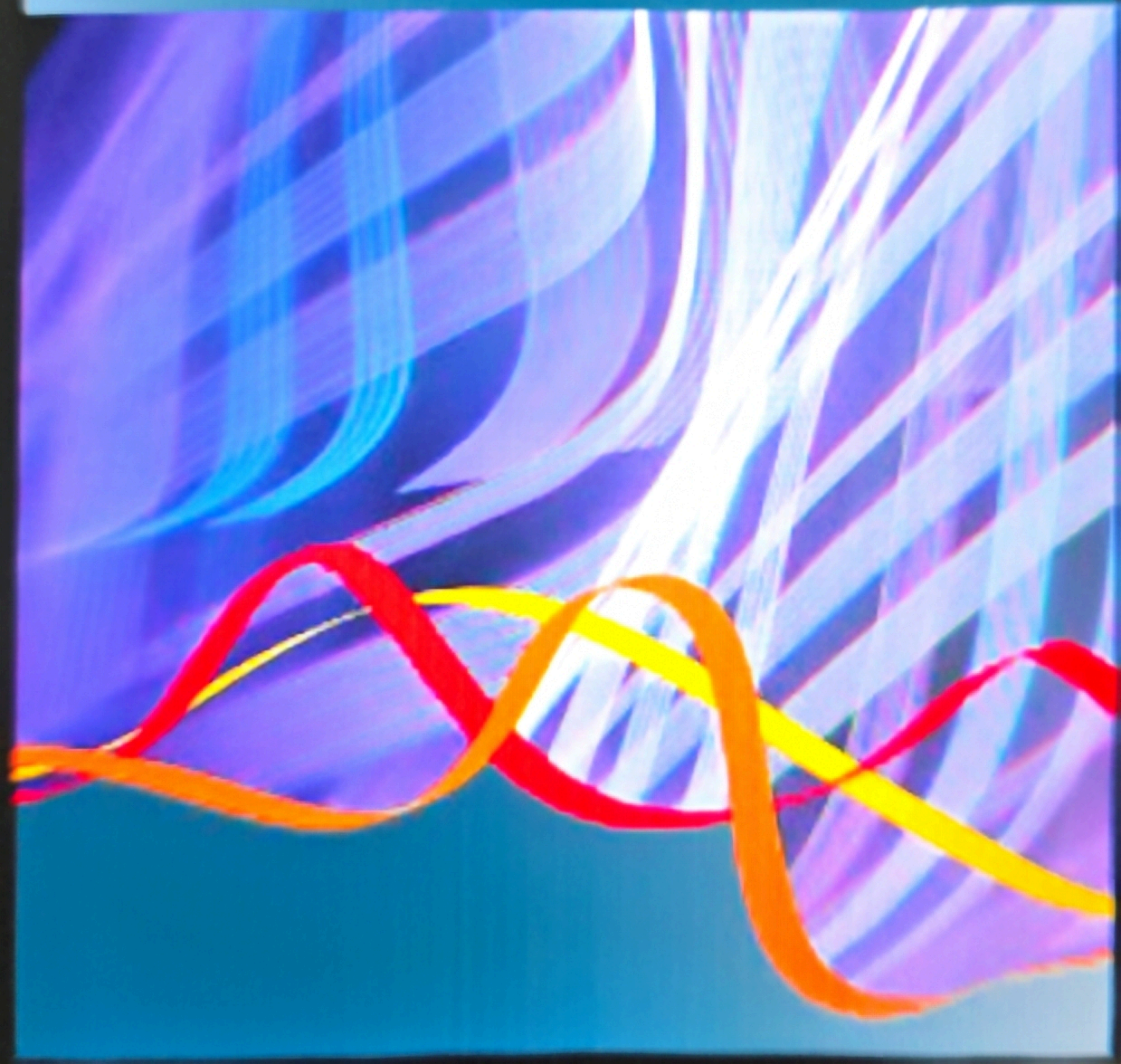


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Deprotonation-mediated vitrification of organic-inorganic hybrid perovskites

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Glasses made from organic-inorganic hybrid perovskites are emerging non-crystalline semiconducting materials whose versatile composition and exceptional processability allow for manifold applications. The current melt-quenching approaches are compatible to only few perovskite crystals, as most undergo irreversible decomposition prior to melting. Herein, we report a general flux-mediated approach to produce a library of high-quality perovskite glasses with different organic molecules, metal centers, and framework dimensionality. We show that the partial deprotonation of organic moiety by alkali metal hydroxide flux modulates the intermolecular interactions in perovskites, and activates the flow unit, which depresses the fusion temperature ranging 21–77 °C with flux ratio <1.1 wt%. More critically, the utilization of hydroxide flux creates a large melt window, and enables the thermal vitrification of many inherently non-melttable systems, such as (BA)₂PbI₄ (BA = butylamine) and (DGA)PbI₄ (DGA = 1,1-dimethylbiguanide). Using these semiconductive glasses, we realize high-performance X-ray detection devices showing a sensitivity of 35674.1 $\mu\text{C Gy}_{\text{air}}^{-1} \text{cm}^{-2}$ under 500 V mm⁻¹ electric field, and a high-resolution X-ray imaging system via its seamless integration on commercial thin-film transistor backplanes.

Glass is defined as a non-equilibrium, amorphous substance, which is usually regarded as a frozen liquid and undergoes reversible transformation near the glass transition temperature (T_g). Glassy semiconductors, characterized by the absence of long-range atomic order^{1–6}, are indispensable building blocks in modern electronics and photonics^{7–10}. Recently, the discovery of organic-inorganic hybrid perovskite glass unlocked unprecedented potential for systematically engineering the chemical composition, electronic structure and functionality of semiconductive glasses^{11–14}. These perovskite glasses properly inherit the physical properties from the crystalline analogues, while gaining extended functionality, like melt-processing and integration versatility for potential applications in radiation detection,

rewritable optical storage, phase change memory and display technologies^{15–23}.

Although perovskite structure offers unlimited chemical and structural diversity²⁴, only a few semiconductive perovskite materials thus far have presented the capability to undergo the crystal-to-glass transformation via the common melt-quenching procedure^{11,14}, and the resulting glasses often suffer from serious material quality issues, like bubble entrapment, microcracking, and partial decomposition. The fundamental difficulty is that the thermal decomposition at the decomposition temperature (T_d) inevitably occurs below the fusion temperature (T_m) for most perovskites upon the thermal vitrification process⁴. Recent development of perovskite glass materials

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