Clustering & Global Challenges

April 07-09, 2021

Online international conference

Health
Smart Energy
Quantum Tech.
Advanced Materials

Exciton-exciton interaction beyond the hydrogenic picture in a $MoSe_2$ monolayer in the strong light-matter coupling regime

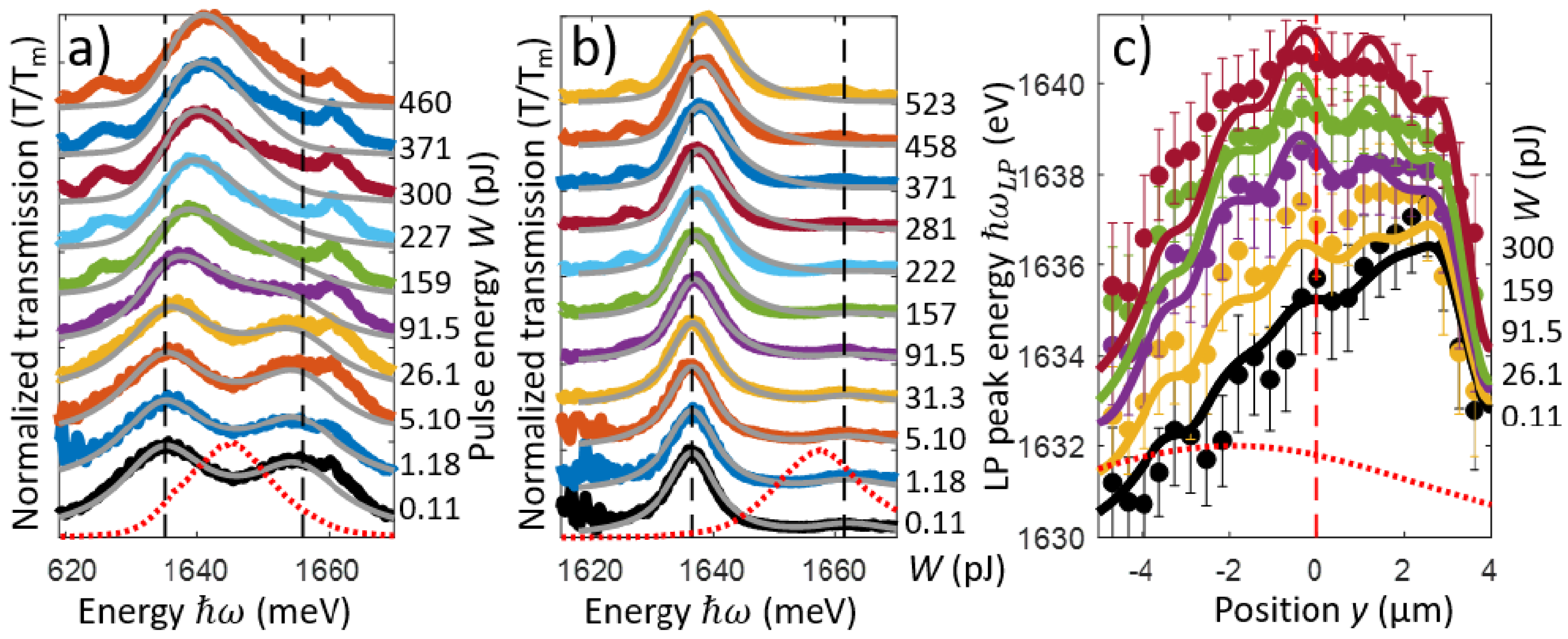
Petr Stepanov[2], **Amit Vashisht[1]**, Martin Klaas[3], Nils Lundt[3], Sefaattin Tongay[4], Mark Blei[4], Sven Höfling[3], Thomas Volz[5,6], Anna Minguzzi[1], Julien Renard[2], Christian Schneider[3] and Maxime Richard[2]

1 Univ. Grenoble Alpes, CNRS, LPMMC, 38000 Grenoble, France
2 Univ. Grenoble Alpes, CNRS, Grenoble INP, Institut Néel, 38000 Grenoble, France
3 Technische Physik and Wilhelm Conrad Röntgen Research Center for Complex Material Systems, Physikalisches Institut, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany
4 Arizona State University, Tempe, Arizona 85287 USA
5 Department of Physics and Astronomy, Macquarie University, NSW, 2109, Australia
6 ARC Centre of Excellence for Engineered Quantum Systems, Macquarie University, NSW, 2109, Australia

Abstract

In transition metal dichalcogenides layers of atomic scale thickness, the electron-hole Coulomb interaction potential is strongly influenced by the sharp discontinuity of the dielectric function across the layer plane. This feature results in peculiar non-hydrogenic excitonic states, in which exciton-mediated optical nonlinearities are predicted to be enhanced as compared to their hydrogenic counterpart. To demonstrate this enhancement, our experimental collaborators performed optical transmission spectroscopy of a MoSe2 monolayer placed in the strong coupling regime with the mode of an optical microcavity, and we analyzed the results quantitatively with a nonlinear input-output theory. We found an enhancement of both the exciton-exciton interaction and of the excitonic fermionic saturation with respect to realistic values expected in the hydrogenic picture. Such results demonstrate that unconventional excitons in MoSe2 are highly favourable for the implementation of large exciton-mediated optical nonlinearities, potentially working up to room temperature.

Nonlinear Transmission Spectroscopy



- The Optical response of excitons in strong light-matter coupling is characterized by **Exciton Polariton resonances**.
- Measured normalized transmission spectra T(ω)/Max(T) at T= 127K and T= 105K are shown in (a) and (b). The spectra are stacked from the lowest used pulse energy, W (bottom) to the highest (top). The laser pulse spectrum is shown in (a, b) as a red dotted line.
- The dashed vertical black lines in (a, b) highlight the polaritonic resonances in the linear regime. The theoretical fits are shown as solid gray lines.
- (c) shows spatially resolved lower-polariton transmission peak energy measured at T= 127K, across the excitation spot diameter, for increasing W(same colour code as in (a)). The spatial laser intensity profile is shown as a red dotted line. The spectra in (a) have been measured at y= 0 (dashed vertical line).

Input Output Theory

Coupled mean-field equations of motion in the exciton and photon basis

$$
\begin{aligned}
i\partial_t\psi_c &= \left(\omega_{c,0} - \frac{\hbar}{2m}\nabla^2 - i\frac{\gamma_c}{2} + V_c(\mathbf{r})\right)\psi_c \\
&\quad + \left(\frac{\Omega}{2} - \frac{\tilde{g}_s(\theta)}{2}|\psi_x|^2\right)\psi_x + \sqrt{2\gamma_{in}}A_{in} \\
i\partial_t\psi_x &= \left(\omega_{x,0} - i\frac{\gamma_x}{2} + V_x(\mathbf{r}) + \tilde{g}_x(\theta)|\psi_x|^2\right)\psi_x \\
&\quad + \left(\frac{\Omega}{2} - \tilde{g}_s(\theta)|\psi_x|^2\right)\psi_c - \frac{\tilde{g}_s(\theta)}{2}\psi_x^2\psi_c^*,
\end{aligned}
$$

$\tilde{g}_x =$ Dipole-Dipole interaction constant

$\tilde{g}_s =$ Fermionic Saturation interaction constant

$\gamma_{x,c} =$ dissipation constants

$V_{x,c} =$ spatial potential

$\Omega =$ Rabi Splitting

$A_{in} = \sqrt{I_{las}(t,\mathbf{r})}$

Input Laser field (Gaussian mode)

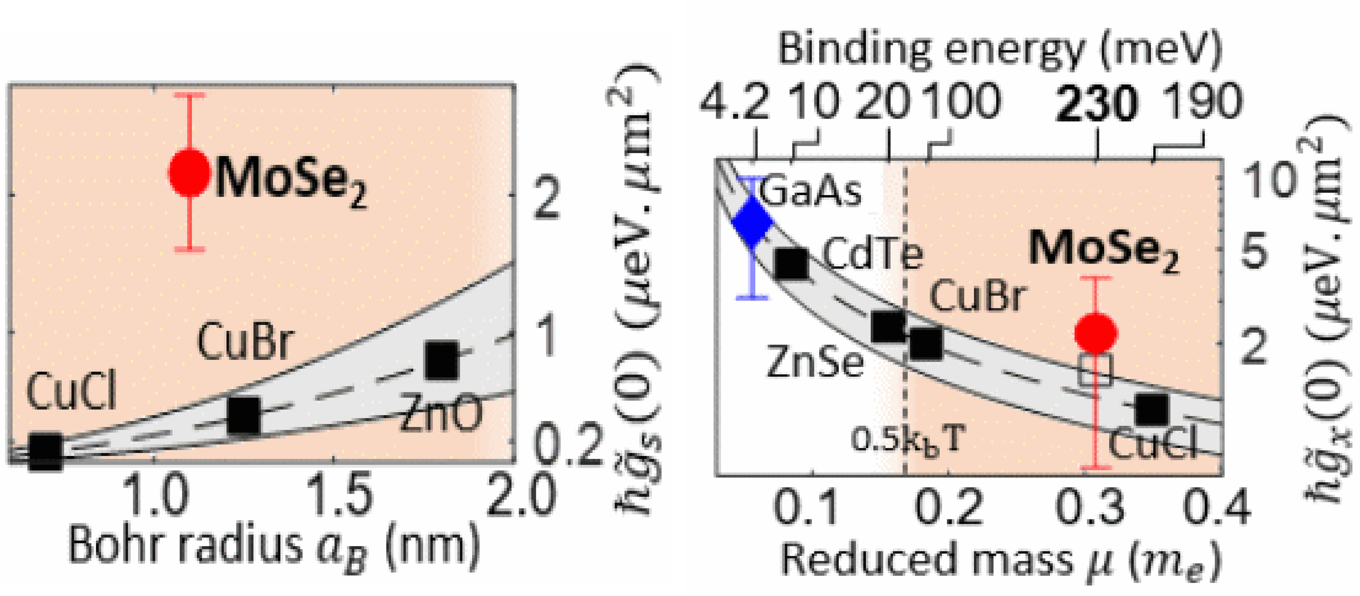
$A_{out} = \sqrt{2\gamma_{out}}\,\psi_c(t,\mathbf{r})$

Transmission spectrum is calculated by

$$T(\omega,\mathbf{r}) = |A_{out}(\omega,\mathbf{r})|^2/|A_{in}(\omega,\mathbf{r})|^2$$

Results

Our measured $\tilde{g}_x$ found to moderately exceed Hydrogenic Quantum Well Exciton's theory (gray area) while $\tilde{g}_s$ exceeds Hydrogenic Excitons's theory by a large factor of 7±2.



Conclusions

We have shown that a $MoSe_2$ monolayer in the strong coupling regime displays enhanced exciton-mediated optical nonlinearity as compared to comparable Quantum Well excitons, in particular via the excitonic saturation mechanism. Our results demonstrate that non-hydrogenic exciton in $MoSe_2$, offer new perspectives for the engineering of exciton-mediated optical nonlinearities.

Acknowledgements

European Union Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 754303.

CONTACT PERSON

Email:
amit.vashisht@lpmmc.cnrs.fr

REFERENCES

[1] Stepanov et al, Arxiv 2007.00431 (Accepted in PRL)

Clustering & Global Challenges

April 07-09, 2021

Online international conference