Supplementary information Section 1:

Theory:

(a) Calculation of the Rabi splitting for the exciton-photon coupling:

We use the expression for the Rabi splitting (1): \( \hbar \Omega_{rabi} = 2 \sqrt{\frac{v^2_1}{4} - \frac{1}{4} (\hbar \Omega_{ex} - \hbar \Omega_{cav})^2} \), where, \( \hbar \Omega_{ex} = 3.5 \text{ meV} \), and \( \hbar \Omega_{cav} = 1.5 \text{ meV} \) are the half width at half maximum (HWHM) of the exciton and cavity photon respectively. From fitting to the data we find an exciton-photon coupling \( v_1 = 13 \text{ meV} \) which results in a Rabi energy of 25.9 meV. Note that the strong coupling condition: \( v_1^2 > \left( (\hbar \Omega_{ex})^2 + (\hbar \Omega_{cav})^2 \right)/2 \) is also satisfied.

(b) Detail calculation of the model

As mentioned in the main text, the full Hamiltonian of the system is given by,

\[
H = \sum_k \Delta_k c_k^+ c_k + \sum_{k,l} E_k^{l} r_{ik}^l r_{ik}^{l*} + \sum_k \omega_k^{tr} \psi_k^+ \psi_k + v_2 \sum_{k,l} (a_k c_q \psi_{k+q}^+ + \psi_{k+q} a_{k}^*)
\]

\[
+ v_3 \sum_{k,l} (f_{2,k} c_q \psi_{k+q}^+ + \psi_{k+q} f_{2,k}^*)
\]

where, \( c_k^+, a_k^+, \psi_k^+, \) and \( r_{ik}^l \) are the electron, photon, trion, upper \((l = 1)\) and middle \((l = 2)\) polariton creation operators respectively; \( \Delta_k \), \( \omega_k^{tr} \), \( E_k^1 \), \( E_k^2 \), are the bare electron, trion, upper, and middle polariton resonance energies respectively with \( v_2, v_3 \) being the coupling constants (see Figure S1 for the corresponding vertex diagram). Important in this approximation is that the trion’s interaction with the cavity is weak with respect to the trion’s interaction with the dispersed exciton derived polariton branch and it is this later coupling that drives the physics in our model. We have also neglected the lattice structure of MoSe2 which may be important for a more
quantitatively correct calculation.

Due to strong coupling of exciton with cavity photon there is no free excitons exist in the cavity. Therefore, it is a good approximation to have an effective field theory model where the trions interact with the lower polariton branch ($f_{2,k}$) given by last term in eq. (1).

Since in the experimental parameter regime, free electrons do not exist at lower $k_{\parallel}$ (or their number are negligible), we now integrate them out as this would mean that free electrons comprise high energy states. There are three such contributions at the leading order i.e. second order, perturbation theory. These are:

1. $O(V_2^2)$: This has the form:

$$H_{eff}^{2} = v_2^{2} \left[ \sum_{k,k',q} \frac{1}{E - \Delta_q^{el}} a_k^+ a_{k'+q} \psi_{k'+q} + \sum_{k,q} \left( \frac{1}{E - \Delta_{k'-q}^{el}} \right) \psi_{k'}^{+} \psi_k \right]$$  \hspace{1cm} (2)

The second term is a renormalization of the trion energy while the first term represents density-density interaction between the photons and the trions.

2. $O(V_3^2)$: This has the form:

$$H_{eff}^{2} = v_3^{2} \sum_{k,k',q} \frac{1}{E - \Delta_q^{el}} (\Gamma_{2,k}^{el} \Gamma_{2,k'} + \delta_{kk'}) \psi_{k'+q}^{+} \psi_{k+q}$$

$$= v_3^{2} \left[ \sum_{k,k',q} \frac{1}{E - \Delta_q^{el}} \Gamma_{2,k}^{el} \Gamma_{2,k'} \psi_{k'+q}^{+} \psi_{k+q} + \sum_{k,q} \left( \frac{1}{E - \Delta_{k'-q}^{el}} \right) \psi_{k'}^{+} \psi_k \right]$$  \hspace{1cm} (3)

Like in the previous expression, the second term denotes renormalization of the trion energy. However, the first term now stands for density-density interaction between the trion and the lower polariton.

3. $O(V_2 V_3)$: This has the form:

$$H_{eff}^{3} = v_2 v_3 \sum_{k,k',q} \frac{1}{E - \Delta_q^{el}} a_k^+ \Gamma_{2,k} \psi_{k'+q}^{+} \psi_{k+q} + h. c.$$  \hspace{1cm} (4)
Collecting the 2nd term from eq. (8) and (9), gives the renormalized energy of the trion:

$$A_k = \omega_k^{tr} + \sum_q \frac{v_q^2 + v_q^2}{E - A_q^{el}} \quad (5)$$

Note that, with the zeroth order approximation we are using only the exciton-photon coupled oscillator model to get the polariton basis. A better approximation can be made by iteration or a self-consistent technique.

**(c) The upper and middle polariton basis**

Inside the cavity, the photon dispersion, *i.e.* bare dispersion of the cavity is given by,

$$\omega_k^{ph} = \frac{hc}{n} \sqrt{\left(\frac{2\pi}{\lambda_c}\right)^2 + k^2}, \text{ where } n \text{ is refractive index of the cavity material.} \quad (6)$$

The exciton-photon interaction Hamiltonian can be written as,

$$H_{ph} + H_{ex} + H_{ex-ph} = \sum_k [a_k^\dagger, \phi_k^\dagger] \begin{bmatrix} \omega_k^{ph} & v_1 \\ v_1 & \omega_k^{ex} \end{bmatrix} \begin{bmatrix} a_k \\ \phi_k \end{bmatrix} \quad (7)$$

Where, $\phi_k^\dagger$ is the exciton creation operator with energy $\omega_k^{ex}$.

Defining:

$$\begin{bmatrix} I_{1,k} \\ I_{2,k} \end{bmatrix} = \begin{bmatrix} \alpha_k & \beta_k \\ \beta_k & -\alpha_k \end{bmatrix} \begin{bmatrix} a_k \\ \phi_k \end{bmatrix} \quad (8)$$

Where, $\alpha_k = \frac{1}{\sqrt{2}} \left(1 + \frac{\epsilon_k}{\sqrt{\epsilon_k^2 + v_1^2}}\right)$; $\beta_k = \frac{1}{\sqrt{2}} \sqrt{1 - \frac{\epsilon_k}{\sqrt{\epsilon_k^2 + v_1^2}}}$

And, $\epsilon_k = \frac{\omega_k^{ph} - \omega_k^{ex}}{2}$

Also note, $\begin{bmatrix} a_k \\ \phi_k \end{bmatrix} = \begin{bmatrix} \alpha_k & \beta_k \\ \beta_k & -\alpha_k \end{bmatrix} \begin{bmatrix} I_{1,k} \\ I_{2,k} \end{bmatrix}$

Eigen values of eq. (12):

$$E_k^1 = \epsilon_k^+ + \sqrt{\epsilon_k^2 - 2 + v_1^2}, \text{ and } E_k^2 = \epsilon_k^+ - \sqrt{\epsilon_k^2 - 2 + v_1^2} \quad (11)$$

where, $\epsilon_k^+ = \frac{\omega_k^{ph} + \omega_k^{ex}}{2}$. 
By performing the polariton transformation which replaces the old basis $a_k$ and $\phi_k$ with polariton basis: $\Gamma_{1,k}$ and $\Gamma_{2,k}$, we get:

$$
H_{eff}^1 = v_2^2 \sum_{k,k',q} \frac{\psi_{k',q}^+ \Gamma_{1,k'} \Gamma_{2,k} \psi_{k+q}}{E - \Delta_q^\ell} \left[ \alpha_k \alpha_{k'} \Gamma_{1,k}^\dagger \Gamma_{1,k'}^\dagger + \beta_k \beta_{k'} \Gamma_{2,k}^\dagger \Gamma_{2,k'}^\dagger + \alpha_k \beta_{k'} \Gamma_{1,k}^\dagger \Gamma_{2,k'}^\dagger + \beta_k \alpha_{k'} \Gamma_{2,k}^\dagger \Gamma_{1,k'}^\dagger \right]
$$

$$
H_{eff}^2 = v_3^2 \sum_{k,k',q} \frac{\Gamma_{1,k}^\dagger \Gamma_{2,k'} \Gamma_{2,k} \psi_{k'\,+\,q} \psi_{k\,+\,q}}{E - \Delta_q^\ell}
$$

$$
H_{eff}^3 = v_2 v_3 \sum_{k,k',q} \frac{\alpha_k \Gamma_{2,k} \Gamma_{1,k'\,+\,q} \psi_{k\,+\,q}}{E - \Delta_q^\ell} + h.c.
$$

(12)

(13)

(14)

(d) **Mean Field Theory:**

We treat the interaction within mean field theory with the non-zero mean fields being

$$
<\psi_{k'}^+ \psi_k> = \delta_{k'k} n_{tk}
$$

$$
<\Gamma_{a,k}^\dagger \Gamma_{b,k'}^\dagger> = \delta_{kk'} \delta_{ab} n_{r_{a,k}}
$$

Equations (15) and (16) describes the average occupancy of trions and polaritons as $n_{tk}$ and $n_{r_{a,k}}$. Note that in this experiment, the system could be highly in non-equilibrium state, as reaching a state of thermal equilibrium with polariton gas is not trivial. Therefore, the statistics for Bosons and Fermions at equilibrium will not be applicable here. Therefore, we take the average occupancy of the non-equilibrium polariton gas as fitting parameters and neglect their k-dependence. The ratio of $n_r$ and $n_{r_{a,k}}$ is used as one of the fitting parameters in our model. We have assumed the trion occupancy number $n_{tk} \sim 1.0$ for the following reason: We assumed that all the free electrons in the system is used up to form the trions, therefore the trion concentration is the same as the free electron concentration in the system. Fermi wave vector for trions are larger than the maximum wave vector ($5 \times 10^6 / m$) we are considering. Which implies that the trion energies are much below the Fermi energy and hence the trion occupancy number of 1 is a reasonable approximation in the model.

The data in Fig. 1d in the main manuscript is fit by the results of our model by taking the ratio of the upper and lower polariton occupancy numbers as a fitting parameter which is around 1.3. However, to obtain the values of trion coupling parameters ($v_2$ and $v_3$) which depends on the exact average occupancy numbers of the exciton-polaritons we need to consider a non-equilibrium statistical mechanics which is beyond the scope of the current model. The model nevertheless, can find the value of exciton-polariton coupling strength ($v_1$) which is almost independent of the occupancy numbers.

Thus we have from (12), (13) and (14),
Collecting the quadratic terms, we can now write the complete mean field Hamiltonian as:

\[ H_{MF} = \sum_k \psi_k^T \mathbf{H}_k \psi_k \]  

Where, \( \psi_k = \begin{bmatrix} \Gamma_{1,k}^+ \\ \Gamma_{2,k}^+ \end{bmatrix} \),

And, \( \mathbf{H}_k = \begin{bmatrix} E_k + \rho_{s1k} + \rho_{s2k} + \rho_{s3k} & \tau_{1k} + \tau_{2k} + \tau_{3k} & 0 \\ \tau_{1k} + \tau_{2k} + \tau_{3k} & E_k + \eta_{1k} + \eta_{2k} + \eta_{3k} & 0 \\ 0 & 0 & \Lambda_k + \lambda_{1k} + \lambda_{2k} + \lambda_{3k} \end{bmatrix} \)
\begin{align*}
\rho_{1k} &= -\frac{v_2^2}{v} \alpha_k^2 n_{tk} \quad (23) \\
\rho_{2k} &= 0 \\
\rho_{3k} &= 0 \\
\eta_{1k} &= -\frac{v_2^2}{v} \beta_k^2 n_{tk} \quad (24) \\
\eta_{2k} &= -\frac{v_2^2}{v} n_{tk} \\
\eta_{3k} &= -\frac{2v_2v_3}{v} \beta_k n_{tk} \\
\tau_{1k} &= -\frac{v_2^2}{v} \alpha_k \beta_k n_{tk} \quad (25) \\
\tau_{2k} &= 0 \\
\tau_{3k} &= -\frac{v_2v_3}{v} \alpha_k n_{tk}
\end{align*}

We can now write down the trion energy as:

\[ E_{tr}(k) = \omega_k^{tr} - \frac{1}{v} \left[ v_2^2 \left\{ 1 + \alpha_k^2 n_{r_1} + \beta_k^2 n_{r_2} \right\} + v_3^2 \left\{ 1 + n_{r_2} \right\} + 2v_2v_3 \beta_k n_{r_2} \right] \quad (26) \]

In the limit \( k \parallel \rightarrow k_{\text{large}} \), \( \alpha_{k_{\text{large}}} \rightarrow 2 \), and \( \beta_{k_{\text{large}}} \rightarrow 0 \), the 2nd term in Eq. (26) becomes:

\[ E_{\text{renorm}} = -\frac{1}{v} \left[ v_2^2 \left\{ 1 + 4n_{r_1} \right\} + v_3^2 \left\{ 1 + n_{r_2} \right\} \right] \]

(27)

The physical significance of the term \( E_{\text{renorm}} \) is that it is the renormalized trion energy because of the many-body interaction effect. The bare trion energy can be written as:

\[ E_{tr}^0 = \omega_k^{tr} + E_{\text{renorm}} \quad (28) \]

Now Eq. (26) can be written as:

\[ E_{tr}(k) = E_{tr}^0 - E(\alpha_k, \beta_k) \quad (29) \]

Where, \( E(\alpha_k, \beta_k) = \frac{1}{v} \left[ v_2^2 \left\{ 1 + \alpha_k^2 n_{r_1} + \beta_k^2 n_{r_2} \right\} + v_3^2 \left\{ 1 + n_{r_2} \right\} + 2v_2v_3 \beta_k n_{r_2} \right] + E_{\text{renorm}} \) \quad (30)

Note that the sign of the function in Eq. (30) depends on the relative sign of \( v_2 \) and \( v_3 \). From fitting we obtain opposite sign (see Table S1) for the two coupling constants (\( v_2, v_3 \)).

Using the values of fitting parameters we obtain the value of \( E_{\text{renorm}} \approx -5 \text{ meV} \), the binding energy of trion in the cavity is around 27 meV. Therefore, the bare trion binding energy we estimate \( \approx 32 \text{ meV} \), which matches with our experimental results nicely.
Table S1. Values of fitting parameters

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Supplementary Information Section 2:

Figure S2: The raw data for the angle resolved PL spectrum from the sample:
The data points presented in the manuscript of Figure 1d is obtained from the raw data shown below (PL intensity is plotted in the log scale). The vertical line cuts at different $k_\parallel$ points give the PL spectrum for the corresponding point. We fit each of the line cut PLs with three Lorentzian line shapes to obtain the exact location of the polariton resonance in the energy axis and the resultant plot is presented in Figure 1d in the manuscript. Figure 3b in the manuscript is obtained by differentiating the raw data in order to enhance the contrast.
Supplementary Information Section 3:

Figure S3: LPB effective mass as a function of temperature:
Fig (a) presents the effective mass of the trion-polariton branch/LPB calculated from the fit to the data in the Fig 4 (a) from manuscript for different temperatures. Fig (a) is the zoomed out version of Fig 4(b) in the main text. For the trion-polariton, the effective mass is negative near the vicinity of zero in-plane momentum for all temperatures. Comparing the effective mass of trion-polariton/LPB (Fig. b) and exciton-polariton/MPB (Fig. c), we see that the effective mass is positive below the inflection point and negative above it (Fig c). However the behavior of the trion-polariton is opposite to that (Fig b).

(a) LPB effective mass as a function of temperature. (b) the LPB effective mass compared with (c) the MPB effective mass at T=6K.

References: