Supporting Information

Thickness-Dependent Thermal Conductivity of Suspended Two-Dimensional Single-Crystal In_2Se_3 Layers Grown by Chemical Vapor Deposition

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Determination of the full-width-at-half-maximum (FWHM) of the 632 nm laser beam

In a previous study,[S1] we used an auto-correlation method to measure the FWHM (~ 445 nm) of the 517 nm laser beam focused through the same 100 X objective used here on a blank SiO_2/Si substrate. As the FWHM is proportional to the wavelength, we obtained a FWHM (~ 544 nm) for the 632 nm laser beam.

Table S1 Material parameters used in simulations

	Thermal conductivity (W/m*K)	In_2Se_3 Interface Thermal conductance (MW/m^{2}K)
Si	130	
Si_3N_4	30	50
Air	0.0257	0.127

Simulated temperature

The average simulated temperature (T_m) was obtained through the following equation,[S2] $T_m \approx \frac{\int_0^R T_1(r)q(r)rdr}{\int_0^R q(r)rdr}$. $T_1(r)$ is the temperature profile inside the hole, R is the beam width, and $q(r)$ is the volumetric Gaussian beam heating:[2] $q(r) =$

$\frac{P_{abs}}{\pi r_0^2 t}\exp\left(-\frac{r^2}{r_0^2}\right)$, where r_0 is half of the beam width, and P_{abs} is the absorbed laser power given by $P_0(1-e^{-\alpha t})$, with P_0 as the incident laser power, α as the measured absorption coefficient for the 632-nm laser, and *t* as the layer thickness. The matching between this temperature and the temperature measured by the Raman spectroscopy allowed for the determination of $k_{//}$.

Callaway Model:

The following Callaway model equations were used for fitting.

$$k(\mathrm{T}) = \frac{k_B}{2\pi^2 c}\left(\frac{k_B T}{\hbar}\right)^3 \int_0^{\theta_D/T} \tau_c \frac{x^4 e^x}{(e^x-1)^2} dx \quad (1)$$

$$\tau_c^{-1} = \frac{c}{t} + A\omega^4 + B\omega^2 T e^{-\frac{\theta_D}{3T}} \quad (2)$$

where ω is the phonon frequency, k_B is the Boltzman constant, θ_D is the Debye temperature, $\hbar$ is the reduced Plank constant, x is given by $\hbar\omega/k_B T$, T is the absolute temperature, *c* is the velocity of sound (average *c* = 2500 m/s), and *t* is layer thickness.[S3] As θ_D is unknown for In_2Se_3, we used the θ_D in silicon (645 K). The fitting of the experimental results was obtained by optimizing the values of *A* and *B*, which are approximately 2.7×10^{-43} s^3 and 1.5×10^{-20} s/K, respectively.

References:

[S1] Tao, X.; Mafi, E.; Gu, Y. Synthesis and Ultrafast Carrier Dynamics of Single-Crystal Two-Dimensional $CuInSe_2$ Nanosheets. *J. Phys. Chem. Lett.* **2014,** 5, 2857-2862.

[S2] Lee, J. U.; Yoon, D.; Kim, H.; Lee, S. W.; Cheong, H. Thermal Conductivity of Suspended Pristine Graphene Measured by Raman Spectroscopy. *Phys. Rev. B* **2011,** *83*, 081419-4.

[S3] Park, N. W.; Lee, W. Y.; Kim, J. A.; Song, K.; Lim, H.; Kim, W. D.; Yoon, S. G.; Lee, S. K. Reduced temperature-dependent thermal conductivity of magnetite thin films by controlling film thickness. *Nanoscale Res. Lett.* **2014**, 9, 96-8.