Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Material

Composition-dependent metallic glass alloys correlate atomic mobility with collective glass surface dynamics

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Cluster size analysis Diameters of clusters were measured following the procedure in ref. [10] in the main text. For each surface, 100 random points were generated in the image being analyzed and clusters closest to the points were measured. The cluster diameters were taken as the average of the measured width and length in x and y directions. The diameters in nm were then converted to the atomic weighted diameter (AWD) (or glass-forming unit). For calculations of the AWD, the following atomic diameters were used: 0.187 nm for La, 0.124 nm for Ni, 0.143 nm for Al and 0.128 nm for Cu. The AWDs for $La_{60}Ni_{15}Al_{25}$, $La_{50}Ni_{15}Al_{25}Cu_{10}$, $La_{60}Ni_{10}Al_{25}Cu_5$ and $La_{60}Ni_{15}Al_{15}Cu_{10}$ are 0.333 nm, 0.321 nm, 0.334 nm and 0.330, respectively. The cluster size distributions were calculated by histogramming all the measured diameters, with binning of 1 AWD.

Average rate analysis Average rate was analyzed by counting number of hops per unit time per unit area. For each surface, ≈ 15 movies were analyzed.

Movie S1: Complete STM movie for the cluster in Fig. 3c. Scanning conditions: 10 pA, 1 V. STM image sizes: 10.4 nm x 10.4 nm. 41 s/frame. Playback rate: 1 s/frame.

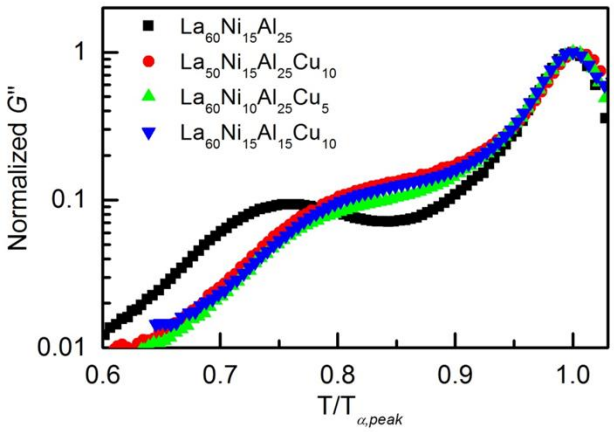

Figure S1: Isochronal DMA. Same as in Fig. 1 but T is normalized to $T_{\alpha,peak}$.

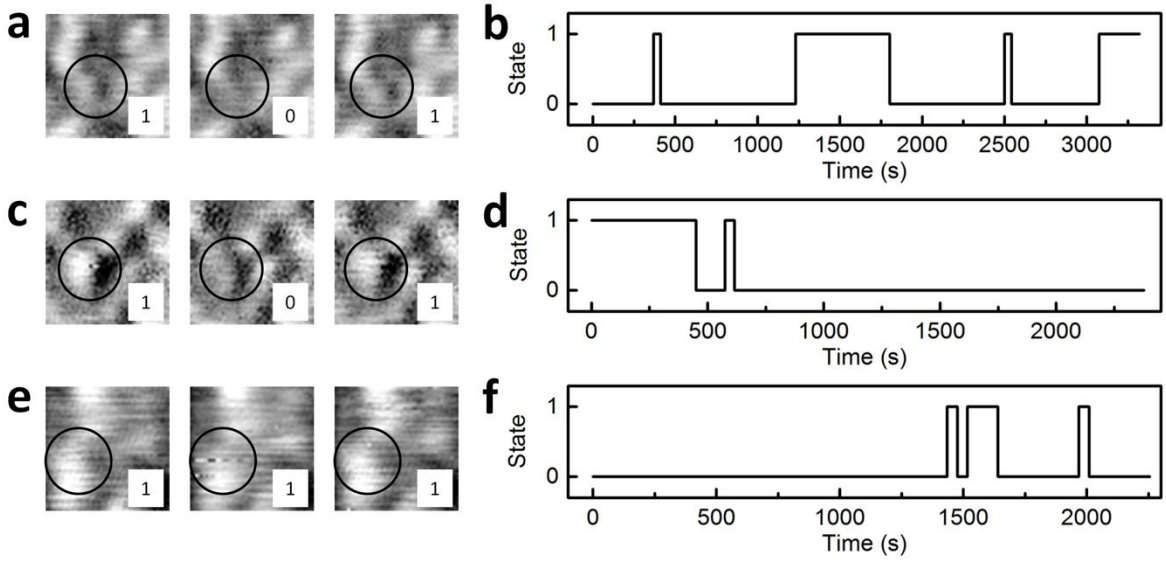

Figure S2: Two-state clusters and time traces on the surface of other compositions. (a)-(b) $La_{60}Ni_{15}Al_{25}$ (as-cast), (c)-(d) $La_{50}Ni_{15}Al_{25}Cu_{10}$ (as-cast), (e)-(f) $La_{60}Ni_{10}Al_{25}Cu_5$ (as-cast). Scanning conditions: 10 pA, 1 V. All STM derivative image sizes: 6 nm x 6 nm.

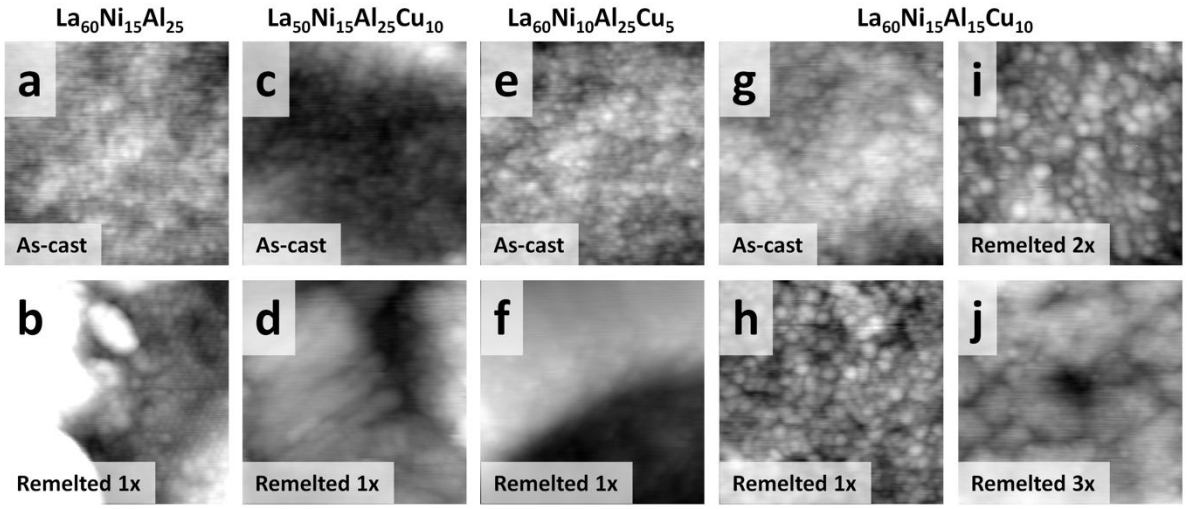

Figure S3: The corresponding topographic version of Fig. 4.

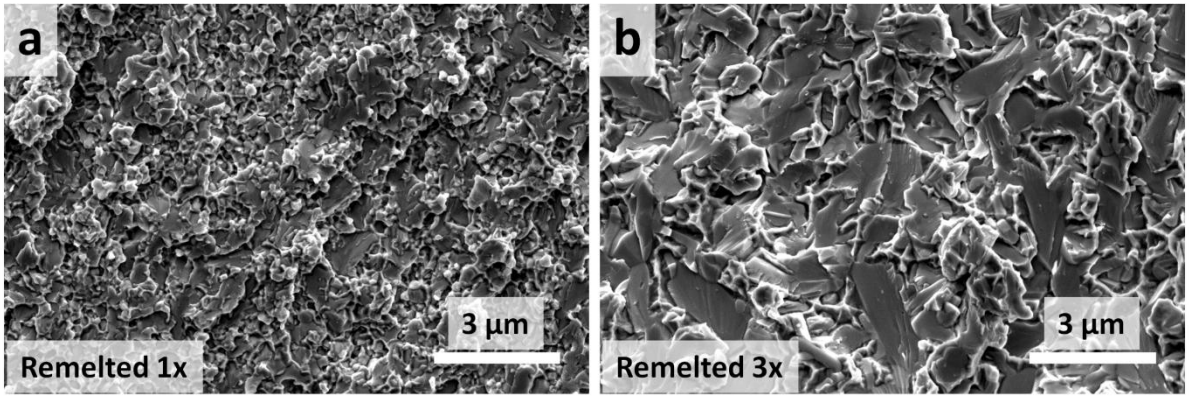

Figure S4: SEM images of the bulk fracture surfaces of the $La_{60}Ni_{15}Al_{15}Cu_{10}$ samples after remelting (a) 1 cycle and (b) 3 cycles. The crystalline grain size increases after successive remelting.