## **Supplementary Material**

## Sonication-Assisted Liquid-Phase Exfoliated $\alpha$ -GeTe: A Two-

## Dimensional Material with high Fe<sup>3+</sup> Sensitivity

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| Table S1. Surface tension of selected solvents. |                           |  |  |  |  |  |  |
|---|---------------------------|--|--|--|--|--|--|
| Solvents  | Surface tension           |  |  |  |  |  |  |
|   | (dynes cm <sup>-1</sup> ) |  |  |  |  |  |  |
| Chloroform (TCM)                                | 27.5                      |  |  |  |  |  |  |
| Dimethyl sulfoxide (DMSO)                       | 42.78                     |  |  |  |  |  |  |
| Dimethylformamide (DMF)                         | 35.20                     |  |  |  |  |  |  |
| Acetonitrile (ACN)                              | 28.1                      |  |  |  |  |  |  |
| N-methyl-2-pyrrolidone (NMP)                    | 40.7                      |  |  |  |  |  |  |
| deionized (DI W)                                | 72                        |  |  |  |  |  |  |
| Ethanol (EtOH)                                  | 22.18                     |  |  |  |  |  |  |

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Fig. S1 Dispersion of bulk  $\alpha$ -GeTe powder in various solvents after 3 h of sonication (left) and after one-day rest (right) with the solvents indicated on the lid of the bottles.

It is well known that successful exfoliation of layered materials by sonication-assisted liquid-phase exfoliation depends on several factors, including the type of solvent, ultrasonication time, and rate of centrifugation. Among these factors, the type of solvent is the most crucial factor, and it determines the efficiency of the exfoliation process and the quality of the exfoliated nanosheets.

If the surface energy of the solvent is close to that of the bulk layered material, the energy cost of the exfoliation is lowest, and the quality of the  $\alpha$ -GeTe nanosheets prepared by sonication-assisted liquidphase exfoliation can then be optimized. In this work, since the surface energy of  $\alpha$ -GeTe is unknown, we tested several solvents with different surface tensions for the sonication-assisted liquid-phase exfoliation of bulk  $\alpha$ -GeTe, including chloroform (TCM), dimethyl sulfoxide (DMSO), dimethylformamide (DMF), acetonitrile (ACN), N-methyl-2-pyrrolidone (NMP), deionized water (DI W), and ethanol (EtOH). The digital photographs show  $\alpha$ -GeTe dispersions with a concentration of 2 mg mL<sup>-1</sup> after sonication for 3 h and after one-day rest in different solvents. The results demonstrate that ethanol provides the best dispersion stability for  $\alpha$ -GeTe, as indicated by the absence of obvious particles or sediments.



Fig. S2 Representative wide-area AFM images of  $\alpha$ -GeTe nanosheets prepared in different time from 2h to12 h (a-f). Statistical analysis of

height (g), aspect ratio (h) and area (i) of  $\alpha$ -GeTe nanosheets prepared in different time from 2h to12 h.



Fig. S3 Representative wide-area AFM images of  $\alpha$ -GeTe nanosheets prepared at different centrifugal speed from 500 rpm/min to5000 rpm/min (a-d). Statistical analysis of height (e), area (f) and aspect ratio (g) of  $\alpha$ -GeTe nanosheets prepared at different centrifugal speed from 500 rpm/min to 5000 rpm/min.

Once the solvent is selected, other experimental factors may have influences on the thickness of nanosheets prepared by sonication-assisted liquid-phase exfoliation. Among others, sonication time and centrifugation speed are important factors.

Figure S2 shows the effect of the sonication time on the thickness of  $\alpha$ -GeTe nanosheets. We characterized  $\alpha$ -GeTe nanosheets prepared in different ultrasonic time from 2h to12 h using AFM (Fig. S2 a-f) and the height, aspect ratio and area were statistically analyzed. The results show that thickness of  $\alpha$ -GeTe nanosheets is decreasing with the increase of ultrasonic time, more important, the change of thickness is very slight after ultrasound for 12 h. The height of nanosheets is ~5 nm, the aspect ratio of nanosheets is ~1.38 and the area of nanosheets is ~1.4×10<sup>6</sup> nm<sup>2</sup> after 12 h ultrasound. Therefore we determined the optimized ultrasonic time is 12 h.

Figure S3 shows the effect of the centrifugation speed on the thickness of  $\alpha$ -GeTe nanosheets. We characterized  $\alpha$ -GeTe nanosheets prepared at different centrifugal speed from 500 rpm/min to 5000 rpm/min and the same

sonication time (12 h) using AFM (Fig. S3 a-f) and the height, area and aspect ratio were statistically analyzed. Among them, thickness of  $\alpha$ -GeTe nanosheets prepared by 3000 rpm/min and 5000 rpm/min centrifugation speed is more uniform and thinner compared with others. Therefore we mainly analyzed the properties of  $\alpha$ -GeTe nanosheets prepared by 3000 rpm/min and 5000 rpm/min centrifugation speed in the following discussions.

**Table S2** Structure parameters of bulk  $\alpha$ -GeTe and monolayer  $\alpha$ -GeTe, in which a is the lattice constant, d is the height of the buckled layer, diagonal line length is the width of hexagonal ring with diagonal direction, bond length Ge-Te and bond angle Ge-Te-Ge are also given, the layer distance is the distance between layer and layer.

| Models gr | Space                | а    | d    | Diagonal line length | O(2)  | Bond length | Layer distance |
|-----------|----------------------|------|------|----------------------|-------|-------------|----------------|
|           | group                | (Å)  | (Å)  | (Å)                  | Ø(')  | (Å)         | (Å)            |
| α-GeTe    | D2M1                 | 2.06 | 1.57 | 4.57                 | 91.17 | 2.77        |                |
| (mono)    | PSIMI                | 5.90 |      |                      |       |             |                |
| α-GeTe    | α-GeTe<br>(bulk) R3M | 4 22 | 1 49 | 1 81                 | 95.40 | 2.86        | 2 59           |
| (bulk)    |                      | 4.23 | 1.40 | 4.04                 |       | 2.00        | 5.58           |



Fig. S4 Phonon band dispersion of monolayer a-GeTe, demonstrating the kinetic stability of the material.



Fig. S5 XRD pattern of bulk  $\alpha$ -GeTe.



Fig. S6 SEM images of pristine bulk  $\alpha$ -GeTe powder (a) and  $\alpha$ -GeTe powder after sonication (b).



Fig. S7 Photograph of  $\alpha\mbox{-}Ge\mbox{-}Te\mbox{-}Sol\mbox{-}B$  dispersion showing the Faraday–Tyndall effect.



Fig. S8 (a) Concentration-dependent absorption spectra of  $\alpha$ -GeTe nanosheet dispersions. (b) Normalized absorbance intensity over the characteristic length of the cell (A/L) at different concentrations for  $\lambda = 800$  nm. The extinction coefficient ( $\alpha$ ) is extracted by linear fitting and can be used to estimate the concentration of other solutions.



Fig. S9 A histogram of the number of visual observations of flakes as a function of the number of monolayers per flake in Sol-B.

In order to estimate the product yield of monolayer  $\alpha$ -GeTe, we count the number of  $\alpha$ -GeTe nanosheets (shown in Fig. 2f) with different layers and the results were presented in Fig. S9. From this we can measure a number fraction of monolayers (number of monolayers / total number of flakes) of 25%. We can also calculate the mass fraction of monolayer  $\alpha$ -GeTe, (the mass of monolayer  $\alpha$ -GeTe / the mass of all flakes,  $M_{monolayer}/M_{total}$ ) from:

$$\frac{M_{monolayer}}{M_{total}} = \frac{\sum_{individuals} A_{monolayer}}{\sum_{allflakes} N_{monolayer} A_{monolayer}}$$

where  $A_{monolayer}$  is the area of a monolayer and  $N_{monolayer}$  represents

## the number of monolayers in a given

nanosheet. The mass fraction was calculated to be 8.7%. We defined the product yield of monolayer  $\alpha$ -GeTe as the mass of monolayer  $\alpha$ -GeTe in Sol-B divided by the starting mass of  $\alpha$ -GeTe powder, which is equivalent to the

ratio of concentration of monolayer  $\alpha$ -GeTe and starting  $\alpha$ -GeTe powder. The product yield of monolayer  $\alpha$ -GeTe in Sol-B was estimated as 0.026%.



Fig. S10 STEM image of a single  $\alpha$ -GeTe nanosheet (a), and Ge (b) and Te (c) elemental mapping of the nanosheet. (d) The EDS image of single

 $\alpha\text{-}GeTe \text{ nanosheet in (a)}.$ 



Fig. S11 XPS spectra near the Ge-3d (a) and Te-3d (b) of  $\alpha$ -GeTe nanosheets in sol-B and Ge-3d (c) and Te-3d (d) of bulk  $\alpha$ -GeTe. The Ge 3d peak at 29.7 eV and Te 3d peak at 572.5 eV are consistent with the binding energies for Ge<sup>2+</sup> and Te<sup>2+</sup>, respectively. The Ge 3d peak at 33.2 eV is consistent with the binding energy for Ge<sup>4+</sup>, ascribing to oxidation on the  $\alpha$ -GeTe surface. Because oxidation also existed in bulk  $\alpha$ -GeTe, we concluded that  $\alpha$ -GeTe nanosheets oxide when exposed to air and the exfoliation doesn't cause extra-oxidation.



Fig. S12 Raman spectra of  $\alpha$ -GeTe nanosheets in sol-A (a) and in Sol-B (b). Results indicate that peak position of both of the A<sub>1</sub>, A<sub>2</sub> and C shift to high wavenumber region with the decreasing of thickness of  $\alpha$ -GeTe nanosheets; Raman spectra of  $\alpha$ -GeTe nanosheets in sol-B after air exposure for one month (c).



Fig. S13 Electronic band structure of (a) bulk  $\alpha\text{-}GeTe$  and PDOS (b) bulk  $\alpha\text{-}GeTe.$ 



Fig. S14 Brillouin zones of bulk α-GeTe (a) monolayer α-GeTe nanosheets (b) which are hexagonals. We marked the high symmetric points.



Fig. S15 Fluorescence excitation and emission spectra of  $\alpha$ -GeTe Sol-B dispersion.

| Serial number          | 1      | 2     | 3     | 4     | 5     |       |  |
|------------------------|--------|-------|-------|-------|-------|-------|--|
| LOD (µM)               | 0.055  | 0.052 | 0.069 | 0.048 | 0.044 |       |  |
| Serial number          | 6      | 7     | 8     | 9     | 10    | 11    |  |
| LOD (µM)               | 0.058  | 0.063 | 0.054 | 0.046 | 0.055 | 0.050 |  |
| The standard deviation | 0.0071 |       |       |       |       |       |  |
| of LOD (µM)            |        |       |       |       |       |       |  |

Table S3 The detection limits of 11 repeated experiments and their the standard deviation



Fig. S16 UV-vis-NIR spectra for the investigation of the interaction between  $\alpha$ -GeTe nanosheets and Fe<sup>3+</sup>.