Synthesis and Characterization of MoSe2 Nanoscrolls via Pulsed Laser Ablation in Deep Eutectic Solvents. Supplementary Information.

Figure S1. Size distribution of MoSe₂

Size distribution of MoSe₂ shows preference for larger sizes in material ablated in deep eutectic solvent, and also show decreasing size with secondary irradiation. The PLAL samples passes from mono to bimodal distribution. In contrast PLADES synthesized sample passes from bimodal to mono disperse distribution.

Figure S2. ¹HNMR of MoSe₂ produced in reline

Figure S2. ¹HNMR results of the serial washing steps suggesting no change in solvent composition by laser irradiation.
The experimental chemical shift of reline was measured before near infrared (NIR) irradiation figure S-2.a. No change in resonance frequency of reline was identified. The chemical shift of Reline after irradiation of MoSe\textsubscript{2} target (figure S-2.c), and after 1\textsuperscript{st}, 2\textsuperscript{nd} (figure S-2.d) and 3\textsuperscript{rd} (figure S-2.f) washing steps, were compared to the untreated neat reline (figure S-2.b). Chemical shift of Reline before and after irradiation conserve the same values and relative abundance, suggesting that there is no formation of secondary species due to solvent degradation by the interaction of 1064 nm pulsed laser.

Table TS1. $^1$HNMR of MoSe\textsubscript{2} produced in reline

<table>
<thead>
<tr>
<th>Label</th>
<th>Treatment</th>
<th>Washed</th>
<th>H\textsubscript{A}</th>
<th>H\textsubscript{B}</th>
<th>H\textsubscript{C}</th>
<th>H\textsubscript{U}</th>
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<tr>
<td>$\delta_0$</td>
<td>Neat reline, no irradiation</td>
<td>No</td>
<td>3.22</td>
<td>3.53</td>
<td>4.07</td>
<td>5.85</td>
</tr>
<tr>
<td>$\delta_1$</td>
<td>Reline + MoSe\textsubscript{2} NIR laser</td>
<td>No</td>
<td>3.21</td>
<td>3.53</td>
<td>4.07</td>
<td>5.85</td>
</tr>
<tr>
<td>$\delta_2$</td>
<td>Reline + MoSe\textsubscript{2} NIR laser</td>
<td>1X</td>
<td>3.22</td>
<td>3.53</td>
<td>4.07</td>
<td>5.85</td>
</tr>
<tr>
<td>$\delta_3$</td>
<td>Reline + MoSe\textsubscript{2} NIR laser</td>
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<td>3.43</td>
<td>3.98</td>
<td>5.68</td>
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<tr>
<td>$\delta_4$</td>
<td>Reline + MoSe\textsubscript{2} NIR laser</td>
<td>3X</td>
<td>--</td>
<td>--</td>
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</tr>
</tbody>
</table>

Table TS1. Chemical shift of deep eutectic solvent before and after ablation, and after removing reline

The chemical shift remains in the same range as neat reline. Shift upfield after second washing step suggest formation of hydrogen boning with water of the remaining DES molecules, thus increasing the proton shielding.

Figure S3. X-ray photoelectron spectrum after edging

![X-ray photoelectron spectrum](image)

Figure S3. X-ray photoelectron spectra of MoSe\textsubscript{2}, after edging
After 20 seconds of edging, within the Mo3d region, PLAL synthesized MoSe$_2$ structures show presence of Mo$^{6+}$ (metallic Mo) at 227.1 eV in large quantities and small amount of MoSe$_2$ peaking at 228.7 eV (figure S-3.a). In contrast PLADES sample has large peaks of MoSe$_2$ at 228.8eV in comparison to the metallic Mo$^{6+}$ at 227.7 eV (figure S-2.b). Se3d region of PLAL treatment shows a 5/2 spin orbit split at 53.81 eV (figure S-2.c), while PLADeS same feature peaks at 54.65 eV (figure S-2.d). These results suggest lower oxidation during synthesis in deep eutectic solvent, beyond the exposed surface.

**Figure S4. MoSe$_2$ monolayer Se-Se distances**

![Figure S4. MoSe$_2$ monolayer with a interplanar Se-Se distance of 3.3 Å](image)

Representation of the rhombohedral MoSe$_2$ monolayer obtained by pulsed laser ablation in deep eutectic solvent corresponding to TEM and DP analysis, displaying distances between adjacent selenium atoms in the same layer (created using Vesta and ChemDraw 3D).

**Figure S5. Measured Se-Se distances of MoSe$_2$ layer synthesized by PLADES**

![Figure S5. MoSe2 nanosheets with inter atomic distances of Se-Se 3.3 Å, analyzed by FFT and line profile.](image)

HR-TEM micrographs of MoSe$_2$ nanosheets were analysed with line profile at atomic resolution, where recorded distances in between Se-Se atoms on the surface layer is 3.3 in all three directions corresponding to the 1T crystal structure.