Supporting Information

Section 2.1 Materials

The selection of ZnS material was compared and analyzed from two aspects of morphology and roughness. In terms of the morphology, ZnS powder has fluidity, which makes it difficult to observe the changes in its macroscopic morphological characteristics. However, ZnS wafer has a fixed shape, which is convenient for the subsequent morphological observations and product characterization.

The roughness of material surface has a certain influence on the laser irradiation experiment. If the roughness is small, it indicates that the surface distribution of sample was relatively uniform and smooth, which is convenient for subsequent research on the changes in the surface morphology and chemical composition of ZnS. If the roughness is too large, the laser-induced damage threshold experiment will be affected, and it is difficult to observe the influence of the laser on the surface morphology of ZnS.

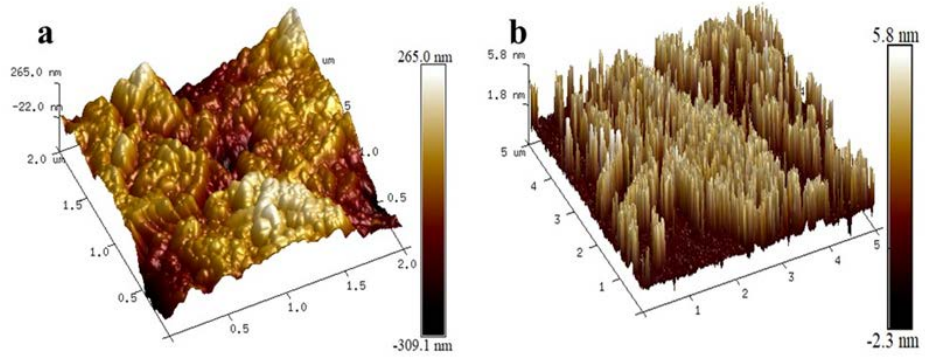

Fig. S1. Atomic force microscopy images of: (a) ZnS powder and (b) ZnS wafer.

Fig. S1 shows that the root mean square roughness (R_q) value of ZnS powder was 79.2 nm and average surface roughness values (R_a) was 62.2 nm, indicating that the surface was not smooth and will have an impact on the laser damage threshold experiment, while the R_q and R_a of ZnS wafer were 1.53 and 1.16 nm, respectively with a relatively small surface roughness. The results show that the ZnS wafer was uniform, densely packed, and smooth, which is more suitable for the laser decomposition of ZnS.

Section 2.4.2. Laser-induced damage threshold experiment

Table S1. Laser parameters used for the laser-induced damage of ZnS.

NO.	F J/cm^2	f/kHz	Scanning speed mm/s	Scanning interval
1	0.18	100	300	0.01
2	0.19	90	300	0.01
3	0.22	80	300	0.01
4	0.27	70	300	0.01
5	0.33	60	300	0.01
6	0.37	100	300	0.01
7	0.43	50	300	0.01
8	0.49	90	300	0.01
9	0.6	80	300	0.01
10	0.8	70	300	0.01

Section 2.4.3. Laser-induced decomposition of ZnS

Table S2. Laser parameters used for the laser-induced damage of ZnS.

NO.	F J/cm^2	f/kHz	Scanning speed mm/s	Scanning interval
1	2.79	20	300	0.01
2	5.57	20	300	0.01
3	7.76	20	300	0.01
4	4.11	60	300	0.01
5	8.33	60	300	0.01
6	15.52	60	300	0.01

Section 3.1. The optimal absorption wavelength of ZnS

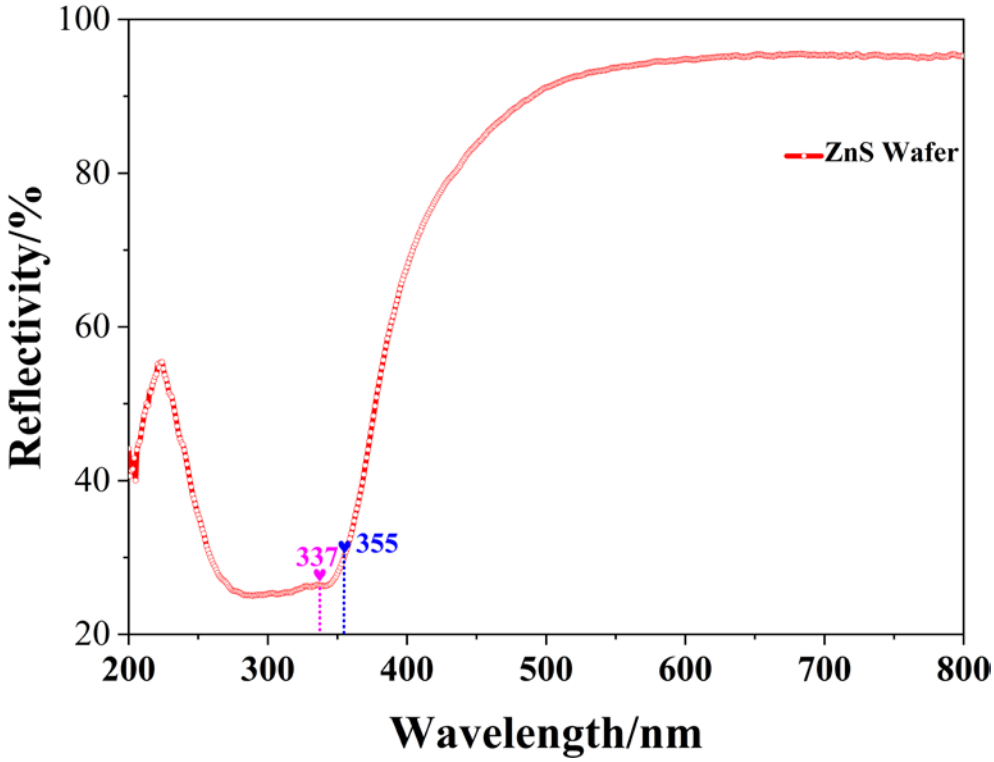

Fig. S2. The UV visible diffuse reflectance spectrum obtained for ZnS.

Section 3.2 The laser-induced damage of ZnS.

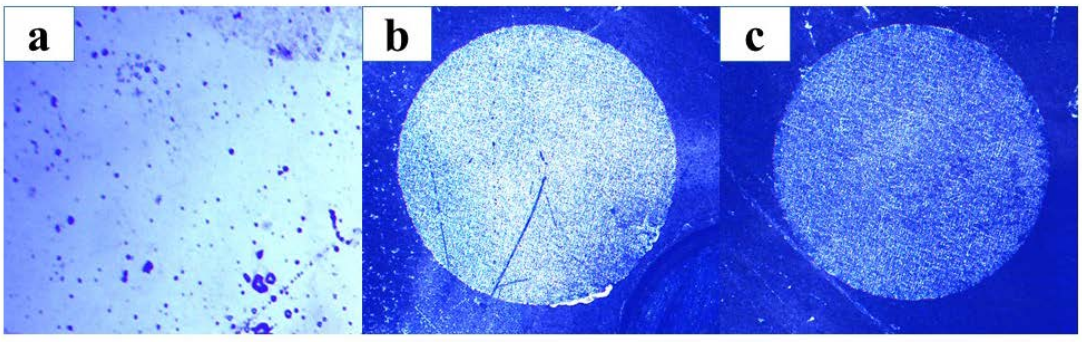

Fig. S3. The micro-morphology of the laser-induced damage of ZnS at different laser fluence: (a) 0.33, (b) 0.37, and (c) 0.43 J/cm^2.

Section 3.3.2. XPS analysis and generation mechanism for the products

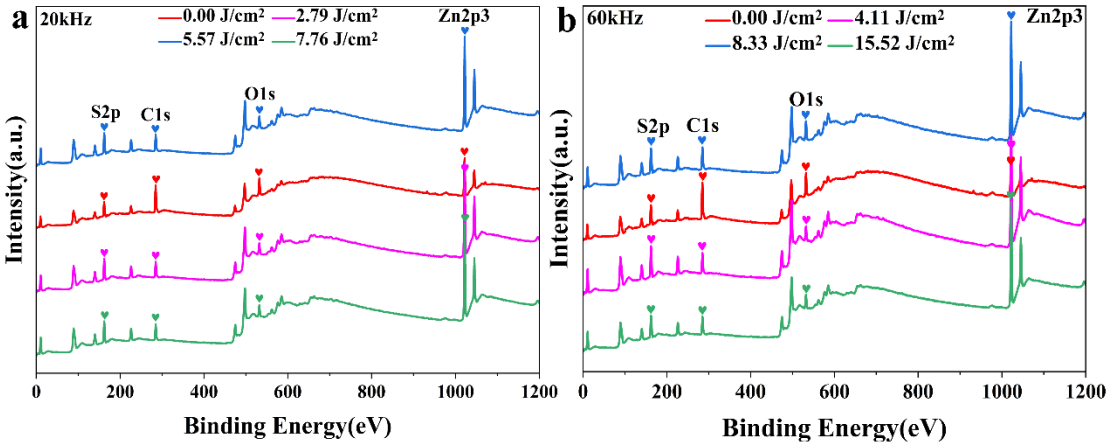

Fig. S4. XPS survey spectra obtained for ZnS at a (a) pulse repetition frequency of 20 kHz and laser fluence of 0.00, 2.79, 5.57, and 7.76 J/cm^2 and (b) pulse repetition frequency of 60 kHz and laser fluence of 0.00, 4.11, 8.33, and 15.52 J/cm^2.

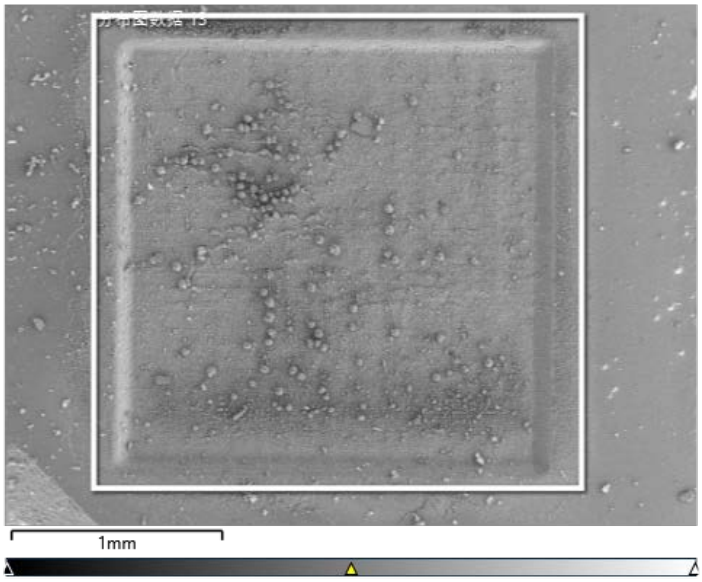

Fig. S5. The surface micro-morphology of ZnS after laser irradiation at a laser fluence of 15.52 J/cm^2.

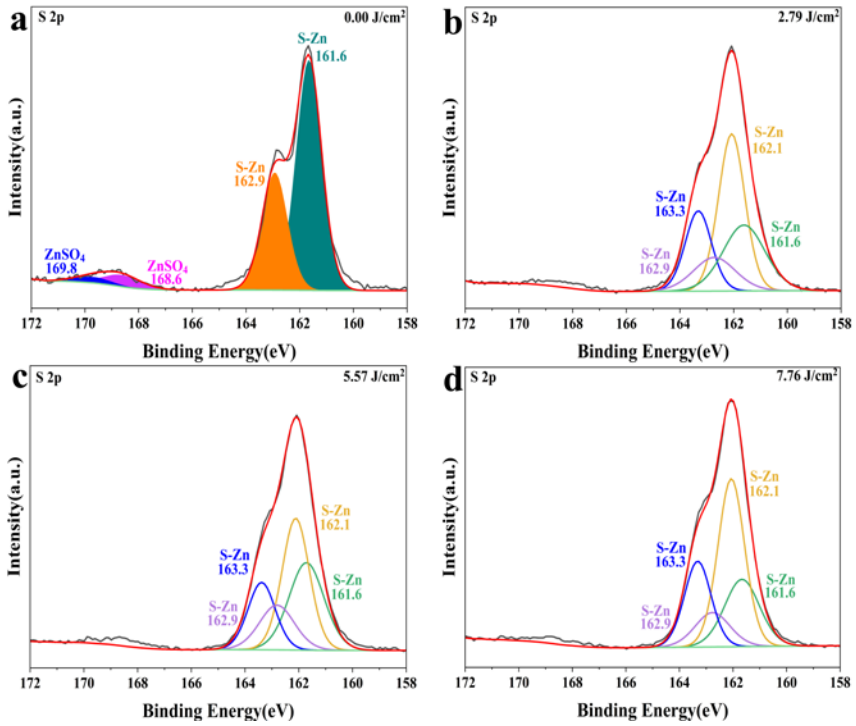

Fig. S6. XPS spectra obtained for S at a pulse repetition frequency of 20 kHz with different laser fluence: (a) S 2p, 0.00 J/cm^2; (b) S 2p, 2.79 J/cm^2; (c) S 2p, 5.57 J/cm^2; (d) S 2p, 7.76 J/cm^2.

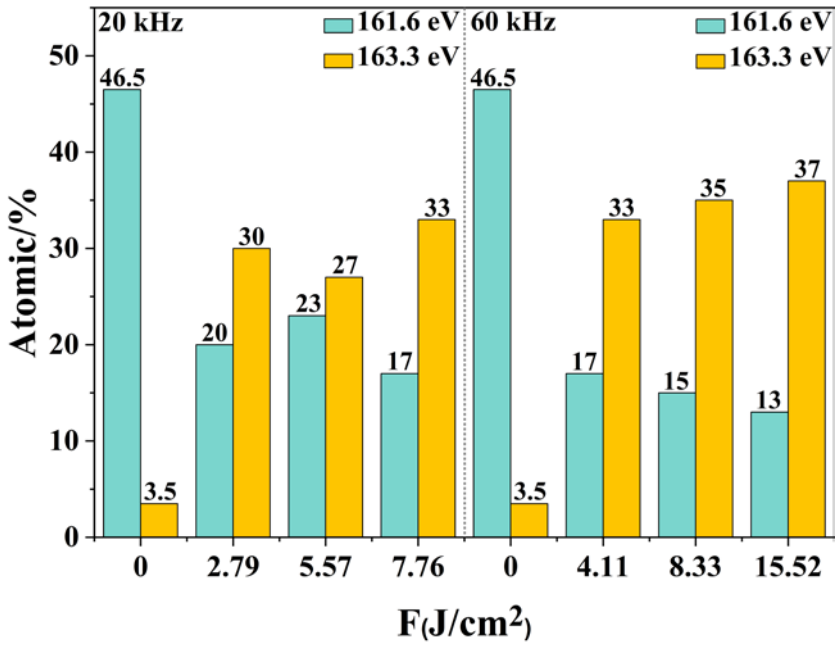

Fig.S7. Diagram of the atomic ratios of the spin-orbit splitting of the S 2p region