Supporting Information:

Organic-component dependent thermal conductivity reduction in ALD/MLD grown ZnO:organic superlattice thin films

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Thermal Conductivity Measurement Details

The reported thermal conductivity values were measured in the cross-plane direction using a time-domain thermoreflectance (TDTR) technique. This optical pump-probe technique has been widely applied to the measurement of thermal properties of thin films and structures, including our previous works on inorganic-organic superlattices. In summary, a relatively high-energy ‘pump’ pulse excites the surface of the sample, while a mechanically-delayed, low-energy ‘probe’ pulse operates as an optical thermometer and monitors the transient cooling dynamics of the sample surface.[1-4] By fitting the experimental cooling curve to a multilayer thermal model, we can accurately extract the thermal conductivity of the ZnO:organic superlattices. An example of our TDTR data and associated fit of the thermal model is shown in Figure S1 for a ZnO:hydroquinone SL thin film with 6 organics layers.

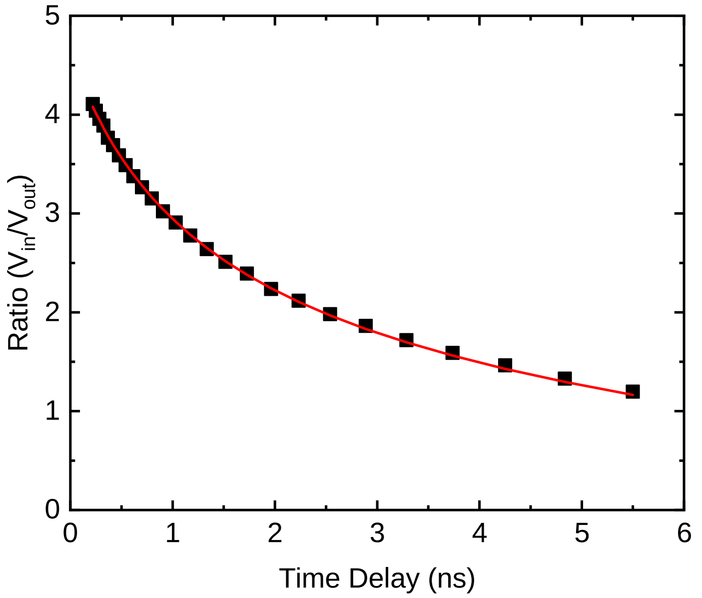

Figure S1. TDTR data (black squares) and associated multilayer thermal model fit (red line) for a ZnO:hydroquinone SL thin film with 6 organic layers.

We deposit an additional 80 nm Al layer on top of the SL of interest (e.g. 80 nm Al/SL/sapphire substrate structure) by electron beam evaporation, which operates as an ideal thermo-optical transducer for our TDTR measurements. The reported error bars account for the standard deviation associated with repeated measurements across the sample surface to account for any sample inhomogeneities, as well the propagation of the uncertainty associated with the error in our Al transducer thickness (80 +/- 2.5 nm). We note that due to the extremely high sensitivity of the SL/sapphire TBC, we assume a nominal value of 125 W m^{-2} K^{-1} for all samples; this value should be constant across all SLs as each interface is a ZnO/sapphire interface and should thus be identical regardless of SL geometry. In addition to fitting for the thermal conductivity of the SL itself, we also measured the thermal boundary conductance of the Al/SL interfaces, finding a TBC of 61 W m^{-2} K^{-1}.

X-ray Reflectivity (XRR) Patterns Fitting Protocol

The initial models were made with the following layers from the bottom to the top: Si (substrate), SiO_2, ZnO, X number of (Organic + ZnO), H_2O, CO_2. After the initial model construction the following process was implemented to achieve the best possible results:

1. All the ZnO layers were initially linked to decrease the number of the variables.
2. Density of Si was kept fixed initially due to its drastic effect on the fittings.
3. Segmented fitting method was used with only the clear section of the pattern as an input.
4. Fitting was started with a margin error of 40% and proceeded till the margin error of 10%.
5. Si density was added to the fitting process.
6. All the ZnO layers were delinked and fitted individually.
7. Fitting range was increased to the full range with the clear section as the initial fit for the segmented fit.
8. Instrumental factors were added to the fitting process to be modified.
9. Fitting for the full range was started with 40% margin error and proceeded till the fitting margin of 10%.
10. Final fittings were double checked with combining Genetic Algorithm with Segmented fitting method.

In the end the average of all the similar layers was taken as the final value for the organic and ZnO layers.

References

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