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Abstract. X-Ray Fluorescence (XRF) is an analytical technique that furnishes complex elemental spectra for element identification. Its non-invasiveness and portable nature have given the technique a broad application across various fields, each of which shows an idiosyncratic spectra type, resulting in a landscape where full automation of XRF analysis is challenging for Artificial Intelligence (AI) techniques.
In this contribution, we make use of recent results and hypothesis on the performance and interpretability of AI networks (superposition theory) to explore the prospects of using AI techniques to overcome the bottleneck of XRF automation analysis. In particular, we suggest that an autoencoder of XRF spectra whose (monosemantic) latent space dimensions match the number of elemental lines present in the input should have improved performance and interpretability.
In addition, we will discuss some of the implications and difficulties in this process, as well as some very preliminary results in this direction.

Keywords: X-Ray Fluorescence (XRF), Synthetic Dataset Generation, Deep Learning, Interpretability, Superposition, Polysemanticity

1 Introduction

X-Ray Fluorescence (XRF) exploits the distinct radiation signatures of elements and it is widely utilised across various fields due to its advantages: portability, non-invasiveness, cost-effectiveness, and powerful analytical capabilities [1], [2].

Although this radiation process is universal at the atomic level, tracing this universality in the final measured XRF spectra can be extremely challenging. This difficulty stems from the complexity of accounting for all the characteristics involved in the wide variability of XRF spectra, which reflect different configurations of material states (liquid, gas, or solid), chemical compositions, bonding, and macroscopic preparations. In very complex configurations, the interactions among these parameters can make the task of tracing back the universal properties exceptionally difficult.

Given the universality of the radiation and the challenges of its traceability, a natural question arises: Is it possible to develop an Artificial Intelligence (AI) algorithm that can exploit the commonalities in the radiation process and automatically learn the corresponding pathways for traceability, thereby automating the analysis process?

In principle, the answer is affirmative, making this an interesting and relevant prospect, as mentioned in [3]. Nevertheless, several other questions arise: What will the architecture and training of such an algorithm entail? What data will it require? How universal can this algorithm be? What will be the main limiting factors, risks, and biases? Why, and under what circumstances might it fail?

In this contribution, we will attempt to address and discuss these questions. Based on new results and recent hypotheses on interpretability [4], we argue that autoencoding XRF spectra, where the latent space dimensions are constrained to be spanned by the elemental lines basis, may achieve the desired outcomes. However, a dedicated dataset (likely synthetic) and several learning methods, such as domain adaptation [5], should be incorporated into the pipeline.

Finally, we present some preliminary results in this direction by training only the encoder part of this architecture.

1.1 A X-Ray Fluorescence Primer

Just as atomic nuclei have distinctive features that differentiate elements, so do their electronic orbitals, which are substantially more accessible for observation due to their energetic properties [1],[2].

The XRF technique leverages these electronic orbital properties for elemental analysis. It involves the stimulated emission of characteristic photons of fixed wavelength (**Fig**.1), resulting from the excitation of inner electrons in the targeted atoms by external X-ray irradiation. These emitted photons are used to identify and, in some circumstances, quantify the elements present in the sample.

This process does not require sample pretreatment, and can be performed with a portable, cost-effective device. Furthermore, it is non-invasive. These features have led to XRF being extensively utilised across various fields, including astrophysics [6], material science [7], environmental monitoring [8],[9],[10] cultural heritage [11], [12], and industrial quality control [13] .

1.2 AI Challenges on XRF Analysis

Despite the fact that XRF emission lines (characterising each element) are universal (a restricted set of these lines - up to 30 KeV - can be seen at **Fig**. 1, corresponding to the set of elements shown in **Fig**. 2), the final form of the spectra detected by a XRF apparatus depends on the complex physics of photon-matter interactions. To fully understand and characterise such process, a multi-faceted approach is necessary. This involves studying the primary excitation source, the sample's composition, and the geometry of the detection system, among other factors. In more detail, all the photon-matter interactions should be known *a*

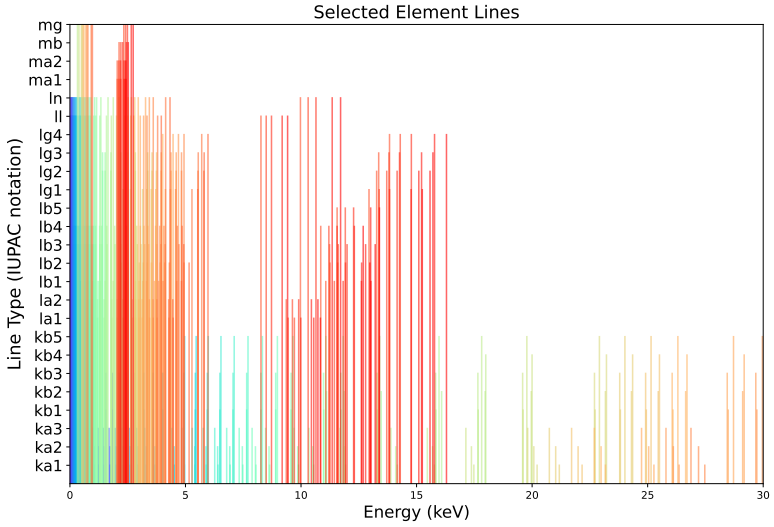

Fig. 1. Distribution of selected lines in energy. The X-axis represents energy, whereas the Y-axis has only labelling information, corresponding to the orbital of the emission line with IUPAC (International Union of Pure and Applied Chemistry) notation. One can view XRF spectra as a function of this set of lines, and the goal for automatic element detection will be to establish a bidirectional relation between the ensemble basis and observed XRF spectra.

priori and accounted for, including absorption, scattering, and fluorescence parameters. This complex scheme is deeply influenced by sample properties such as density, composition, physical and chemical states, in addition to the geometric and matrix effects[4] (see [14], and references therein).

One of the consequences of this photon-matter interaction is the significant signal attenuation suffered by emission fluorescence lines below 5 KeV[5], making it challenging to analyse low atomic number elements ($Z < 10 \div 12$) with XRF technique.

Therefore, we are in a regime where, even though the final analysis object comes from a universal known common basis, the traceability and accountability of such are extremely difficult, due to the multiple processes involved.

[4] I.e., the influence (excitation/absroption) of one element XRF spectra over another one.

[5] The reader may note in **Fig**. 1 that this is the most dense line region, making it an extremely relevant zone.

2 Methods

2.1 Related works

Given the situation discussed in the previous section, the majority of Machine Learning (ML) and AI techniques applied on XRF spectra have reasonably focused on trying to model and describe them for concrete, limited purposes. These techniques include: Dimensionality Reduction [15], [16], [17], Classification [18], [19],[20], Autoencoding [21],[22], [23], Denoising [24], Superresolution [25], etc.

Despite the advancement these techniques represent and the benefits they provide, the work is still very limited due to many factors, e.g., lower statistics, the incorporation of idiosyncratic features to the AI algorithms (in order to characterise a particular field or phenomenon), restricting their application to other scenarios, even though some of them are already incorporating fundamental parameters in their approaches (see [21]). As pointed out in [3]: "[...] *models are only relevant within the minimum and maximum observed values during training*"[6], meaning that only data within the training distribution set characteristics are correctly elaborated by the ML algorithms.

With this perspective, the aforementioned specialised scenario is highly comprehensible, since both dedicated algorithms and dataset for these tasks are presumably extremely challenging to elaborate.

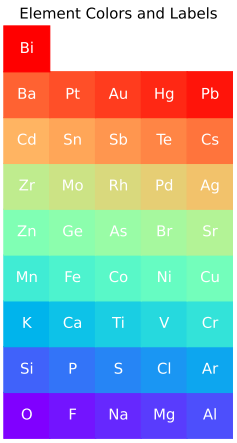

Fig. 2. Colour map for the elements in fig.1.

2.2 Proposed AI Architectures

To develop a comprehensive algorithm capable of overcoming these challenges, we must consider a few fundamental questions: What form will the artificial neural network (ANN) architecture take? Is there a simple ANN architecture that can still provide significant improvements?

It is clear that, if some physical parameters must be included, we should have a hybrid model combining them with ML techniques. Since one of our goals is to separate the XRF inputs into its constituents, sparse autoencoder are ideal candidates for the task [4]. Furthermore, in light of recent hypothesis [4], making the latent space of such autoencoder match the dimensions of the desired features can have great impact in terms of interpretability (which is highly desirable in such a complex algorithm). That is, having the latent space match basic emission element lines basis will theoretically allow neurons of the last layer of the encoder network to be superposition-free, that is *monosemantic*

[6] Even with techniques such as domain adaptation, that extend this applicability range.

(i.e., to activate only when the line is present). In other words, we hypothesise that emission elemental lines constitute an overcomplete basis for encoding XRF spectra that can be at the same time a *privileged basis*[4] (i.e. features align with neurons).

It is clear that the elements of such basis are not linear independent, since lines from the same elements are (highly) correlated. The choice of individual emission lines, instead of the element presence, lies in a presumption of simplicity and robustness of the former[7].

Ideally, in this scenario, the complex photon-matter interactions would be modelled in the outer encoder/decoder layers in such a way that different directions in their activation space will correspond to different matter and experimental configurations. Therefore, in principle, one could obtain the equivalent spectra for a certain elemental composition just by changing direction in the activation space.

3 Results

Table 1. Emission Lines Autoencoder Performance On Synthetic XRF Spectra Dataset. Pearson represents its correlation coefficient between predictions and true labels, and Guess-R denotes the guess ratio.

AI Performance Metrics for Synthetic Spectra

ANN Architecture	Pearson	Sensitivity	Specificity	Guess-R	Precision
MLP Encoder	0.76	0.65	0.99	0.95	0.92

3.1 Preliminary Results

As a toy model, we trained a classifier to serve as the encoder part of such an architecture. The model is a two layered Multi-Layer Perceptron (MLP), classifying the 556 emission elemental lines (**Fig**. 1) of the selected elements[8] in the $0 \div 30$ KeV energetic interval. The hidden layer size[9] was determined with a Bayesian hyperparameter optimisation algorithm (using Optuna [26]), resulting in 900 hidden nodes[10]. The loss function was chosen to be the Kullback-Leibler Divergence (KLD) with respect to the predicted and true emission elemental lines.

[7] Element presence will lie in a more internal layer with respect to element emission lines. Nevertheless, introducing a deeper, learnable encoding layer for element presence and concentration may give the desired classification outcome as well.

[8] Most frequent elements present on Cultural Heritage Analysis.

[9] Along with the following parameters: learning rate, gradient optimiser, dropout value, activation function and batch size.

[10] Model class and weights are available upon request at https://baltig.infn.it/fgarciaa/multiel_spectra_app and soon will be included in the python package.

The dataset used for training consists in 1.2 Million synthetic XRF spectra generated with the MULTIEL_SPECTRA python package[11]. Using a 70% for training and the rest for testing.

The performance of the model is reported in **Table** 1.

4 Discussion

X-ray Fluorescence (XRF) spectra are widely variable, complex objects reflecting the intricate underlying photon-matter interactions of atoms under X-ray irradiation. They can be seen as composed of a universal basis changed and distorted through several transformations.

As in many other fields, the existing input variability has resulted in a great number of specific AI algorithms focusing on partial aspects, with restricted applicability. We presented possible approaches and architectures that could have a wider range of applicability, in light of new interpretable hypothesis. Only the desired architecture and functionality are discussed, i.e., a sparse autoencoder with emission lines corresponding to the latent space basis.

Finally, we show the performance of a toy model of only the encoder part of such an architecture on an synthetic dataset. We have shown that even though the network can have some bias towards present lines (high specificity with respect to the sensitivity), the task is feasible. Yet, more work is needed in order to further progress in the proposed path.

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[11] https://pypi.org/project/multiel-spectra/ . Input spectra is 300 bin in the 0-30 KeV range.

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