Supporting Information

Room-Temperature Electron-Hole Liquid in Monolayer MoS$_2$

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S1. Reproducibility of the anomalous power dependence of the PL

The observed evolution of the PL’s power-dependence is very reproducible. We have performed measurement on numerous samples, and always observed similarly dramatic changes in the power dependence at certain transition range. Fig. S2 shows the power dependent PL spectra from one of those samples. In order to make sure that the observed phenomena are not due to defects or photoinduced degradation, we also monitor the evolution of the PL by sweeping the incident power from low to high at the same suspended MoS$_2$ for multiple times and confirm that the observed PL spatial and spectral features are highly reproducible at each of the sweeps, as shown in Fig. S3. Additionally, we have measured Raman spectra before and after the power-dependent PL measurements and ensure negligible difference in the Raman spectra after the measurements (Fig. S4).

Fig. S1. Optical images of typical suspended monolayer MoS$_2$.

Fig. S2. Power-dependent PL spectra from another sample. The incident power density is listed as shown.
Fig. S3. Reproducible measurement results from multiple sweeps. (a) Evolution of the integrated PL intensity with incident power of 1st sweep (red) and 2nd sweep (blue). (b) The PL spectra at laser intensity of 0.57kw/cm² at the beginning of the 1st sweep (red) and 2nd sweep (blue).

Fig. S4. Raman spectra collected from the same suspended monolayer MoS₂ before and after the measurement showing the formation of EHL.

S2. Evaluation temperature and its effect during the measurement

The information of temperature during the measurement is important. Two different methods are used to find out the temperatures of the sample under illumination of different laser intensities. Both methods give rise to similar values. The first method is based on the temperature effect on peak shifting and broadening. The peak and the FWHM of PL tend to change with temperature by linear relationship. We may find out the temperature by comparing the observed position and FWHM with the calibrated temperature-dependence of the peak position and FWHM. We first calibrate the temperature dependence by measuring the PL of a suspended MoS₂ monolayer in a temperature-controlled chamber under illumination of a 532 nm laser. The incident power is set to be very low (40 μW) to ensure no
obvious laser heating effect. The position and FWHM are plotted as a function of temperature as shown in Fig. S5.

We may then find out the temperature of the suspended monolayer MoS$_2$ in our experiments by comparing the measured peak position and FWHM of the PL from the A exciton with the result given in Fig. S5. Sometime to find out the PL from the exciton might request fitting since it could merge with the emission of other species. Fig. S6 shows the peak position and FWHM for the A exciton as a function of the incident laser power density. Based on the results given in Fig. S5-S6, we may obtain the temperature of the suspended monolayer at different incident powers.

![Graph showing temperature dependence of peak position and FWHM of PL](image)

**Fig. S5.** Temperature dependence of peak position and FWHM of the PL collected from suspended monolayer MoS$_2$. (a) Peak position and (b) FWHM plotted as function of temperature. Both show linear dependence on the temperature as indicated by the fitting equations given in the figure.
Fig. S6. Peak position and FWHM of the PL from the A exciton in suspended monolayer MoS$_2$ under illumination of different incident powers. (a) Peak position (red dots) (b) FWHM (blue dots) of the A exciton PL as a function of incident powers. The dash lines represent the temperature of the monolayer obtained from comparison with the calibrated temperature coefficient shown in Fig. S5.

The second method is based on the low-energy exponential tail of the PL (Fig. S7). The PL of EHP is known to have an exponential tail at the low energy end due to the random potential fluctuations from electron-hole Brownian motion. The electron-hole Brownian motion results from thermally acceleration, so it can directly indicates temperature.$^{1}$ Fig. S7a shows the tail of the PL spectra at the low energy end, and the result is plotted in logarithm scale. These tails can be fitted as $I \propto \exp(-E/E_{\exp})$, and the temperature can be obtained from the fitting parameter of $E_{\exp}$ based on its linear relationship with the temperature. For instance, the PL at the power density 0.57 kW/cm$^2$ has a fitting value of 43 meV, while the one at 2.50 kW/cm$^2$ has a value of 66 meV. By assuming the monolayer at the power density 0.57 kW/cm$^2$ is room temperature, 293 K, we may have the temperature at 2.50 kW/cm$^2$ as $T= 293 \times 66 \text{meV}/43 \text{meV} = 450 \text{K}$ at intensity 2.50 kW/cm$^2$. The $E_{\exp}$ become almost constant in the high power region, which suggests a constant temperature. The temperatures estimated from the low-energy exponential tail match what obtained from the first method very well. We may also find a linear relationship between the fitting parameter and the temperature as $E_{\exp}/k_B T = 1.7 \pm 0.1$, which is excellent agreement with what previously reported.$^{1}$
Fig. S7. Temperature of monolayer MoS$_2$ estimated from exponential tail fitting. (a) Exponential tail of the PL at the low energy end edge tail. The lines indicate fitting to the experimental measurement as $I \propto \exp(-E/E_{exp})$. (b) Fitting parameter $E_{exp}$ and the temperature derived from the fitting parameter as function of incident power density. The dashed line indicates a linear relationship between the fitting parameter and the temperature as $E_{exp}/k_B T = 1.7 \pm 0.1$.

With the information of the temperatures, we are able to evaluate how thermal effect may affect the power-dependence of PL. First, we can exclude out any major role of the thermal effect in the red shift and broadening of PL spectra (about 200meV peak shift and 200meV broadening) as observed in Fig. 1 of the main text. The temperature at 2.50kW/cm$^2$ and above is around 450K. The temperature has increased by 207K, which is expected to give rise to a redshift of 50 meV and a broadening of 40 meV according to the calibrated temperature coefficient given in Fig. S3. It is much less than what been observed in the laser intensity range 0.57kW/cm$^2$-2.50kW/cm$^2$. We may also estimate the effect of the temperature on band gap based on the calibrated temperature-dependent bandgap, which is around 50 meV at the critical power density, which may be reasonably ignored for simplicity as it is one order of magnitude smaller than the chemical potential.

S3. Theoretical calculation for the EHP and EHL

Ground state free energy. The chemical potential of a many interacting fermion system is determined by the electron and hole Fermi energies relative to the edge of the renormalized bandgap. If using the edge of the original bandgap as the ground state, the chemical potential is the sum of the Fermi energies of electrons $E_{F,e}(n)$ and holes $E_{F,h}(n)$ and the band renormalization $\Delta E_{BG}$ as $\mu = E_{F,e} + E_{F,h} + \Delta E_{BG}$. The Fermi energies in monolayer MoS$_2$ is related with the charge density $n$ as $E_{F,e}(n) = n\hbar^2/2m_e$ and $E_{F,h}(n) = n\hbar^2/2m_h$, in which $\hbar$ is the Planck’s constant,
$m_e$ and $m_h$ are the effective mass of electrons and holes, respectively. The bandgap renormalization is caused by the exchange and correlation energies of charges and, for 2D structures, can be related to the charge density by an empirical equation as
\[ \Delta E_{BG} = -3.1(na^2)^{1/3}E_b, \]
where $a$ and $E_b$ are the Bohr radius and binding energy of excitons. Using the value we found from our own experiments ($a = 0.65$ nm, and $E_b = 0.42$ eV) and in references ($m_e = 0.35 \ m_0$, and $m_h = 0.46 \ m_0$), we can calculate the chemical potential in monolayer MoS$_2$ as a function of the charge density as shown in Fig. 4b. For simplicity, we ignore the effect of temperature in the calculation. This is reasonable because previous studies have demonstrated that the Fermi energy of monolayer MoS$_2$ calculated without considering the effect of temperature shows reasonable agreement with the experimental measurements at room temperatures. It has also been well known that the bandgap renormalization is insensitive to temperature.

**Bandgap renormalization.** The bandgap renormalization $\Delta E_{BG}$ can be figured out as
\[ \Delta E_{BG} = E_{BG} - \Delta E_T - E'_{BG}, \]
where $E_{BG}$ is the electronic bandgap of unexcited monolayer MoS$_2$, $\Delta E_T$ is the bandgap change due to temperature increase, and $E'_{BG}$ is the renormalized bandgap that is indicated by the low energy band bottom of the PL emission as illustrated in Fig. 4a. Without losing generality, we focus on the PL spectra measured 3.00 kW/cm$^2$. We may find the band bottom to be 1.57 eV (Fig. 4a). Additionally, the bandgap change caused by temperature increase is estimated to be around 70 meV (see §S1). We can calculate the charge density to be 3.40 $\times 10^{13}$ cm$^{-2}$ at the EHL phase.

**PL calculation for EHL.** Different from the emission of excitons, which only involves the charges at the band edges, the luminescence of EHL involves all the occupied electronic states at the conduction band and the hole states at the valence band. It can be described as
\[ I(h\nu) = C \int_0^\infty (1 + e^{(E=E_{F,c})/k_BT})^{-1}(1 + e^{(h\nu-E-E_{F,h})/k_BT})^{-1} dE, \]
where $C$ is a term involving the oscillation strength and the density of states.

![Fig. S8. PL at the edge and center of the luminescence ring, including PL extracted from the edge of the ring (red), and PL extracted from the center of the ring at the incident power of 3.75 kW/cm$^2$ (yellow) and 4.00 kW/cm$^2$ (blue). The dashed horizontal line $Tc$ marks the critical temperature for the EHL state. The red and blue dots indicate the band bottoms for the PL spectra collected.](image-url)
References:


