1	Supplemental Materials
2	<b>For</b>
3 1	Pressure tuned photoluminscence and dand gap in two-dimensional layered g-C <sub>3</sub> N <sub>4</sub> : the
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36 Fig. S1 (a) Typical XRD pattern of FL-CN; (b) TGA curve of FL-CN measured under N<sub>2</sub>
37 atmosphere.

The (100) diffraction peak in the Fig. S1a corresponds to the periodic packing of heptazines in the conjugated planes, while the (200) peak corresponds to the interlayer stacking of aromatic CN units.<sup>1,2</sup> The TGA curve (Fig. S1b) suggests a high purity of the g- $C_3N_4$ .<sup>3</sup>



## 41

42 Fig. S2 The optical images of the recovered sample after compression and grinding.

	The peak position (nm)			The peak intensity		
	P1	P2	P3	P1	P2	Р3
FL-CN	432	452	491	6313	8472	4066
6MPa-CN	434	454	491	10969	16224	8941

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44 Table S1. The peak position and intensity of FL-CN and 6MPa-CN.



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46 Fig. S3 SEM image of the ground sample.

47 Our SEM observation on the released sample after uniaxial compression suggests a planarization of g-C<sub>3</sub>N<sub>4</sub> layers, from which we can see that no obvious wavy layers but 48 49 instead flatten layered structures can be observed in some areas after compression. For comparison, the sample after grinding shows that C<sub>3</sub>N<sub>4</sub> mainly forms particle-like aggregation 50 which is similar to the bulk g-C<sub>3</sub>N<sub>4</sub>, but with decreased size. From the recorded XRD patterns 51 in Fig. 3e (main text), we can see that the intensity of the (002) diffraction peak from 52 intralayer of g-C<sub>3</sub>N<sub>4</sub> increases obviously both after uniaxial compression and grinding, 53 54 compared with the pristine sample, suggesting that the periodic arrangement of g-C<sub>3</sub>N<sub>4</sub> layers may become better. 55



57 Fig. S4 PL spectra of the sample (a) at selected pressures upon compression and (b) upon58 decompression to ambient pressure.





61 AA-and AB stackings, respectively.

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