

This is the accepted manuscript made available via CHORUS. The article has been published as:

Magneto-optical measurements of the negatively charged s exciton in WSe_2

J. C. Sell, J. R. Vannucci, D. G. Suárez-Forero, B. Cao, D. W. Session, H.-J. Chuang, K. M. McCreary, M. R. Rosenberger, B. T. Jonker, S. Mittal, and M. Hafezi

Phys. Rev. B **106**, L081409 — Published 23 August 2022

DOI: [10.1103/PhysRevB.106.L081409](https://doi.org/10.1103/PhysRevB.106.L081409)

Magneto-Optical Measurements of the Negatively Charged $2s$ Exciton in WSe_2

J.C. Sell*,¹ J.R. Vannucci*,¹ D.G. Suárez-Forero,¹ B. Cao,¹ D.W. Session,¹ H.-J. Chuang,²
K.M. McCreary,³ M.R. Rosenberger,⁴ B.T. Jonker,³ S. Mittal,⁵ and M. Hafezi^{1,6}

¹*Joint Quantum Institute, NIST and University of Maryland, College Park, MD, 20742, USA**

²*Nova Research, Inc., Washington, DC 20375, USA[†]*

³*Naval Research Laboratory, Washington, DC 20375, USA*

⁴*Department of Aerospace and Mechanical Engineering,
University of Notre Dame, Notre Dame, IN 46556, USA*

⁵*Department of Electrical and Computer Engineering,
Northeastern University, Boston, MA 02115, USA*

⁶*Department of Electrical Engineering and Institute for Research in Electronics and Applied Physics,
University of Maryland, College Park, MD 20742, USA*

Monolayer transition metal dichalcogenides host a variety of optically excited quasiparticles species that stem from two-dimensional confinement combined with relatively large carrier effective masses and reduced dielectric screening. The magnetic response of these quasiparticles gives information on their spin and valley configurations, nuanced carrier interactions, and insight into the underlying band structure. Recently, there have been several reports of $2s/3s$ charged excitons in TMDs, but very little is still known about their response to external magnetic fields. Using photoluminescence excitation spectroscopy, we observe the presence of the $2s$ charged exciton and report for the first time its response to an applied magnetic field. We benchmark this response against the neutral exciton and find that both the $2s$ neutral and charged excitons exhibit similar behavior with g -factors of $g_{X_0^{2s}} = -5.20 \pm 0.11$ and $g_{X_{\pm}^{2s}} = -4.98 \pm 0.11$, respectively.

Monolayer semiconductor transition metal dichalcogenides (TMDs) have attracted significant attention in the last decade due to their unique optical properties. Similar to graphene, but with a three-layer (staggered) honeycomb lattice, TMDs host direct-gap transitions at their $\pm K$ valleys and exhibit circular-dichroism due to their finite Berry curvature [1–3]. The reduced dimensionality of materials in this system, coupled with techniques like hexagonal boron nitride (hBN) encapsulation, lead to enhanced Coulomb interaction and excitons with large binding energies ($E_B \approx 150 - 500$ meV) [4–6].

When there is excess charge present in the system during exciton formation, the exciton may lower its energy by capturing an electron or hole and form a bound, charged three-body state referred to as a charged exciton [7, 8]. Charged excitons are a ubiquitous feature of semiconductors, but are difficult to observe in traditional systems – like GaAs/AlGaAs quantum wells – due to their small binding energies (1-2 meV) [9–11]. In TMDs, however, both singlet and triplet charged species have been discovered with $E_B \approx 20 - 40$ meV [8, 12–15]. In the high carrier density regime, these resonances have been alternatively interpreted as many-body polaron states [16–19].

In analogy to the hydrogen atom, excitons are known to form a Rydberg series of higher energy states [20]. In TMDs, they have been observed through a variety of different optical techniques up to principal quantum number $n = 11$ [4, 5, 21–24]. However, even in the presence

of excess charge, a corresponding series for the charged exciton has remained elusive. Lack of experimental observations of these states has been thought of analogously to the H^- ion, for which there exists no bound excited state [25]. More recent theoretical work [26–29] has detailed scenarios in which these higher n charged excitons could exist, but that work does not match with a series of compelling experimental reports of metastable $2s/3s$ charged excitons in TMDs [30–33]. Significant work remains to reconcile experimental results with theoretical understanding.

The difficulty in observing these higher n states is two-fold: (I) the weak radiative decay rate of excitons with higher n makes them increasingly dim in typical photoluminescence (PL) measurements [34] and (II) even once the state is observed optically, further carrier-density and magnetic field dependent measurements are needed to correctly identify the exciton species. In particular, magneto-optical characterization of exciton species has proven to be an important tool for distinguishing between particle configurations [35, 36]. It also gives valuable information about the spin-valley character of each excitation [13, 14, 37–39], many-body interaction [16, 40, 41], and the underlying band structure of the materials themselves [42–46].

In this work, we confirm the presence of negatively charged $2s$ -exciton ($X_{\pm}^{2s}$) in WSe_2 via photoluminescence excitation measurements (PLE). In PLE, we monitor the emission from the $1s$ (lowest energy) exciton species while the excitation laser’s energy was swept in the energy regime needed to resonantly probe higher n states. This provides a superior signal-to-noise ratio compared to PL.

* These two authors contributed equally

[†] At the time of submission, H.-J.C. has transitioned to a position at the Naval Research Laboratory

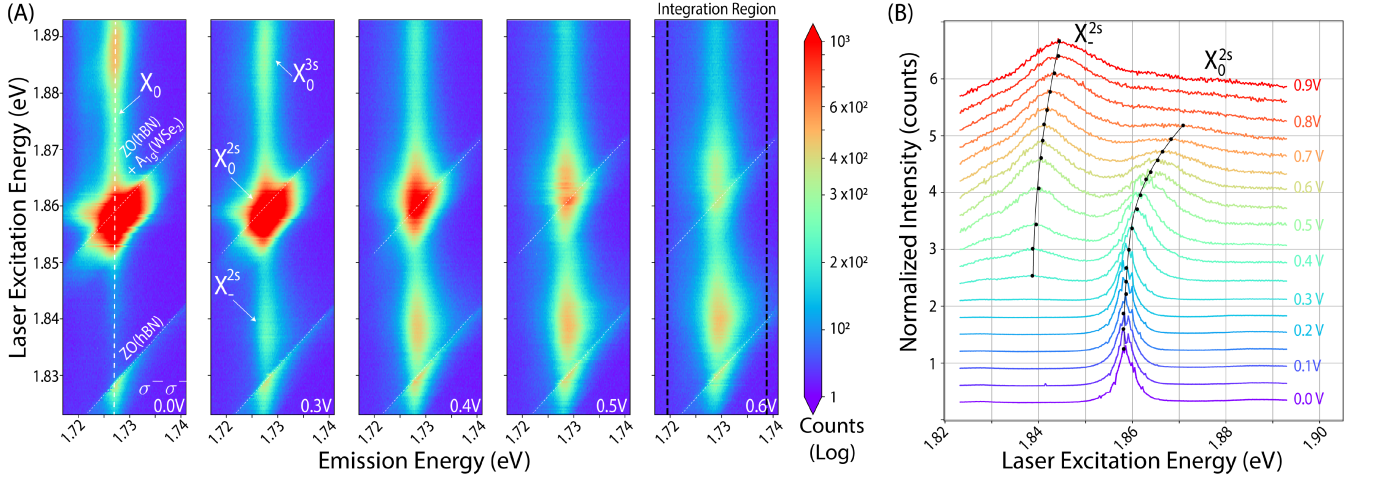


FIG. 2. (A) PLE data with increasing n_e -doping while monitoring the X_0 recombination channel in the $-K$ -valley ($\sigma^- \sigma^-$). (B) Waterfall plot of vertical cross-sections from $V_g = 0 - 0.9$ V. The integration region is annotated in panel (A). The counts were summed over the emission width for each excitation energy.

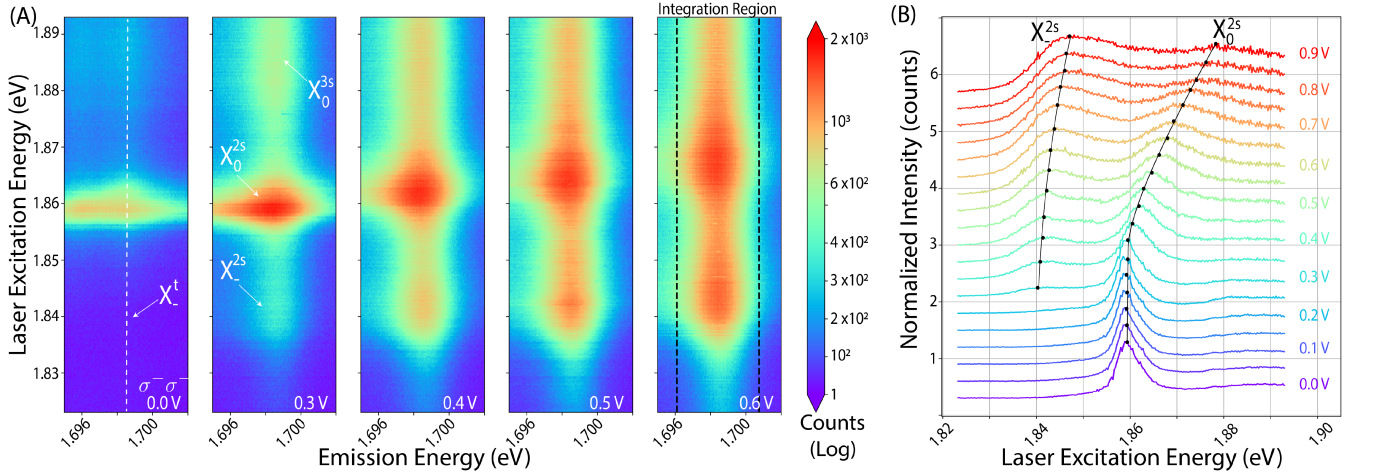


FIG. 3. (A) PLE data with increasing n_e -doping while monitoring the X_t^t recombination channel ($\sigma^- \sigma^-$). (B) As in Fig. 2, the waterfall plot corresponds to vertical cross-sections from $V_g = 0 - 0.9$ V.

gence of the negatively charged $1s$ excitons X_-^t/X_-^s (see Ref. [49] for $1s$ PL data). The X_-^{2s} resonance displays a similar onset at $V_g = 0.3$ V indicating a similar negative charge character. (II) When the X_-^{2s} resonance first appears at $V_g = 0.3$ V, we find that $\Delta E_{(X_0^{2s}-X_-^{2s})} = 21$ meV while $\Delta E_{(X_0-X_-^t)} = 29$ meV and $\Delta E_{(X_0-X_-^s)} = 35$ meV. This reduction indicates that the $2s$ charged exciton is less tightly bound than its $1s$ state counterpart. This is in accordance with other observations in the literature [30–33] and consistent with the fact that Rydberg states display a reduction in relative binding energy with each increasing n .

Since the $2s$ charged exciton is expected to be a doublet, as observed for the $1s$ charged excitons, the extracted position of X_-^{2s} is an average. X_-^t and X_-^s have a narrow linewidth and a strong intervalley exchange inter-

action that splits them (≈ 6 meV [15, 75]) which allows us to spectrally resolve them. However, the broadness of the $2s$ states combined with a reduced intervalley exchange energy (theoretically predicted to be ≈ 1 meV [32, 75]), prevents us from resolving the doublet of the $2s$ charged exciton. There is, however, indication of the two states in the asymmetric lineshape of the X_-^{2s} peak (see Ref. [49]).

In Fig. 2(B), we see the spectral dependence of X_0^{2s} and X_-^{2s} with carrier density. As the n_e -doping increases with increasing gate voltage, the X_0^{2s} resonance broadens, decreases in intensity, and spectrally blueshifts. The broadening and loss of spectral intensity are consistent with more rapid decoherence from interaction with the Fermi sea. The blueshift results from the competing effects of band gap and binding energy renormalization due to decreased $e^- - e^-$ and $e^- - h^+$ interaction from screen-

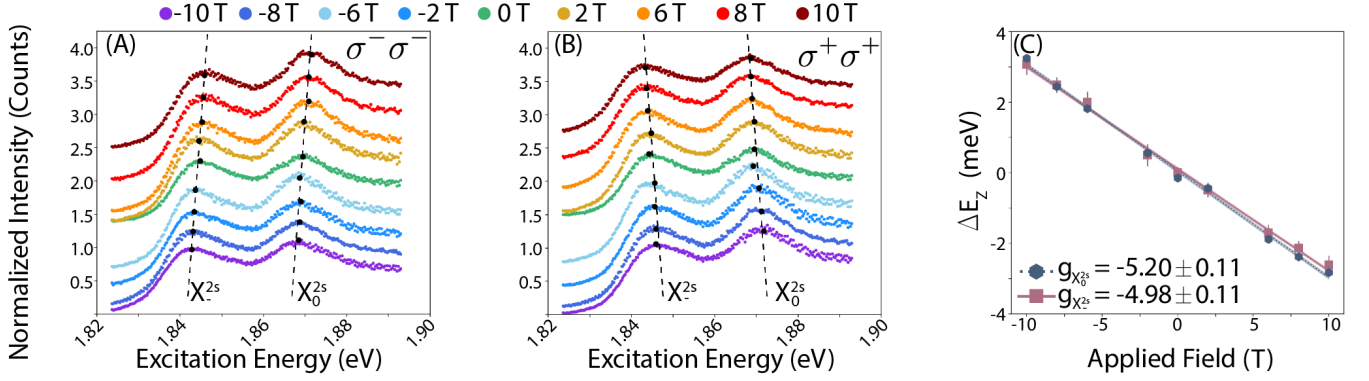


FIG. 4. Vertical cross-sections from the X^t emission channel as a function of field for (A) $(\sigma^- \sigma^-)$ and (B) $(\sigma^+ \sigma^+)$ marked with the corresponding peak positions (black dots) for the X_0^{2s} and X_{-}^{2s} states from fitting with the dashed line serving as a guide to the eye. (C) Extracted g -factors for X_0^{2s} and X_{-}^{2s} states. The thickness of the fit line in panel (C) corresponds to the error in the fit.

ing by the Fermi sea [33, 76, 77].

In contrast, X_{-}^{2s} peak grows in intensity and experiences minimal spectral drift with increased carrier density. In the case of a three-body quasiparticle, one expects a redshift that is linearly dependent on the charge concentration in the system resulting from momentum conservation [12, 19, 76]. This competes with the effects of band gap and binding energy renormalization previously discussed for the neutral excitons that favor a blueshift [76], and leads to the minimal spectral drift observed. Both the increase in intensity and small spectral shift are consistent with the behavior of $1s$ and $2s$ charged excitons previously observed [33, 76, 77].

Since X_{-}^{2s} emerges in the n_e -doped regime, we expect X^t and X_{-}^s to be the most prominent emission channels for $2s$ exciton species (see Ref. [49]). To verify this, we monitor the X^t emission channel in a similar manner to X_0 and show the results as a function of V_g in Fig. 3 (the results for X_{-}^s can be found in Ref. [49]). We confirm that the behavior (spectral position, shift with gate, etc.) of X_0^{2s} and X_{-}^{2s} is independent of the monitored decay channel.

We turn our attention to extracting the behavior of the X_0^{2s} and X_{-}^{2s} with applied magnetic field. We chose to take the data at $V_g = 0.6$ V because both the neutral and charged exciton have similar intensity. Integrated vertical cross-sections of the X^t emission channel presented in Fig. 4 (A)/(B) show the response of the $-K(\sigma^- \sigma^-)/+K(\sigma^+ \sigma^+)$ valleys, respectively, with magnetic field. The extracted peak centers from fitting are marked with black dots. Applying a magnetic field breaks the time-reversal symmetry in the system, and results in a red(blue) shift with positive field for the $+K(-K)$ valley and vice versa with applied negative field [13, 16].

Using the definition for the Zeeman splitting in terms of polarization components, $\Delta E_Z = E^{\sigma^+ \sigma^+} - E^{\sigma^- \sigma^-} = g\mu_B B$, we fit a linear model to our data and extract a

g -factor of -5.20 ± 0.11 and -4.98 ± 0.11 for X_0^{2s} and X_{-}^{2s} , respectively. This fit and extracted difference is shown in Fig. 4(C). Results that agreed within experimental error were found for both X_0^{2s} and X_{-}^{2s} for a similar analysis of the X_{-}^s emission channel (see Ref. [49]).

Frequently, a single-particle model is used to interpret the g -factor for $1s$ excitons. In this model, the contributions to the Zeeman splitting are defined as $\Delta E_Z = -\vec{\mu} \cdot \vec{B}$. The magnetic moment $\vec{\mu}$ is composed of additive terms for the orbital and spin contributions (intracellular components μ_O, μ_S) along with a correction for the effects of the finite Berry curvature in the system (intercellular component μ_V) [38, 39, 44, 75, 78] in each relevant band. Within this interpretation, we expect $g_{X_0} \approx -4.4$ and $-11 \lesssim g_{X_{-}^{t/s}} \lesssim -4$ (depending on the method used to calculate μ_V , and whether the doublet is resolved [13, 14, 47]).

To serve as a reference point between the literature and our $2s$ results, we also extracted the g -factors for X_0 and $X_{-}^{t/s}$. These values are $g_{X_0} = -4.22 \pm 0.04$, $g_{X_{-}^t} = -4.12 \pm 0.04$, and $g_{X_{-}^s} = -3.86 \pm 0.05$ in our system at $V_g = 0.6$ V. They are consistent with the results from the single particle interpretation, but highlight a distinct increase in our $2s$ g -factors with respect to the corresponding $1s$ states. We discuss two possible contributions to this enhancement.

(I) Enhancement of the g -factor for the $2s$ neutral exciton has been observed in magnetic Rydberg measurements in both intrinsic and electrostatically neutral samples [21, 23, 24]. Since the observation in neutral samples rules out doping effects, the divergence from $g_{X_0} \approx -4.4$ has been attributed to enhanced intercellular contributions arising from the increased k -space localization of the wavefunctions with each subsequent n [24]. Extending this technique to charged excitons gives an intercellular component that *decreases* as the Bohr radius *increases*. This is compounded by an increased k -space

localization of the charged exciton (see Ref. [49]). While this model could explain the results for X_0^{2s} it would underestimate the g -factor for X_-^{2s} .

(II) A second possibility is the onset of many-body interaction (polaron picture) between the excitons and the emerging Fermi sea from electrostatic gating. Many-body interactions are expected to be very favorable in WSe_2 which has a Wigner-Seitz radius greater than 1 even at extremely high densities [41, 79]. The interaction strength will vary with the Fermi sea's population and the Bohr radius, and induce Fermi sea polarization. Carrier dependent enhancement of the g -factor in TMDs has been documented for many materials/quasiparticles, with the strength of enhancement dictated by the degree of the induced Fermi sea polarization [16, 37, 40, 41].

In the many-body picture, it has been observed that as doping levels are varied there is a convergence of the g -factor between competing quasiparticles (e.g. X_0 and X^-) in regions in which they coexist. In analogy to the Kondo effect, the impurity (exciton) is dressed with either an attractive or repulsive interaction with the Fermi sea. As carrier density increases, the state dressing will become more similar for all exciton species – regardless of the type of interaction – resulting in a convergence of the g -factors [40] for X_0 -like and X_- -like excitons. Such behavior is not expected to be limited to the $1s$ state excitons and can explain the convergence of our extracted values of g for the X_0^{2s} and X_-^{2s} within experimental error.

Our results serve as the first marker in mapping the behavior of the $2s$ charged state, X_-^{2s} , with magnetic field in TMDs. Experimental quantification of the g -factor serves as another physical benchmark for future theoretical models of stable $2s$ charged states. Additionally, the stability of the X_-^{2s} state offers a possible medium for studying the cross-over from exciton Rydberg physics to the quantum Hall regime for charged species at high magnetic fields. Recent work by Klein *et al.* used carrier density dependent g -factor measurements to demonstrate tunable many-body physics through all $1s$ exciton species in MoS_2 [40]. Our initial results indicate that it would be possible to produce this type of map for $2s$ species with access to higher magnetic fields and devices with larger dynamic carrier density range. This opens up a unique opportunity to study many-body interactions in higher energy exciton species that is generally limited in traditional semiconductor systems with smaller exciton binding – like GaAs quantum wells.

The authors acknowledge fruitful discussions with A. Friedman, T. O'Haver, G. Solomon, A. Srivastava, and E. Waks. The work at Maryland was supported by AFOSR FA95502010223, NSF PHY1820938, and NSF DMR-2019444, ARL W911NF1920181, ARO W911NF2010232, Simons and Minta Martin Foundations. J.C.S. acknowledges additional support from the ARCS Scholar program. The work done at NRL was supported by core programs funding.

-
- [1] K. Mak, K. McGill, J. Park, and P. McEuen, *Science* **344**, 1489 (2014).
 - [2] D. Xiao, G. Liu, W. Feng, X. Xu, and W. Yao, *Physical Review Letters* **108**, 196802 (2012).
 - [3] K. Mak, C. Lee, J. Hone, J. Shan, and T. Heinz, *Physical Review Letters* **105**, 136805 (2010).
 - [4] A. Stier, K. McCreary, B. Jonker, J. Kono, and S. Crooker, *Nature Communications* **7**, 10643 (2016).
 - [5] A. Chernikov, T. Berkelbach, H. Hill, A. Rigosi, Y. Li, O. Aslan, D. Reichman, M. Hybertsen, and T. Heinz, *Physical Review Letters* **113**, 076802 (2014).
 - [6] K. He, N. Kumar, L. Zhao, Z. Wang, K. Mak, H. Zhao, and J. Shan, *Physical Review Letters* **113**, 026903 (2014).
 - [7] M. Lampert, *Physical Review Letters* **1**, 450 (1958).
 - [8] J. Ross, S. Wu, H. Yu, N. Ghimire, A. Jones, G. Aivazian, J. Yan, D. Mandrus, D. Xiao, W. Yao, and X. Xu, *Nature Communications* **4**, 1474 (2013).
 - [9] K. Kheng, R. Cox, M. Y. d'Aubigné, F. Bassani, K. Saminadayar, and S. Tatarenko, *Physical review letters* **71**, 1752 (1993).
 - [10] G. Finkelstein, H. Shtrikman, and I. Bar-Joseph, *Physical Review B* **53**, R1709 (1996).
 - [11] V. Solovyev and I. Kukushkin, *Physical Review B* **79**, 233306 (2009).
 - [12] K. Mak, K. He, C. Lee, G. Lee, J. Hone, T. Heinz, and J. Shan, *Nature Materials* **12**, 207 (2013).
 - [13] A. Srivastava, M. Sidler, A. Allain, D. Lembke, A. Kis, and A. Imamoglu, *Nature Physics* **11**, 141 (2015).
 - [14] T. Lyons, S. Dufferwiel, M. Brooks, F. Withers, T. Taniguchi, K. Watanabe, K. Novoselov, G. Burkard, and A. Tartakovsky, *Nature Communications* **10**, 2330 (2019).
 - [15] E. Courtade, M. Semina, M. Manca, M. Glazov, C. Robert, F. Cadiz, G. Wang, T. Taniguchi, K. Watanabe, M. Pierre, W. Escoffier, E. Ivchenko, P. Renucci, X. Marie, T. Amand, and B. Urbaszek, *Physical Review B* **96**, 085302 (2017).
 - [16] P. Back, M. Sidler, O. Cotlet, A. Srivastava, N. Take-mura, M. Froner, and A. Imamoglu, *Physical Review Letters* **118**, 237404 (2017).
 - [17] M. Sidler, P. Back, O. Cotlet, A. Srivastava, T. Fink, M. Kroner, E. Demler, and A. Imamoglu, *Nature Physics* **13**, 255 (2017).
 - [18] M. Glazov, *Journal of Chemical Physics* **153**, 034703 (2020).
 - [19] D. Efimkin and A. MacDonald, *Physical Review B* **95**, 035417 (2017).
 - [20] T. Kazimierzczuk, D. Frölich, S. Scheel, H. Stolz, and M. Bayer, *Nature* **514**, 343 (2014).
 - [21] T. Wang, Z. Li, Y. Li, Z. Lu, S. Miao, Z. Lian, Y. Meng, M. Blei, T. Taniguchi, K. Watanabe, S. Tongay, D. Smirnov, C. Zhang, and S.-F. Shi, *Nano Letters* **20**, 7635 (2020).
 - [22] A. Stier, N. Wilson, K. Velizhanin, J. Kono, X. Xu, and S. Crooker, *Physical Review Letters* **120**, 057405 (2018).
 - [23] M. Goryca, J. Li, A. Stier, T. Taniguchi, K. Watanabe, E. Courtade, S. Shree, C. Robert, B. Urbaszek, X. Marie, and S. Crooker, *Nature Communications* **10**, 4172 (2019).
 - [24] S.-Y. Chen, Z. Lu, T. Goldstein, J. Tong, A. Chaves, J. Kunstmann, T. Cavalcante, L.S.R. in Woźniak, G. Seifert, D. Reichman, T. Taniguchi, K. Watanabe,

- D. Smirnov, and J. Yan, *Nano Letters* **19**, 2464 (2019).
- [25] R. N. Hill, *Physical Review Letters* **38**, 643 (1977).
- [26] C. Fey, P. Schmelcher, A. Imamoglu, and R. Schmidt, *Physical Review B* **101**, 195417 (2020).
- [27] J. Yan and K. Varga, *Physical Review B* **101**, 235435 (2020).
- [28] A. Rau, *Journal of Astrophysics and Astronomy* **17**, 113 (1996).
- [29] Shiau-S.Y., M. Combescot, and Y.-C. Chang, *Physical Review B* **86**, 115210 (2021).
- [30] T. Goldstein, Y.-C. Wu, S.-Y. Chen, T. Taniguchi, K. Watanabe, K. Varga, and J. Yan, *The Journal of Chemical Physics* **153**, 071101 (2020).
- [31] E. Liu, J. van Baren, Z. Lu, T. Taniguchi, K. Watanabe, D. Smirnov, Y.-C. Chang, and C. Lui, *Nature Communications* **12**, 6131 (2021).
- [32] A. Arora, T. Deilmann, T. Reichenauer, J. Kern, S. de Vasconcellos, M. Rohlfing, and Bratschitsch, *Physical Review Letters*, 167401 (2019).
- [33] K. Wagner, E. Wietek, J. Ziegler, M. Semina, T. Taniguchi, K. Watanabe, J. Zipfel, M. Glazov, and A. Chernikov, *Physical Review Letters* **125**, 267401 (2020).
- [34] H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, 4th ed. (World Scientific, 2004).
- [35] E. Liu, J. van Baren, T. Taniguchi, K. watanabe, Y.-C. Chang, and C. Lui, *Physical Review Research* **1**, 032007 (2019).
- [36] M. He, P. Rivera, D. Van Tuan, N. Wilson, M. Yang, T. Taniguchi, K. Watanabe, J. Yan, D. Mandrus, H. Yu, H. Dery, W. Yao, and X. Xu, *Nature Communications* **11**, 618 (2020).
- [37] Z. Li, T. Wang, C. Jin, Z. Lu, Z. Lian, Y. Meng, M. Blei, S. Gao, T. Taniguchi, K. Watanabe, T. Ren, S. Tongay, L. Yang, D. Smirnov, T. Cao, and S.-F. Shi, *Nature Communications* **10**, 2469 (2019).
- [38] D. MacNeill, C. Keikes, K. Mak, Z. Anderson, A. Kormányos, V. Zólyomi, J. Park, and D. Ralph, *Physical Review Letters* **2014**, 037401 (2015).
- [39] Y. Li, J. Ludwig, T. Low, A. Chernikov, X. Cui, G. Arefe, Y. Kim, A. van der Zande, A. Rigosi, H. Hill, S. Kim, J. Hone, Z. Li, S. Smirnov, and T. Heinz, *Physical Review Letters* **113**, 266804 (2014).
- [40] J. Klein, A. Hotger, M. Florian, A. Steinhoff, A. Delhomme, T. Taniguchi, K. Watanabe, F. Jahnke, A. Holleitner, M. Potemski, C. Faugeras, J. Finley, and A. Stier, *Physical Review Research* **3**, L022009 (2021).
- [41] Z. Wang, K. Mak, and J. Shan, *Physical Review Letters* **120**, 066402 (2018).
- [42] G. Plechinger, P. Nagler, A. Arora, A. Granados del Águila, M. Ballotín, T. Frank, P. Steinleitner, M. Gmitra, J. Fabian, P. Christianen, R. Bratschitsch, C. Schüller, and T. Korn, *Nano Letters* **16**, 7899 (2016).
- [43] T. Deilmann, P. Krüger, and M. Rohlfing, *Physical Review Letters* **124**, 226402 (2020).
- [44] J. Förste, N. Tepliakov, S. Kruchinin, J. Lindau, V. Funk, M. Förg, K. Watanabe, T. Taniguchi, A. Baimuratov, and A. Högele, *Nature Communications* **11**, 4539 (2020).
- [45] T. Woźniak, F. J. P.E., G. Seifert, A. Chaves, and J. Kunstmann, *Physical Review B* **101**, 235408 (2020).
- [46] C. Robert, H. Dery, L. Ren, D. Van Tuan, E. Courtade, M. Yang, B. Urbaszek, D. Largarde, K. Watanabe, T. Taniguchi, T. Amand, and X. Marie, *Physical Review Letters* **126**, 067403 (2021).
- [47] E. Liu, J. van Baren, Z. Lu, M. Altaïary, T. Taniguchi, K. Watanabe, D. Smirnov, and C. Lui, *Physical Review Letters* **123**, 027401 (2019).
- [48] M. Barbone, A. Montblanch, D. Kara, C. Palacios-Berraquero, A. Cadore, D. De Fazio, B. Pingault, E. Mostaani, H. Li, B. Chen, K. Watanabe, T. Taniguchi, S. Tongay, G. Wang, A. Ferrari, and M. Atature, *Nature Communications* **9**, 1 (2018).
- [49] See Supplemental Material at [URL will be inserted by publisher] for additional information on experimental setup, calibration measurements, supplemental measurements, and fitting.
- [50] C. Jin, J. Kim, J. Suh, Z. Shi, B. Chen, X. Fan, M. Kam, K. Watanabe, T. Taniguchi, S. Tongay, A. Zettl, J. Wu, and F. Wang, *Nature Physics* **13**, 127 (2017).
- [51] J. Jadczyk, J. Kutrowska-Girzycka, J. Schindler, J. Debuss, K. Watanabe, T. Taniguchi, C.-H. Ho, and L. Bryja, *Materials* **14**, 399 (2021).
- [52] M. Rosenberger, H.-J. Chuang, K. McCreary, A. Hanbicki, S. Sivaram, and B. Jonker, *ACS Applied Materials and Interfaces* **10**, 10379 (2018).
- [53] A. Steinhoff, M. Florian, A. Singh, K. Tran, M. Kolarczik, S. Helmrich, A. W. Achstein, U. Woggon, N. Owschmikow, F. Jahnke, and X. Li, *Nature Physics* **14**, 1199 (2018).
- [54] E. Liu, J. van Baren, C.-T. Liang, T. Taniguchi, K. Watanabe, N. Gabor, Y.-C. Chang, and C. Lui, *Physical Review Letters* **124**, 196802 (2020).
- [55] M. Molas, C. Faugeras, A. Slobodeniuk, K. Nogajewski, M. Bartos, D. Basko, and M. Potemski, *2D Materials* **4**, 021003 (2017).
- [56] X. Zhang, T. Cao, Z. Lu, Y.-C. Li, F. Zhang, Y. Wang, L. Zhiqiang, J. Hone, J. Robinson, D. Smirnov, S. Louie, and T. Heinz, *Nature Nanotechnology* **12**, 883 (2017).
- [57] A. Kormányos, G. Nurkard, M. Gmitra, J. Fabian, V. Zólyomi, N. Drummond, and V. Fal'ko, *2D Materials* **2**, 022001 (2015).
- [58] M. Manca, M. Glazov, C. Robert, F. Cadiz, T. Taniguchi, K. Watanabe, E. Courtade, T. Amand, P. Renucci, . Marie, G. Wang, and Urbaszek, *Nature Communications* **8**, 14927 (2017).
- [59] T. O'Haver, *A Pragmatic Introduction to Signal Processing*, july 2021 ed.
- [60] P. Dagupta, Y. Chen, C. Serrno, G. Guiochon, H. Liu, J. Fairchild, and R. Shalliker, *Analytical Chemistry* **82**, 10143 (2010).
- [61] M. Wahab, F. Gritti, T. O'Haver, G. Hellinghausen, and D. Armstrong, *Chromatographia* **82**, 211 (2019).
- [62] O. Ajayi, J. Ardelean, G. Shepard, J. Wang, A. Antony, T. Taniguchi, K. Watanabe, T. Heinz, S. Strauf, X.-Y. Zhu, and J. Hone, *2D Materials* **4**, 031011 (2017).
- [63] Y. Lin, X. Ling, L. Yu, S. Huang, A. Hsu, Y.-H. Lee, J. Kong, M. Dresselhaus, and T. Palacios, *Nano Letters* **14**, 5569 (2014).
- [64] X. Zhu, N. Monahan, Z. Gong, H. Zhu, K. Williams, and C. Nelson, *Journal of the American Chemical Society* **137**, 8313 (2015).
- [65] M. Newville, T. Stensitzki, D. B. Allen, and A. Ingargiola, *Astrophysics Data System* (2014).
- [66] K. Levenberg, *Quarterly of Applied Mathematics* **2**, 164 (1944).
- [67] C. Chow, H. Yu, A. Jones, J. Yan, D. Mandrus,

- T. Taniguchi, K. Watanabe, W. Yao, and X. Xu, Nano letters **17**, 1194 (2017).
- [68] J. Serrano, A. Bosak, R. Arenal, M. Krisch, K. Watanabe, T. Taniguchi, H. Kanda, A. Rubio, and L. Wirtz, Physical Review Letters **98**, 095503 (2007).
- [69] Z. Jin, X. Li, J. Mullen, and K. Kim, Physical Review B **90**, 045422 (2014).
- [70] G. Liu, W. Shan, Y. Yao, W. Yao, and D. Xiao, Physical Review B **88**, 085433 (2013).
- [71] C. Dean, A. Young, I. Meric, C. Lee, L. Wang, S. Sorgenfrei, K. Watanabe, T. Taniguchi, P. Kim, K. Shepard, and J. Hone, Nature Nanotechnology **5**, 722 (2010).
- [72] E. Liu, J. van Baren, T. Taniguchi, K. Watanabe, Y.-C. Chang, and C. H. Lui, Physical Review B **99**, 205420 (2019).
- [73] Z. Li, T. Wang, Z. Lu, M. Khatoniar, Z. Lian, Y. Meng, M. Blei, T. Taniguchi, K. Watanabe, S. McGill, S. Tongay, V. Menon, D. Smirnov, and S.-F. Shi, Nano Letters **19**, 6886 (2019).
- [74] Z. Wang, L. Zhao, K. Mak, and J. Shan, Nano letters **17**, 740 (2017).
- [75] H. Yu, G.-B. Liu, P. Gong, X. Xu, and W. Yao, Nature Communications **5**, 3876 (2014).
- [76] A. Chernikov, A. van der Zande, A. Hill, H.M. anf Rigosi, A. Velauthapillai, J. Hone, and T. Heinz, Physical Review Letters **115**, 126802 (2015).
- [77] J. Roch, N. Leisgang, G. Froelicher, P. Makk, K. Watanabe, T. Taniguchi, C. Schönenberger, and R. Warburton, Nano Letters , 1070 (2018).
- [78] M. Koperski, M. Molas, A. Arora, K. Nogajewski, M. Bartos, J. Wyzula, D. Vaclavkova, P. Kossacki, and M. Potemski, 2D Materials **6**, 015001 (2018).
- [79] S. Das Sarma, E. Hwang, and Q. Li, Physical Review B **80**, 121303 (2009).