Supplementary Information

Density-wave-like gap evolution in $La_3Ni_2O_7$ under high pressure revealed by ultrafast optical spectroscopy

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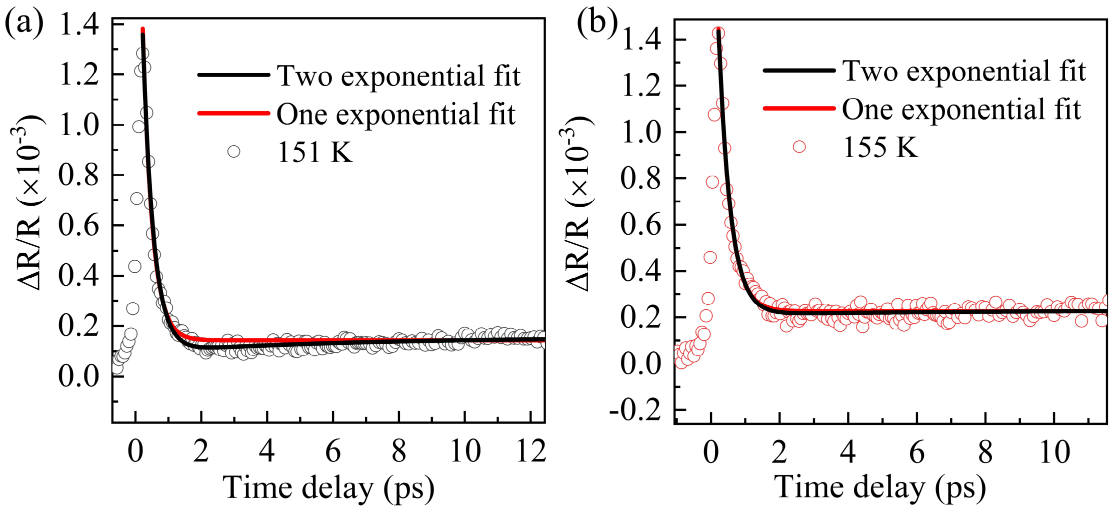
Including

1. Fitting curves for 151 K and 155 K for $La_3Ni_2O_7$ at ambient pressure.
2. Illustration of the inhomogeneity of the sample. Including pump probe data of the sample in the article at different positions, and Raman spectra at different locations together with the picture of the loaded sample
3. Extracted A_s and τ_s at different pressure and fitted by RT-model.

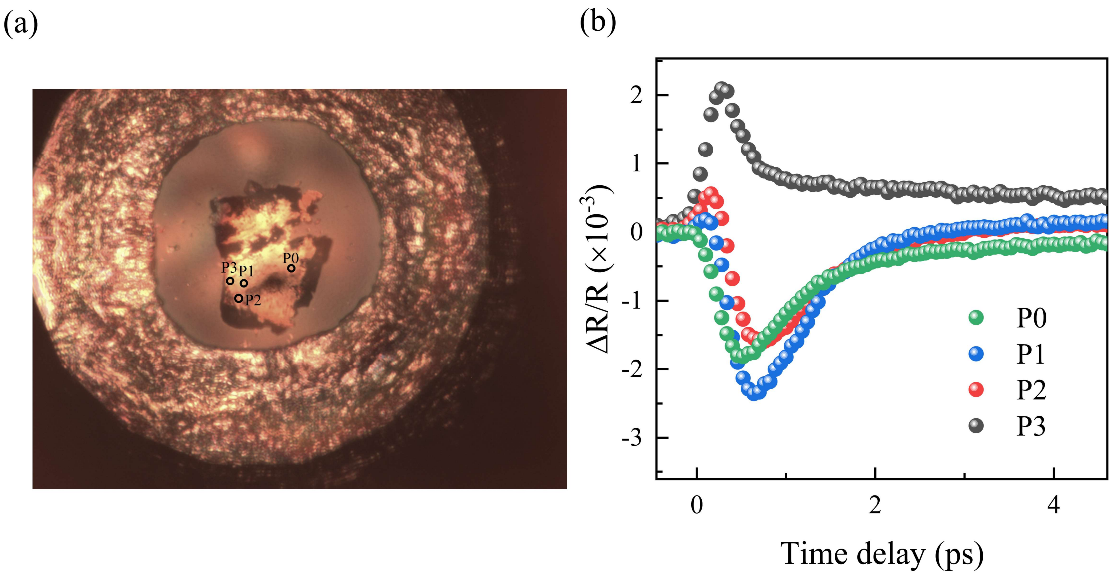
1. Fitting curves for 151 K and 155 K for $La_3Ni_2O_7$ near ambient pressure.



Supplementary Fig. 1 Exponential fittings for pump probe data near ambient pressure. (a) At 151 K, there is a difference that two exponential fits batter than one exponential fit. (b) At 155 K, two fitting curves nearly overlap with each other.

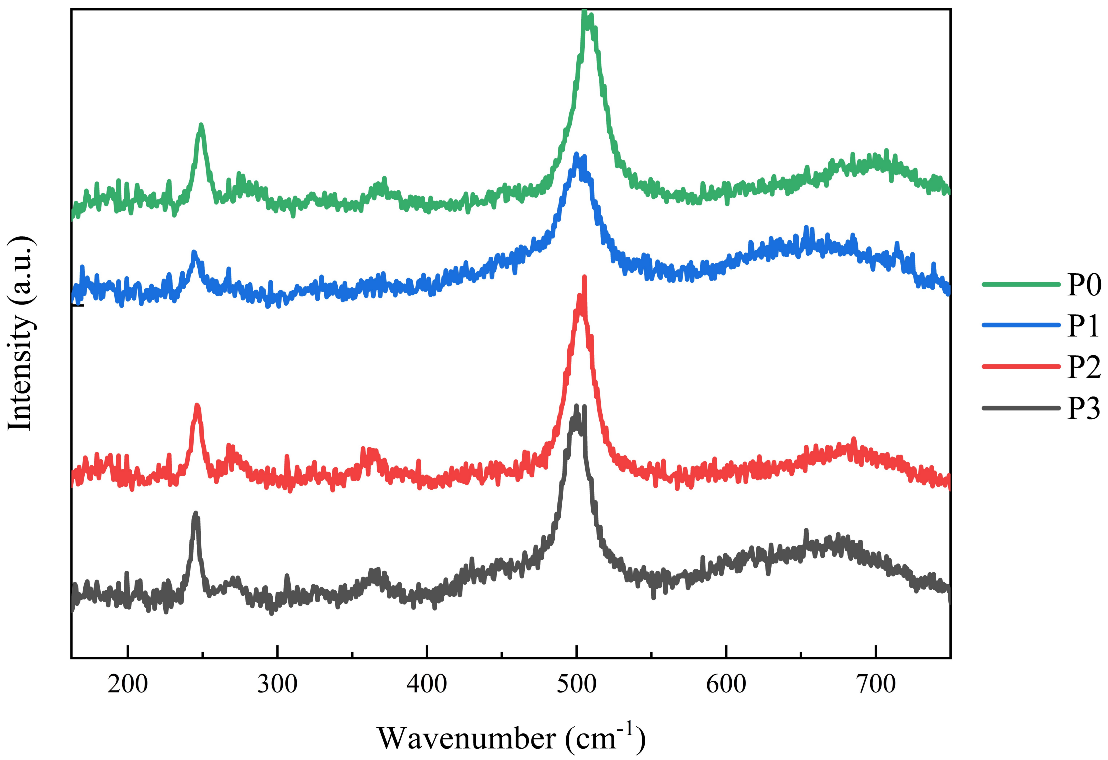
We fit the pump probe data at 155 K and 151 K with one exponential decay and two exponential decay respectively using the expression $\Delta R/R = Ae^{-t/\tau} + C$ and $\Delta R/R = A_f e^{-t/\tau_f} - A_s e^{-t/\tau_s} + C$. The $\Delta R/R$ signal at 151 K near ambient pressure can be well fitted using two exponential decay component with $\tau_f = 0.32$ ps and $\tau_s = 5.91$ ps. Using one exponential decay alone $\tau = 0.29$ ps can also fit the experimental data but miss the feature of the small dip feature around 2 ps as shown in Supplementary Fig. 1(a). The data at 155 K can be well fitted by both methods with $\tau_f = 0.34$ ps, $\tau_s = 7.04$ ps and $\tau = 0.33$ ps, as depicted in Supplementary Fig. 1(b). These two fitting curves nearly overlap with each other. Therefore we fit the experimental data above 155 K using one exponential decay component.

2. Inhomogeneous nature of $La_3Ni_2O_7$ crystal.



Supplementary Fig. 2 Illustration of inhomogeneous nature of the sample. (a) Photograph of the loaded sample and Ruby. (b)Pump probe spectra obtained from different positions as indicated in (a) at 20 K near ambient pressure.

Supplementary Fig. 2(a) shows the photograph of the loaded crystal and ruby. The circles marked as P0, P1, P2, P3 are the positions where the transient reflectivity signals were collected. The pump probe spectra collected from different positions on the same sample at 20 K near ambient pressure are shown in Supplementary Fig. 2(b). P0 is the position in the main article. It is seen that the spectra collected from different positions can be distinguished into two groups, For P0 to P2, the spectra have similar waveforms with different amplitudes and decay times. The data in the main text was collected at position P0. In ref. [1, 2], different structures and oxygen doping levels are observed, which explains the inhomogeneous behavior of the sample. For P3, only positive relaxation processes exist, which is similar to the results reported in ref. [3].



Supplementary Fig. 3 Raman spectra of $La_3Ni_2O_7$ at 34.2 GPa at room temperature.

Raman spectroscopy was employed to investigate the inhomogeneous nature of the $La_3Ni_2O_7$ sample as shown in Fig. S3. Raman spectra were collected on a Monovista Raman Spectroscopy system. A 532 nm laser with 2 μm spot size was employed. Four sharp Raman peaks around 240 cm^{-1}, 260 cm^{-1}, 360 cm^{-1} and 500 cm^{-1}, together with a broad band located between 600 and 700 cm^{-1} with two wide peaks are observed. It is obvious that the shapes vary at different positions within the sample:

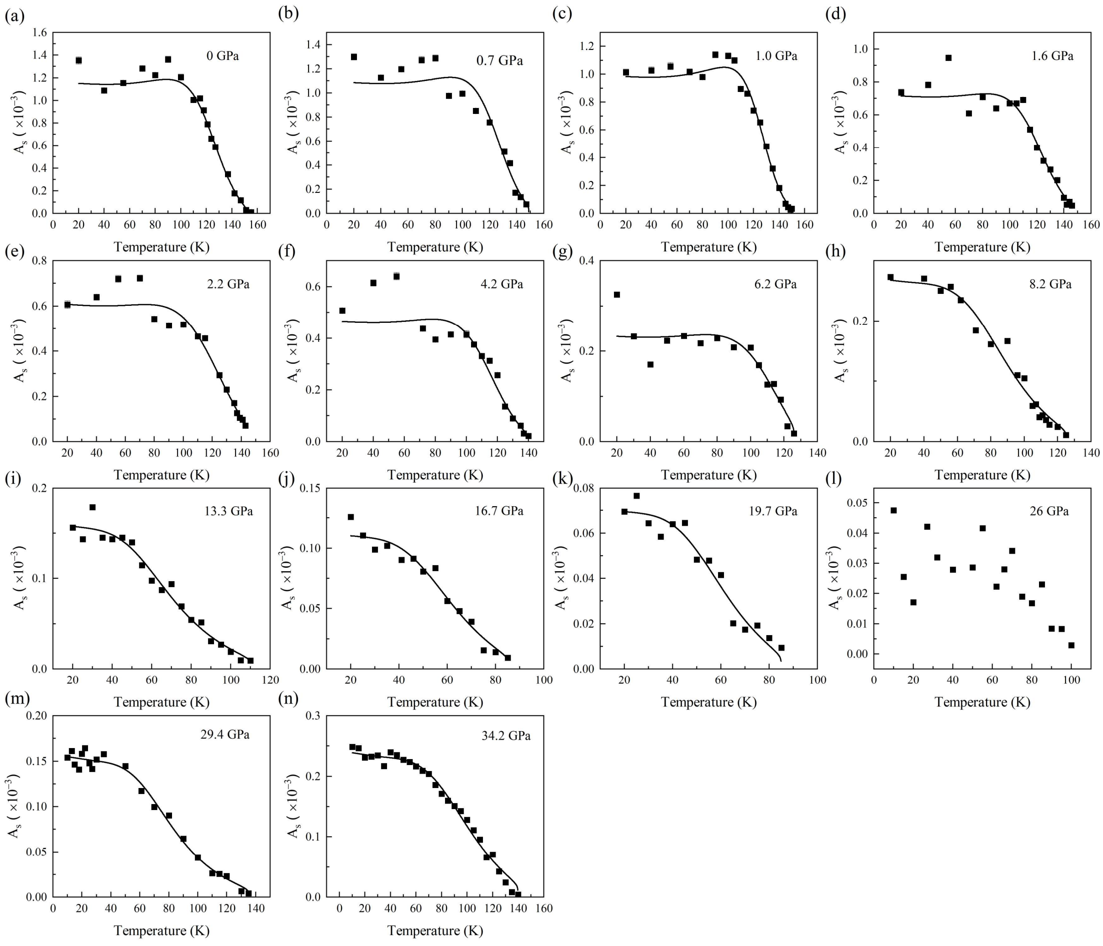
1. The broad band between 600 and 700 cm^{-1} varies in different samples. For P0, P2, and P3, the peak near 700 cm^{-1} is the dominant feature, whereas, for P1,

the two peaks are so broad that they form a large enveloping band covering the entire 600-700 cm^{-1} region.

2. The Full Width at Half Maximum (FWHM) near 500 cm^{-1} and the relative heights of these peaks compared to the broad band near 600-700 cm^{-1} differ among the samples.
3. For P1 and P3, a "stage" or plateau is observed between 400 and 500 cm^{-1}, while for P0 and P2, this feature is less distinct.
4. The relative heights of the peaks around 240 cm^{-1} and 260 cm^{-1} vary, as well as the separation between these peaks.
5. The peak around 360 cm^{-1} is insignificant for P2.

In cuprates and iron pnictides, different doping levels have been shown to lead to variations in Raman peak heights, FWHM, relative peak distances and peak positions [4, 5]. Combining the STEM results in ref. [1, 2], the observed variation in the $La_3Ni_2O_7$ sample suggests the presence of different doping levels at various positions.

3. Extracted A_s and τ_s at different pressure and fitted by RT-model.



Supplementary Fig. 4 Fitting results of A_s using RT model. The solid lines are the fitting results using Eq (S1).

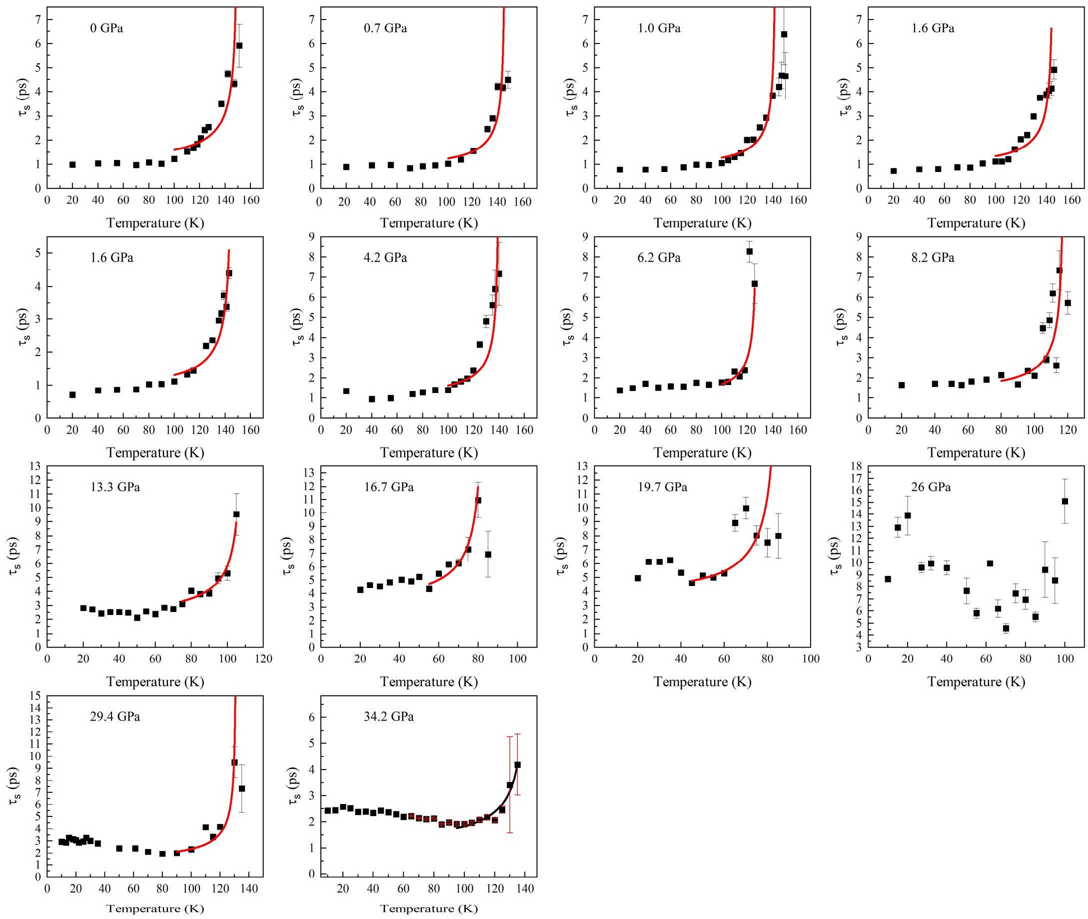
RT analysis was applied to the temperature dependence of the slow relaxation A_s

(Supplementary Fig. 4) and τ_s (Supplementary Fig. 5) under various pressures using:

$$A(T) \propto \frac{\Phi/(\Delta(T)+k_BT/2)}{1+\gamma\sqrt{2k_BT/\Delta(T)}\exp[-\Delta(T)/k_BT]} \qquad (S1)$$

$$\tau^{-1}(T) \propto \Delta(T) \qquad (S2)$$

Where $\Delta(T)$ takes BCS temperature dependence $\Delta(T) \approx \Delta(0)\tanh\left(1.74\sqrt{T_c/T-1}\right)$. We didn't fit the data at 26.0 GPa using RT model, since no divergent behavior of τ_s was observed. The extracted transition temperatures T_{DW} and the energy gaps $\Delta(0)$ under various pressure are summarized in Fig.4 in the main text.



Supplementary Fig. 5 Fitting results of τ_s using RT model. The solid lines are the fitting results using Eq (S1).

Supplementary References

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