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On optical dipole moment and radiative recombination lifetime of excitons in WSe2

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KEYWORDS

WSe2, Delocalized exciton, Dipole moment, Radiative lifetime, Dark state.

ABSTRACT

Optical dipole moment is the key parameter of optical transitions, as it directly determines the strength of light-matter interaction such as intrinsic radiative lifetime. However, experimental determination of these fundamental properties of excitons in monolayer WSe2 is largely limited, because that commonly used measurement, such as (time-resolve) photoluminescence, are inherently difficult to probe the intrinsic properties. For example, dark states below bright exciton can change the photoluminescence emission rate by orders of magnitude and gives an "effective" radiative lifetime distinctive from the intrinsic one. On the other hand, such "effective" radiative lifetime becomes important itself because it describes how dark states affect exciton dynamics. Unfortunately, the "effective" radiative lifetime in monolayer WSe2 is also not determined as it requires photoluminescence measurement with resonant excitation, which is technically difficult. These difficulties are overcome here to obtain both the "intrinsic" and "effective" radiative lifetime experimentally. A framework is developed to determine the dipole moment and "intrinsic" radiative lifetime of delocalized excitons in monolayer WSe2 from the absorption measurements. In addition, the "effective" radiative lifetime in WSe2 is obtained through time-resolved photoluminescence and absolute quantum-yield measurement at resonant excitation. These results provide helpful information for fundamental understanding of exciton light-matter interaction in WSe2.

1. Introduction

Monolayer transition metal dichalcogenides (MX_2) have been intensively studied as a model system of two-dimensional (2D) semiconductors. The remarkably strong light-matter interaction in atomically thin MX_2 can lead to new optical phenomena and enable novel optoelectronic applications. Strong optical responses in MX_2 originate from exciton resonances. It has been shown that excitons in MX_2 monolayers can have a binding energy up to hundreds of meV and a Bohr radius down to 1nm due to reduced dielectric screening in 2D systems.[1-3] Although there have been many experimental studies on optical properties of monolayer MX_2, several fundamental questions regarding to excitons in MX_2, such as the exciton transition dipole moment and the intrinsic radiative recombination lifetime, are still not well understood.

Previously, time-resolved photoluminescence (TRPL) measurements were used to estimate the exciton radiative lifetime in MX_2.[4-7] However, photoluminescence (PL) varies significantly with sample qualities (such as defects and impurities) and excitation condition. Furthermore, the potential existence of dark states below bright exciton can dramatically change its population decay dynamics. For example, low energy dark excitons can significantly reduce the number of bright excitons at thermal equilibrium; and therefore PL measurement gives an "effective" radiative lifetime orders of magnitude longer than the intrinsic radiative lifetime of bright exciton.[8-10] Therefore, it is very challenging to obtain intrinsic exciton dipole moment and radiative lifetime by PL measurement alone. On the other hand, the "effective" radiative lifetime itself is essential to probe the existence of dark states as well as quantitatively study their effects on exciton dynamics. Nevertheless, experimental determination of "effective" radiative lifetime is again not simple, as recent photoluminescence excitation (PLE) spectroscopy study shows that quantum yield (QY) of monolayer WSe2 varies significantly with the excitation energy above A

exciton resonance.[11] To avoid such complication, one needs use photoexcitation exactly at A exciton resonance, which is experimentally demanding since the PL and excitation light have the same energy and thus cannot be spectrally separated.

Here we develop a theoretical framework to obtain the intrinsic dipole moment and radiative lifetime of excitons in monolayer WSe2 from absorption measurement. In contrast to PL, optical absorption is rather insensitive to different perturbations such as defects/impurities, dark states, and non-radiative recombination channels. Combined with absorbance measurement, we obtain an intrinsic radiative lifetime of 4ns for monolayer WSe2 at room temperature. In addition, we successfully measure "effective" radiative lifetime through TRPL and absolute QY measurement with resonant excitation exactly at A exciton energy, by careful rejection of excitation light with combination of a spatial and polarization filter. By comparing the intrinsic and "effective" radiative lifetime, we find only ~4% of excited excitons stay at bright A exciton state. Our work not only provides complete theoretical picture to relate absorption and emission processes for an exciton in MX_2; but also unambiguously reveal the important role of dark states in PL dynamics even for high quality WSe2 at room temperature.

2. Results and Discussions

2.1. Determination of intrinsic dipole moment and radiative lifetime

Transition dipole moment governs the strength of light-matter interaction in both absorption and emission. The relation between dipole moment and optical processes is most transparent in atomic systems. For a hydrogen atom, an electron is "localized" in a range described by Bohr radius a_B, and the dipole moment μ for an allowed transition is comparable to ea_B, where e is

electron charge. The total frequency-integrated absorption cross-section σ^I and spontaneous emission rate A of this transition are then directly determined by the dipole moment through $\sigma^I = \pi\omega\mu^2/(3\varepsilon_0\hbar c)$ and $A = \omega^3\mu^2/(3\pi\varepsilon_0\hbar c^3)$, respectively.[12] Here $\hbar\omega$ is the transition energy. Ensembles of hydrogen atoms can often be described by a sum of non-interacting individual atoms, where the absorption is proportional to the number of atoms, but the radiative recombination lifetime remains the same.

In crystalline solids, optical absorption and emission from exciton transitions are also intrinsically determined by the transition dipole moment, however excitons in solids (including 2D MX_2) are delocalized in space. Such delocalization modifies the exact relation between optical absorption cross-section, radiative recombination lifetime, and transition dipole moment: delocalized excitons can have a dipole moment much larger than the exciton "Bohr radius" and lead to faster spontaneous emission rate.[13,14] Such effects in monolayer MX_2 have not been investigated. We construct below a theoretical framework to connect the optical dipole moment, absorption cross-section and intrinsic radiative lifetime in monolayer WSe2; and perform absorption measurement to obtain them reliably in experiment. Note that the scheme developed here is particularly useful for excitonic transition in MX2 because absorption spectra directly probe transition dipole moment without complication from various relaxation processes. One extreme example is the difference between monolayer and bilayer MX_2: the absorption cross section of A-exciton resonance in bilayer MX_2 is approximately twice that of a monolayer, whereas PL in monolayer and bilayer MX2 can be different by orders of magnitude due to the direct to indirect bandgap transition.[15,16]

The optical absorption of monolayer WSe2 at room temperature is measured through reflection spectroscopy of exfoliated WSe_2 flakes on quartz substrates. The optical absorbance α of a

freestanding monolayer WSe2 is directly related to its differential reflectance ($\Delta R/R$) on a transparent quartz substrate by $\alpha = \frac{n_s^2-1}{4}\frac{\Delta R}{R}$, where $n_s = 1.47$ is the refractive index of quartz.[17,18] **Figure 1**a shows the extracted absorbance spectrum of a freestanding monolayer WSe2. The two prominent absorption peaks at 1.65eV and 2.06 eV correspond to the A and B exciton resonances, respectively. Here we focus on the A exciton state.

The total frequency-integrated absorption cross section of the A excitons per unit area (${\sigma^I}_{tot}$) can be obtained experimentally from the integration of the A-exciton absorption peak over angular frequency in the absorbance spectrum, which yields the value ${\sigma^I}_{tot} = 7.8meV/\hbar$ for a freestanding monolayer WSe2. The total absorption cross section is related to the behavior of individual excitons through the two-dimensional density of active exciton states (supplementary information S1):

$$ {\sigma^I}_{tot} = n_{2D}\sigma_A^I = n_{2D}\frac{\pi\omega_A}{\varepsilon_0\hbar c}\mu_A^2 \quad (1) $$

Here n_{2D} is the two-dimensional density of exciton states that can absorb light, σ_A^I is the frequency-integrated absorption cross section of a single exciton, $\hbar\omega_A = 1.65\text{eV}$ is the A-exciton transition energy, and $\mu_A$is the A-exciton transition dipole moment.

The density of exciton states that can absorb light (n_{2D}) can be obtained in the following way. Figure 1b illustrates the absorption process in momentum space, where G is the ground state and A is the A-exciton band, which has an energy dispersion over the center-of-mass momentum k, and the exciton mass $m_A = m_e + m_h = 1.05m_0$.[19] The finite homogeneous linewidth $\Delta(T)$, which is determined by the temperature-dependent dephasing rate, leads to an uncertainty of the exciton momentum $\Delta k(T)$. At most temperatures, $\Delta(T)$ is dominated by exciton-carrier and

exciton-phonon interaction, and takes the form $\Delta(T) = \gamma T$.[20,21] The oscillator strength of the $k = 0$ bright exciton is shared by all states within $\Delta k(T)$,[22,23] as highlighted by yellow area in Figure 1b. Consequently, excitons within the momentum $\Delta k(T) = \sqrt{2m_A\gamma T}/\hbar$ are active states in light absorption, and it has a density of

$$n_{2D}(T) = \frac{\pi\Delta k^2(T)}{(2\pi)^2} = \frac{m_A\gamma T}{2\pi\hbar^2} \quad (2)$$

This density can be understood physically in a real space picture as shown in Figure 1c, where delocalized excitons coherently absorb and emit light within the coherent length $\Lambda(T) = h/\sqrt{2\pi m_A\Delta(T)}$, and the exciton density is simply $n_{2D}(T) = 1/\Lambda(T)^2$.

We can determine the dipole moment of delocalized A exciton by combining Equation 1 and 2, and obtain $\mu_A(T) = 400\sqrt{k_B/\gamma T}$ (Debye $\cdot$ K$^{\frac{1}{2}}$), which varies strongly with the temperature. Upon photoexcitation, hot excitons quickly form a thermal equilibrium, and only the fraction within $\Delta(T)$ will contribute to radiative recombination[22,23], giving an "intrinsic" radiative life time in freestanding WSe_2

$$\tau_R^A(T) = \frac{3\pi\varepsilon_0\hbar c^3}{\omega_A^3\mu_A^2(T)}/(1 - e^{-\frac{\Delta(T)}{k_BT}}) = 13\,T\ (\text{ps}\cdot\text{K}^{-1}) \quad (3)$$

which is about 4ns at room temperature. Here we determined $\Delta = 50$meV at room temperature from our absorption and photoluminescence excitation measurement (Supplementary Information S5). At lower temperature, the "intrinsic" radiative lifetime can be much shorter with reduced resonance linewidth.

The above framework relates the absorption cross-section, the dipole moment, and the radiative recombination lifetime of delocalized excitons in a solid state system. We note that an alternative

emission picture has been employed to obtain the linear dependence of radiative lifetime on temperature in semiconductor quantum well, where a thermal average of radiative lifetime among all excitons with different momentum was used.[21,24] However, a consistent absorption picture is absent. We provide in Supplementary Information S2 a corresponding absorption picture and show that such alternative framework yields the equivalent results as equation 3.

2.2. Determination of effective radiative lifetime

Having obtained the "intrinsic" radiative lifetime from light absorption in monolayer WSe2, next we compare it to the "effective" radiative lifetime determined from light emission to probe the existence and effects of dark states. In principle, radiative lifetime of A exciton in emission process can be obtained as $\tau_A' = \tau_A^{R'}/\mathrm{QY}$. Here τ_A' represents the population decay (i.e. total lifetime) of the A exciton, which can be measured from time-resolved PL or pump-probe spectroscopy;[6,7,25,26] QY is the quantum yield, defined as the ratio between emitted photon and excited A exciton number.[11,16,27] However, it was reported that light emission efficiency can depend sensitively on the optical excitation energy in MX_2, presumably because that excitons excited with large excess energies will have different relaxation pathways, and only a proportion will relax to A exciton.[11] Therefore, the PL efficiency measured with large excitation energy contains convoluted information and cannot be used to determine "effective" radiative lifetime of A exciton. To eliminate such complication, a resonant excitation at A exciton energy is required, since A excitons are then directly excited.

To measure PL emission at resonant excitation, a 50um-sized exfoliated monolayer WSe2 on SiO2/Si is used to reduce scattering of excitation light, combining with spatial and polarization filter (see supplementary information S3). We perform time-resolved PL measurements using the

Time-Correlated Single Photon Counting (TCSPC) technique. **Figure 2** shows the time-resolved PL spectrum with resonant excitation at 1.65 eV. The red curve is the experimental decay curve, and it has a decay lifetime much longer than the instrument response function (black curve). After deconvolution we obtain a PL decay dominated by a single decay exponent with a time constant of $\tau_A' = 400$ ps. This PL decay lifetime is more than one order longer than typical values reported from CVD grown samples,[28] indicating a rather high quality of our exfoliated monolayer WSe_2. The lifetime τ_A' obtained here represents the population decay (i.e. total lifetime) of the A exciton.

To obtain quantum yield, we measured PL intensity with different incident excitation energy across the A exciton resonance (see Supplementary Information S4) at room temperature. **Figure 3**a shows 2D color plot of photoluminescence excitation spectroscopy results, where vertical, horizontal axis and color scale correspond to excitation energy, emission energy and PL intensity, respectively. PL spectra for a specific energy can be obtained by horizontal cuts of the 2D plot. PL spectra for 1.65eV (A exciton resonance) and 1.85eV excitations are shown in Figure 3b. The emission peak has the same energy as the A-exciton resonance in absorption, and exhibits an almost constant spectral lineshape as we scan the excitation energy (supplementary information S5). It clearly shows that the PL originates from the A exciton. On the other hand, the PL intensity is over 10 times higher at resonant excitation. This increased PL intensity arises from two factors: an increased light absorption and a higher quantum yield with resonance excitation. Figure 3c shows the variation of quantum yield for freestanding monolayer WSe_2 as a function of the excitation energy after correction for the local field effect and the objective collection efficiency (Supplementary Information S4). We find that the quantum yield at resonant excitation is indeed significantly higher than non-resonant excitation. However QY is low even

for resonant excitation (4×10^{-3}), indicating that majority of photo-excited excitons still decay through non-radiative channel.

Combining a total decay lifetime $\tau_A' = 400$ps and a PL QY $= 4\times10^{-3}$ at resonant excitation yields an "effective" radiative recombination lifetime $\tau_A^{R'} = 100$ns for freestanding monolayer WSe2. The "effective" radiative recombination lifetime is ~ 25 times longer than the "intrinsic" radiative recombination lifetime estimated from light absorption. This large discrepancy indicates that a simple model with only a bright exciton band is insufficient to describe light emission from high quality exfoliated WSe2 samples at room temperature. Apparently only a small portion of excited excitons (~ 4%) stay in bright A-exciton state even with resonant excitation. All the other excitons are not emitting, either trapped in a "dark" exciton state, or trapped by impurities and defects. Obviously, some of these trapping processes are very efficient, with the trapping time constant significantly shorter than the instrument response time (~100ps) in our measurement; otherwise we will observe an initial fast decay component in the TRPL curve. On the other hand, a fraction of excitons can stay in the bright state for an extended time rather than completely disappear due to trapping into dark states. It suggests that these fast-trapping dark states are "shallow" in energy and are able to repopulate the A-exciton state to reach a quasi-equilibrium. As a consequence of the quasi-equilibrium between "shallow" dark states and bright A-exciton states, about 4% excitons remain in the bright A-exciton state and give rise to a well behaved single exponential decay of 400ps. See Supplementary Information S6 for a phenomenological model including fast-trapping "shallow" dark states to describe the exciton dynamics in monolayer WSe_2.

3. Conclusion

The capability of dark states to repopulate indicates that they must lie within ~ 100meV of the A-exciton. In addition, it cannot efficiently interact with photon, which excludes the possibility of other bright states such as trion. Potential candidates of the dark states are triplet excitons due to flipped electron/hole spins, finite-momentum excitons due to the non-parabolic dispersion of conduction and valance bands in monolayer WSe_2,[29] and shallow trap states around impurities and defects, which deserves further studies. Our results unambiguously reveal the existence of dark states in high quality monolayer WSe2, and quantitatively determine its strong effect on PL emission that only 4% of excited states stay at A exciton level. Consequently, the population lifetime measured in time-resolved PL and transient absorption cannot be compared to the intrinsic radiative lifetime directly even after considering quantum yield.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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FIGURES

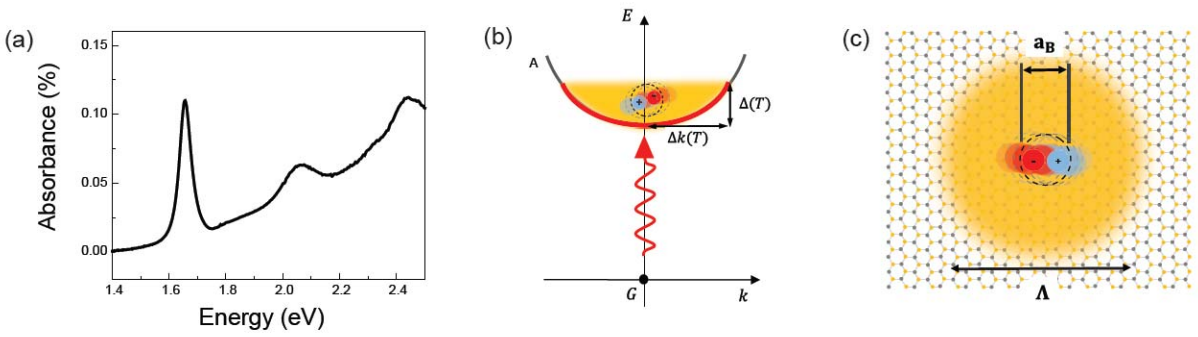

Figure 1: Determining exciton dipole moment and "intrinsic" radiative lifetime from absorption measurement for monolayer WSe_2. (a) Optical absorbance spectrum of a freestanding monolayer WSe2. The two prominent absorption peaks at 1.65 eV and 2.06 eV correspond to A and B exciton resonances, respectively. (**b) Absorption process of delocalized excitons in momentum space.** Delocalized A excitons form a band (A). The yellow area represents states within homogeneous linewidth $\Delta(T)$, which can absorb and emit light. (**c) Absorption process of delocalized excitons in real space.** Delocalized exciton coherently absorb and emit light within the coherence length Λ (yellow circle), which can be significantly larger than exciton Bohr radius a_B.

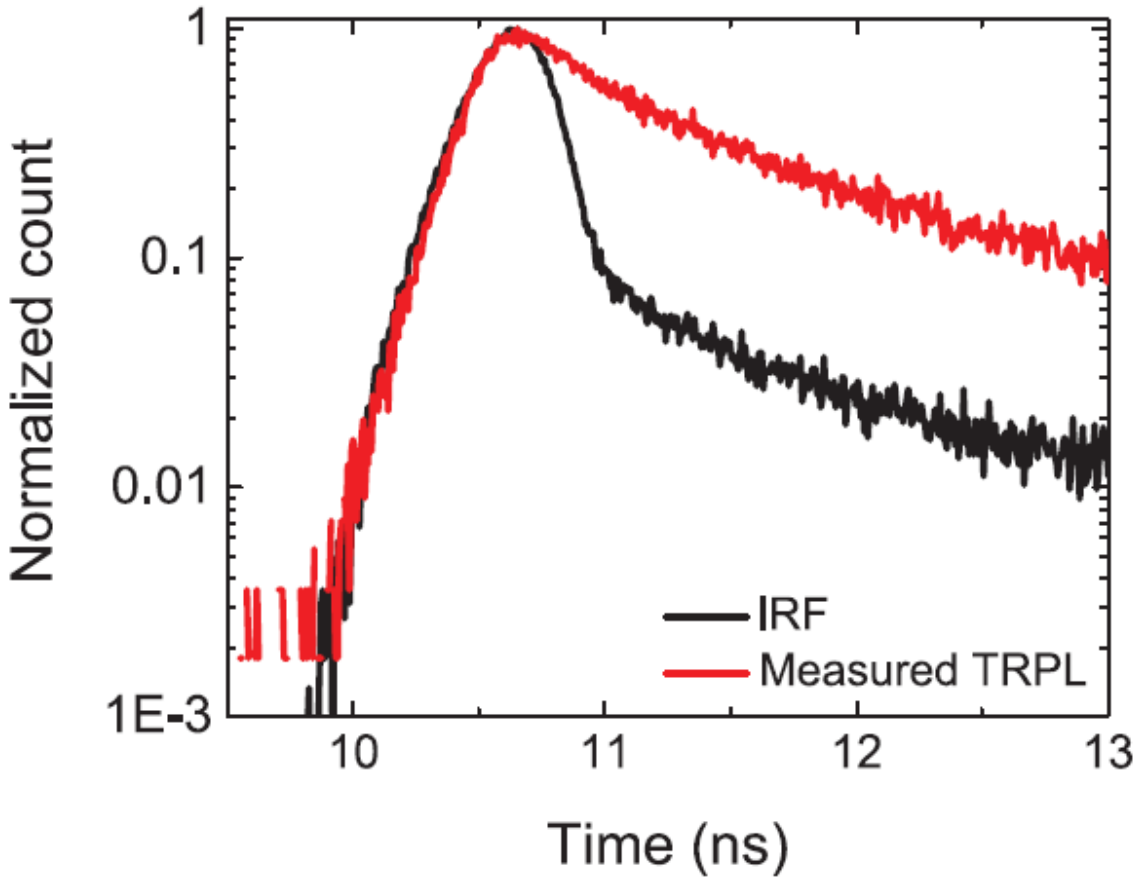

Figure 2: Time-resolved PL of monolayer WSe2 with resonant excitation at 1.65 eV. Red and black curves are experimentally measured decay curve and instrumental response functions (IRF), respectively. The PL decay is dominated by an exponent with a time constant of $\tau_A' =$ 400 ps.

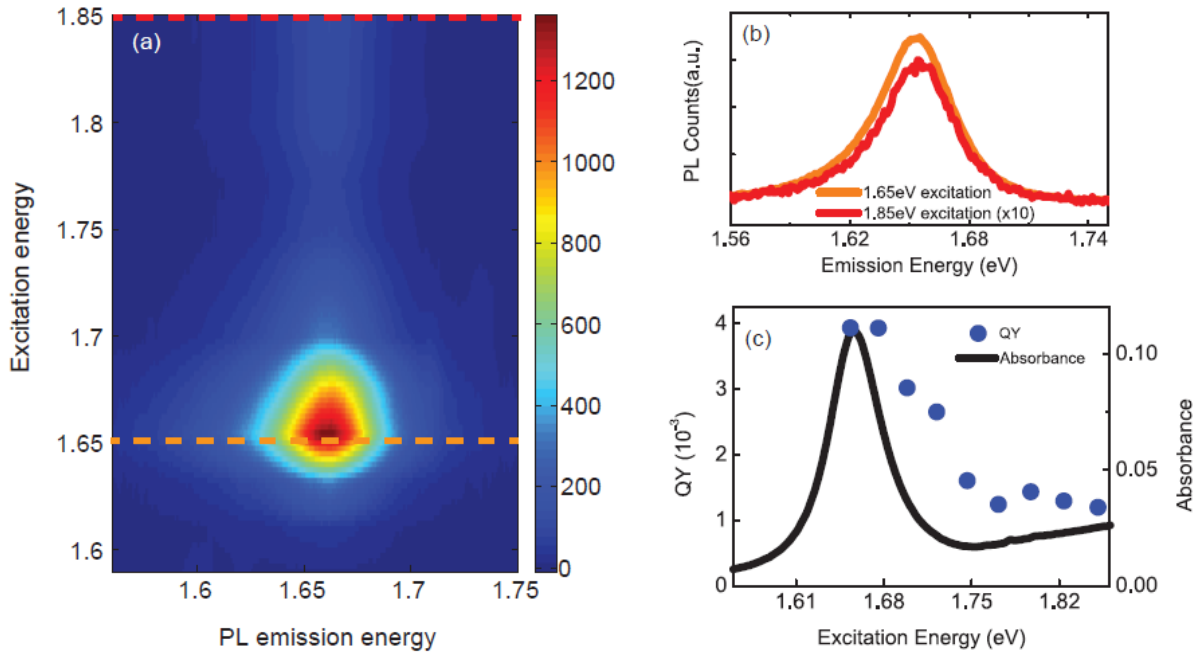

Figure 3: Determining "effective" radiative lifetime from emission measurement for monolayer WSe_2. (a) Two-dimensional mapping of photoluminescence excitation spectrum (PLE). Two representative excitation conditions at 1.65eV (resonant) and 1.85eV (non-resonant) are labeled with orange and red dashed lines, with corresponding PL spectra shown in **(b)**. **(c) Excitation energy dependence of absolute quantum yield.** Absorbance spectrum (black line) is also shown for reference.

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The dipole moment and "intrinsic" radiative lifetime of excitons in WSe2 are determined from absorption measurement; The "effective" radiative lifetime of excitons is also obtained through time-resolved photoluminescence and absolute quantum-yield measurement with resonant excitation. The framework developed here provides helpful information to determine fundamental quantities of exciton light matter interaction, and to understand dynamics of delocalized excitons in solids.

Keyword

WSe2, Delocalized exciton, Dipole moment, Radiative lifetime, Dark state.

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