Supporting Information for
Observation of Superconductivity in Pure 1T’-MoS₂ Nanosheets

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Supplementary Figures

Figure S1. The structure of LiMoS$_2$ crystal observed from (a) [110] direction and (b) [001] direction. In LiMoS$_2$ structure, Li ions are intercalated orderly between the MoS$_2$ layers; the MoS$_2$ basal plane shows a 2$a$$\times$2$a$ superstructure and Mo–Mo diamond-like chains along the [110] direction. Blue, yellow and green spheres represent Mo, S and Li ions, respectively.

The structure of LiMoS$_2$ had been unknown for a long time due to the broad and overlapped XRD peaks resulted from the short structural coherence inside the material. Until 2002, the nanocrystalline structure in the disordered LiMoS$_2$ was determined by a pair distribution function (PDF) analysis. Based on the previous reports, the crystal structure of LiMoS$_2$ is triclinic, belonging to the P$\overline{1}$ space group, and the cell parameters are $a = 6.963$ Å, $b = 6.386$ Å, $c = 6.250$ Å, $\alpha = 88.60^\circ$, $\beta = 89.07^\circ$, $\gamma = 120.06^\circ$, respectively. Li ions are arranged orderly between S–Mo–S layers and play an important role in stabilizing the structure of Mo–S basal planes. Further, the S atoms adopt an ABAB stacking sequence (Figure S1a), and each Mo species is coordinated by a distorted octahedron of six S atoms. In the S–Mo–S layers, four neighboring Mo ions are aggregated together and form a diamond-like cluster. Through the single bonding of edge-site Mo ions, Mo clusters are connected into chains along [110] direction (Figure S1b).

Figure S2. (a) The XRD of the as-synthesized LiMoS$_2$ crystals, showing pure phase with high crystallinity; (b) The SEM image of the as-synthesized LiMoS$_2$ crystals, which displays obviously hexagonal shape.

In this work, LiMoS$_2$ crystals were synthesized by a simple solid-state reaction with stoichiometric MoS$_2$, Li$_2$S and Mo in an evacuated quartz tube. LiMoS$_2$ crystals were characterized by X-ray Diffraction (XRD) and scanning electron microscopy (SEM). As illustrated in Figure S2a, all the peaks in the XRD pattern fit well with the JCPDS card No. #44-1078, and the sharp profile of the peaks
indicates high crystallinity of the LiMoS\(_2\) crystal. The SEM image in Figure S2b shows a nearly perfect hexagon shape of LiMoS\(_2\) single crystals with the lateral size of about 4 \(\mu\)m.

**Figure S3.** (a) The illustration of exfoliation process by using LiMoS\(_2\) crystals as precursors; (b) A homogenous dispersed colloid of exfoliated MoS\(_2\) nanosheets with obvious Tyndall effect.

**Figure S4.** The AFM image of exfoliated MoS\(_2\) nanosheets obtained by LiMoS\(_2\) crystals, corresponding to a thickness of \(-1\) nm, which proves the single layer characteristics.
Figure S5. (a) XRD pattern of n-butyllithium-treated Li$_x$MoS$_2$ powders, the peaks labeled by ** are formed by the intercalation of H$_2$O into the interlayers. Expect the main peak at 14.0 ° and two peaks caused by the intercalation of water, other peaks are too broad to identify, ascribed to the short coherence in its structure. (b) The SEM image of the Li$_x$MoS$_2$ powders prepared by the n-butyl lithium method, showing rough surfaces and edges, which are damaged by the etching effect of the n-butyl lithium.

Figure S6. The STEM image of the MoS$_2$ nanosheets obtained by using the butyl lithium method, exhibiting mixed phases (1H, 1T and 1T’) and abundant defects.
Figure S7. The phase transition of the 1T’ filtrated films annealed at various temperatures. (a) XRD of 1T’ MoS$_2$ filtrated films annealed at various temperatures. The shift of the main peak from 14.8° (d=5.99 Å) to 14.4° (d=6.16 Å) indicates the phase transition from 1T’ to 2H; (b) Raman spectra of the restacked 1T’-MoS$_2$ nanosheets annealed at various temperatures. The Raman characteristic peak of 2H-MoS$_2$ E$_{1g}$ around 383 cm$^{-1}$ strengthens gradually, while J$_1$, J$_2$, J$_3$ modes corresponding to 1T’ phase weakens slowly. (c) XPS spectra for the restacked 1T’-MoS$_2$ nanosheets with different annealing temperatures. The Mo 3d peaks were deconvoluted to show the 2H and 1T/1T’ contributions, represented by blue and red plots, respectively. The relative content of 2H phase increases, while that of 1T/1T’ phase decreases with increasing annealing temperature.

Figure S8. Temperature dependence of magnetic susceptibility of 1T’-MoS$_2$ powders under the
magnetic field of 5 Oe, with zero-field-cooling curve (black) and field-cooling curve (white)

Figure S9. Magnetic and electrical measurements of MoS$_2$ nanosheets obtained from n-butyl lithium method. a. Magnetic measurements; b. Electrical measurements.