Learning to Pre-process Laser Induced Breakdown Spectroscopy Signals Without Clean Data

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Abstract

This work tests whether deep neural networks can clean laser induced breakdown spectroscopy (LIBS) signals by using only uncleaned raw measurements. Our view of this problem considers a disentanglement of the effects of the target of interest from those of the nuisance factors (with non-zero mean) by leveraging the vast amounts of redundancies in LIBS data and our proposed learning formulation. This later aims at promoting consistency between repeated measurement views of a target while simultaneously removing consistencies with all other LIBS measurements taken throughout the history of the instrument. Evaluations on real data from the ChemCam instrument onboard the Martian Curiosity rover show a superior performance in cleaning LIBS signals compared to the standard approaches being used by the ChemCam team.

1. Introduction

Laser induced breakdown spectroscopy (LIBS) is one of the core remote sensing technologies used to acquire the chemical composition of geomaterials (e.g., rocks, soil). On Mars, the ChemCam LIBS based instrument is capable of investigating 1 mm size samples by laser excited samples from up to 7 m distances. It is equipped with a 1064 nm laser and ultraviolet (UV), visible (VIO) and near infrared (NIR) band spectrometers; which altogether are capable of collecting the sample’s spectral signatures occurring between 240-905 nm. It is through the analysis of such spectral signatures that chemical composition information of a sample (i.e., calibration) can be readily extracted. Unfortunately, nuisance factors introduce measurement corruptions that can cause erroneous predictions if these remain untreated [1]. Examples of these include the sensor effects (e.g., dark currents, white noise, electron continuum), environment and conditions (e.g., Martian dust) and laser (e.g., time of flight, angle of incidence).

To address this, a pre-processing of the LIBS signal is applied resulting in a "clean" representation used throughout all downstream tasks of interest. The work of the ChemCam team [2] for example proposes a multi-step sequential approach applying methods for background subtraction, wavelet Gaussian denoising, continuum removal, match filtering, adjustment of instrument response, and sensor-to-target distance corrections. Such a method currently stands as the preferred choice by the ChemCam team for pre-processing LIBS signals. All of these sequentially applied sub-methods are however, based on manually selected features, require human tuning and still present issues as treatments are far from perfect. Recently, the work of [3, 4] demonstrated the capability of deep convolutional neural networks (CNN’s) to learn the pre-processing functions as performed by [2] in a single feed forward pass and with added computational benefits. Unfortunately, [3, 4] learns by supervision and is thus limited by the quality of the ‘cleaned’ signals used as ground truth. Other related lines of work, show promising venues through the use of unsupervised learning (e.g., VAE, normalizing flows) to extract representations that are, in addition to unsupervised, also amenable for uncertainty quantification (UQ) [5, 6]. In these, a
latent representation is learned by imposing a prior on latent space structure which in turn enables UQ constructs on the latent distributions. Both works, however, focus on the task of calibration and are thus different in that they are applied on top of a ‘cleaned’ signal using [2].

Here, we propose a new self-supervised learning (SSL) approach to clean LIBS signals by training only with raw uncleaned inputs. The idea is based on the work noise2noise [7] which demonstrated the ability of deep neural networks (DNN’s) to denoise images of zero mean noise and without ground truth. We apply this idea here, but on 1D spectrally resolved LIBS signals and test whether DNN’s can disentangle the effects of the target from those of the non-zero mean nuisance factors in the measured signal. We propose a learning formulation that seeks to simultaneously: (1) promote consistency between repeated measurements of a target while also (2) reducing consistency with respect to every other measurement collected throughout the history of the instrument. In what follows, Section 2 describes our proposed learning formulation. Section 3 details our experiments and finally Section 4 summarizes our results.

2. Approach

2.1. Data generation model

The data generation model we consider is composed of modules each describing the physical mechanisms that have an effect on the measured LIBS signal. Fig. 1 illustrates the modules (shown as circles) describing the data generation model we adopt. A laser pulse of short duration and controlled power travels forth and back a certain distance through a medium (e.g., environment, planet) and excites a target sample. The returned waveform is measured at the sensor where an entanglement between the effects of the laser-target interaction and the nuisance factors (e.g., laser-medium interaction, sensing conditions and sensor) occur. In Fig. 1 two LIBS signal measurements \(x_p', x_p \in \mathbb{R}^N\) share information from the same target and nuisance factors whereas no target information is shared with respect to measurement \(x_n \in \mathbb{R}^N\). Instead, this later shares information mostly about the nuisance factors assuming measurements come from the same instrument. The idea here is to exploit data redundancies informative of the effects of the data generation modules either individually or of combinations thereof in order to isolate the effects of a module of interest. Here, we focus on disentangling the target from the non-zero mean nuisance factors for the purpose of cleaning and assuming the information from the isolated target effects is sufficient for downstream tasks.

![Figure 1: Data generation model](image)

Figure 1: Data generation model: measurements \(x_p', x_p, x_n\) are composed by the effects of ambient conditions, laser, sensor, environment (i.e., Planet), \(x_p'\) and \(x_p\) share the same target.

2.2. Problem formulation

The disentanglement problem is posed here as a deep neural network \(h: \mathbb{R}^N \rightarrow \mathbb{R}^N\) that given raw inputs \(x \in \mathbb{R}^N\) outputs a representation \(z = h(x)\) without the effects of the nuisance factors and probably
leaving only those from the target. To train this network we formulate a learning objective that promotes consistency between representations of repeated target measurements in a set $\mathcal{P}$ while simultaneously reducing it with respect to every other LIBS measurements in a set $\mathcal{N}_\mathcal{P}$ with $\mathcal{N}_\mathcal{P}\cap\mathcal{P} = \emptyset$. Mathematically, this is described as the minimization of the function in Eq. (1) with respect to $z$ as:

$$\ell = -\sum_{p\in\mathcal{P}} \frac{1}{|\mathcal{P}| - 1} \sum_{p'\in\mathcal{P}\setminus\{p\}} \log \frac{\exp(\langle z_p, z_{p'} \rangle)}{\sum_{n\in\mathcal{N}_p} \exp(\langle z_p, z_n \rangle^2)}$$ (1)

where $\langle \cdot, \cdot \rangle$ denotes an inner product. The numerator of the right most term in Eq. (1) is responsible for keeping the target information while its denominator removes the nuisance effects. The squared inner product in Eq. (1) removes a sign ambiguity that if ignored, flips the non-zero mean nuisance effects in the spectrum upside down instead of removing them. Also, division by $\mathcal{P} - 1$ normalizes against the variable number of repeated target shots. In our formulation, the latent representation $z \in \mathbb{R}^N$ is the same size as the input $x$ to keep the spectral line activation to chemical content relationship properties often used by ChemCam analysts for interpretation.

We would like to note that although not the same, similar objective functions to Eq. (1) have been devised. An example is InfoNCE [8, 9]; a learnable mutual information (MI) proxy. Under such perspective, the numerator in Eq. (1) extracts a latent representation that maximizes the mutual information between repeated target measurements while also simultaneously minimizes it with respect to every other measurement by exploiting nuisance effect similarities.

3. Experiments

The ChemCam datasets available at [10] were used as the experimentation benchmark for validation of the proposed approach. The datasets contain raw and pre-processed spectra obtained from Martian targets (e.g., rocks, soil) and from reference calibration standards of known chemical composition. The specific dataset we employ consists of spectrally resolved LIBS signal measurements collected on Earth in a laboratory setting from a set of ~ 408 reference calibration standards [11] of known and certified chemical reference composition. Each reference standard is repeatedly shot (e.g., 50 times) following each time measurements of the full 240-905 nm LIBS signal. This process is repeated at multiple locations for each target which amounts to ~41,000 uncleaned LIBS signals. After collection, wavelengths within the bands [240.811,246.635], [338.457,340.797], [382.13,387.859], [473.184,492.427], [849,905.574] were ignored consistent with practices of the ChemCam team [11].

For representation learning, we use the spectral CNN architecture described in [3] with $D = 18$ Conv+Bn+ReLu layers optimized through SGD with a learning rate (lr) of 0.1, momentum 0.9 and number of epochs 60. At training, the batch sizes consist each of 512 raw uncleaned LIBS signals constructed by batching together the repeated LIBS measurements from a target (~50) and a randomly chosen subset of examples from every other target (with replacement).

Quantitative evaluation of the effectiveness of a representation is performed here on the downstream task of calibration. We do this by testing the ability of a neural network (NN) head to calibrate using LIBS representation inputs. Comparisons are made against three representations: (1) raw, (2) the pre-processing used by the ChemCam team [2], and the proposed. Training and testing is performed independently of the representation network also using the Calib dataset but under a leave one out split. In other words, multiple linear heads are trained where for each we leave all the data pertaining to one calibration standard out and then use it at test time. This process is repeated multiple times from scratch until all standards are covered, for all of the compared representations and for each element oxide. The architecture used is a single linear layer in Fig. [7] that takes as input a LIBS signal of 5485 wavelengths and outputs a scalar $\hat{y}_{\text{oxide}}$ for each oxide $\in \{\text{SiO}_2, \text{TiO}, \text{Al}_2\text{O}_3, \text{FeO}_T, \text{MgO}, \text{CaO}, \text{Na}_2\text{O}, \text{K}_2\text{O}\}$. The
Figure 2: Linear head architecture for downstream calibration.

hyperparameters used are an initial lr of 1.0, decayed after 75 epochs with cosine annealing \cite{12} and with \#epochs 200. Batches are constructed from a set of 64 shot-averaged examples randomized over the whole training set without replacement. The shot-averages are computed by averaging the LIBS signal representations over an individual target and laser shot location. This averaging is consistent with common practices of the ChemCam team \cite{2, 11}.

4. Results

4.1. Pre-processing representation

A few examples illustrating the qualitative performance of our learned representations compared against the raw and pre-processing of \cite{2} are included here. Fig. 3 shows examples of representative calibration standards and patches of the LIBS spectrum. Fig. 3(a-c) presents spectral lines modulated by what appears to be remainders of electron continuum amplified by a system spectral response adjustment. Fig 3(d-f) show instead the representation obtained with our proposed method. Note that in all cases the representations in (d-f) seem to cope better in isolating the effects of the target from the nuisance factors. The spectral content from the non-zero mean dark currents and electron continuum are brought down close to zero better than \cite{2} where remainders of both are visually present in all cases. A similar example included in Fig. 4 shows now the entire LIBS spectrum of a representative example. We observe that the nuisance factor removal properties seen in Fig. 3 are also reflected throughout the entire UV, VIO and VIS spectrum.

4.2. Pre-processing in downstream calibration task.

Although, the proposed aproach yields representations that seem qualitatively ‘cleaner’ compared to the raw uncleaned and the pre-processing of \cite{2}, we also quantitatively evaluate its effectiveness in the downstream task of calibration. For this, we follow the leave one out training and testing strategy described in the last two paragraphs of Section 3.
Performance results are included as plots of prediction (in y-axis) versus ground truth %-oxide (in x-axis) for each of the eight major elements (labeled inside each plot). Fig. 5 columns (a,d), (b,e) and (c,f) are the results of the raw, pre-processing of [2] and the proposed learned representation, respectively. Here, we observe that for all three representations and element oxides the predictions (shown as orange dots) follow in some way or another the ideal 1:1 line in black. The raw representation leads to a higher spread of calibration predictions over the 1:1 line, followed by the pre-processing of [2] while the proposed leads to a relatively better and smaller spread. This is quantified by means of the root mean squared error (RMSE) shown in the legend of each subfigure where the proposed learned representation outperforms all others throughout all element oxide cases.

Our interpretation of the results shown in Fig. 5 is that our proposed framework was effective in isolating the effects of the target while removing those of the nuisance factors, at least in what regards to the eight major element oxides. Moreover, it was able to outperform in a calibration task the current ChemCam’s team method of choice for pre-processing uncleaned LIBS signals. Compared to the raw uncleaned representation where no target information has been lost from the application of pre-processing, we observe that our method is able to probably isolate the effects of the target without losing any meaningful information. With this, we thus corroborate that the isolation of modular effects in LIBS is possible in a self-supervised way using DNN’s and our learning formulation in Eq. (1). This, without assuming any explicit priors or statistical characterizations of the nuisance factors and by leveraging the vast amount of data redundancies that exist in LIBS data. In terms of computational complexity, we would like to note that our proposed method can generate cleaned representations at $\sim$50 Hz on a CPU when in feed-forward mode similar to [3]. This renders our method more computationally efficient than those described in [2, 11].
Figure 5: Effectiveness of pre-processing representation in downstream calibration task. The proposed method resulted in a latent representation that leads to less spread and lower RMSE predictions from the ideal 1:1 line and compared to the raw and the ChemCam team’s pre-processing in [2] representations. Included are the regression results of the 8 major chemical elements in the ∼470 reference standards ‘Calib’ dataset. Corresponding RMSE’s for each element are shown in the legend for each oxide.

5. Conclusion

In this work, we tested the ability of deep neural networks to clean LIBS signals in a self-supervised way using only the raw uncleaned LIBS measured data. Our formulation simultaneously exploits redundant information in the repeated measurements of a target to isolate its effects while disentangling those from the non-zero mean nuisance factors by removing consistencies with respect to every other available LIBS signal measurement. Our experiments on real data show that the learned representation is qualitatively and quantitatively more effective and efficient than the pre-processing methods currently used by the ChemCam team.

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