

Supporting information: Vapor-Deposited Ethylbenzene Glasses Approach “Ideal Glass”
Density

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Fits of ellipsometry data

In Figure S1 we provide an example showing the fitting procedure used with ellipsometry data obtained during temperature-ramping. The figure shows thickness and refractive index data obtained as the temperature is increased for the same data set shown in Figure 2 of the main text. We fit the data to an equation (S1) described by Dalnoki-Veress et al.⁵³

$$h(T) = w \left(\frac{M - G}{2} \right) \ln \left[\cosh \left(\frac{T - T_g}{w} \right) \right] + (T - T_g) \left(\frac{M + G}{2} \right) + c \quad (S1)$$

This procedure is intended to eliminate potential bias associated with manually extracting T_g and T_{onset} values.

Figure S1 illustrates that Equation S1 fits the data well except for the transformation of the as-deposited glass into the supercooled liquid, between 123 K and 125 K. (The non-monotonic behavior displayed in the data over this 2 K range is indicative of transformation via a growth front. The rest of the fit is not affected.) The arrows in Figure S1 indicate the meaning of the values extracted from the fit for T_g , T_{onset} , and h_0 . T_g is the glass transition temperature for the liquid-cooled glass, obtained upon heating; T_{onset} is the beginning of the transformation for the PVD glass; h_0 is the thickness at T_g and is used to normalize the y-axis data in Figure 3. The temperature axis in Figure S1 has been corrected by subtracting the difference between the fitted value of T_g and the reference T_g value of 115.7 K. (See methods for details.)

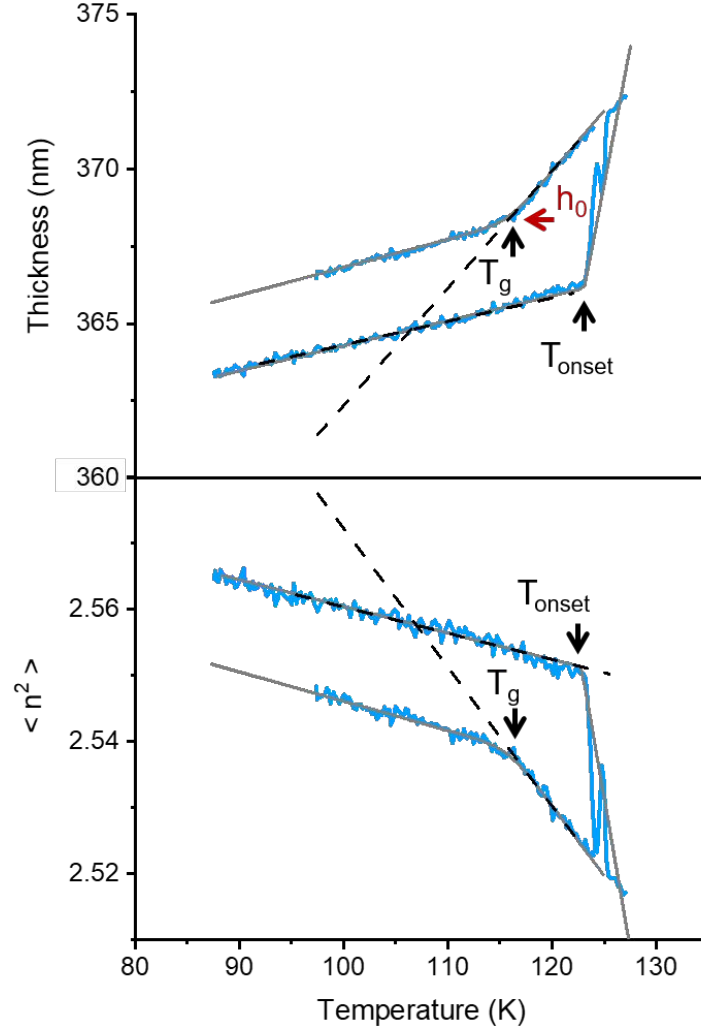


Figure S1. Fits of equation S1 to the heating curves for the as-deposited glass and the liquid-cooled glass for the data presented in Figure 2 of the main text. The blue lines are the thickness and refractive index data, the grey lines are the fits to equation S1, and the black dashed lines are linear extrapolations of the fits to the supercooled liquid data. The intersection between the supercooled liquid extrapolation and the glass data defines the fictive temperature T_f . Arrows indicate values that are extracted from the fits to equation S1.