Multidimensional analysis of excitonic spectra of monolayers of tungsten disulphide: Towards computer vision of physically distinct spatial domains of 2D materials

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Abstract

Despite monolayers holding great promise for a broad range of applications, the research around 2D materials suggests that proliferation of the potential devices and their fulfillment of real-life demands are still far from realization. Experimentally obtainable samples commonly experience a wide range of perturbations (ripples and wrinkles, point and line defects, grain boundaries, strain field, doping, water intercalation, oxidation,
edge reconstructions) significantly deviating crystal structure from idealistic models. These perturbations, in general, can be entangled or occur in groups with each group forming a complex perturbation making the interpretations of observable physical properties and the disentanglement of simultaneously acting effects a highly non-trivial task even for an experienced researcher, and advanced characterisation methods are often desirable. Here we generalise statistical correlation analysis of excitonic spectra of monolayer WS$_2$, acquired by hyperspectral absorption and photoluminescence imaging, to a multidimensional case, and examine multidimensional correlations via unsupervised machine learning algorithms. We are able to distinguish between different sets of perturbations acting on the otherwise ideal crystal structure and reveal multiple heterogeneous regions with an unprecedented level of details. This approach can be applied to any multi-modal imaging data acquired from other 2D materials, and our study paves the way towards advanced, machine-aided, characterisation of monolayer matter.

Abbreviations

2D, PCA, TMdC, PL, DR, CVD, FWHM, SM, CE, SO

Keywords
doping, dendrite, charge, monolayer, spectroscopy, stokes, photoluminescence, differential reflectance, absorption, tungsten disulphide, two-dimensional, exciton, trion, correlation, statistical analysis, transition metal dichalcogenide, spectral median, pattern, machine learning, pca, principal component analysis, k-means, clustering
Introduction

Since exfoliation of a single layer of graphite (graphene) and confirmation of its extraordinary physical properties, a wave of efforts aiming at synthesizing other two-dimensional (2D) materials has naturally emerged. A broad spectrum of experimentally obtained ultra-thin materials covering metals, semimetals, semiconductors, insulators, topological insulators, superconductors, and ferromagnets has been already reported with many others having been theoretically predicted. This has opened an avenue to material engineering in the form of van der Waals heterostructures giving rise to novel potential devices such as single-molecule and DNA sensors, photodiodes, transistors, memory cells, batteries, magnetic field sensors, and spintronic logic gates.

Despite monolayers holding great promise for a broad range of applications, the research around 2D materials suggests that proliferation of the potential devices and their fulfillment of real-life demands are still far from realization. In contrast to theoretical descriptions of the physical properties of various 2D materials, experimentally obtainable samples commonly experience a wide range of perturbations significantly deviating crystal structure from idealistic models, affecting the performance of the devices. Amongst these perturbations are the presence of ripples and wrinkles, point and line defects, grain boundaries, strain field, doping, water intercalation, oxidation, and edge reconstructions. These perturbations, in general, can be entangled or occur in groups with each group forming a complex perturbation. This, in turn, makes the interpretations of observable physical properties and the disentanglement of simultaneously acting effects a highly non-trivial task even for an experienced researcher, and advanced characterisation methods are often desirable.

Due to the monolayer nature of 2D materials, their optical signatures are highly sensitive to fluctuations in the local environment. This sensitivity can be exploited in attempts to identify physically distinct regions distributed across monolayers using unsupervised machine learning algorithms applied to the data acquired from generally multi-modal imaging of 2D materials. Here we consider a semiconducting monolayer of tungsten disulphide ($\text{WS}_2$)
grown via chemical vapour deposition (CVD) on a sapphire substrate. Optical properties of WS$_2$ monolayers are dominated by excitonic effects manifested as intense signatures in their absorption and emission spectra. We, therefore, apply absorption and photoluminescence (PL) hyperspectral imaging to gather data on spatial variations of excitonic properties of the material. Having fully parameterised spatially-resolved excitonic spectra, we then construct a multi-dimensional parametric phase-space (hypercube) where a single data point represents the set of values corresponding to all parameters at a given spatial location on the monolayer sample. This allows us to apply principal component analysis (PCA) to project the multi-dimensional data-cloud onto such a 2D plane that preserves the maximum variance in the data. Finally, we use unsupervised K-means clustering of the data-points in the PCA-plane to recognise optically distinct domains across the monolayer sample. Similar unsupervised learning approach has been practised previously, for example, in astronomy to analyze spectra from celestial objects, cell biology to categorize cell phenotype and genomics for metabolite profiling of proteins. Unsupervised identification of spatially distributed physically distinct domains of 2D materials with the level of details achieved here has not been reported previously, and our study paves the way towards advanced, machine-aided, characterisation of monolayer matter.

**Results and discussion**

Typical absorption and emission spectra of WS$_2$ monolayers are shown in Figure 1a. Absorption spectra are approximated here by differential reflectance and feature two distinct peaks corresponding to spin-orbit split A- and B-exciton transitions occurring at K symmetry points in the first Brillouin zone. Red-shifted PL emission is evident as an asymmetric peak formed as a result of annihilation of excitons and trions. Figure 1b,c shows spatially-resolved peak absorption amplitude and wavelength corresponding to A-exciton transition revealing trigonal-symmetric patterns in their lateral variations. Similar trends are observed
in the spatial maps of PL emission (Figure 1d,e): absorption and emission are blue-shifted in the regions spanning from the center of the flake towards its apexes. This behaviour has been attributed previously to elevated $n$-doping levels in those areas\textsuperscript{37,57} whereas red-shifted absorption and emission peaks in the adjacent regions have been shown to result from larger strain field.\textsuperscript{37} While absorption and emission wavelength maps are somewhat similar, obvious differences are observed between the patterns formed by absorption peak amplitudes (Figure 1b) and emission peak intensities (Figure 1d). First, the edges of the triangular island can be clearly distinguished in the PL emission intensity map where the PL enhancement occurred as a result of combined effects of water intercalation progressing towards interior over time\textsuperscript{38} and oxidation.\textsuperscript{40,41} Second, three bright spots near the center of the flake can be clearly distinguished in the absorption amplitude map. These bright features are believed to represent multilayer WS\textsubscript{2} material formed at the nucleation centers of the monolayer since larger reflectance contrasts have been observed for TMdC multilayers.\textsuperscript{58} Third, the absorption amplitude map features dark stripes within the bright regions, which is not the case of other maps shown in Figure 1b–e. All these observed differences point at complementarity of absorption and PL measurements allowing for observations of nonzero correlations between various parameters.

Statistical correlation analysis has been already proven to be a powerful tool in the studies of optoelectronic properties of 2D materials.\textsuperscript{33,36,59–63} In [59,62], for example, the authors correlated spectral shifts of prominent Raman peaks in graphene and graphene/TMdC heterostructures to disentangle the effects of doping and strain. Another route to solve a similar problem for TMdC monolayers was suggested in our previous publication\textsuperscript{37} where statistical analysis of spatially distributed PL Stokes shifts was used. Correlation analyses also facilitated the recognition of physically distinct edges of triangular TMdC flakes\textsuperscript{60,63} as domains hosting large number of point defects. Hsu \textit{et al.}\textsuperscript{33} used correlation analysis to recognise the effects of strain on optoelectronic properties of various TMdC monolayers and identify the direct-/indirect-bandgap nature of their respective electronic band structures. All these
Figure 1: (a) Integrated absorption and PL emission spectra of WS$_2$ monolayer. (b,c) PL emission peak intensity and wavelength spatial patterns, and (d,e) DR peak amplitude and wavelength corresponding to A-exciton transition. The length of the scalebar in (b) corresponds to 10 µm.

results, however, were based on scatter plots between specifically chosen pairs of parameters missing out other possible correlations, and were not able to recognise the presence of any subtle variations in the data. Here, we generalise statistical correlation analysis to an $N$-dimensional case to acquire more insights into the optoelectronic heterogeneities commonly found in 2D materials.

To make the $N$-dimensional correlation analysis possible we fully parameterise absorption and emission spectra (Figure 2a) and use each of the parameters to represent a dimension of an $N$-dimensional parametric space (Figure 2b) with $N = 17$ in our case. This space, due to the limited ranges of values parameters can take, is represented by an $N$-dimensional hypercube ($N$-cube) encapsulating an $N$-dimensional data-cloud where each data-point $\vec{\gamma}$ is described by a set of $N$ values (coordinates), i.e. $\vec{\gamma} = \{\gamma_1, \gamma_2, ..., \gamma_N\}$. The spatial distributions of the values of four of the parameters, $\gamma_5, \gamma_6, \gamma_{12}, \gamma_{14}$ (see Table 1 in Supporting Information for meanings of the parameters $\gamma_i, i = \{1, 2, ..., 17\}$), are shown in Figure 1b–e, while other spatial distributions corresponding to the rest of the parameters are given in Supporting Information. A specific location (pink point in Figure 1b–e) on the monolayer
Figure 2: (a) Schematic diagram representing absorption (shaded in light blue) and PL emission (shaded in pink) spectra with PL spectrum decomposed into exciton (shaded in yellow) and trion (shaded in green) contributions. All parameters (total 17) used in multidimensional analysis are labeled in pink: PL I (peak) = PL peak intensity; PL I (int.) = PL integrated intensity; SS = Stokes shift; CE = trion charging energy; FWHM = PL full width at half maximum; SM = PL spectral median; PL $\lambda$ (peak) = PL peak wavelength; $\Delta$SM = the difference between the SM and PL $\lambda$ (peak); PL $\lambda$ (X) = exciton emission peak wavelength; PL $\lambda$ (T) = trion emission peak wavelength; PL I (X) = exciton emission peak intensity; PL I (T) = trion emission peak intensity; SO = effective spin-orbit splitting at K symmetry points; DR (A) = differential reflectance peak amplitude of A-exciton; DR (B) = differential reflectance peak amplitude of B-exciton; DR $\lambda$ (A) = differential reflectance peak wavelength of A-exciton; DR $\lambda$ (B) = differential reflectance peak wavelength of B-exciton. (b) Schematic diagram of a multidimensional data-cloud (blue object) within a multidimensional hypercube. Qualitatively different trends (shaded in grey) can be observed depending on the angle of view. Generic parameters $\gamma_1, \gamma_2, \gamma_3, ..., \gamma_N, N = 17$, form dimensions (axes) of the hypercube.

island can, therefore, be assigned a set of $N = 17$ numbers corresponding to the values of the 17 parameters chosen to describe the optical properties of the material.

The natural approach to visualization of the geometry of a multi-dimensional object (data-cloud) is to look at its projections onto 2D planes (Figure 2b). Amongst infinite number of possible planes and projecting angles, a particular case of orthogonal projections onto the sides of the $N$-cube is the simplest to realise. It is this particular case that was considered in the previously reported correlation analyses of optoelectronic properties of 2D materials where certain physical trends and clusters have been identified.
In the general case of multidimensional correlation analysis, investigating trends between physical observables in the multi-dimensional parametric space is equivalent to investigating the geometry of the multidimensional data-object.

We notice that, in the particular case of the WS\(_2\) monolayer shown in Figure 1b–e, due to the existence of the distinct edge as a domain optically different from heterogeneous interior (Figure 3a), a given spatial location within the interior of the monolayer flake can be shown to be a part of a loop (green line), with physical properties being always unique as one travels along the loop. In our analysis the notion of the “physical property” is represented by \(\vec{\gamma}\); therefore, for a proper choice of parameters \(\gamma_1, \gamma_2, \ldots, \gamma_N\) it is possible to obtain a torus-isomorphic object in the \(N\)-dimensional parametric space (Figure 3b). This torus-isomorphic object could be observed in one of the 2D projections provided the proper projecting angle and projection plane are known. This correspondence between the real-space pattern on the flake and a particular geometry of the data-cloud in the multidimensional space may signify that it is possible to classify naturally formed patterns by their representation in a parametric space.

Indeed, after having examined all \(C^2_{17} = 136\) 2D orthogonal projections of the 17-dimensional data we found 16 projections (see Supporting Information) where the density of the scattered data resembles a torus-shaped object (see Supporting Information for data density estimation) with obvious clusters present. Figure 3c shows one of these projections. To clarify the meanings of the observed clusters, we have preliminary separated them by the boundaries connecting those points of the contours that have high negative curvature (Figure 3d, see Supporting Information for estimation of curvatures), and mapped all data-points within each of the clusters back into the real space (Figure 3e). This tentative identification of clusters in the data-cloud allowed us to visualise optically distinct regions on the monolayer flake revealing heterogeneous areas along the distinct edge. Interestingly, it can be shown that one of the reasons contributing to the clarity of the observed torus is the stretching effect of an oblique projection (Figure 2b). Specifically, it has been shown pre-
Figure 3: (a,b) Schematic diagram demonstrating how a torus (b) can be obtained in the multi-dimensional parametric phase space for the case of WS\(_2\) monolayer characterised by the optical spectroscopy in the real space (a). (c) 2D orthogonal projection of the data onto the plane formed by the exciton PL peak intensity and PL \(\Delta SM\) featuring a “shadow” of a torus. The data-density is colorcoded, and the levels of contours are marked on the colorbar. (d,e) Initial mapping of the data represented in the phase space back into the real space. Four heterogeneous domains are identified: heterogeneous interior (red and yellow) and heterogeneous edge (green and purple).

In case this parameter is not considered as a separate dimension then it would correspond to the oblique projection of the data within the framework of a 16-dimensional hypercube (see Supporting Information for details).

Having achieved this clustering result, we realise that a possible fine structure within these clusters could be missed out, and, therefore, a more systematic approach is generally required. In addition, the optically distinct multilayer domains have not been revealed since PL emission from multilayers of TMdC is suppressed;\(^{43}\) moreover, these domains lie in the trigonal-symmetric regions of elevated \(n\)-doping where PL emission is already reduced. Therefore, another 2D projection combining information obtained from both PL emission and absorption measurements has to be chosen. One of the candidates for such a projection could be any orthogonal projection with one axis formed by a parameter derived from PL profile and another axis formed by a certain parameter derived from the absorption spectra (e.g. A-exciton absorption peak amplitude). However, there is no guarantee that the clusters

\(^{43}\)
in a chosen projection will be maximally separated from each other, and there may exist a "better" 2D projection which does not coincide with any of the coordinate isosurfaces. To find the unique solution to the problem of identifying such a 2D projection, we apply the principal component analysis (PCA) which has been proven to be an efficient method allowing to project the data orthogonally on such a 2D plane that preserves the maximum variance in the data amongst all other 2D planes in the multi-dimensional parametric space.

The PCA method has been described elsewhere (see also Supporting Information for details). Shortly, within the framework of the PCA, a new hypercube with axes called principal components (PC1, PC2, ..., PC17) is identified in such a way that the variances \( \Delta_i, i = \{1, ..., 17\} \) of the data along the principal components PC\( i \) are non-increasing, i.e. \( \Delta_1 \geq \Delta_2 \geq ... \geq \Delta_{17} \). It is then possible to project the data into a hypercube of a lower dimensionality retaining the desirable variance present in the data-cloud. In the case of the hypercube defined by the parameters of absorption and emission spectra considered here, the PCA approach showed that it is possible to reduce the number of dimensions from 17 (defined by the parameters \( \gamma_1, ..., \gamma_{17} \)) down to 8 (defined by the parameters PC1, ..., PC8) and still preserve as much as 99.36% of the total variance (see Supporting Information).

To visualize and inspect the data within the new hypercube, we chose the plane spanned by the first two principal components (PC1 and PC2) and orthogonally projected the 17-dimensional data-cloud onto the chosen plane (Figure 4a), preserving 67.88% of the total variance (see Supporting Information).

It is worth noticing that the PC1-component is predominantly defined by PL-wavelength-related parameters \( (\gamma_{2,4,6,8}) \) which have spatial variations mostly in the angular direction (around the center of the flake). In contrast, the PC2-component is predominantly defined by the parameters CE \( (\gamma_{11}) \) and ΔSM \( (\gamma_9) \) which, in addition to angular variations, also exhibit variations in a radial direction (from the center of the flake). This hints at the potential possibility of defining such a local curvilinear reference frame (isomorphic to the polar reference frame) on the surface of the flake, in which four main regions (perturbations)
would be unambiguously separated. The existence of such a reference frame, in turn, could ultimately be predetermined by the growth history of the flake under consideration and various relaxation effects when monolayers are exposed to a "cold" environment after having been extracted from a CVD chamber. This idea is yet to be investigated in more details in our future research.

In order to acquire more insights into the projected data and reveal its fine-structure, we applied the K-means unsupervised learning algorithm as the method of inspection of the data cloud in the 2D PCA-plane. This algorithm, for a given input number $K$, tries to classify the data-set into $K$ labeled clusters (see Supporting Information for details). The value of $K$, however, cannot be automatically identified by the algorithm, and, therefore, cluster identification methods are commonly used. Here we used the so called “elbow” method as one of the most popular methods for identification of the natural number of clusters, if there are any (see Supporting Information). As expected, in the case of the WS$_2$ monolayer considered here, the “elbow” method revealed the presence of four prominent clusters in the data-cloud (see Supporting Information) corresponding to the two heterogeneous interior domains and two heterogeneous edge domains within the flake. However, the method also revealed that there are other natural cluster sets ($K = 2$, $K = 8$ and $K = 12$) present in the data, although they are not as prominent as the set of 4 clusters ($K = 4$). With increasing the number of clusters $K$, additional “shades” are introduced to the most natural cluster set $K = 4$ (see Supporting Information), and the case of $K = 12$ corresponds to the most sophisticated clustering identified in this work (Figure 4a). We note that we excluded multilayer regions (gray data-points in Figure 4a) from the K-means analysis by treating them as “anomalies” (or “outliers”) within an anomaly detection method: the multilayers were identified as data-points with the data-density below a certain threshold in the A- and B-exciton absorption amplitude-amplitude correlation plot (see Supporting Information).

Having identified multilayers via anomaly detection and the rest of the clusters via the K-means analysis, we mapped the data-points in the PCA-plane within each of the clusters back
Figure 4: (a) Phase space: 2D projection of 17-dimensional data-cloud onto the PCA-plane spanned by the parameters PC1 and PC2. Colored areas are the domains separated by the K-means clustering algorithm featuring 4 main clusters (yellow, green, red and purple) and 12 subclusters (shades of yellow, green, red and purple). Gray data-points correspond to multilayers identified by an anomaly detection method. A blue-colored cluster is centered around 0 in the PCA plane and corresponds to a “boundary” between the four main domains. (b) Real-space: all 13 clusters (including multilayers) are mapped back onto the WS$_2$ monolayer flake. Colorbar is labeled in accordance with the previously reported results. A question mark “?” in front of a label indicates an unconfirmed and tentative assignment. The size of the labels’ font symbolically represents the weight of the corresponding perturbation. (c) The PCA-plane with an added reference frame represented by linearly varying codes of an RGB palette: the coordinates PC1, PC2 $\in [-9,9]$ were mapped to an RGB-color $\text{RGB} = \{R,G,B\}$ where red component $R = (\text{PC1} + 9)/18$, green component $G = (\text{PC2} + 9)/18$, and blue component $B = 1 - (\text{PC1} + 9)/18$. (d) Real-space distribution of the RGB-colors defined in (c).
onto the monolayer flake (Figure 4b) revealing fine-structure of the four main clusters mentioned above. These four clusters are assigned here to the domains predominantly affected by the tensile strain field (yellow); elevated $n$-doping (red); combination of tensile strain field and intercalated water (purple), and combination of the elevated $n$-doping and intercalated water (purple). This assignment is based on the previously reported research \cite{36,37,69,70} where the bright regions have been recognised to be affected by the tensile strain \cite{69,70} and the dark regions to be affected by the elevated $n$-doping. \cite{36,37} The bright edges have been previously attributed to water intercalation for the case of WS$_2$ monolayers on sapphire. \cite{38} The net effect of water intercalation is to introduce hole-doping and to increase the number of radiatively recombining excitons, particularly, lowering the intrinsic $n$-doping level at the apexes and making those domains optically distinct. We note that water intercalation does not change appreciably the amount of strain along the edges as evidenced by the data-cloud projected onto the plane spanned by the PL peak wavelength and $\Delta$SM, where the overall trend is preserved despite the water-related splitting of the data-cloud (see Supporting Information). This signifies that on average the strain field vectors are aligned angularly around the center of the monolayer island so that the radially-propagating water intercalation does not release strain. Finally, the assignment of the domains of elevated absorption amplitudes to multilayers is based on the previous measurements of the dependence of the absorption spectra on the number of layers. \cite{58,71–74} We note the presence of the intermediate domain (blue) identified by the K-means learning algorithm, which is located in the phase-space in the middle of the torus (the torus’ “hole”) and connects all four major domains together. Figure 4a,b also reveals that the regions affected by strain (shades of yellow) are well aligned along the PC1-axis, whereas the regions affected by doping (shades of red) are distributed predominantly along the PC2-axis. This reflects that the first two principal components correlate well with these two physical properties and therefore can have physical meanings assigned to them.

Finally, we make a remark that there are not necessarily sharp boundaries between the
identified regions but rather these boundaries are generally smooth and blurred. Nevertheless, the gradients of parameters are larger across these boundaries than within the identified regions. This is evidenced by Figure 4c,d where PC-components were linearly color-coded (Figure 4c) and the colors were mapped back onto the flake (Figure 4d). This mapping also shows that within each of the clusters the physical properties also exhibit variations giving rise to non-zero correlations identified previously.\textsuperscript{37}

Conclusions

As a result of spatial variations of a complex local environment, 2D materials commonly exhibit spatially heterogeneous optoelectronic properties induced by a complex perturbation superimposed on the otherwise ideal crystal structure. These heterogeneities can be manifested as localized charge domains,\textsuperscript{36,37,59,62,75–78} distinct wide edges,\textsuperscript{38,60,63,79} structural/chemical heterogeneities\textsuperscript{80–82} or variations in strain field.\textsuperscript{33,59,62,83} In order to identify various physically distinct domains that are commonly observed for 2D materials, advanced characterisation methods are, therefore, required. A promising approach is to apply various combinations of the existing imaging methods and theoretical fitting models (see Supporting Information for some examples) aiming to extract multiple parameters describing physical properties of 2D materials. A multi-dimensional hypercube can then be formed from these parameters, and the corresponding multi-dimensional data-cloud can be examined. This is the generalisation of the statistical correlation analysis which is reduced to the examination of a general geometry of the data-cloud in the multi-dimensional parametric space. Machine learning algorithms can aid such an examination and, specifically, identification of clusters and trends. Here we used the combination of the PCA, K-means clustering and anomaly detection methods applied to the spatially-distributed absorption and emission spectra measured on a specific WS\textsubscript{2} monolayer flake on sapphire substrate grown via CVD. As a result, various labeled clusters of physically distinct domains have been identified and visualised,
with the meanings of each of the label assigned based on the previous research. Similar approach could be applied to a vast amount of 2D materials in order to gather large amount of data. This data then could be used to train supervised learning algorithms such as neural networks, which would be a powerful study resulting in a possibility of real-time identification of spatially-varying perturbation acting on the crystal structure of a given 2D material. Our clustering results suggest, in addition, that, similar to the 3D cube representing the P-V-T phase diagram of water, it may be, in principle, possible to find a set of parameters that would lead to a hypercube acting as a multi-dimensional phase space where various perturbations would be completely and unambiguously separated.

**Experimental**

**Sample preparation**

The sample preparation was performed in a similar way as described in Ref. \[23\].

**Experimental realization**

The PL and DR imaging setups were implemented in the same way as described in Ref. \[37\].

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**Supporting Information Available**

The following files are available free of charge.
A listing of the contents of each file supplied as Supporting Information should be included. For instructions on what should be included in the Supporting Information as well as how to prepare this material for publications, refer to the journal’s Instructions for Authors.

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Supporting Information
Multidimensional analysis of excitonic spectra of monolayers of tungsten disulphide:
Towards computer vision of physically distinct spatial domains of 2D materials

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S1. Hypercube’s dimensions

Table 1: List of parameters $\gamma_i, i = \{1, ..., 17\}$, used as dimensions for a hypercube. “X” stands for exciton; “T” stands for trion; “A” stands for A-exciton; “B” stands for B-exciton; “SM” stands for spectral median.

| Parameter | Meaning                          | Parameter | Meaning                          |
|-----------|----------------------------------|-----------|----------------------------------|
| $\gamma_1$ | PL peak intensity (X)$^a$        | $\gamma_{10}$ | PL FWHM                          |
| $\gamma_2$ | PL peak wavelength (X)$^a$      | $\gamma_{11}$ | Trion charging energy            |
| $\gamma_3$ | PL peak intensity (T)$^a$        | $\gamma_{12}$ | DR peak intensity (A)            |
| $\gamma_4$ | PL peak wavelength (T)$^a$      | $\gamma_{13}$ | DR peak wavelength (A)           |
| $\gamma_5$ | PL peak intensity               | $\gamma_{14}$ | DR peak intensity (B)            |
| $\gamma_6$ | PL peak wavelength              | $\gamma_{15}$ | DR peak wavelength (B)           |
| $\gamma_7$ | PL integrated intensity         | $\gamma_{16}$ | Effective spin-orbit splitting   |
| $\gamma_8$ | PL spectral median              | $\gamma_{17}$ | PL Stokes shift                  |
| $\gamma_9$ | PL $\Delta$SM                  |           |                                  |

$^a$ These parameters were derived from fittings;

Various parameters extracted from hyperspectral absorption and PL emission imaging can be regarded as dimensions of a parametric phase-space represented by a hypercube. In this work 17 different parameters listed in Table 2 were used to construct a 17-dimensional hypercube. Spatial maps corresponding to each of these dimensions are shown in Figure 1 (except for those given in Figure 1b–e in the main text). As seen from Figure 1, a certain spatial location $\vec{\gamma}$ (purple point) can be represented by a set of 17 numbers (i.e. $\vec{\gamma} = \{\gamma_1, \gamma_2, ..., \gamma_{17}\}$) and, therefore, by a single point in the constructed hypercube. It is possible that several spatial locations on the monolayer flake can be represented by a similar set of the chosen 17 parameters resulting in the points located close to each other in the parametric phase-space forming clusters.
Figure 1: Spatial maps corresponding to each of the dimensions of a multidimensional hypercube. A point $\vec{\gamma}$ (purple circle) on the monolayer flake can be represented by a set of 17 numbers, i.e. $\vec{\gamma} = \{\gamma_1, \gamma_2, \ldots, \gamma_{17}\}$. The length of the scalebar in (a) corresponds to 10 $\mu$m.
S2. Estimation of data densities

To obtain the densities of scattered data-points in each of the 2D projections of a multi-dimensional hypercube, a Gaussian smoothing was used. First, scattered data in a given 2D projection was converted into the $N \times N$ 2D histogram. This ensures that the width of a Gaussian filter will remain the same independent on the nature of the projection under consideration. In other words, each scatter plot was converted into an image $I(x, y)$ of $N \times N$ pixels, thus, standardizing the data analysis between all 2D projections. The value of $I$ corresponds to the number of scattered data-points within a bin. Second, a Gaussian smoothing filter

$$G(x, y) = e^{-\frac{x^2+y^2}{2\sigma^2}}$$

was applied to a $N \times N$ image $I(x, y)$, where $x, y = \{1, 2, ..., N\}$. The smoothing has been performed by the convolution of the image $I(x, y)$ with the Gaussian kernel $G(x, y)$ as follows

$$S(x, y) = \sum_{x'=1}^{N} \sum_{y'=1}^{N} G(x', y')I(x - x', y - y'),$$

where $S(x, y)$ is the smoothed image. The smoothed image was then normalized to the maximum of the data density.

Figure 2 demonstrates the effect of the Gaussian smoothing on the 2D projection formed by the parameters $\gamma_5$ (PL peak intensity) and $\gamma_9$ (PL $\Delta$SM) for different values of $\sigma$ and $N = 400$. In this work, the width of the Gaussian kernel was chosen $\sigma = 12$ unless otherwise specified.
Figure 2: Application of the Gaussian smoothing filter to the projection $(\gamma_5, \gamma_9)$ representing correlation between PL peak intensity and PL ΔSM. (a) Two-dimensional 400x400 histogram $I(x,y)$ of the scattered data with no filter applied. The origin of fringes is the limited spectral resolution of the spectrometer: the distance between the fringes corresponds to the pixels of the spectrometer’s detector. The colorbar calibrates the number of data-points within a bin. (b–i) Data density plots for increasing values of $\sigma$. The kernels $G(x,y)$ and their widths $\sigma$ are shown in insets. The colorbar in (b) is applicable to (c–i).
S3. 2D orthogonal projections featuring structures resembling a torus

Figure 3: 2D orthogonal projections resembling toroidal shapes of the data-density.
S4. Negative contour curvatures as signatures of boundaries between the clusters

Figure 4: (a) Contour plot of the data-density plot formed from the projection $(\gamma_9, \gamma_1)$. (b) An example of how boundaries between the clusters in the data-cloud could be identified by connecting the point of a high negative curvature.

The data-density plots introduced above could be considered as digital elevation models (DEM) of topographic landscapes\textsuperscript{1} (Figure 4a), featuring valleys and channel network. Contours of the data-density plots are closed loops with points having either a negative curvature (concave), a positive curvature (convex) or the zero curvature. The points of a high negative curvature could be a signature of a boundary between the clusters, and connecting such points together (Figure 4b) could render a boundary network. A robust automatic identification of channel network is an ongoing research\textsuperscript{1–3} which could be beneficial in the problems of identification of clusters in data-clouds.
S5. Charging energy as an oblique projection of the data in a hypercube of a lower dimensionality

Figure 5: Schematic diagram of a hypercube (green), a data-cloud (blue) and its orthogonal and oblique projections (gray shadows).

Considering the parameters $\gamma_2^{3D}$ (exciton emission energy), $\gamma_4^{3D}$ (trion emission energy) and $\gamma_1^{3D}$ (exciton emission intensity) of a 3-dimensional cube (or an $N$-dimensional hypercube), a linear superposition of $\gamma_2^{3D}$ and $\gamma_4^{3D}$ can be represented as an oblique projection on a 2-dimensional plane (or an $(N - 1)$-dimensional hypercube). This follows from the matrix representation of an oblique projection

$$
\begin{pmatrix}
\gamma_4^{2D} \\
\gamma_2^{2D} \\
\gamma_1^{2D}
\end{pmatrix} =
\begin{pmatrix}
0 & 0 & 0 \\
-\cot \alpha & 1 & 0 \\
-\cot \beta & 0 & 1
\end{pmatrix}
\begin{pmatrix}
\gamma_4^{3D} \\
\gamma_2^{3D} \\
\gamma_1^{3D}
\end{pmatrix},
$$

(3)

where $\alpha, \beta$ are the angles of obliqueness: $\alpha$ is the angle between the positive direction of $\gamma_2$-axis and the projection lines (dashed orange), projected onto the plane $(\gamma_4, \gamma_2)$ (Figure 5); $\beta$ is the angle between the positive direction of $\gamma_1$-axis and the projection lines, projected onto the plane $(\gamma_4, \gamma_1)$ (in Figure 5, $\beta = \pi/2 \pm \pi n, n \in \mathbb{Z}$); $\gamma_4^{3D}, \gamma_2^{3D}, \gamma_1^{3D}$ are coordinates
of the points of a 3D object, and $\gamma_2^{2D}, \gamma_1^{2D}$ are the coordinates of the corresponding points, projected onto the plane $(\gamma_2, \gamma_1)$. The oblique projection given in Eq. (3) can be reduced to the following two equations:

$$
\gamma_2^{2D} = \gamma_2^{3D} - \gamma_4^{3D} \cot \alpha,
$$

$$
\gamma_1^{2D} = \gamma_1^{3D} - \gamma_4^{3D} \cot \beta. \tag{4}
$$

If $\alpha = \beta = \pi/2 \pm \pi n, n \in Z$, then the oblique projection becomes degenerate and represents the orthogonal projection of the 3D object onto the plane $(\gamma_2, \gamma_1)$. One can notice that in cases when $\alpha = \pm \pi/4 \pm \pi n, n \in Z$, and $\beta = \pi/2 \pm \pi n, n \in Z$, Eqs. (4) further reduce to

$$
\gamma_2^{2D} = \gamma_2^{3D} \mp \gamma_4^{3D},
$$

$$
\gamma_1^{2D} = \gamma_1^{3D}. \tag{5}
$$

The first expression in Eqs. (5), in the case of “−” sign, is a definition of the trion charging energy $CE (\gamma_2^{2D})$ and therefore this parameter can be considered as a result of an oblique projection. Charging energy, therefore, adopts the resultant non-trivial stretching effect and makes trends and clusters easier to observe.
S6. Principal component analysis

Principle component analysis (PCA) has been described elsewhere.\textsuperscript{4–6} Shortly, the procedure was reduced to the following steps.

1. First, the $m \times N$ matrix $\tilde{X}$ representing $m$ data-points in an $N$-dimensional hypercube was normalised so that the normalised dataset $X$ had zero mean. To be more specific, each element $\tilde{x}_{i,j} \in \tilde{X}, i = \{1, \ldots, m\}, j = \{1, \ldots, N\}$ was transformed into the element $x_{i,j}$ as follows:

\[
x_{i,j} = \frac{\tilde{x}_{i,j} - \bar{\mu}_j}{\bar{\sigma}_j},
\]

where $\bar{\mu}_j$ and $\bar{\sigma}_j$ are the mean and the standard deviation of the data along the dimension $j$, respectively.

2. Second, the normalised dataset $X$ was presented as the product of three matrices via the singular value decomposition (SVD) method\textsuperscript{7} as

\[
X = USU,
\]

where $U$ is the $N \times N$ matrix of eigenvectors $u_i$ (principal components (PC)), and $S$ is the diagonal $N \times N$ matrix of eigenvalues $s_{ii}, i = \{1, \ldots, N\}$. The columns $u_i$ of the matrix $U$ define unit vectors of a new $N$-dimensional hypercube which is a rotated version of the initial hypercube, and, therefore, are linear combinations of unit-vectors $\gamma_i, i = \{1, \ldots, N\}$ (see Table 2–3 and Figure 7). The eigenvalues $s_{ii}$ describe the variance of the data along the axis $u_i$ (see Table 4).

3. Finally, the data $X$ has been projected onto the 2D plane defined by the first two principal components along which the variance of the data is the largest compared to that along the rest of the principle components (Figure 6):

\[
Z = XU_2,
\]
where $Z$ is the projection of the $N$-dimensional data $X$ onto the 2D plane spanned by the unit-vectors $u_1$ and $u_2$, and $U_2$ is $N \times 2$ matrix of these eigenvectors.

Figure 6: (a) Variances of the data along each of the principal components. 99.36% of the overall variance is retained in 8-dimensional PC-hypercube (green); 67.88% of the overall variance is retained in 2D PC-plane (red) used for visualization of data. (b) 2D histogram representing the data in the plane spanned by the first two principle components.

Figure 7: Graphical representation of the eigenvectors $u_i$ determined from PCA. (a) Each eigenvector is a linear combination of unit vectors $\gamma_j$ of the initial hypercube axes with coefficients $c_{i,j}$ (the weights of the parameters $\gamma_j$) color-coded in the shades of gray. (b) Squared values of parameter weights.

We note that we have not excluded multilayers from our PCA-analysis due to two reasons: (i) the number of corresponding data-points is much smaller compared to the overall number
Table 2: Eigenvectors \((u_1 - u_9)\) found by singular value decomposition of the 17-dimensional data-cloud. The numbers are coefficients \(c_{i,j}\) of a linear superposition \(u_i = \sum_{j=1}^{N} c_{i,j} \gamma_j\).

| \(\gamma_1\) | \(u_1\)  | \(u_2\)  | \(u_3\)  | \(u_4\)  | \(u_5\)  | \(u_6\)  | \(u_7\)  | \(u_8\)  | \(u_9\)  |
|----------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| -0.2941  | 0.2496 | -0.1963 | 0.0696 | -0.0005 | 0.2089 | -0.0188 | -0.0302 | -0.3029 |
| -0.3349  | -0.1978 | 0.1051 | -0.1026 | 0.0618 | -0.1032 | -0.0828 | 0.0871 | 0.0562 |
| -0.3139  | 0.2035 | -0.0875 | 0.0216 | 0.0582 | 0.3269 | 0.2815 | -0.0559 | 0.3383 |
| -0.3258  | 0.1491 | -0.0300 | -0.0477 | -0.0788 | -0.4028 | -0.4178 | -0.1040 | -0.0753 |
| -0.3077  | 0.2325 | -0.1703 | 0.0582 | 0.0182 | 0.2500 | 0.0575 | -0.0263 | -0.1430 |
| -0.3377  | -0.1888 | 0.1191 | -0.1068 | 0.0614 | -0.0971 | -0.0529 | 0.0476 | 0.0838 |
| -0.3028  | 0.2520 | -0.1547 | 0.0213 | 0.0258 | 0.1988 | 0.1063 | 0.0028 | -0.1404 |
| -0.3540  | -0.1358 | 0.1040 | -0.1147 | 0.0510 | -0.1356 | -0.0566 | 0.0754 | 0.2183 |
| 0.0872   | 0.4183 | -0.1544 | 0.0110 | -0.0954 | -0.1765 | 0.0067 | 0.1398 | 0.7552 |
| -0.0043  | 0.3129 | 0.1192 | -0.3680 | 0.0965 | -0.4881 | 0.6196 | 0.2037 | -0.2707 |
| 0.0450   | 0.4071 | -0.1616 | 0.0727 | -0.1643 | -0.3253 | -0.3677 | -0.2323 | -0.1508 |
| 0.0659   | -0.1400 | -0.6187 | 0.0111 | 0.3560 | -0.0786 | -0.1687 | 0.6345 | -0.0677 |
| 0.0111   | -0.2580 | -0.5042 | -0.1197 | 0.3326 | -0.2069 | 0.2131 | -0.6668 | 0.1043 |
| -0.3074  | -0.2369 | 0.0074 | 0.1731 | -0.1498 | -0.1839 | 0.0988 | 0.0666 | 0.0491 |
| -0.1777  | -0.2571 | -0.2126 | -0.5310 | -0.0078 | 0.0577 | 0.0631 | 0.0241 |
| -0.1730  | 0.0044 | 0.2524 | 0.5592 | 0.4269 | -0.2170 | 0.0573 | 0.0054 | 0.0302 |
| -0.0267  | 0.1275 | 0.2319 | -0.6038 | 0.4581 | 0.2040 | -0.3302 | -0.0494 | 0.0671 |

Table 3: Eigenvectors \((u_{10} - u_{17})\) determined from singular value decomposition of the 17-dimensional data-cloud. The numbers are coefficients \(c_{i,j}\) of a linear superposition \(u_i = \sum_{j=1}^{N} c_{i,j} \gamma_j\).

| \(\gamma_1\) | \(u_{10}\) | \(u_{11}\) | \(u_{12}\) | \(u_{13}\) | \(u_{14}\) | \(u_{15}\) | \(u_{16}\) | \(u_{17}\) |
|----------|-----|-----|-----|-----|-----|-----|-----|-----|
| 0.4008   | 0.2574 | 0.2928 | -0.5924 | -0.0684 | -0.0516 | -0.0155 | 0 |
| 0.1709   | -0.5604 | 0.2335 | 0.0154 | -0.1046 | -0.3906 | -0.4832 | 0 |
| -0.6812  | -0.0957 | 0.1699 | -0.2054 | -0.0194 | -0.0147 | -0.0090 | 0 |
| -0.1099  | -0.3835 | 0.1142 | -0.0944 | 0.0979 | 0.3582 | 0.4358 | 0 |
| 0.1496   | 0.0824 | 0.3334 | 0.7585 | 0.0894 | 0.0744 | 0.0495 | 0 |
| -0.0236  | 0.3006 | -0.1065 | 0.0598 | -0.2310 | -0.2185 | 0.2360 | -0.7326 |
| 0.1353   | -0.2676 | -0.8110 | 0.0252 | -0.0047 | -0.0103 | -0.0281 | 0 |
| 0.3834   | 0.3296 | -0.1141 | 0.0638 | -0.2452 | -0.2321 | 0.2510 | 0.6712 |
| 0.3816   | 0.0089 | 0.0126 | -0.0090 | 0.0415 | 0.0383 | -0.0396 | -0.1128 |
| -0.0372  | 0.0109 | 0.0688 | 0.0007 | 0.0006 | 0.0014 | 0.0008 | 0 |
| -0.3295  | 0.2715 | -0.0889 | 0.0957 | -0.0846 | -0.3179 | -0.3908 | 0 |
| -0.1559  | 0.0164 | 0.0026 | -0.0030 | 0.0011 | -0.0001 | -0.0008 | 0 |
| 0.1020   | -0.0089 | -0.0039 | 0.0029 | -0.0022 | 0.0003 | 0.0006 | 0 |
| -0.0177  | 0.2242 | -0.0680 | 0.0026 | -0.1752 | 0.6486 | -0.4898 | 0 |
| -0.0270  | 0.1370 | -0.0481 | -0.0497 | 0.6562 | -0.1409 | -0.0372 | 0 |
| 0.0090   | 0.1190 | -0.0446 | -0.0432 | 0.5709 | -0.1226 | -0.0324 | 0 |
| 0.0011   | 0.1688 | -0.0531 | -0.0236 | 0.2385 | 0.2255 | -0.2403 | 0 |

Table 4: Eigenvalues \(s_{ii}\) determined from singular value decomposition of the 17-dimensional data-cloud.

| \(s_{11}\) | \(s_{55}\) | \(s_{99}\) | \(s_{13,13}\) | \(s_{17,17}\) |
|--------|--------|--------|----------|----------|
| 6.8075 | 0.8670 | 0.0763 | 8\cdot10^{-5} |
| 4.7278 | 0.5968 | 0.0305 | 2\cdot10^{-5} |
| 1.8281 | 0.3550 | 0.0010 | 9\cdot10^{-6} |
| 1.5733 | 0.1307 | 0.0006 | 6\cdot10^{-6} |
of data-points, so that the presence of multilayer-related data-points influence eigenvectors and eigenvalues negligibly, and (ii) we wished to capture multilayers in the PC1-PC2 plane for the sake of generality.
S7. Identification of multilayers as anomalies (outliers)

The orthogonal projections of the data on the sides of the $N$-cube with the $y$-axis formed by either A- or B-exciton absorption peak amplitudes feature data-points lying outside of and above the main body of the data (high data-density regions). Two of such projections are shown in Figure 8. The data-points above the high data-density body correspond to the regions on the monolayer flake with large absorption amplitude and reflect the dependence of the absorption peak amplitudes on the number of layers reported previously.\textsuperscript{8–12} Therefore, we assign these domains and corresponding data-points to multilayer WS$_2$ material since the absorption peak amplitudes change discontinuously within those regions. It is worth noting that multilayer data-points in Figure 8 feature a wavelength-dependent behaviour having larger wavelengths in the bright interior regions and lower wavelength in the dark regions which is likely due to coupling effects between adjacent layers.

In this work, to identify the multilayer domains we considered A-B-exciton amplitude-amplitude correlation plot (Figure 9) since both amplitudes increase with the number of layers. Within unsupervised learning approach multilayers were considered as anomalies in a method similar to the anomaly detection methods described elsewhere.\textsuperscript{13} The core idea of an anomaly detection algorithm is to fit the data with a probability distribution function and label those data-points as anomalies that correspond to low values (below a specified threshold) of this function. Here, instead of fitting the data, we apply a Gaussian smoothing filter (Eq. (2)) with $\sigma = 12$ to the projection leaving the data-cloud featureless and resembling a multivariate Gaussian distribution. The result then was normalised and a threshold of 0.065 was chosen to detect “anomalous” data-points. This detection was performed in a directional way as shown in Figure 9: only those points were considered as anomalies which lie within the yellow-shaded area. This area was chosen to be bound by two semi-infinite lines starting at the maximum of the data-cloud with each line passing through the point of a negative curvature of the outermost contour.
Figure 8: 2D projections of a hypercube formed by (a) A- and (b) B-exciton absorption peak amplitudes and PL peak wavelength. The vertical gray line splits the data-points in two parts with one part (left) originating from dark regions on the monolayer flake and another part originating from the bright regions. Dashed pink lines are guidelines separating different multilayer clusters.

Figure 9: (a) 2D histogram showing correlation between A- and B-exciton absorption peak amplitudes. (b) 2D histogram after application of the Gaussian smoothing filter with $\sigma = 7$. The result was normalised, and the values below the threshold of 0.065 were colorcoded in pink. The yellow area corresponds to the area where multilayers were searched in the anomaly detection method. Two pink points correspond to the points of negative curvatures of the contours defining the silhouette of the data-cloud. Black lines in (a–b) represent diagonals.
S8. Determination of the number of clusters for K-means clustering

It is well-known that K-means algorithm requires the value of $K$ as an input defined by a user having some *a priori* knowledge/insight about the nature of the data-cloud, and it is not always straightforward to identify $K$. In this work, two methods have been considered in efforts to identify the number of clusters present in the PCA-projection of the data shown in Figure 6b.

(i) Elbow method

Elbow method is, perhaps, the most popular method for determination of optimal number of clusters in a K-means clustering method and was described elsewhere.\(^\text{14}\) Shortly, for an input number of clusters $K$, the K-means clustering algorithm iteratively tries to minimise the following objective function $J$:

$$J(C_1, ..., C_m, \mu_1, ..., \mu_K) = \sum_{i=1}^{m} \left[ (x_{i,1} - \mu_{C_i,1})^2 + (x_{i,2} - \mu_{C_i,2})^2 \right],$$  \hspace{1cm} (9)

where $K$ is the number of clusters; $C_i = \{1, ..., K\}$ is the index of the cluster in the PCA-plane to which the data-point $x_i = \{x_{i,1}, x_{i,2}\}$ belongs to; $\mu_k = \{\mu_{k,1}, \mu_{k,2}\}, k = \{1, ..., K\}$ is the point in the PCA-plane representing the centroid of the cluster $k$; $\mu_{C_i} = \{\mu_{C_i,1}, \mu_{C_i,2}\}$ is the point in the PCA-plane representing the centroid of the cluster $C_i$ to which the data-point $x_i$ has been assigned; $m$ is the number of data-points.

Elbow method is the method of inspection of the dependence $J(K)$ of the minimised objective function $J$ on the number of clusters $K$. If for a given dataset a natural set of clusters exists then the graph $J(K)$ will feature an obvious “elbow” (Figure 10a) formed by the change of slope of the function $J(K)$. In cases when there exist several natural sets of clusters then there may be several “elbows” present in the plot $J(K)$. In the case of WS$_2$ monolayer considered in this work, there are at least two elbows present in the plot $J(K)$,
as evidenced in the logarithmic plot of the function $J(K)$ shown in Figure 10b.

Each point in Figure 10a,b was calculated as follows. To make sure that $J$ corresponds to the global minimum (as opposed to a local minimum), for a given $K$, 5000 random initialisations of the positions of $K$ centroids were performed, and for each of these initialisations the function $J$ was calculated. Amongst all $J$-functions for the given $K$ the minimum value was found and plotted as a point in the graph $J(K)$. This procedure was repeated for all $K = \{1, \ldots, 25\}$.

To identify the optimal numbers of clusters $K$ more precisely and in an automatic manner we used the following procedure.

1. For each segment $[K, K + 1], K = \{1, \ldots, 24\}$, calculate the slope of the segment $[\log_{10}(J(K)), \log_{10}(J(K + 1))]$ (Figure 10c).

2. Calculate the elbow strengths as differences between the slopes at $K$ and $K - 1$ for all $K = \{2, \ldots, 24\}$. Assign the elbow strength to zero at $K = 1$ (Figure 10d).

3. Compute the squares of elbow strengths and find local maxima above a certain threshold ($0.0004$ in our case) in the resulting plot (Figure 11).

This procedure allowed us to identify 4 natural cluster sets (purple points in Figure 11) that can be used to fit the data-cloud in the PCA-plane using K-means clustering: these sets correspond to $K = 2, 4, 8, 12$ with the most natural set consisting of 4 clusters ($K = 4$).

(ii) Method of local maxima in the data density landscape

For a naturally-identifiable cluster set, it is also possible to find the value of $K$ by smoothing the scattered data with a Gaussian kernel of a certain width and finding the local maxima of the resultant landscape (Figure 12b,d,f,h). The procedure of finding the local maxima was the following. For a constant threshold of the data density ($0.1$ in this work) the width $\sigma$ of the Gaussian smoothing filter (Eqs. 1–2) was kept increasing by an increment of 0.1 until the desired number of local maxima was identified. Starting from $\sigma = 7.0$, the case of $K = 12$
occurred at $\sigma = 9.7$, the case of $K = 8$ occurred at $\sigma = 11.7$, the case of $K = 4$ occurred at $\sigma = 17$, and the case of $K = 2$ occurred at $\sigma = 33$. In Figure 12, the comparison between the positions of the centroids that minimise the objective function $J$ (Figure 12a,c,e,g) and the positions of local maxima of the data-density landscape (Figure 12b,d,f,h) is shown. Compared to the “elbow” method, the method of local maxima in the data-density plot allows to initialise centroids for a given $K$ in a straightforward and intuitive manner without performing multiple random centroid initialisations. In Figure 10a,b, red points correspond to the values of $J$ where centroids were initialised at these local maxima in the K-means calculations showing that the difference is of the order of $10^{-5} - 10^{-4}$. 
Figure 10: (a) Dependence $J(K)$ of the objective function $J$ on the number of centroids $K$. (b) Dependence $J(K)$ represented on a logarithmic scale. (c) Slopes of each of the line segments in (b) versus the number of centroids. (d) Elbow strengths versus the number of centroids $K$. In (a,b) red points correspond to the case where prior to K-means clustering centroids were initialised at the local maxima of the data-density plots (see the local maxima method below). The differences $\Delta J$ between the two objective functions evaluated for $K = 2, 4, 8, 12$ were found to be $0, 8.6 \cdot 10^{-5}, 8.7 \cdot 10^{-4}$ and $6.3 \cdot 10^{-4}$, respectively. In (b), three colored lines serve as guidelines showing different slopes of the function $\log_{10}(J(K))$. Purple points in (c–d) mark the number of clusters identified from calculations of elbow strengths; the red dim points correspond to the case where centroids were initialized at the local maxima of the data-density plot.
Figure 11: Dependence of the squared values of elbow strengths on the number of clusters. Purple points mark the local maxima at $K = 2, 4, 8, 12$ located above the threshold of 0.0004. The dependence of the squared “elbow” strength on the number of clusters initiated at the local maxima of the data-density plot is shown in dim red. This dependence was normalized to the maximum of the blue curve.
Figure 12: Comparison between the positions of centroids leading to the global minimum of the objective function $J$ (left column: a,c,e,g) and the initial positions of centroids identified from the local maxima method (right column: b,d,f,h). The width of the smoothing filters in the cases of $K = 2, 4, 8, 12$ were $\sigma = 33, 17, 11.7, 9.7$ for $K = 2, 4, 8, 12$, respectively.
S9. K-means algorithm and the results for $K = 2, 4, 8, 12$

K-means clustering algorithm has been described elsewhere.\textsuperscript{15–17} Simply, it consists of the following steps.

1. For an initialised set of centroids $\mu_k = \{\mu_{k,1}, \mu_{k,2}\}$, $k = \{1, \ldots, K\}$, split all data-points into $K$ different clusters in accordance with whether a data-point is closer to the centroid $\mu_1, \mu_2, \ldots, \mu_{K-1}$ or $\mu_K$.

2. Compute the averages between the data-points assigned to same clusters, and use these averages as new centroid positions.

3. Repeat the steps 1 and 2 until the centroids converge to a minimum of the objective function $J$ (Eq. (9)).

The number of iterations (step 3) in this work was chosen to be 30.

We excluded multilayer-related data points from the K-means clustering analysis, because in this case these data-points affect the result significantly.

Figure 13 shows the clustering results for $K = 2, 4, 8, 12$ in the cases when the function $J(K)$ is at its absolute minimum (Figure 13a,b,e,f,i,j,m,n) and when centroids were initialised at the local maxima of the data-density plot (Figure 13c,d,g,h,k,l,o,p). It is seen that for the case of $K = 2$, two main domains corresponding to the dark and bright regions on the monolayer flake have been clearly separated. A fine structure is introduced in the case of $K = 4$ clusters revealing the distinct and heterogeneous edge. In the case of $K = 8$, more “shades” of optoelectronic properties are introduced revealing a domain (blue) that lies between the four main domains in the phase and real spaces. Further refinement can be observed for the case of $K = 12$.

Both centroid initialisation methods described above produced identical clustering results for $K = 2$, and nearly identical results for $K = 4$. For $K = 8$, however, centroids initialised by the local maxima method (Figure 13k,l) did not lead to the identification of the central
blue domain as in the case of the absolutely minimised function $J(K)$ (Figure 13i,j); instead, a shade of green at the apexes has been introduced. In the case of the absolute minimum of $J(K)$, this shade is introduced only for $K = 12$ (Figure 13m,n). For $K = 12$, the differences between the two approaches of centroid initialisation is the most obvious (Figure 13m–p), however, qualitatively, the domains have been successfully identified in both cases.

Figure 13: K-means clustering results: a comparison between the two methods of cluster initialisation. Two columns (a–m) and (b–n) on the left correspond to the case of the initial cluster locations that correspond to the global minimum of the function $J$. The two columns (c–o) and (d–p) on the right shows the clustering results when centroids were initialised at the local maxima of the data density plots. The cases of (a–d) $K = 2$, (e–h) $K = 4$, (i–l) $K = 8$ and (m–p) $K = 12$ are shown.
S10. Evidence that water intercalation has not released strain

Figure 14: Orthogonal projection of the multi-dimensional data-cloud onto the plane spanned by the parameters $\gamma_6$ (PL peak wavelength) and $\gamma_9$ ($\Delta SM$). Two regions has been drawn approximately following trenches in the data-density landscape separating high- and low-density areas. (b) Mapping of the data-points back onto the monolayer flake demonstrates that water intercalation has not released intrinsic strain present in the crystal structure.
S11. Other approaches capable of introducing dimensions to a hypercube
Table 5: Other approaches capable of introducing dimensions to a hypercube.

| #  | Method                                      | Dimension(s)                                           | Refs. |
|----|---------------------------------------------|--------------------------------------------------------|-------|
| 1  | Raman imaging                              | Parameters of vibrational modes (intensity, frequency) | [18]  |
| 2  | Polarization-resolved PL<sup>a</sup>        | Degree of the circular polarization                     | [19]  |
| 3  | SNOM<sup>b</sup> PL<sup>a</sup>/absorption  | PL<sup>a</sup>/absorption spectral parameters          | [20–22] |
| 4  | Electric-field assisted SNOM<sup>b</sup> PL<sup>a</sup> | Bias voltage                                            | [23]  |
| 5  | Opto-valleytronic imaging                  | Valley polarization, valley coherence                   | [24]  |
| 6  | micro-PLE<sup>c</sup> spectroscopy         | PLE<sup>c</sup> spectral parameters                    | [25]  |
| 7  | PSHG/SHG<sup>d</sup> imaging and fitting models | Orientation of an armchair direction, crystal orientation, strain field parameters (amplitude and direction) | [26–28] |
| 8  | CARS<sup>e</sup> microscopy               | CARS<sup>e</sup> intensity                             | [29,30] |
| 9  | FWM<sup>f</sup> microscopy                 | Exciton radiative/dephasing lifetimes, degree of the circular polarization, doping level | [31]  |
| 10 | SF-2DES<sup>g</sup>                        | Characteristics of 0Q, 1Q, 2Q spectra (e.g. coupling strength between quantum states) | [32]  |
| 11 | Nanoscale ARPES<sup>i</sup>                | Parameters of electronic dispersion E(k)<sup>j</sup> (bandwidth, effective mass, band alignment) | [33,34] |
| 12 | KPFM<sup>k</sup>                           | Electronic surface potential                            | [34,35] |
| 13 | AFM<sup<l</sup>                            | Height above a substrate AFM<sup<l</sup> phase          | [35]  |
| 14 | Nanoscale XPS<sup>m</sup>                  | Defect density                                          | [34]  |
| 15 | Temperature-dependent trion fitting model  | Length of the trion spectral tail due to electron recoil effects | [36]  |
| 16 | TEM<sup>n</sup> and GPA<sup>o</sup>         | Strain field parameters (orientation and amplitude)     | [37]  |
| 17 | TR-PEEM<sup>p</sup>                        | Carrier decay time constants                            | [38]  |

<sup>a</sup> PL = Photoluminescence; <sup>b</sup> SNOM = Scanning near-field optical microscopy; <sup>c</sup> PLE = Photoluminescence excitation; <sup>d</sup> (P)SHG = (Polarization-resolved) second harmonic generation; <sup>e</sup> CARS = Coherent anti-Stokes Raman scattering; <sup>f</sup> FWM = Four-wave mixing; <sup>g</sup> SF-2DES = Spatially-resolved fluorescence-detected two-dimensional electronic spectroscopy; <sup>h</sup> 0Q = Zero-quantum; 1Q = One-quantum; 2Q = Two-quantum; <sup>i</sup> ARPES = Angle-resolved photoemission spectroscopy; <sup>j</sup> E is the binding energy; <sup>k</sup> k is the wavevector; <sup>l</sup> KPFM = Kelvin probe force microscopy; <sup>m</sup> AFM = Atomic force microscopy; <sup>n</sup> XPS = X-ray photoelectron spectroscopy; <sup>o</sup> TEM = Transmission electron microscopy; <sup>p</sup> GPA = Geometrical phase analysis; <sup>p</sup> TR-PEEM = Time-resolved photoemission electron microscopy.
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