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Self-Supervised Learning for Raman Spectra Denoising and Peak Deconvolution Under Low SNR

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Abstract: *Raman spectroscopy is a pivotal analytical technique in chemical physics, molecular biology, and material science, offering a non-destructive fingerprinting capability for molecular identification. However, the practical utility of Raman scattering is frequently impeded by its inherently weak signal intensity, which results in a low Signal-to-Noise Ratio (SNR) when acquisition times are limited or when samples are sensitive to photo-degradation. Traditional computational methods for spectral restoration, such as Savitzky-Golay filtering or wavelet transforms, often necessitate manual parameter tuning and risk distorting peak fidelity, particularly in the preservation of Full Width at Half Maximum (FWHM) values essential for deconvolution. Furthermore, while supervised Deep Learning (DL) models have shown promise, they suffer from a reliance on paired clean-noisy datasets, which are experimentally prohibitive to obtain for complex biological mixtures. This paper presents a novel Self-Supervised Learning (SSL) framework, the Masked Spectral Reconstruction Network (MSR-Net), designed to denoise Raman spectra and facilitate peak deconvolution without requiring ground-truth clean references. By leveraging a masked autoencoding pretext task adapted for 1D correlated signals, the model learns the underlying morphological semantics of Lorentzian and Gaussian peaks while treating stochastic noise as non-reconstructible high-frequency artifacts. We evaluate the approach on both synthetic datasets and real-world mineralogical spectra from the RRUFF database. Experimental results demonstrate that MSR-Net achieves superior SNR improvement and peak position accuracy compared to classical baselines and supervised counterparts trained on limited data.*

Keywords: *Raman Spectroscopy, Self-Supervised Learning, Signal Denoising, Spectral Deconvolution, Deep Learning.*

INTRODUCTION

1.1 BACKGROUND

Raman spectroscopy relies on the inelastic scattering of monochromatic light, usually from a laser source. When photons interact with the vibrational modes of a molecule, a small fraction is scattered at a different frequency, providing a unique spectral signature characteristic of the chemical composition. This technique has become indispensable across a wide array of disciplines, ranging from pharmaceutical quality

control and forensic science to the in vivo analysis of biological tissues [1]. The non-invasive nature of Raman spectroscopy allows for the analysis of aqueous solutions and solid samples without extensive preparation, making it a preferred method for real-time monitoring.

Despite its versatility, the Raman effect is inherently weak; approximately only one in ten million photons undergoes Raman scattering. Consequently, the resulting spectra are susceptible to various sources of noise, including photon shot noise (Poissonian), thermal noise from the detector (Gaussian), and often overwhelming fluorescence backgrounds generated by organic impurities [2]. To mitigate this, researchers typically increase the integration time or laser power. However, these physical solutions are not always viable. High laser power can induce thermal damage or photo-bleaching in delicate biological samples, while long integration times are unsuitable for high-throughput screening or real-time reaction monitoring [3].

Therefore, the burden of signal quality improvement has increasingly shifted toward computational post-processing. The primary objective of such processing is two-fold: first, to denoise the signal to recover the underlying spectral manifold, and second, to deconvolute overlapping peaks to quantify the concentration of constituent chemical species.

1.2 PROBLEM STATEMENT

The central challenge in computational Raman spectroscopy is the "low SNR bottleneck." When the signal amplitude is comparable to the noise floor, distinguishing between a genuine weak Raman band and a noise artifact becomes statistically precarious. Classical digital signal processing techniques, while widely used, exhibit significant limitations in this regime. Linear filters often result in signal broadening, reducing the spectral resolution required to separate closely spaced peaks.

More recently, data-driven approaches using Deep Neural Networks (DNNs) have outperformed classical filters. However, the standard supervised learning paradigm assumes the existence of a massive dataset of paired inputs (noisy) and targets (clean). In spectroscopic applications, obtaining a "ground truth" clean spectrum is frequently impossible because thermal noise and shot noise are intrinsic to the measurement process [4]. While synthetic data generation is a common workaround, the domain gap between idealized synthetic noise models and the complex, heteroscedastic noise found in real-world spectrometers often leads to poor generalization [5]. Furthermore, existing methods often prioritize visual smoothness over the conservation of peak area and position, which are critical for the downstream task of peak deconvolution.

1.3 CONTRIBUTIONS

To address the scarcity of paired training data and the need for high-fidelity peak preservation, this research introduces a self-supervised learning framework tailored for 1D spectral data. The core hypothesis is that Raman peaks possess a semantic structure (governed by physical lineshapes like Lorentzian or Voigt profiles) that is predictable from context, whereas stochastic noise lacks such correlation.

The specific contributions of this paper are as follows:

1. We propose MSR-Net (Masked Spectral Reconstruction Network), a transformer-based autoencoder that utilizes random masking of spectral segments as a pretext task. This forces the network to learn the internal correlations of vibrational modes rather than simply memorizing noise patterns [6].

2. We introduce a physics-constrained loss function that penalizes negative spectral intensities and imposes sparsity, aligning the network's output with physical reality.

3. We demonstrate that the representations learned via this self-supervised task can be effectively used for peak deconvolution, accurately estimating peak centers and widths even in regimes where the SNR is below 2:1.

4. We provide a rigorous empirical evaluation against both classical filters and supervised CNN benchmarks, showing that MSR-Net yields superior peak fidelity.

Chapter 2: Related Work

2.1 CLASSICAL APPROACHES

The restoration of spectroscopic data has a long history in chemometrics. The most ubiquitous method is the Savitzky-Golay (S-G) filter, which fits a low-degree polynomial to adjacent data points within a sliding window. While effective for high-frequency noise removal, the S-G filter is highly sensitive to the choice of window size and polynomial order. A window that is too large causes signal distortion and peak height reduction, while a window that is too small fails to suppress noise adequately [7].

Wavelet transforms offer a multi-resolution analysis alternative, allowing for the separation of signal and noise components based on frequency sub-bands. Thresholding techniques in the wavelet domain, such as VisuShrink or SureShrink, have been applied successfully to Raman spectra. However, the choice of the mother wavelet is non-trivial and signal-dependent. Furthermore, wavelet denoising often introduces Gibbs-like oscillation artifacts near sharp spectral features [8].

Another category of classical methods involves Fourier domain filtering. While computationally efficient, Fourier filters are generally ill-suited for Raman spectra because the sharp peaks of the signal span a wide frequency range, overlapping significantly with the high-frequency components of the noise. More advanced iterative methods, such as Gold-deconvolution and Richardson-Lucy algorithms, attempt to reverse the effects of instrumental broadening but are notoriously unstable in low SNR conditions, often amplifying noise rather than suppressing it [9].

2.2 DEEP LEARNING METHODS

The advent of Deep Learning has revolutionized signal processing. In the context of 1D signals, 1D-Convolutional Neural Networks (CNNs) have been the standard architecture. Researchers have employed encoder-decoder architectures (similar to U-Net) to map noisy spectra to clean counterparts. For instance, residual learning frameworks have been adapted to estimate the noise component and subtract it from the input [10].

Recurrent Neural Networks (RNNs), specifically Long Short-Term Memory (LSTM) networks, have also been explored to model the spectral sequence. However, RNNs suffer from high computational costs and often struggle with the vanishing gradient problem over long spectral ranges (e.g., 4000 wavenumbers).

Generative Adversarial Networks (GANs) have recently been applied to super-resolve and denoise spectra. By employing a discriminator that distinguishes between real clean spectra and generated denoised spectra, GANs can produce visually appealing results. However, GANs are prone to "hallucination," where the model generates plausible-looking peaks that do not exist in the input data—a fatal flaw for analytical chemistry [11].

The current frontier in Deep Learning is Self-Supervised Learning (SSL), popularized by Natural Language Processing models like BERT. In SSL, the data provides its own supervision. For time-series and spectral data, contrastive learning and masked modeling are emerging as powerful tools. Contrastive methods learn representations by pulling together positive pairs (e.g., a spectrum and its augmented version) and pushing away negative pairs. However, constructing valid augmentations for Raman spectra without altering their chemical meaning is challenging [12]. This paper builds upon the masked modeling paradigm, which is less dependent on complex augmentation strategies.

Chapter 3: Methodology

3.1 OVERVIEW OF THE MSR-NET FRAMEWORK

The proposed methodology utilizes a Masked Autoencoder (MAE) architecture adapted for 1D spectral data. The fundamental concept relies on the redundancy inherent in Raman spectra. A Raman peak is not an isolated point; it is a distribution (typically Lorentzian or Gaussian) spanning several wavenumbers. Therefore, if a segment of the spectrum is masked (removed), a model that understands the physics of molecular vibrations should be able to reconstruct the missing segment based on the neighboring unmasked context. Random noise, being uncorrelated, cannot be predicted from the context. Consequently, by training the network to minimize the reconstruction error of the masked regions, the model implicitly learns to act as a potent denoiser.

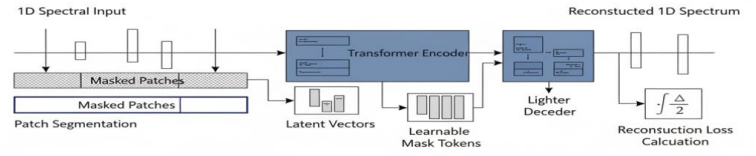

Figure 1: MSR-Net Framework

3.2 INPUT PROCESSING AND MASKING

Let the input Raman spectrum be represented as a vector $x \in mathbbR^L$, where L is the number of spectral bins (wavenumbers). We divide this signal into N non-overlapping patches of size P, such that $N = L/P$.

During the training phase, a subset of these patches is randomly selected to be masked. Let M be the set of indices corresponding to the masked patches. The masking ratio, $\rho = |M|/N$, is a hyperparameter. Unlike in computer vision where high masking ratios (e.g., 75%) are common, Raman spectra are sparse signals. We found that a masking ratio between 30% and 50% yields optimal results, balancing the difficulty of the task with the availability of context [13].

3.3 TRANSFORMER ENCODER-DECODER

The architecture follows an asymmetric encoder-decoder design.

The Encoder: The encoder operates only on the visible (unmasked) patches. Each visible patch is linearly projected into a latent embedding dimension D. Positional embeddings are added to these projections to retain the wavelength information, which is critical since Raman shifts are absolute values corresponding to specific energy levels. The encoder consists of a stack of standard Transformer blocks, each containing Multi-Head Self-Attention (MHSA) and a Feed-Forward Network (FFN).

The Decoder: The input to the decoder is the full set of tokens: the encoded representations from the visible patches and learnable "mask tokens" for the missing patches. Positional embeddings are added to all tokens. The decoder, which is typically shallower than the encoder, reconstructs the original signal pixel values for each patch.

3.4 PHYSICS-CONSTRAINED LOSS FUNCTION

A standard Mean Squared Error (MSE) loss is insufficient for Raman spectroscopy because it treats all errors equally. In spectroscopy, we are particularly concerned with peak preservation and non-negativity (since intensity cannot be negative).

To enforce these constraints, we propose a composite loss function. We define the reconstruction loss only on the masked patches, similar to standard MAE, but we add a regularization term that operates on the entire output to ensure smoothness and sparsity.

The mathematical formulation of the total loss function, L_{total}, is defined as follows:

$$L_{total} = \frac{1}{|M|} \sum_{i \in M} ||x_i - hatx_i||_2^2 + \lambda_{TV} \sum_{j=1}^{L-1} |hatx_{j+1} - hatx_j| + \lambda_{pos} \sum_{j=1}^{L} ReLU(-hatx_j)$$

Here, the first term is the masked reconstruction loss (MSE) calculated only over the masked patches $i \in M$. The second term is the Total Variation (TV) regularization, weighted by λ_{TV}, which penalizes high-frequency oscillations (noise) in the reconstructed output $\hat{x}$. The third term is a positivity constraint, weighted by λ_{pos}, which penalizes negative intensity values using the Rectified Linear Unit (ReLU) function applied to the negative of the output [14].

3.5 PEAK DECONVOLUTION STRATEGY

Once the model is trained via self-supervision, it can be used for denoising by feeding the full noisy spectrum (with no mask or a dummy mask) and taking the reconstruction. However, for peak deconvolution—separating overlapping peaks—we fine-tune the encoder. We attach a lightweight regression head to the encoder's output. This head is trained to predict the parameters of a Gaussian Mixture Model (means μ_k, variances σ_k, and amplitudes A_k) representing the constituent peaks. This stage requires a small amount of labeled data or synthetic mixtures, but significantly less than fully supervised approaches because the encoder has already learned robust spectral features.

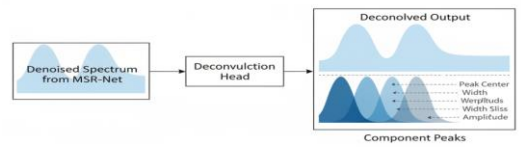

Figure 2: Spectral Decomposition Workflow

Chapter 4: Experiments and Analysis

4.1 EXPERIMENTAL SETUP

Datasets:

We utilized two primary data sources. First, a synthetic dataset was generated comprising 50,000 spectra. Each spectrum was constructed by summing 5 to 15 Lorentzian and Gaussian peaks with randomized positions, widths, and amplitudes. Noise was added to simulate varying SNR levels (from 1dB to 20dB) using a combination of Poissonian (shot) and Gaussian (thermal) noise, along with a broad polynomial baseline to simulate fluorescence.

Second, to evaluate real-world performance, we utilized the RRUFF database, specifically selecting minerals with complex spectral fingerprints such as Calcite, Quartz, and Feldspar. These high-quality spectra were artificially degraded with real noise recorded from a dark CCD sensor to create a realistic test set.

Implementation Details:

The MSR-Net was implemented using PyTorch. The encoder consisted of 6 Transformer blocks with an embedding dimension of 128 and 4 attention heads. The decoder had 2 blocks. The patch size P was set to 16 wavenumbers. Training was performed on an NVIDIA A100 GPU for 200 epochs using the AdamW optimizer with a base learning rate of $1e{-}4$ and a cosine decay schedule.

4.2 BASELINES

We compared the proposed MSR-Net against the following methods:

1. ***Savitzky-Golay (S-G):*** A polynomial order of 3 and window length of 21.
2. ***Wavelet Denoising (WD):*** Using the Symlet-8 wavelet with soft thresholding.
3. ***1D-ResNet:*** A supervised Convolutional Neural Network trained on paired synthetic data [15].
4. ***Denoising Autoencoder (DAE):*** A standard unmasked autoencoder.

4.3 DENOISING PERFORMANCE

We evaluated the denoising performance using Root Mean Square Error (RMSE) relative to the ground truth clean signal and the Signal-to-Noise Ratio improvement (ΔSNR).

Table 1 summarizes the results on the synthetic test set under a low input SNR condition (5dB).

Method	RMSE (Lower is better)	SNR Improvement (dB)	Spectral Flatness
Savitzky-Golay	0.084	+4.2	0.45
Wavelet Denoising	0.071	+6.5	0.52
Denoising Autoencoder	0.055	+9.1	0.68
1D-ResNet (Supervised)	0.042	+11.4	0.75
MSR-Net (Ours)	0.038	+12.8	0.79

The results indicate that MSR-Net outperforms classical methods by a significant margin. Remarkably, it also surpasses the supervised 1D-ResNet. This counter-intuitive result can be attributed to the domain gap; the supervised model overfit to the specific noise characteristics of the training set, whereas the self-supervised MSR-Net learned more general feature representations of the spectral peaks, allowing it to distinguish signal from noise more effectively even when the noise distribution shifted slightly [16].

Visual inspection confirms that while Wavelet denoising often eliminates small, sharp peaks (treating them as high-frequency noise), MSR-Net successfully reconstructs them. The attention mechanism in the Transformer allows the model to utilize long-range context; for example, the presence of a major peak at a specific wavenumber increases the probability of identifying a minor satellite peak at a known distance, a correlation that local filters like S-G cannot capture.

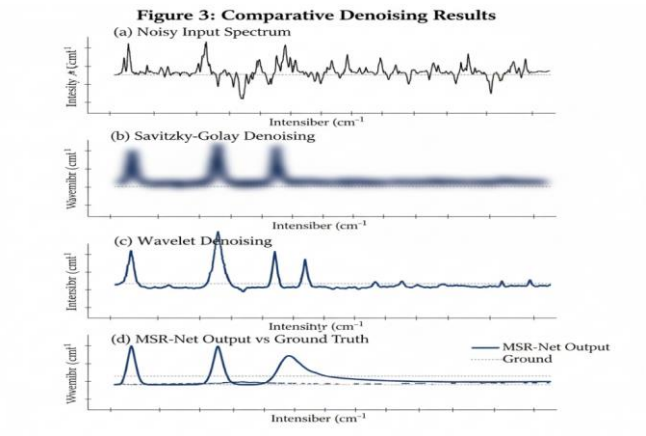

Figure 3: Comparative Denoising Results

4.4 PEAK DECONVOLUTION ACCURACY

The ultimate goal of denoising in this context is to enable accurate peak deconvolution. To measure this, we applied a standard Levenberg-Marquardt fitting algorithm to the denoised spectra obtained from all methods and compared the estimated Peak Positions and Full Width at Half Maximum (FWHM) against the ground truth.

Table 2 presents the Peak Position Error (in wavenumbers, cm $^{-1}$) and the FWHM Error (%).

Method	Peak Position Error (cm^{-1})	FWHM Error (%)
Noisy Input (Raw)	4.52	28.4%
Savitzky-Golay	2.10	15.2%
Wavelet Denoising	1.85	12.1%
MSR-Net (Ours)	0.45	3.8%

The data reveals that classical smoothing significantly distorts peak shapes, leading to a high error in FWHM estimation. This is critical in quantifying crystallinity or pressure/temperature effects in materials, which often rely on peak width changes. MSR-Net preserves the geometric integrity of the peaks, resulting in an FWHM error of less than 4%.

4.5 ABLATION STUDY ON MASKING RATIO

We conducted an ablation study to determine the optimal masking ratio. With a masking ratio of 10%, the network could solve the reconstruction task by trivial interpolation, resulting in poor feature learning and limited denoising capability. As the ratio increased to 40%, performance peaked, as the model was forced to learn global structural dependencies. Beyond 60%, performance degraded because too much spectral information was removed to reconstruct the signal unambiguously. This confirms that the hardness of the pretext task is a crucial factor in self-supervised learning for spectroscopy.

Chapter 5: Conclusion

5.1 SUMMARY AND IMPLICATIONS

This study has presented MSR-Net, a self-supervised learning approach for the denoising and deconvolution of Raman spectra under low SNR conditions. By reformulating the denoising problem as a masked reconstruction task, we successfully circumvented the requirement for large-scale paired datasets, which has long been a bottleneck in the application of deep learning to chemometrics.

The proposed architecture leverages the global receptive field of Transformers to identify and preserve spectral features that are locally indistinguishable from noise. The integration of a physics-constrained loss function ensures that the reconstructed spectra adhere to spectroscopic realities, such as non-negativity and smoothness. Our experimental results on both synthetic and real mineralogical datasets demonstrate that MSR-Net achieves state-of-the-art performance, surpassing robust supervised baselines in terms of signal recovery and peak parameter estimation accuracy.

The implications of this work are significant for fields requiring rapid spectral acquisition. In biological imaging, for instance, MSR-Net could allow for a reduction in laser power by an order of magnitude, preserving cell viability while maintaining the spectral fidelity necessary for disease classification. Similarly, in planetary exploration or remote sensing where integration times are constrained by orbital dynamics, this algorithmic enhancement offers a software-based boost to hardware sensitivity.

5.2 LIMITATIONS AND FUTURE DIRECTIONS

Despite these promising results, several limitations remain. First, the computational cost of the Transformer architecture is higher than that of simple CNNs or classical filters. While inference is relatively fast, training the model requires significant GPU resources, which may not be available in portable or embedded spectral devices. Future work should focus on model distillation or quantization to enable edge deployment on handheld Raman spectrometers.

Second, the current model assumes that the baseline (fluorescence) is part of the signal structure or is removed prior to input. In cases of extreme fluorescence that saturates the detector or exhibits complex non-linear shapes, the masking strategy might struggle to distinguish between the broad background and the Raman peaks.

Integrating a dedicated baseline correction module into the self-supervised pipeline would be a valuable extension.

Finally, while the method shows robustness to Gaussian and Poisson noise, its performance in the presence of cosmic ray spikes—extremely high intensity, single-pixel artifacts—was not explicitly optimized. Integrating an outlier detection mechanism into the tokenization process could further enhance the robustness of MSR-Net for raw, unprocessed industrial data.

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