Baseline Correction and Denoising of Raman Spectra by Deep Residual CNN

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Abstract: A practical deep residual CNN model for the improvement of Raman spectral analysis is proposed in this work. Through the feature transformation in the deep residual blocks, it successfully denoises and precisely corrects the baseline.

1. Introduction

Raman spectral analysis is widely applied in various fields, including material inspection, biomedical engineering, and analytic chemistry. Through the peak information of the Raman spectra, we can identify the ingredients of various samples, estimate the lattice stress, and evaluate the quality of crystallinity. There are three key properties of Raman spectra. First, the peak value indicates the intensity. Second, the full width at half maximum (FWHM) indicates the lattice quality. Third and the most important one, the peak position indicates the "fingerprint" of the sample. However, several types of noise may deteriorate the quality of the spectra. According to the previous reports [1–3], there are two major types of noise, the fluorescence background noise and random noise. Thus, many methods regarding denoising and baseline correction of Raman spectra were proposed. The wavelet transformation (WT) is the prevalent one [1,2], which determines the baseline and random noise by using time-frequency analysis to correct the spectra. In this paper, we propose employing a deep residual convolutional neural network (DRCNN) for baseline correction and denoising. Residual learning is an outstanding technique in neural networks. The most significant advantage of residual learning is that the residue would prevent the gradient from vanishing during the learning process. Therefore, the networks can be made deeper than conventional CNNs. Through feature transformations in the residual blocks (Res-Block), the exact information we care can be extracted precisely.

2. The Proposed Method

2.1. Spectral Model and Data Preparation

Let s denote the measured Raman spectrum. The spectral model can be written as $s = g + b + n$, where g, b, and n refer to the ideal Raman spectrum, the baseline, and the random noise, respectively [1–3]. For these three components, g can be expressed as a sequence of Lorentz kernel functions for gases and liquids, or as Gaussian kernel functions for solid samples [4]. That is,

$$g = \sum_{i=1}^{m} \frac{2A_i}{\pi} \frac{w_i}{4(x-x_i)^2 + w_i^2} + \sum_{j=1}^{n} A_j \exp\left(\frac{(x-x_j)^2}{2\sigma^2}\right). \quad (1)$$

We assume that the fluorescence background noise b is of low frequency. Thus, we use a polynomial to emulate it [1]. In this work, we write $b = a_0x^3 + a_1x^2 + a_2x + a_3$, where a_i are different uniformly distributed random variables. The random noise n considered in this work is Gaussian noise. Others like shot noise, readout noise, and pink noise will not be discussed in this paper. The noise n is emulated by the Gaussian function with its noise level related to the amplitude of the lowest peak of g. We employ 55000 training data and 5000 test data for our DRCNN. All are generated by this spectral model. We also use some RRUFF^{TM} [5] experimentally measured Raman spectra to evaluate the performance of our method.

2.2. Architecture of the DRCNN

We use the PyTorch tool to implement our DRCNN. Fig. 1. illustrates the architecture and parameters of the proposed DRCNN. It contains 10 convolutional layers and 18 Res-Blocks. Notice that we use an additional shortcut to provide the extra residue [6]. The maximal value of each data set is normalized to one in preprocessing. After 100 epochs of training, the MSE loss function converges to the order of 10^{-5}.

3. Results and Discussions

We use the emulated test data and real data [5] to evaluate the performance of our DRCNN. Table 1. shows the performance of our model on the emulated test data. It can be seen that our method slightly outperforms the method of Chen *et al.* [1] regarding estimated SNR (eSNR). As to the peak position error (P_{error}), our method leads to approximate a half error of that by Hu *et al.* [2]. Note that our model is able to deal with the spectra with unknown noise levels (i.e., blind denoising). Furthermore, the model performance on real spectra from RRUFF [5] is evaluated. Fig. 2. shows the raw spectrum with a high noise level (black line) and the recovered spectrum by our method (red line). Our DRCNN eliminates the noises with high noise levels effectively. Moreover, Fig. 3. shows the model performance on real spectra with two different baselines. The blue lines are the difference between the black and the red lines, which are the estimated noises and baselines. Our model can correct diverse baselines without losing details of peak information. The errors of peak positions in these figures are negligible. In addition, the shapes of these peaks remain. The FWHMs are nearly equal to the raw samples.

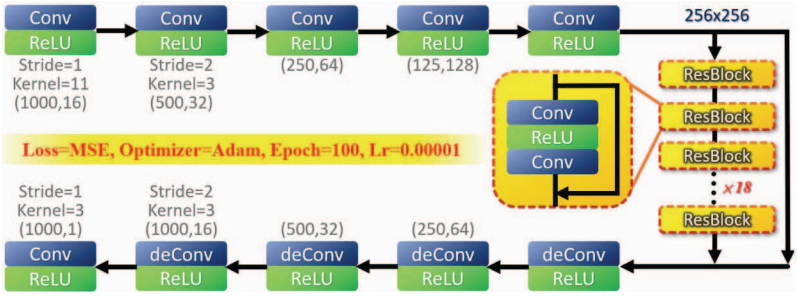

Fig. 1. The DRCNN.

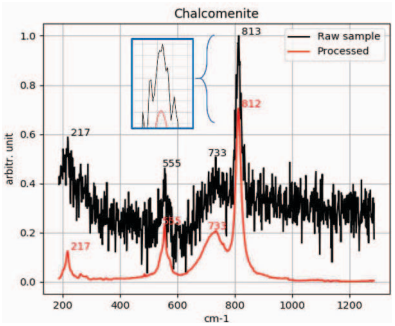

Fig. 2. The RRUFF real spectrum with a high noise level.

Table 1. Compare to other methods

Methods	Noise level	eSNR (dB)	P_{error} (cm^{-1})
Proposed	10~30%	33.50	0.791
Chen [1]	30%	28.03	N/A
Chen	20%	31.38	N/A
DWT [1]	20%	31.11	N/A
Hu [2]	0%	N/A	1.333

*DWT = discrete wavelet transform.

**$\text{eSNR} = 10\log(\frac{\Sigma g_{est}^2}{\Sigma(g-g_{est})^2})$.

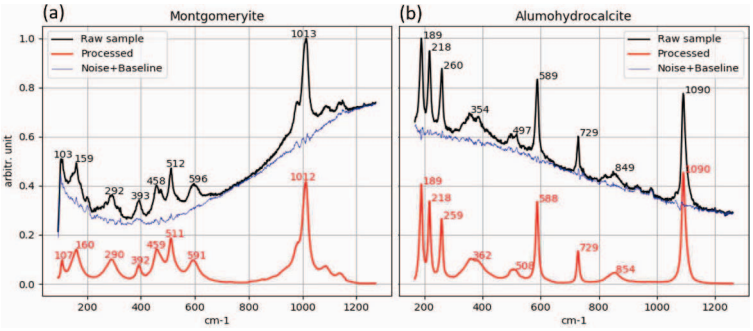

Fig. 3. RRUFF real spectra with different baseline drifts.

4. Conclusions

We proposed a novel, effective, and robust method to extract the key information of the spectral peaks from diverse baselines and noises utilizing the DRCNN. In our method, a priori information regarding noise levels and baselines is unnecessary. Our DRCNN method not only can improve the quality of Raman signals but also can be applied to other spectral analyses.

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