Having a good vibe: Strong coupling to the electronic system creates incoherent and coherent phonons in 1L-TMDCs

Research data repository documentation

Tim Völzer

1 General Information

Dataset title:	Having a good vibe: Strong coupling to the electronic system creates incoherent and coherent phonons in 1L-TMDCs
Authors/Creators:	Tim Völzer, Marvin Krupp, Julian Schröer, Annika Bergmann-Iwe, Tobias Korn, and Stefan Lochbrunner
Affiliations:	Institute of Physics and Department "Life, Light and Matter", University of Rostock, Albert-Einstein-Str. 23-25, 18059 Rostock, Germany
E-Mail:	stefan.lochbrunner@uni-rostock.de
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2 Description

This document provides supplementary information for the data belonging to the publication [1].

3 Data Format

All files are provided in an ASCII-based ".dat" format and can thus be accessed by practically any software. They are built up as 2D arrays containing either integer or decimal floating point values using decimal points. The individual columns are separated either by tabs or commas. Rows are terminated via line breaks. The only exceptions to this structure are the files containing _globalFitParameters in their name. However, their structure is explained further below.

4 Archive Structure

Here, we describe the structure of the presented ".zip" archive as well as the interior structure and value assignment for the different types of files. Generally, the data either stems from Raman or transient absorption (TA) measurements, see the following subsections.

4.1 Raman spectra

Raman spectra were collected for all materials, using a silicon wafer reference each to calibrate the wavenumber axis, so that the silicon peak lies at $520.8\,\mathrm{cm}^{-1}$. The file names contain the sample material, the excitation power and wavelength, the integration time as well as a whether it is the silicon reference. The interior structure is the following:

- <material>_Raman_<power>_<wavel.>_<int.time>(_Si-Reference).dat: Three column table.

Part	**Quantity [unit]**	**Comment**
Header (Rows 1-3)		Describes which quantity is displayed in each column.
Column 1	Wavelength [nm]	As obtained from spectrometer.
Column 2	Wavenumber [cm^{-1}]	Relative to excitation laser line, calibrated to silicon Raman peak.
Column 3	Intensity [Counts]	As obtained from spectrometer.

4.2 Transient absorption spectroscopy

For the TA data, multiple processing and analysis steps exist, which will be described in detail in the following. The corresponding folder contains four subdirectories, one for each TMDC material plus one for the PR:PMMA sample used to quantify the time resolution in figure S2 of the supplementary material (SM) of [1]. The latter contains only a raw and a processes TA data file, which are explained in detail below. The former, in contrast, are again subdivided with each folder comprising data from one particular measurement. Hence, the characteristic parameters are given in the folder name, *i.e.* excitation fluence and wavelength as well as whether the time axis covers the whole range of 100 ps using a logarithmic spacing (`log`) or focuses on the first few picoseconds with a equidistant spacing (`lin`).

Folder name = `<fluence>_<exc.wavel.>_<time axis>`

Generally, all these files are named starting with the material and the applied fluence. While excitation at 530 nm is seen as the default settings, we explicitly note a pump wavelength of 595 nm if applied. Similarly, we define a logarithmic time axis as the standard and add `lin` for the equidistant spacing. Aforementioned elements are followed by `TA` and further terms (`<appendix>`) to specify the various stages of analysis conducted and information extracted.

`<material>_<fluence>(_595nm)(_lin)_TA<appendix>.dat`

In the following, the interior of each file type in dependence of the `<appendix>` as well as the link to other files and the occurrence in the figures of the publication are described.

- `<appendix>` = `_raw`:
  2D array as obtained from measurement program, containing the unprocessed, raw TA data.

Part	**Quantity [unit]**	**Comment**
Row 1	Probe delay [ps]	As obtained from the position of the delay stage in the TA setup.
Rest	$\frac{T^*}{T_0}$	Ratio of transmitted probe intensity with versus without preceding pump pulse.

  Note that this file contains multiple scans of the time axis simply appended to one another. Hence, the first row exhibits a repetitive pattern of time points, as does the rest of data.

- `<appendix>` contains `_cleaned`:
  In some measurements, a few single time points are substantially offset in relation to the signal baseline. In these cases, we modified the "`raw`" file by deleting those entries, creating the corresponding "`_cleaned`" file. The latter was then used for all further analysis and figures.

- `<appendix>` is empty (or consists only of `_cleaned`, see above):
  Processed TA data. Obtained from the above by averaging multiple scans, correcting for the probe chirp (see [1], SM, section 3.4), minor baseline deviations and appending the wavelength axis. Finally, the TA signal was converted to $\Delta A = -\log_{10} \frac{T^*}{T_0}$.

Part	Quantity [unit]	Comment
Column 1	Probe delay [ps]	Chirp-corrected and unified for all individual scans.
Row 1	Wavelength [nm]	As obtained from wavelength calibration, but cut to display only the region with a decent signal-to-noise ratio.
Rest	ΔA	Change in absorbance corrected in aforementioned fashion.

This data or parts of it are shown in figures 1, 3 (a), and 5 (b) of the main body as well as figures S3, S4, and S6 of the SM [1]. On top, it forms the basis for fitting the general TA dynamics.

- `<appendix>` = `_globalFit`:
  Fit to the processed TA data. Obtained from the latter by global fitting, as described in [1], SM, section 2.3.

Part	Quantity [unit]	Comment
Row 1	Probe delay [ps]	As in processed TA data, but cut before 0.2 ps.
Column 1	Wavelength [nm]	As obtained from processed (cleaned) TA data.
Rest	ΔA_{fit}	Fitted change in absorbance.

Parts of this data are shown in figure S6 of the SM [1].

- `<appendix>` = `_globalFitParameters`:
  Fit parameters to the processed TA data. Obtained from the latter by global fitting, as described in [1], SM, section 2.3. The structure deviates from the usual one, consisting of rows that each state a parameter name as well as the corresponding numeric value. The number of used parameters depends on the material, as we used only three instead of four exponentials in the case of WSe_2. All parameters that are not mentioned in the following are not included in the fit and thus set to zero.

Part	Quantity [unit]	Comment
`sigma`	σ [a u]	Fit deviation from data.
`fwhmCC`	FWHM time resolution [ps]	Set to 0.04.
`t0`	t_0 [ps]	Time zero, set to 0.
`A_i` ($i = 1-4$ (1-3))	Amplitude scaling factor	Equals 1 to indicate that this part of the decay function is used in fitting.
Next entry after `A_i`	τ_i [ps]	Decay time constant (regardless of name in the file)
`O1_A`	Amplitude scaling factor for oscillation	Equals 1 to indicate that this part of the decay function is used in fitting.
`O1_tau`	τ_{osc} [ps]	Decay time constant for oscillation.
`O1_wn`	$\tilde{\nu}_{\text{osc}} = \frac{f_{\text{osc}}}{c}$ [cm^{-1}]	Wavenumber for oscillation.
`O1_phi`	α_{osc} [rad]	Phase shift for oscillation.

Parts of this data are shown in figures 2 of the main body and figures S7-8 of the SM [1].

- `<appendix>` = `_globalFitDAS`:
Decay-associated amplitude spectra (DAS) to the processed TA data. Obtained from the latter by global fitting, as described in [1], SM, section 2.3. The exact number of columns depends on the material, as we used only three instead of four exponentials in the case of WSe_2.

Part	Quantity [unit]	Comment
Header (Rows 1-4)		Describes which quantity or DAS is displayed in each column and notes used data file and creation date.
Column 1	Wavelength [nm]	As obtained from processed (cleaned) TA data.
Columns 2-5 (2-4)	$A_{1-4(1-3)}(\lambda)$ [a u]	DAS 1-4 (1-3).
Last column [6 (5)]	$A_{\text{osc}}(\lambda)$ [a u]	DAS of the oscillation.

Parts of this data are shown in figures 2 of the main body and figures S7-8 of the SM [1].

- `<appendix>` = `_FitResult`:
Fit to the processed TA data. Obtained from the latter by fitting three exponential decays plus a signal echo separately for each wavelength, as described in [1], SM, section 3.1.

Part	Quantity [unit]	Comment
Row 1	Probe delay [ps]	As in processed TA data, but cut before 0.2 ps (and after 3 ps for logarithmic time axes).
Column 1	Wavelength [nm]	As obtained from processed (cleaned) TA data.
Rest	ΔA_{fit}	Fitted change in absorbance.

- `<appendix>` = `_FitParameters`:
  Fit parameters to the processed TA data. Obtained from the latter by fitting three exponential decays plus a signal echo separately for each wavelength, as described in [1], SM, section 3.1.

Part	Quantity [unit]	Comment
Column 1	Wavelength [nm]	As obtained from processed (cleaned) TA data.
Column 2	$A_1(\lambda)$	DAS 1.
Column 3	$\tau_1(\lambda)$ [ps]	Decay 1 time constant.
Column 4	$A_2(\lambda)$	DAS 2.
Column 5	$\tau_2(\lambda)$ [ps]	Decay 2 time constant.
Column 6	$A_3(\lambda)$	DAS 3.
Column 7	$\tau_3(\lambda)$ [ps]	Decay 3 time constant.
Column 8	$a_{\text{echo}}(\lambda)$	Relative amplitude of echo.
Column 9	$\tau_{\text{echo}}(\lambda)$ [ps]	Time of echo.
Column 10	$w_{\text{echo}}(\lambda)$ [ps]	Temporal width of echo.

- `<appendix>` = `_residue`:
  Residue of the processed TA data. Obtained from the latter by subtracting the fit discussed above.

Part	Quantity [unit]	Comment
Row 1	Probe delay [ps]	As in processed TA data, but cut before 0.2 ps (and after 3 ps for logarithmic time axes).
Column 1	Wavelength [nm]	As obtained from processed (cleaned) TA data.
Rest	ΔA_{res}	Absorbance residue.

  This data is shown in figures 3 (b) and 5 (c-d) of the main body and figure S9 (a) of the SM [1]. It serves as the basis for all further analyses of the oscillatory part of the transient dynamics, *i.e.* also all of the following data sets.

- `<appendix>` contains `_smoothed`:

In the case of the detailed scan on WS_2 with equidistant time points, we smoothed the "`_residue`" data before performing some of the further analysis. To this end, we simply calculated every new, smoothed residue value as the mean of a 7x7 subarray of the residue data centered around the position of the original data point.
This smoothed data is shown in figure 5 (e) of the main body and figure S6 (b) of the SM [1]. It is further used for all analysis regarding this particular scan on WS_2 that do not intrinsically include spectral averaging.

- `<appendix>` = `_residue(_smoothed)_2DFFT_Ampl`:
  Fast Fourier transform (FFT) amplitude (*i.e.* absolute value of the complex FFT result) of the (smoothed) residue data. The FFT is performed for each time trace, *i.e.* for each wavelength separately.

Part	**Quantity [unit]**	**Comment**
Row 1	f [Hz]	Resulting FFT frequency axis.
Column 1	Wavelength [nm]	As in (smoothed) TA residue data.
Rest	FFT amplitude [a u]	

  This data is shown in figures 3 (c) of the main body and figure S10 (a-c) of the SM [1].

- `<appendix>` = `_residue(_smoothed)_2DFFT_Phase`:
  Fast Fourier transform (FFT) phase (*i.e.* argument of the complex FFT result) of the (smoothed) residue data. The FFT is performed for each time trace, *i.e.* for each wavelength separately.

Part	**Quantity [unit]**	**Comment**
Row 1	f [Hz]	Resulting FFT frequency axis.
Column 1	Wavelength [nm]	As in (smoothed) TA residue data.
Rest	FFT phase [rad]	

  This data is shown in figure S10 (d-f) of the SM [1].

- `<appendix>` = `_residue(_smoothed)_2DFit_Result`:
  Fit to the TA (smoothed) residue data. Obtained from the latter by fitting exponentially decaying cosines separately for each wavelength, as described in [1], SM, section 3.4.

Part	**Quantity [unit]**	**Comment**
Column 1	Probe delay [ps]	As in processed TA data, but cut before 0.2 ps (and after 3 ps for logarithmic time axes).
Row 1	Wavelength [nm]	As in (smoothed) TA residue data.
Rest	$\Delta A_{\mathrm{res,fit}}$	Fitted residual change in absorbance.

- `<appendix>` = `_residue(_smoothed)_2DFit_Parameters`:

Fit parameters to the TA (smoothed) residue data. Obtained from the latter by fitting exponentially decaying cosines separately for each wavelength, as described in [1], SM, section 3.4.

Part	Quantity [unit]	Comment
Column 1	Wavelength [nm]	As in (smoothed) TA residue data.
Column 2	$A_{\text{osc}}(\lambda)$	DAS of the oscillation.
Column 3	$\tau_{\text{osc}}(\lambda)$ [ps]	Oscillation decay time constant.
Column 4	$T_{\text{osc}}(\lambda)$ [ps]	Oscillation period.
Column 5	$\alpha_{\text{osc}}(\lambda)$	Oscillation phase.

The phase $a_{\text{osc}}(\lambda)$ is depicted in figure S15 of the SM [1].

- `<appendix>` = `_residue_AvrgTraceFit_Curves`:
Spectrally averaged residual time traces and their fits. The former are obtained by integrating the "`_residue`" data as described in [1], SM, section 3.3.

Part	Quantity [unit]	Comment
Column 1	Probe delay [ps]	As in processed TA data, but cut before 0.2 ps (and after 3 ps for logarithmic time axes).
Column 2	$\Delta A_{\text{res,avrg}}$	Averaged residual absorbance.
Column 3	$\Delta A_{\text{res,avrg,fit}}$	Fit to the above.

This data is shown in figure S11 (a-c) of the SM [1]. The averaged curves also serve as the input for a FFT analysis, see below.

- `<appendix>` = `_residue_AvrgTraceFit_Parameters`:
Parameters of the fit to the spectrally averaged residual time traces. Obtained from the latter by fitting an exponentially decaying cosine. These files are 4x4 arrays, indicating fitted value, standard deviation, and t-statistics for each parameter.

Part	Quantity [unit]	Comment
Column 1	x_{fit}	Fitted value
Column 2	Δx_{fit}	Standard deviation
Column 3	$t = \frac{x_{\text{fit}}}{\Delta x_{\text{fit}}}$	t-value of the null hypothesis test: Distance of the fitted value to zero, in units of standard deviations
Column 4	p-value of the null hypothesis test	Probability of the parameter being zero.
Column 5	$\Delta x_{95\%}$	Parameter uncertainty for a 95% confidence level.
Row 1	A_{osc}	Oscillation amplitude.
Row 2	τ_{osc}	Oscillation decay time constant.
Row 3	T_{osc}	Oscillation period.
Row 4	α_{osc}	Oscillation phase.

This data is shown in figure 6 of the main body as well as figure S13 of the SM [1].

- `<appendix> = _residue_AvrgTraceFit_<parameter_name>Varied_ValueRange`:
Value and uncertainty of <parameter_name> (period, amplitude, or lifetime) for the fit to the spectrally averaged residual time traces, estimated using the method described in the SM, section 3.3 and figure S12. These files contain five values in a row, indicating fitted value, limits of the confidence interval, as well as positive and negative uncertainty of the respective parameter.

Part	**Quantity [unit]**	**Comment**
Entry 1	x_{fit}	Fitted optimal value
Entry 2/3	$x_{\mathrm{l}}/x_{\mathrm{u}}$	Lower/Upper limit for the parameter
Entry 4/5	$\Delta x_{\mathrm{l}}/\Delta x_{\mathrm{u}}$	Corresponding uncertainty for the parameter in negative/positive direction

This data is shown in figure 6 of the main body as well as figure S13 of the SM [1].

- `<appendix> = _residue_AvrgTraceFit_<parameter_name>Varied_SSR`:
Evolution of the sum of squared residuals (SSR) under variation of <parameter_name> (period, amplitude, or lifetime) for the fit to the spectrally averaged residual time traces, estimated using the method described in the SM, section 3.3 and figure S12. These files contain two columns, indicating parameter value and the SSR, respectively.

Part	**Quantity [unit]**	**Comment**
Column 1		Varied parameter: Period, amplitude, or lifetime
Column 2	SSR	Sum of squared residuals from fitting the other parameters

Some of this data is shown in figure S12 (a) of the SM [1].

- `<appendix> = _residue_AvrgTraceFit_<parameter_name>Varied_residuals`:
Residual curves of the fit to the spectrally averaged residual time traces, for the optimal and boundary values for <parameter_name> (period, amplitude, or lifetime), following the method described in the SM, section 3.3 and figure S12. These files contain four columns, indicating time axis and the residuals, respectively.

Column 1	Probe delay [ps]	As in processed TA data, but cut before 0.2 ps (and after 3 ps for logarithmic time axes).
Column 2	$\Delta A_{\mathrm{res}^2,\mathrm{opt}}$	Residue of the fit to the averaged residual absorbance for optimal parameters.
Column 3/4	$\Delta A_{\mathrm{res}^2,\mathrm{opt}}$	Residue of the fit to the averaged residual absorbance for value limits of <parameter_name>.

Some of this data is shown in figure S12 (b) of the SM [1].

- <appendix> = _residue_AvrgTraceFFT_Ampl:
  Fast Fourier transform (FFT) amplitude (*i.e.* absolute value of the complex FFT result) of the spectrally averaged residue data.

Part	**Quantity [unit]**	**Comment**
Column 1	f [Hz]	Resulting FFT frequency axis.
Column 2	FFT amplitude [a u]	

  This data is shown in figure 4 of the main body as well as figure S11 and S14 of the SM [1].

- <appendix> = _residue_AvrgTraceFFT_Phase:
  Fast Fourier transform (FFT) phase (*i.e.* argument of the complex FFT result) of the spectrally averaged residue data.

Part	**Quantity [unit]**	**Comment**
Column 1	f [Hz]	Resulting FFT frequency axis.
Column 2	FFT phase [rad]	

  This data is shown in figure S14 of the SM [1].

References

[1] Tim Völzer, Marvin Krupp, Julian Schröer, Annika Bergmann-Iwe, Tobias Korn, and Stefan Lochbrunner, *Having a good vibe: Strong coupling to the electronic system creates incoherent and coherent phonons in 1L-TMDCs*, 2D Materials, 2025. https://doi.org/10.1088/2053-1583/addf5a