

# Supporting Information

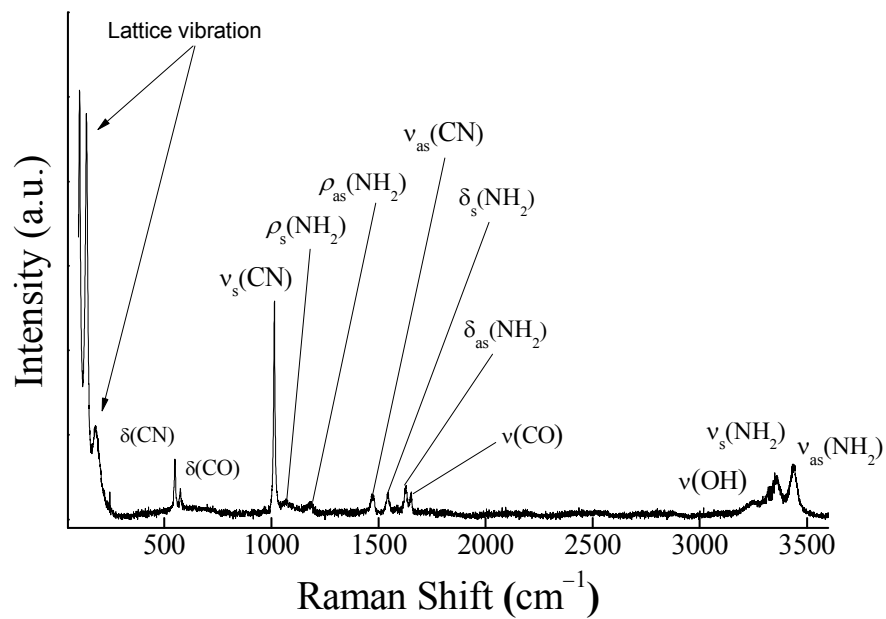
## Molecular Paradigm Dependant Nucleation in Urea

### Aqueous Solution

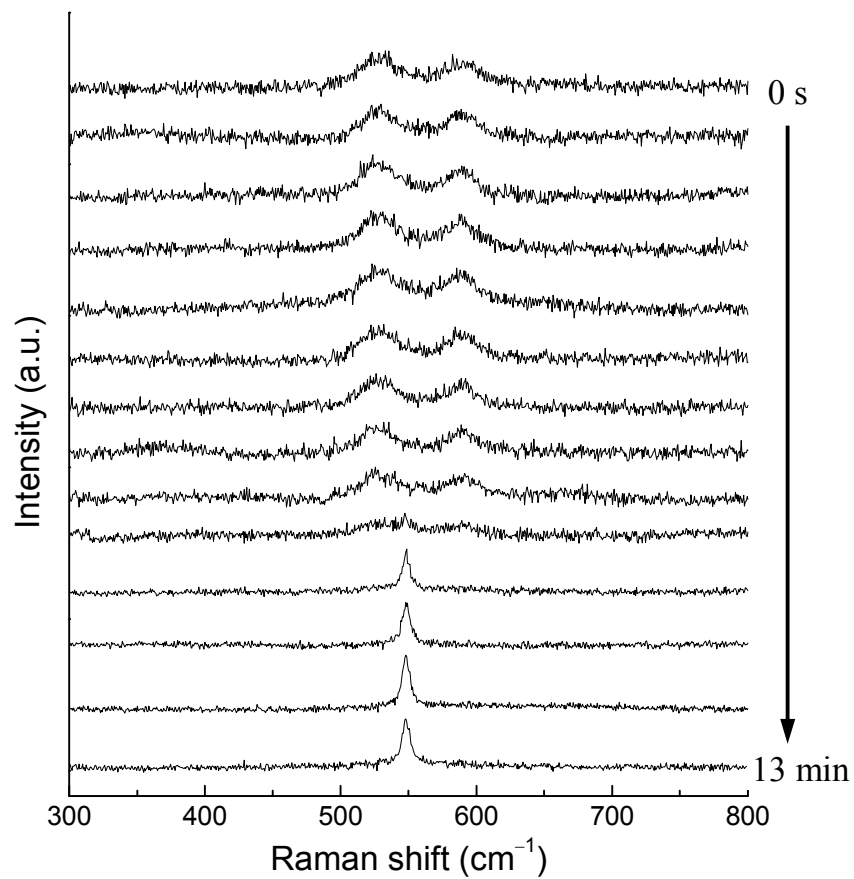
Xiaoyan Chen,<sup>†,‡</sup> Congting Sun,<sup>‡</sup> Sixin Wu,<sup>\*,†</sup> Yingning Yu,<sup>‡</sup> and Dongfeng Xue<sup>\*,‡</sup>

<sup>†</sup>The Key Laboratory for Special Functional Materials of MOE, Henan University, Kaifeng, Henan 475004, China

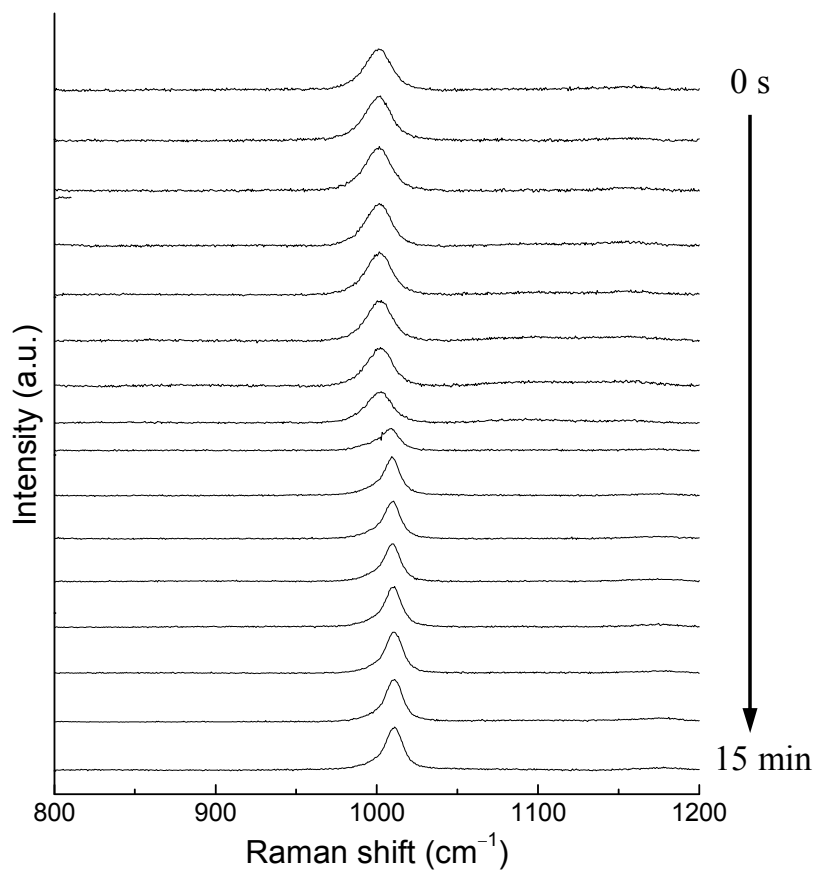
<sup>‡</sup>State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022, China



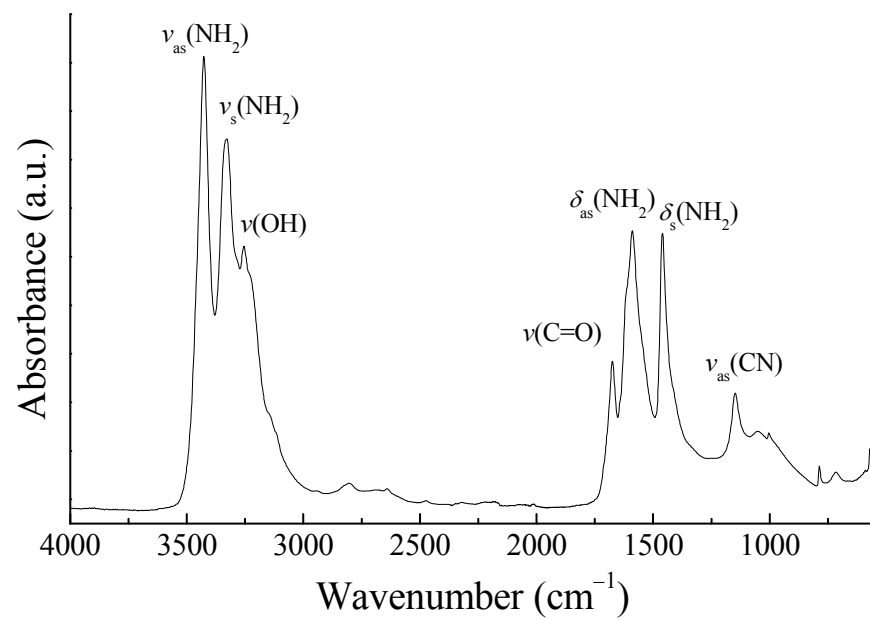
**Figure S1 Raman spectra of urea powder in the region of 3600–50 cm<sup>-1</sup>.**



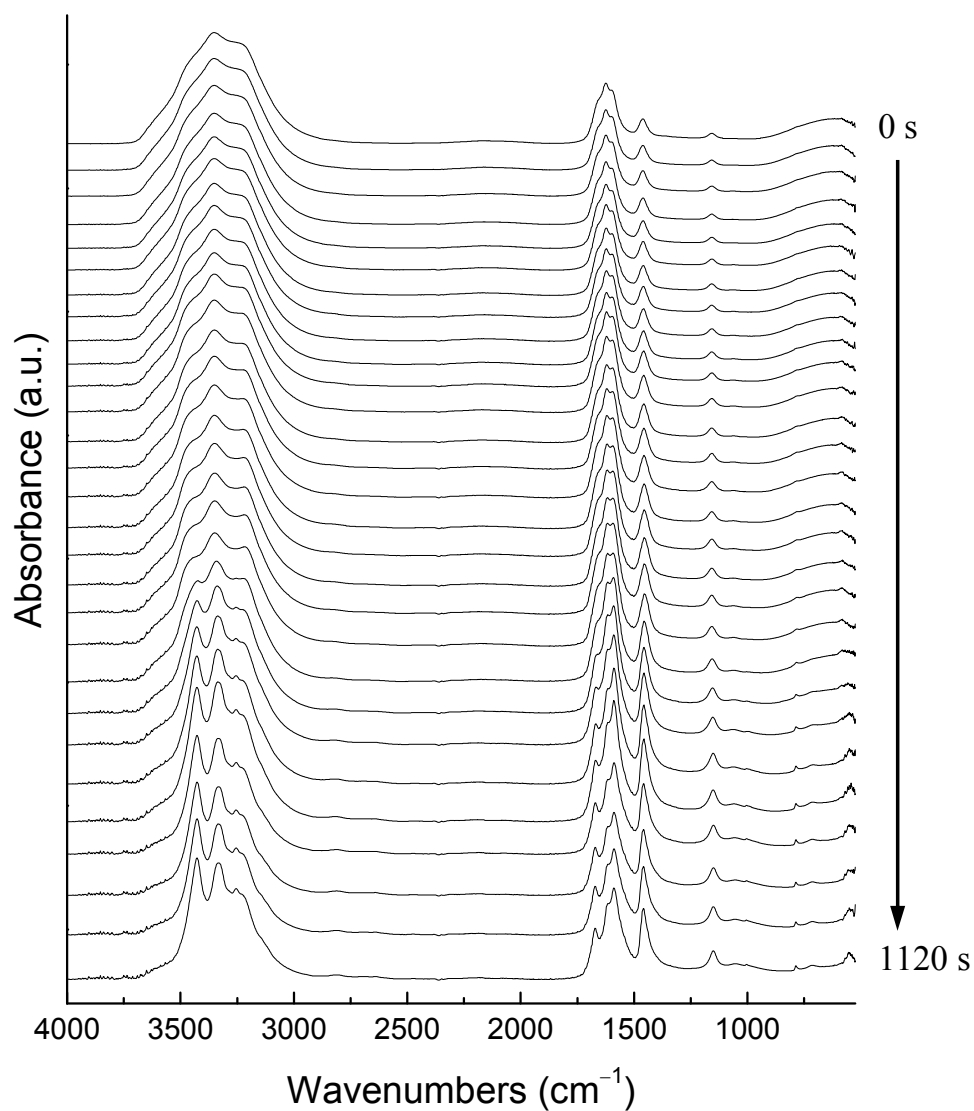
**Figure S2** Time-dependent Raman spectra of urea aqueous solution in the region of 300–800 cm<sup>-1</sup>. The successive Raman measurements were carried out Mapping mode per 1 minute.



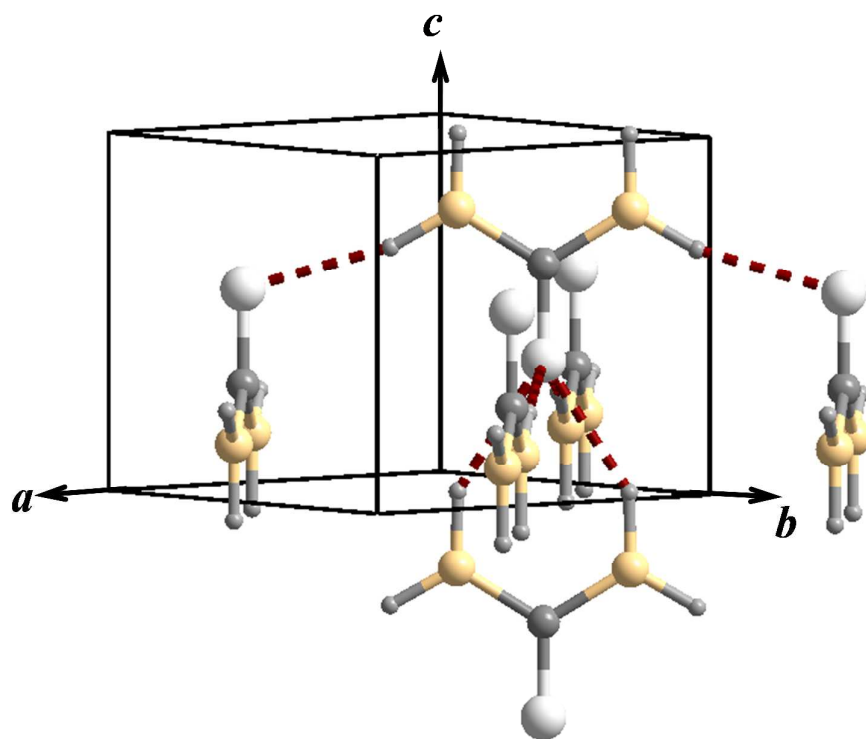
**Figure S3 Time-dependent Raman spectra of urea aqueous solution in the region of 800–1200 cm<sup>-1</sup>.** The successive Raman measurements were carried out Mapping mode per 1 minute.



**Figure S4** IR spectra of urea powder in the region of 4000–550 cm<sup>-1</sup>.



**Figure S5** In situ ATR-IR observation of nucleation process of urea aqueous solution in the region of 4000–550 cm<sup>-1</sup>. Successive ATR-IR spectra can be obtained with increasing the measurement time until the urea aqueous solution transforms into crystalline solid completely. The collection time per ATR-IR spectrum is 40 s.



**Figure S6** Unit cell of crystalline urea. There are two types of  $\text{C}=\text{O}\cdots\text{H}-\text{N}-\text{C}$  hydrogen bonding between urea molecules, one links urea molecules along coaxial direction, and the other one links urea molecules in the same crystallographic  $a-b$  plane.