SUPPLEMENTAL MATERIALS

Quantitative Analysis of the Local Phase Transitions Induced by Laser Heating

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Detailed description of Principal component analysis (PCA)

To allow PCA on the Raman 3-dimensional spectroscopic dataset consisting of *NxN* spatial and *M* spectral points is transformed into set of N^2 vectors A_i, each has a length *M*.

Further PCA represents each vector as a superposition of the orthogonal, linearly uncorrelated eigenvectors v_k such that

$$A_i = l_{ik} v_k , \tag{1}$$

Where l_{ik} are expansion coefficients, or component loadings, the eigenvectors v_{k} and the corresponding eigenvalues λ_{k} can be found from the singular value decomposition of the covariance matrix, $\mathbf{C} = \mathbf{A}\mathbf{A}^T$, where $\mathbf{A}$ is the matrix of all experimental data points $\mathbf{A}_{ij}$ i.e. the columns of $\mathbf{A}$ correspond to spectral vectors ($j = 1,..,M$), and rows correspond to spatial points, $i = 1,..,N^2$. The eigenvectors v_{k} (Fig. 5a) are orthogonal and are arranged such that the corresponding eigenvalues are placed in a descending order by variance. This means that first eigenvector v_1 contains the most important spectral information; the second v_2 contains the most important response after the subtraction of variance from the first one, and so on. In this manner the most relevant information about Raman spectra can be characterized by a few first eigenvectors v_k while remaining eigenvectors are dominated by noise.

Results of the PCA of the Raman data on $Cu_{0.74}In_{0.26}P_2S_6$ sample are presented on figure S2. Significant contribution into the Raman spectra can be seen in 2-3 components only.

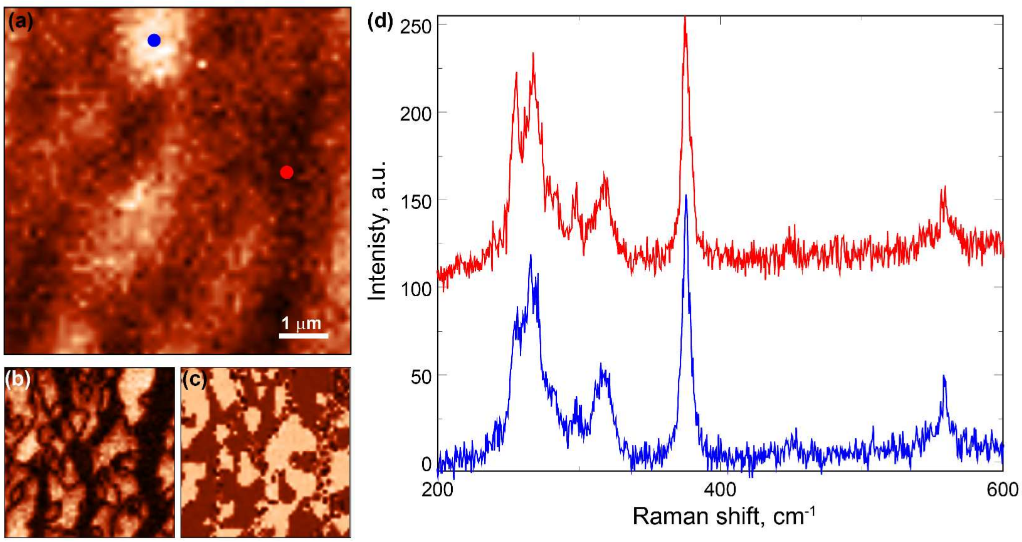

Figure S1 Simultaneous PFM and confocal Raman spectroscopy mapping in $Cu_{0.77}In_{1.12}P_2S_6$ sample. Maps of integral spectral intensity (a), amplitude (b) and phase (c) of piezoresponse. (d) Raman spectra in two points labeled on (a).

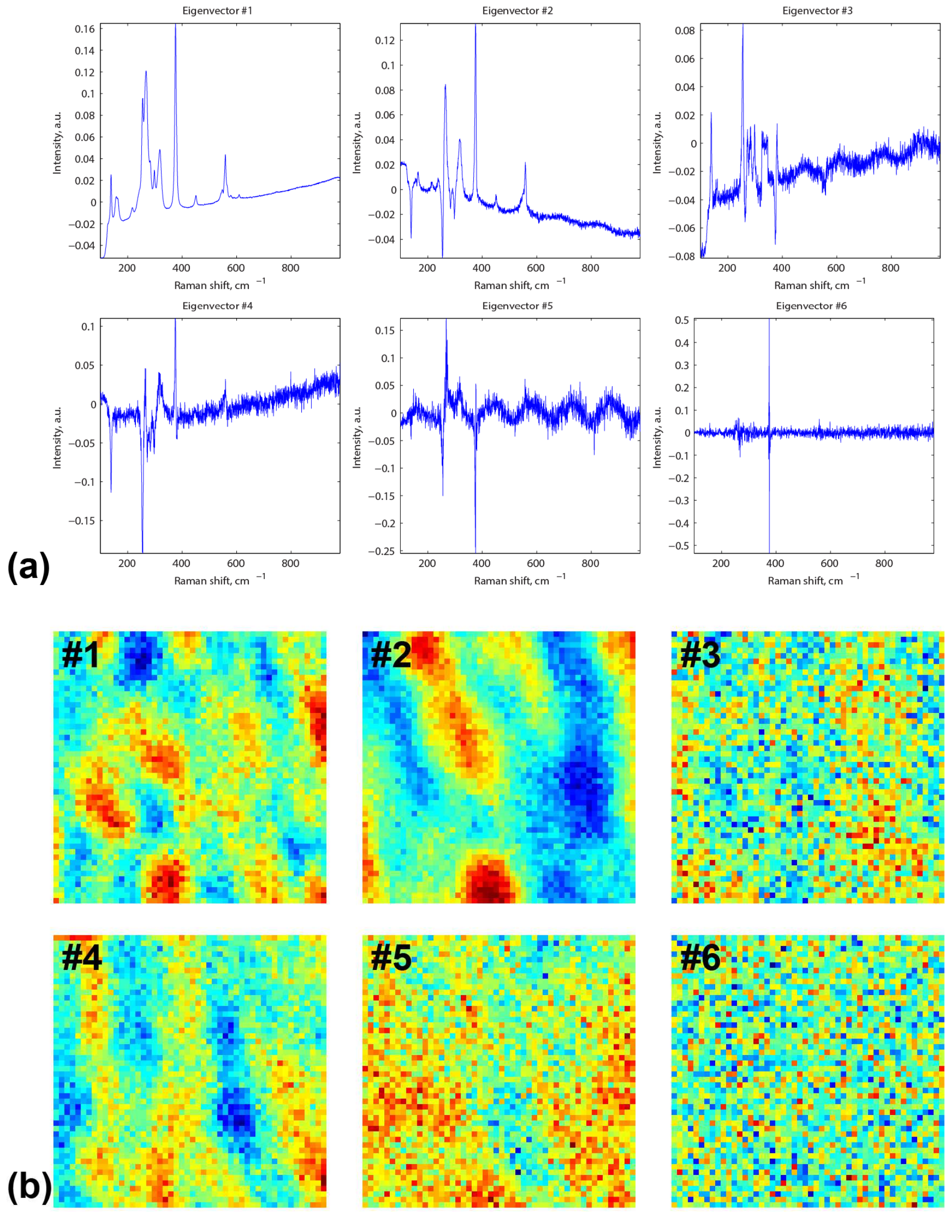

Figure S2 Principal component analysis on a Raman spectroscopy dataset measured on $Cu_{0.77}In_{1.12}P_2S_6$ sample. (a) First 6 eigenvectors and (b) corresponding loading maps.

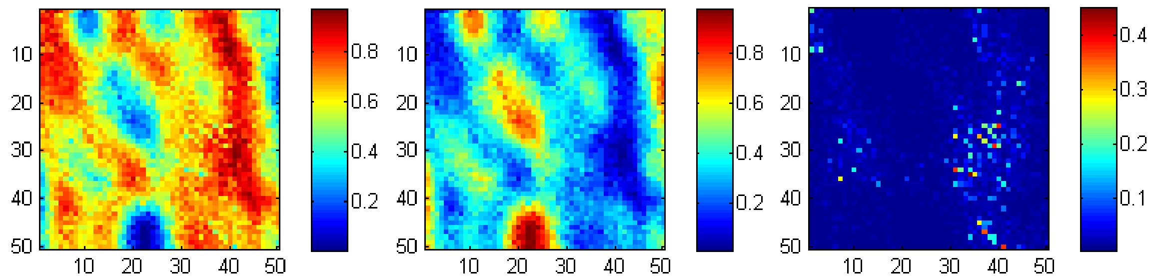

Figure S3 Abundance maps Bayesian linear unmixing of Raman spectroscopy dataset measured on $Cu_{0.77}In_{1.12}P_2S_6$ sample on 3 components.

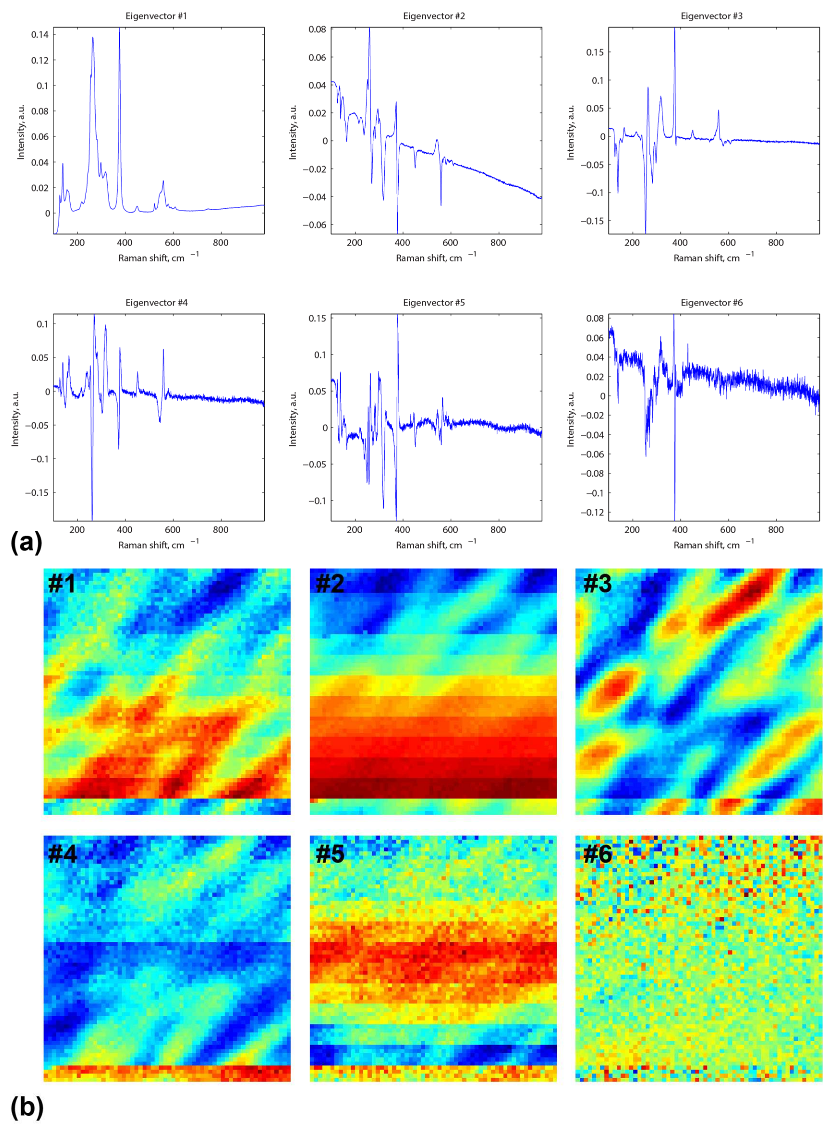

Figure S4 Principal component analysis on the normalized Raman spectroscopy dataset measured on $Cu_{0.77}In_{1.12}P_2S_6$ sample with different power of the excitation laser. (a) First 6 eigenvectors and (b) corresponding loading maps.

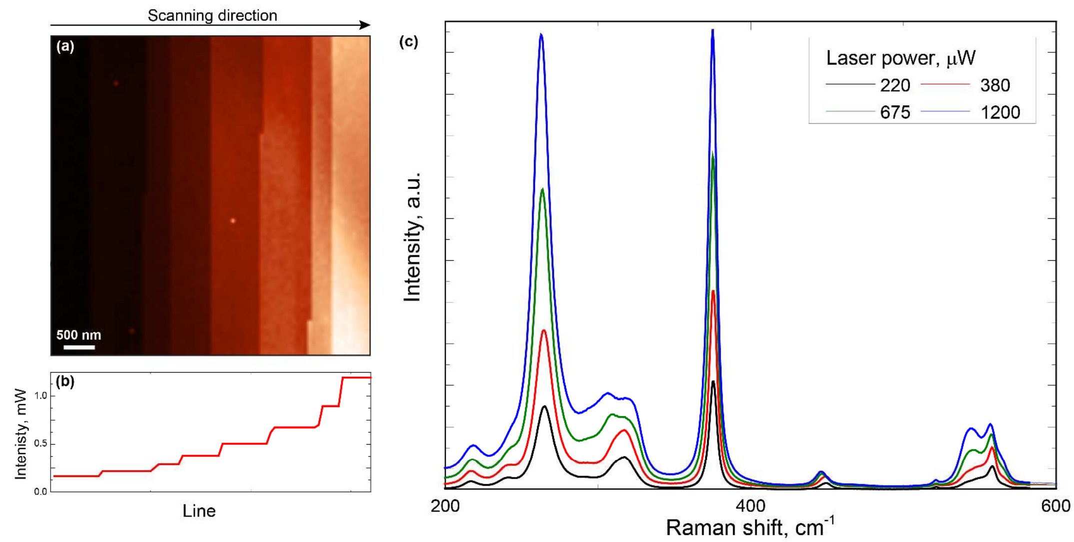

Figure S5 Raman spectra mapping with different intensities of the pumping laser in $Cu_{0.99}In_{0.01}P_2S_6$. (a) Map of the integral Raman intensity. (b) Plot of the laser power. (c) Raman spectra averaged over regions with fixed irradiation power.

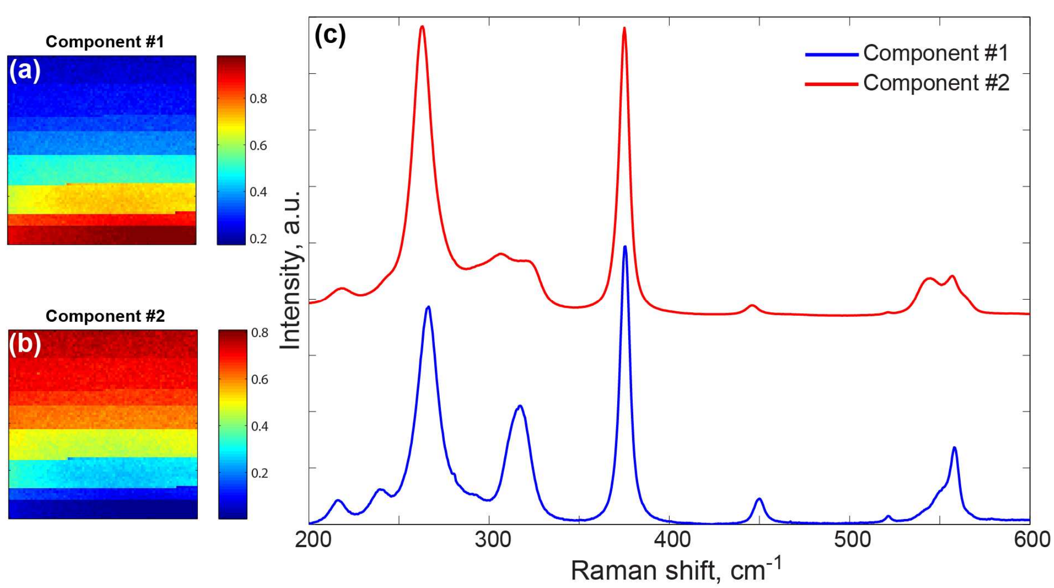

Figure S6 BLU of normalized Raman spectroscopic data of $Cu_{0.99}In_{0.01}P_2S_6$ (mapping with different values excitation laser power).

Abundance maps (a-c) and corresponding endmember spectra (d).