada, K. Takahashi, and M. Schmidt, Journal of Laser Micro Nanoengineering **8**, 65 (2013).
[18] S. Quan, J. Hong-Bing, L. Yi, Z. Yong-Heng, Y. Hong, and G. Qi-Huang, Chinese Physics Letters **23**, 189 (2006).
[19] M. Li, S. Menon, J. P. Nibarger, and G. N. Gibson, Phys. Rev. Lett. **82**, 2394 (1999).
[20] A. Horn, H. Khajehnouri, E.-W. Kreutz, and R. Poprawe (2003) pp. 393–400.
[21] C. Mauclair, K. Mishchik, A. Mermillod-Blondin, A. Rosenfeld, I.-V. Hertel, E. Audouard, and R. Stoian, Physics Procedia **12**, 76 (2011).
[22] P. K. Velpula, M. K. Bhuyan, F. Courvoisier, H. Zhang, J. P. Colombier, and R. Stoian, Laser & Photonics Reviews **10**, 230 (2016).
[23] T. Doualle, L. Gallais, P. Cormont, D. Hébert, P. Combis, and J.-L. Rullier, Journal of Applied Physics **119**, 113106 (2016).
[24] R. Kraus, S. Stewart, D. Swift, C. Bolme, R. Smith, S. Hamel, B. Hammel, D. Spaulding, D. Hicks, J. Eggert, *et al.*, Journal of Geophysical Research: Planets **117**, 1 (2012).
[25] J. Zhao, J. Sullivan, J. Zayac, and T. D. Bennett, Journal of Applied Physics **95**, 5475 (2004).
[26] M. I. Ojovan, Physics and Chemistry of Glasses-European Journal of Glass Science and Technology Part B **53**, 143 (2012).
[27] W. Martienssen and H. Warlimont, *Springer handbook of condensed matter and materials data* (Springer Science & Business Media, 2006).
[28] G. Urbain, Y. Bottinga, and P. Richet, Geochimica et Cosmochimica Acta **46**, 1061 (1982).
[29] R. H. Doremus, Journal of Applied Physics **92**, 7619 (2002).
[30] J. A. Billo, J. Jones, W. Asghar, R. L. Carter, and S. M. Iqbal, Applied Physics Letters **100**, 233107 (2012).