Chemical Vapor Deposition of High-Optical-Quality Large-Area Monolayer Janus Transition Metal Dichalcogenides

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1. Introduction

The physical properties of monolayer (ML) transition metal dichalcogenides (TMD) can be tailored for specific applications by stacking them in heterostructures, alloying, or more recently by preparing ML Janus TMDs. Due to the difference in electronegativity of the top and bottom chalcogen layers, Janus TMDs possess an in-built electric dipole that breaks the out-of-plane plane mirror symmetry, leading to a manifold of novel physical phenomena such as, for example, strong Rashba splitting, piezoelectric, pyroelectric, novel excitonic, and valleytronic phenomena.

One-pot chemical vapor deposition (CVD) growth of large-area Janus SeMoS monolayers is reported, with the asymmetric top (Se) and bottom (S) chalcogen atomic planes with respect to the central transition metal (Mo) atoms. The formation of these 2D semiconductor monolayers takes place upon the thermodynamic-equilibrium-driven exchange of the bottom Se atoms of the initially grown MoSe$_2$ single crystals on gold foils with S atoms. The growth process is characterized by complementary experimental techniques including Raman and X-ray photoelectron spectroscopy, transmission electron microscopy, and the growth mechanisms are rationalized by first principle calculations. The remarkably high optical quality of the synthesized Janus monolayers is demonstrated by optical and magneto-optical measurements which reveal the strong exciton-phonon coupling and enable an exciton $g$-factor of $-3.3$. 

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However, the existing synthetic routes for ML Janus TMDs, which depend on the plasma induced replacement of the top chalcogen layer of a parent TMD, are rather complex and it remains challenging to obtain large-area samples with high quality.[4,11,12] Therefore, to realize the potential of Janus TMDs for science and applications, technologically relevant methods for their fabrication still need to be established.

Here, we present a scalable and highly reproducible one-pot synthesis of high quality ML Janus SeMoS via chemical vapor deposition (CVD). It is based on a thermodynamic equilibrium approach resulting in replacement of the bottom Se layer of ML MoSe₂ grown on Au foil with S atoms at \( \approx 700 \) °C. The Au substrate plays a crucial role in the formation of Janus SeMoS by acting as a substrate which promotes adsorption and dissociation of S clusters from the vapor phase. The formation of ML Janus SeMoS is demonstrated by angle-resolved X-ray photoelectron and Raman spectroscopy, and the growth mechanisms are confirmed by first principles calculations. Encapsulation in hBN reveals spectrally narrow excitonic emission linewidths below 20 meV at cryogenic temperatures. As a result of circularly polarized laser excitation, the free exciton peak shows strong circular polarization up to 25% that persists above liquid nitrogen temperatures while temperature dependent linewidth analysis indicates enhanced exciton–phonon coupling. In applied longitudinal magnetic fields, we obtain an exciton g-factor of \(-3.3\).

2. Results and Discussion

The synthesis method of ML Janus SeMoS is schematically illustrated in Figure 1A. There are two-steps in our one-pot synthesis.

![Figure 1. Synthesis and microscopic characterization of ML Janus SeMoS. A) Schematic illustration of the process for synthesizing ML Janus SeMoS. Initially, ML MoSe₂ is synthesized on Au foils by CVD, followed by a conversion process in which the MoSe₂ crystals were exposed to S vapor at \( 700 \) °C, replacing the bottom-layer Se atoms of MoSe₂, which is in contact with the Au surface, by S atoms to form the ML Janus SeMoS. The Au surface absorbs and catalytically dissociates the S clusters. The dissociated S atoms are mobile on the Au surface at high temperature which migrates underneath the ML MoSe₂ and exchanges with the Se atoms in the bottom layer. On the other hand, replacement of the top Se layer is not energetically favorable as the S clusters elastically bounced back after colliding with the top Se layer, rather than dissociating and reacting. B,C) Optical microscopy images of ML Janus SeMoS, as grown on Au foil (B) and transferred on to SiO₂/Si substrate (C). D) AFM topography image of transferred ML Janus SeMoS. The thickness of the ML Janus SeMoS is estimated as \( 0.8 \pm 0.2 \) nm from the height profile shown in the inset. E) Raman spectra recorded at room temperature using 532 nm excitation wavelength on pristine ML MoSe₂ and ML MoSe₂ exposed to S vapor at different temperatures. The optimum sulfurization temperature for obtaining Janus SeMoS is \( 700 \) °C.](https://www.advancedsciencenews.com/doi/10.1002/adma.202205226)
method. i) Our growth procedure begins with the CVD growth of ML MoSe₂ single crystals.[13] Subsequently, the sample is cooled down to room temperature (RT). ii) The conversion of the MoSe₂ into Janus SeMoS takes place upon the temperature increase and annealing at 700 °C in the presence of S vapor. The gold substrate plays an important role in the conversion, as it results in the dissociation of S molecules constituting the vapor phase[24] (Sn with n = 2, 3, 4, 6, 7, and 8) and chemisorption of the S atoms on the gold substrate.[15] Upon their diffusion underneath the MoSe₂ crystals, they replace the bottom Se layer thereby forming an ML Janus SeMoS. A detailed description of the experimental procedures and a schematic diagram of the CVD process is given in Figure S1 and Note S1, Supporting Information, respectively. A comparison of our synthetic approach with the previously reported methods is provided in Note S2, Supporting Information.

The above-described mechanism of ML Janus SeMoS synthesis can be understood by analyzing the energetics of the exchange processes in density-functional theory (DFT) calculations[16] (see Note S3 and Figures S2–S5, Supporting Information). We consider the adsorption and migration of S and Se atoms on the Au(111) surface as well as their interaction with the free-standing and supported MoSe₂ monolayer. Our calculations show that it is energetically favorable for S molecules to split upon adsorption on Au(111). The formed S adatoms are highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV). Note that it costs highly mobile at the temperatures used during the synthesis (0.4 eV).

In Figure 1B, an optical microscopy image of as-grown ML Janus SeMoS crystals is presented. A high density and homogeneous distribution of the triangular crystals can be clearly recognized. Figure 1C shows the Janus MLs after their transfer onto a SiO₂/Si wafer by electrochemical delamination transfer technique.[15] The atomic force microscopy (AFM) topography image of Janus SeMoS on SiO₂/Si is provided in Figure 1D. The AFM height profile (insert to Figure 1D) and the height distribution histogram (Figure S6, Supporting Information) demonstrate an average thickness of 0.8 ± 0.2 nm as expected for the ML Janus SeMoS.[4]

To better understand and optimize the conversion process, we performed a series of experiments at different sulfurization temperatures, ranging from 650 °C to 800 °C using the sulfuration vapor phase[14] (S₂). Figures S8–S11, Supporting Information, show the X-ray photoelectron spectroscopy (XPS) of as-grown samples on Au foils, Figure 2; Figures S11 and S12, Supporting Information. From the binding energies (BEs) and intensities of the respective XP Mo 3d, S 2p, and Se 3d peaks, the formation of ML Janus SeMoS is clearly confirmed (see Figure 2A; Note S7 and Table S1, Supporting Information). The XP spectra were measured at various photoelectron emission angles, θ, as illustrated in the inset of Figure 2B, taking advantage of the enhanced surface sensitivity for larger θ. For each element (Se, Mo, S, and Au), the relative intensity (RI) of the XP components were calculated as a function of θ using the equation

\[ RI = \frac{I(x, θ)}{I(x, 0)} \]

From the variation of the RI values for Se, Mo, and S, it is seen that the Se layer is on top, followed by Mo and S at the bottom. Thus, ARXPS confirms the exchange of the bottom Se layer in ML MoSe₂ with S atoms, in agreement with the energetics analysis from DFT. The atomic ratio of Se:Mo:S is estimated as 0.9 ± 0.2:1:0.1:1.6 ± 0.2 indicating an excess amount of S atoms chemisorbed on the Au substrate. After the transfer of the ML Janus SeMoS onto an SiO₂/Si substrate, the obtained ratio is 0.8 ± 0.2:1:0.1:1.3 ± 0.2, which corresponds to the nearly ideal stoichiometry and suggests a loss of the excess S upon the transfer.

The narrow full width at half maximum (FWHM) of the A₁ peak (4.5 ± 0.2 cm⁻¹) indicates a high crystalline quality of the formed ML Janus SeMoS, which is consistent with our low-temperature Raman data (see below).[19] As shown in Figure S7, Supporting Information, the sulfurization at 700 °C with time <10 min resulted in the incomplete conversion (see also Note S4, Supporting Information). With further increase of temperature (green and purple spectra), significant changes in Raman spectra are observed: diminishing peak intensity, peak broadening as well as appearing of additional peaks characteristic for MoS₂. This behavior corresponds to the formation of a random MoS₂[1−x]Seₓ alloy[17,20] as expected for replacement with S of both bottom and top Se layers; for temperatures ≥800 °C, the Raman spectra are typical of highly defective ML MoS₂.[21] A comparison of the Raman spectra for as-grown Janus SeMoS MLs acquired directly on Au and after the transfer on SiO₂/Si wafers reveals identical spectroscopic characteristics showing that the transfer process can be applied in a non-invasive manner, Figure S8A, Supporting Information. A Raman map recorded on the transferred Janus ML demonstrates a uniform intensity distribution, hence proving the material homogeneity, Figure S8B,C, Supporting Information. Note that besides the individual Janus ML crystals as shown in Figure 1, also millimeter-scale homogeneous Janus ML films were synthesized by tuning the growth conditions (see Figure S9 and Note S5, Supporting Information). Our attempt to convert ML MoSe₂ into ML Janus SeMoS on commonly used SiO₂/Si substrate under the identical experimental conditions as used on Au foils resulted only in the formation of MoS₂[1−x]Seₓ alloys (see Figure S10 and Note S6, Supporting Information). This observation further shows the crucial role of the Au surface for the successful conversion.

To analyze the chemical composition and asymmetric structure of synthesized ML Janus SeMoS, we performed angle-resolved X-ray photoelectron spectroscopy (ARXPS)[22] of as-grown samples on Au foils, Figure S2; Figures S11 and S12, Supporting Information. From the binding energies (BEs) and intensities of the respective XP Mo 3d, S 2p, and Se 3d peaks, the formation of ML Janus SeMoS is clearly confirmed (see Figure 2A; Note S7 and Table S1, Supporting Information). The XP spectra were measured at various photoelectron emission angles, θ, as illustrated in the inset of Figure 2B, taking advantage of the enhanced surface sensitivity for larger θ. For each element (Se, Mo, S, and Au), the relative intensity (RI) of the XP components were calculated as a function of θ using the equation

\[ RI = \frac{I(x, θ)}{I(x, 0)} \]

From the variation of the RI values for Se, Mo, and S, it is seen that the Se layer is on top, followed by Mo and S at the bottom. Thus, ARXPS confirms the exchange of the bottom Se layer in ML MoSe₂ with S atoms, in agreement with the energetics analysis from DFT. The atomic ratio of Se:Mo:S is estimated as 0.9 ± 0.2:1:0.1:1.6 ± 0.2 indicating an excess amount of S atoms chemisorbed on the Au substrate. After the transfer of the ML Janus SeMoS onto an SiO₂/Si substrate, the obtained ratio is 0.8 ± 0.2:1:0.1:1.3 ± 0.2, which corresponds to the nearly ideal stoichiometry and suggests a loss of the excess S upon the transfer.
Figure 2. Angle-resolved XPS and TEM investigation of ML Janus SeMoS. A) High-resolution Mo 3d, S 2p, and Se 3d XP spectra of as-grown ML Janus SeMoS on Au foil measured at an emission angle (θ) of 0° (normal emission, top) and 75° (bottom), respectively. The fitted components have been named in the figures and discussed in detail in Note S7, Supporting Information. For better representation, the intensities of the spectra were multiplied by the factors represented in the respective figures. 

B) Relative intensities (RIs) of the Janus SeMoS components represented by Mo 3d, S 2p, and Se 3d peaks as well as the substrate reference Au 4f peak calculated according to the formula written in the inset are plotted for certain emission angles. Thereby, \( I(x, \theta) \), \( I(x, 0°) \), \( I(Au, \theta) \), and \( I(Au, 0°) \) are the intensities of each element \( x \) at \( \theta \) and at normal emission as well as the intensities of Au at \( \theta \) and at normal emission. In addition in the inset, two schemes are shown to illustrate the set-up for ARXPS (θ = 0° and 60°), whereby \( d \) is the information depth.

C) Atomically resolved cross-sectional HAADF-STEM (Z-contrast) image of the interface of Janus structure on SiO\(_2\)/Si substrate. Two bright layers (Se and Mo) above a third layer (S) are visible on top of the native SiO\(_2\) layer and the crystalline Si. The arrows mark the three different layers. 

D) EDX elemental maps of S, Se, and Mo together with the corresponding HAADF reference image showing three layers. The composed RGB colormap and the integrated linescan of the EDX intensity shows the ordering structure of the Janus from bottom interface to the top surface as S/Mo/Se.
Transmission electron microscopy (TEM) together with spatially resolved energy-dispersive X-ray (EDX) spectroscopy were applied to further verify the Janus SeMoS structure. The sample grown on the Au substrate was transferred to a Si wafer with a native SiO2 layer acting as an atomically flat substrate for the cross-sectional investigation (see Figure S13, Supporting Information for the lamella preparation details). Figure 2C shows a high-angle annular dark-field (HAADF) scanning (S) TEM image of the SeMoS/SiO2/Si interfaces. The image is dominated by Z-contrast where the intensity roughly scales with the power of two with heavier elements showing much brighter contrast than light elements. Two brighter layers together with the layer of lower intensity (similar contrast as the atomic columns of the Si substrate) are visible indicating the presence of two layers of heavier elements (Se and Mo) and one lighter element (S) with a similar Z-number as Si. The SeMoS structure is visible as three continuous layers instead of atomic columns corresponding to each atomic layer because there is no epitaxial relationship with the SiO2/Si substrate. Figure S14, Supporting Information, shows an additional high resolution (HR) TEM image in the cross-sectional view. The three individual atomic layers of the Janus SeMoS structure are recognized; however, because of the different contrast mechanism of HRTEM (interference contrast); the S-, Mo-, and Se-layer appear only with very slight contrast differences. EDX elemental maps of the interface regions are shown in Figure 2D. Together with the corresponding HAADF reference map (top), three individual layers of S, Mo, and Se are clearly identified. The RGB colormap and the corresponding line profiles of the individual EDX signals show that the three layers are slightly separated from each other. The expected elemental ordering from substrate to the surface as S/ Mo/Se is obtained. The local quantification of the atomic content shows a ratio of S: Mo: Se of almost 1:1:1 (Figure S15, Supporting Information). To verify also the crystalline nature of the layer of lower intensity (similar contrast as the atomic column of the Si substrate) proving the high crystallinity of the lattice of a TMD monolayer, we have applied this approach to our ML Janus SeMoS as sketched in Figure A, we show power dependent PL spectra using an HeNe laser (λ = 633 nm), and in Figure 4B, we plot the integrated PL intensity Iph as a function of laser power P. The X peak intensity increases linearly with power, which is the signature of a free exciton peak, whereas for the L transition, the contributing states become saturated. We show in Figure 2B the photoluminescence (PL) from the ML Janus SeMoS in the temperature range from 5 K to RT, where the bottom (top) hBN layer is typically 50 nm (10 nm) thick. We show in Figure 2B the photoluminescence (PL) from the ML Janus SeMoS in the temperature range from 5 K to RT (see Note S1, Supporting Information for experimental details). At T = 5 K, the spectrum is dominated by a spectrally narrow peak (FWHM 18 meV) marked as X, visible up to RT. At low energy centered around 1.5 eV, we detect a spectrally broad transition marked L, only visible for temperatures up to 150 K.

In Figure 3C, we plot the emission energy of the X peak (exciton) as a function of temperature. Fitting the characteristic redshift of the band gap (see Note S8, Supporting Information) allows us to extract an average phonon energy \( \langle \hbar \omega \rangle = (38.9 \pm 0.7) \text{ meV} \) that we use in the fit of the transition linewidth as a function of temperature.

\[
\gamma = \gamma(0) + a \cdot T + (b/(e^{(\hbar \omega/k_B T)} - 1)), \text{ where, } \gamma(0) = (18.5 \pm 0.1) \text{ meV}
\]

is the linewidth of X at \( T = 0 \) K, \( a = (24 \pm 5) \mu \text{eV K}^{-1} \) is the linear broadening due to acoustic phonons and \( b = (124 \pm 2) \text{ meV} \) is the strength of the phonon coupling. The value of \( \beta \) found here is considerably larger than the values reported previously for MoSe2 and MoS2 monolayers. This value could be evidence of stronger exciton–phonon coupling for Janus layers as compared to TMD monolayers due to the intrinsic electric dipole.

Janus TMD MLs are expected to possess momentum dependent spin splitting and to obey chiral optical selection rules. In Figure 3B, we plot the PL emission both “and counter-” polarized with respect to the excitation laser polarization. We extract the circular polarization degree

\[
P_c = (I_{\sigma^+} - I_{\sigma^{-}})/(I_{\sigma^+} + I_{\sigma^{-}}) \]

of the X emission. For an excitation photon energy lying \( \approx 180 \text{ meV} \) above the X emission energy, we measure a high polarization of \( P_c = 25\% \) for temperatures up to 100 K (Figure 3D) as chiral selection rules hold for the direct transition at the non-equivalent K-points at the Brillouin zone edge.\[4,27]\] We find a drastic drop in \( P_c \) for temperatures \( T > 100 \text{ K} \) and at 200 K we find \( P_c \approx 0 \). For comparison, the PL emission from localized states L is unpolarized over the investigated temperature range in Figure 3B. It is striking that for the X peak, the sudden decrease in \( P_c \) occurs over the same temperature range that sees drastic changes in the peak position and linewidth plotted in Figure 3C (see Note S8, Supporting Information, for discussion).

A key feature for ML Janus TMDs is the valley Zeeman splitting of the two exciton states in magnetic fields,\[6,28\] which was so far not accessible in samples with larger optical transition linewidth. In Figure 3E, we observe a clear energy difference \( \Delta \) between the \( \sigma^+ \) and the \( \sigma^- \) polarized PL components (see Note S8, Supporting Information) of \( \approx 1.7 \text{ meV} \), at \( B = 9 \text{ T} \), which corresponds to an exciton Landé \( g \)-factor of \( \approx 3.3 \). Interestingly, this value lies in between the Landé \( g \)-factor of the A-exciton in MoS2, which is reported to be between \( -2 \) to \( -4 \), depending on background doping\[6\] and the A-exciton \( g \)-factor of MoSe2 of \( -4.28 \).\[29\] In Figure 3E, the PL intensity is higher from the lower Zeeman branch, which indicates a relaxation toward the energetically lower valley branch during the exciton lifetime.

In Figure 4A, we show power dependent PL spectra using an HeNe laser (\( \lambda = 633 \text{ nm} \)), and in Figure 4B, we plot the integrated PL intensity \( I_{\text{ph}} \) as a function of laser power \( P \). The X peak intensity increases linearly with power, which is the signature of a free exciton peak, whereas for the L transition, the contributing states become saturated. We show in Figure 2B the photoluminescence (PL) from the ML Janus SeMoS in the temperature range from 5 K to RT, where the bottom (top) hBN layer is typically 50 nm (10 nm) thick. We show in Figure 2B the photoluminescence (PL) from the ML Janus SeMoS in the temperature range from 5 K to RT (see Note S1, Supporting Information for experimental details). At \( T = 5 \text{ K} \), the spectrum is dominated by a spectrally narrow peak (FWHM 18 meV) marked as X, visible up to RT. At low energy centered around 1.5 eV, we detect a spectrally broad transition marked L, only visible for temperatures up to 150 K.

In Figure 3C, we plot the emission energy of the X peak (exciton) as a function of temperature. Fitting the characteristic redshift of the band gap (see Note S8, Supporting Information) allows us to extract an average phonon energy \( \langle \hbar \omega \rangle = (38.9 \pm 0.7) \text{ meV} \) that we use in the fit of the transition linewidth as a function of temperature.\[24\]

\[
\gamma = \gamma(0) + a \cdot T + (b/(e^{(\hbar \omega/k_B T)} - 1)), \text{ where, } \gamma(0) = (18.5 \pm 0.1) \text{ meV}
\]

is the linewidth of X at \( T = 0 \) K, \( a = (24 \pm 5) \mu \text{eV K}^{-1} \) is the linear broadening due to acoustic phonons and \( b = (124 \pm 2) \text{ meV} \) is the strength of the phonon coupling. The value of \( \beta \) found here is considerably larger than the values reported previously for MoSe2 and MoS2 monolayers. This value could be evidence of stronger exciton–phonon coupling for Janus layers as compared to TMD monolayers due to the intrinsic electric dipole.\[26\]
Our data shows the main Raman modes reported for Janus SeMoS\cite{18} in both co- and cross-polarized detection under linear excitation and several additional, yet to be identified features. We note that the Raman peaks at $\approx 155 \text{ cm}^{-1}$ and $\approx 175 \text{ cm}^{-1}$, linked to the presence of defects,\cite{18} are comparatively weak for our samples, which is an indication of high structural quality.

We also plot in Figure 4C, the measurements in a random alloy MoSSe monolayer (hBN encapsulated) measured under the same conditions. The Raman spectra of the random alloy and the ordered Janus layer are clearly different. For the alloy, we find the MoS$_2$-like doublet at 400 cm$^{-1}$ reported before in the literature.\cite{17,20} Interestingly, the average phonon energy $\langle \hbar \omega \rangle = (38.9 \pm 0.7) \text{ meV}$ (corresponds to $\approx 313 \text{ cm}^{-1}$) extracted in temperature dependent measurements in Figure 3C,D, falls within the range of the main phonon energies we find in Raman spectroscopy. For further information on the electronic structure, we present in Figure 4D,E, PL measurements as a function of excitation laser energy (PLE). The data reveals a strong resonance in absorption at around 1.965 eV. By comparing with reflectivity measurements (Figure 4A), we ascribe this resonance to the B-exciton of the Janus SeMoS monolayer. Our experiments reveal that the HeNe laser excitation (633 nm; 1.96 eV) is nearly resonant with the B-exciton at $T = 4 \text{K}$, which explains the comparatively high PL signal and the very rich structure in the Raman data as compared to 532 nm excitation. The PL data also shows underlying periodic features that are at a fixed energy with respect to the laser energy, likely linked to phonon assisted absorption and/or emission (Figure 4D, compare with Ref.\cite{31}). Please note that these periodic features are absent for the random alloy monolayer sample. We plot for comparison the PLE data for the alloy monolayer, for which we find a resonance.
at a very similar energy position but the PLE spectrum at higher energies (>1.97 eV) is very different compared to the Janus monolayer due to the different band structure.

3. Conclusion

We have developed a simple, reproducible one-pot CVD synthesis of large-area Janus SeMoS MLs on Au foils. Their high optical quality is confirmed by a spectrally narrow excitonic emission from the Janus SeMoS MLs encapsulated in hBN. The free exciton peak shows strong circular polarization up to 25% and we extract in applied longitudinal magnetic fields an exciton valley g-factor of ~3.3. Temperature dependent linewidth analysis indicates enhanced exciton–phonon coupling.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Z.G., I.P., and A.E.-R. contributed equally to this work. Z.G., A.G., and A.T. designed the synthesis of Janus TMDs. A.T. directed the research. Z.G. synthesized Janus TMDs and performed basic material characterizations. E.N. and Z.G. performed the Raman spectroscopy and analysis. J.P. and C.N. performed AR-XPS and analysis. J.B., M.M., R.L., and U.K. performed the TEM investigation and analyzed the results. F.D. and A.V.K. performed the DFT calculations and contributed to the interpretation of experimental results. I.P. and A.E.R. performed encapsulation, temperature-dependent optical spectroscopy experiments, and spectral analysis. B.U., C.R., P.W., and X.M. designed the low-temperature optical spectroscopy experiments. K.W. and T.T. provided the hBN. Z.G., A.G., I.P., B.U. and A.T. wrote the manuscript furnished with input from all co-authors.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

2D materials, exciton–phonon coupling, high optical quality, Janus transition metal dichalcogenides, monolayers

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