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Ph.D. THESIS SUMMARY

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CONTRIBUȚII LA IMPLEMENTAREA UNUI SISTEM DE PROCESARE ȘI ANALIZĂ DE SEMNAL UTILIZAT ÎN SPECTROSCOPIA DE REZONANȚĂ NUCLEARĂ CUADRUPOLARĂ

CONTRIBUTIONS TO THE IMPLEMENTATION OF A SIGNAL PROCESSING AND ANALYSIS SYSTEM FOR NUCLEAR QUADRUPOLE RESONANCE SPECTROSCOPY

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Content

Chapter 1. Introduction

Recent decades have seen an unprecedented increase in global terrorism and organized crime, especially regarding drug trafficking [1,2]. In this context, numerous systems for detecting prohibited substance have been developed and installed in airports, customs, post offices and border checkpoints to combat illegal activity and increase the safety of citizens.

1.1 Presentation of the field of the thesis

The detection of prohibited substances (explosives, drugs, toxic substances) has an important place in the field of security. Therefore, a wide range of techniques and equipment has been developed for the identification of landmines or narcotics and explosives hidden in luggage or clothing.

Nuclear quadrupole resonance (NQR) is a spectroscopy technique used to analyze solids. The frequency of the response signal is strongly influenced by the chemical structure of the substance, which gives the NQR technique a very high specificity, allowing the detection of a substance by its unique radio "fingerprint" [3]. It can be applied to substances that contain quadrupolar nuclei, such as the ^{14}N isotope found in most explosives, narcotics, drugs and toxic substances.

The challenges of detecting prohibited substances by nuclear quadrupole resonance are related to the physical principle used, but also to the requirements imposed by the operational environment in which the equipment based on this technique is installed.

1.2 Aim of the thesis

The purpose of this paper is to propose hardware and software solutions to improve the detection of prohibited substances based on the nuclear quadrupole resonance technique.

From a hardware point of view, several solutions are being researched to reduce noise and compensate the factors that influence NQR detection. From a software point of view, the aim is to improve the detection by applying signal pre-processing, processing and analysis techniques. These solutions are implemented in an equipment that ensures the processing and analysis of the NQR response signal, intended for detecting illicit substances in security checkpoints.

1.3 Contents of the thesis

This paper is structured in seven chapters. The **current chap.** has an introductory role and is intended to present the topic and structure of the thesis.

Chap. 2 is a bibliographic study of the methods and equipment for the acquisition and analysis of signals for the detection of prohibited substances.

Chap. 3 focuses on signal processing and analysis in nuclear quadrupole resonance detection by performing a bibliographic study of the signal pre-processing, post-processing and analysis techniques.

Chap. 4 presents a comparative study of the signal processing and analysis algorithms in order to verify and create its own benchmark for the performance indicators.

Chap. 5 aims to present contributions regarding the development of signal analysis algorithms based on machine learning in order to improve the detection of substances using the NQR technique. The optimization of excitation sequences by using black-box techniques is also being investigated.

Chap. 6 presents the signal processing and analysis system developed for NQR detection.

Chap. 7 concludes this paper by presenting the final conclusions of the research performed and summarizing the contributions. Also, it presents the list of publications and contributions, and finally the perspectives for future development.

Chapter 2. Methods and equipment for the acquisition and analysis of signals for the detection of prohibited substances

This chapter aims to perform a bibliographic study of the methods and equipment for the acquisition and analysis of signals for the detection of prohibited substances, starting from the general framework of the detection theory and focusing on nuclear quadrupole resonance spectroscopy.

2.1 Signal detection and estimation theory

Signal detection theory, also called detection theory, is a field that aims to identify the occurrence of an event with a certain degree of certainty and to extract information about it [4].

The correct identification of a signal from an observation is called success. The success rate is defined as the ratio between the number of correct identifications and the total number of trials. Similarly, the false alarm rate is defined as the ratio of the number of false alarms to the total number of trials. By graphically displaying the success rate depending on the false alarm rate for different values of the decision criterion, the receiver operating characteristic (ROC) is obtained.

2.2 Techniques for detecting prohibited substances

The detection of prohibited substances is an application of the detection theory and has an important place in the field of security. The detection of explosives is of greater importance due to the material damage and casualties that they can cause either accidentally or when used for terrorist purposes. The techniques used for the detection of explosives are classified according to the target sought, in techniques for bulk and trace detection, respectively [5].

2.3 Nuclear quadrupole resonance spectroscopy

The nuclear quadrupole resonance technique is applied in the field of security for the bulk detection of prohibited substances. The NQR phenomenon occurs in solids

(powders or crystalline substances) composed of atoms with quadrupolar nuclei having a spin quantum number greater than 1.

The technique is based on the excitation of the scanned substance with a train of electromagnetic pulses and the reception of the response signal emitted by it when the excitation ceases. The applied energy determines the reorientation of the quadrupolar nuclei, and when the excitation ceases, they return to the initial states, releasing the excess energy in the form of radio frequency (RF) radiation having unique characteristics for each substance. The process is illustrated in Fig. 2.1.

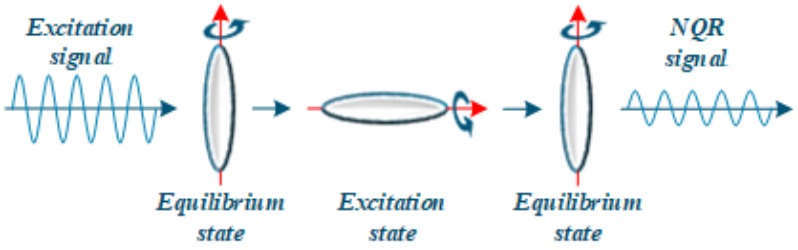

Fig. 2.1 *NQR excitation process*

The response signal generated by the nuclei is called free induction decay (FID) and is characterized by the transversal relaxation time (T_2). The time required for the excited nuclei's orientations to return to their initials states is denoted T_1 and is called longitudinal relaxation time. In order to obtain a stronger response, pulse excitation aims to receive the echo signals generated after the excitation with several successive pulses of different durations.

Taking into account the NQR principle, several factors that affect the signal detection can be mentioned, such as: detection distance, excitation frequency, receiver noise, substance temperature, substance nature, substance quantity, RF interferences, quality factor of the detection circuit.

As illustrated in Fig. 2.2, the NQR signal acquisition (or detection) methods are classified into two categories, depending on the response intensity of the investigated substances: direct and indirect detection methods [6]. Pulse methods are the most used, because they increase the signal-to-noise ratio (SNR) and decrease the acquisition time. They are based on pulse sequences that are fine-tuned to each substance in order to maximize the response signal. The types of sequences used are classified according to the number of pulses in FID and multi-pulse sequences. The latter, in turn, are classified in SLSE (Spin-Locked Spin Echo) and SSFP (Steady-State Free Precession) sequences, respectively.

There are many types of equipment used in NQR spectroscopy, consisting of laboratory equipment, detectors made at the laboratory or prototype level, as well as commercial equipment. Among the laboratory modules, it is worth mentioning the excitation and acquisition systems Spincore [7] and Redstone [8], produced by the American companies SpinCore Technologies and Tecmag, respectively.

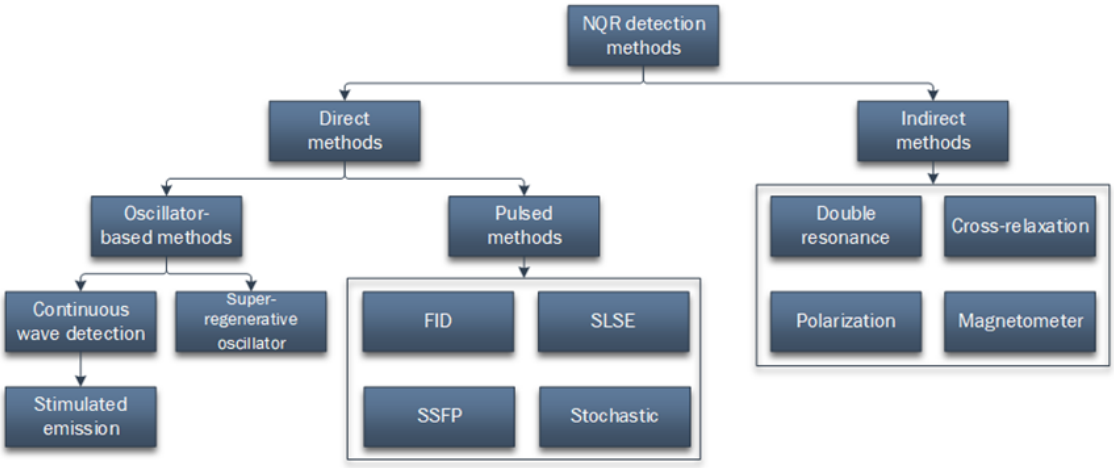

Fig. 2.2 *NQR detection methods (FID - free induction decay, SLSE - spin-locked spin echo, SSFP - steady-state free precession)*

At the laboratory level, several portable equipment have been developed for mine detection [9,10], drug validation [11,12] and identification of explosive substances [13,**Error! Reference source not found.**]. At the commercial level, the following detectors can be mentioned: QScan QR 500, developed by the American company InVision Technologies, T-3-03, made by the Australian company QRSciences, NQR-15, made by the Russian company Logys, the detector produced by the Chinese company Anhui Qilootech Photoelectric Technology and SEEQR, developed by the American company SEEQR Security [6]. However, existing solutions are developed at the laboratory level and may not meet actual operational requirements. Also, there are currently no commercially available systems on the global market because there is no up-to-date information about them and some manufacturers no longer exist.

Chapter 3. Signal processing and analysis techniques used in nuclear quadrupole resonance

This chapter aims to perform a bibliographic study of the signal pre-processing, post-processing and analysis techniques used in nuclear quadrupole resonance detection applications.

3.1 Classification of signal processing and analysis techniques

The signal processing and analysis techniques used in nuclear quadrupole resonance generally aim to increase the signal-to-noise ratio and to identify the response signal. Fig. 3.1 illustrates the classification of the signal processing and analysis techniques based on the location in the reception chain where they are implemented.

The pre-processing techniques aim to increase the signal-to-noise ratio during signal acquisition, while post-processing techniques use the signal obtained after pre-processing and analyze it in order to extract data from it, such as the presence of the response signal or its characteristics.

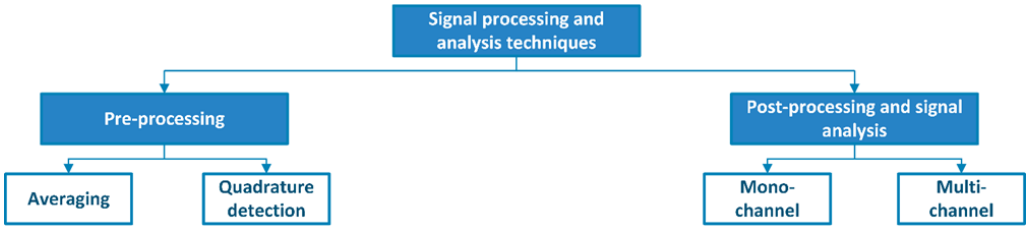

Fig. 3.1 *Classification of the signal processing and analysis techniques*

3.2 Pre-processing techniques

The most used pre-processing techniques are: signal averaging and quadrature detection. Signal averaging is utilized to separate a repetitive signal from noise without distorting it [14]. This involves the averaging of the substance's responses and is based on the fact that the useful signal will add coherently and the noise, incoherently. The technique is effective if the noise is random, with zero average value and uncorrelated with the useful signal, in which case the SNR increases with the square root of the number of scans.

Quadrature detection is used to distinguish the response signal's components with frequencies lower or higher than the excitation frequency. It also offers the advantage of spectrally limiting the noise by low pass filtering.

3.3 Post-processing and analysis techniques

These techniques are classified into several main categories. Spectral estimation algorithms aim to estimate the power spectrum in the case of a small number of data sets. Initially, statistical algorithms were proposed as an alternative to signal energy-based detectors. They use a data model and aim to estimate its unknown parameters. Their development was supported by the need to increase the robustness to interferences and uncertainties regarding unknown parameters, respectively to exploit more information from the received signal. Interference rejection techniques aim to reduce the influence of disturbing signals, performing or not performing the detection. In the latter case, they are complemented by a detection algorithm which is usually a statistical solution. Adaptive filtering techniques also aim to reduce noise and are mentioned separately to distinguish the classic algorithms applied in NQR from the new ones that were developed specifically for this application. Machine learning classification techniques have been proposed relatively recently for the recognition of the NQR response in different noise conditions. They aim to identify some signal features in order to associate it to a detection class [15].

Chapter 4. Modeling and simulation of signal processing and analysis algorithms

This chapter aims to carry out a comparative study of the signal processing and analysis algorithms used in nuclear quadrupole resonance detection applications, in order to verify and create its own benchmark for the performance indicators.

4.1 Modeling of signals in nuclear quadrupole resonance spectroscopy

The FID signal model was proposed in [16] as a sum of damped sinusoids in noise. Similarly, the echo model is defined as two symmetric FID signals [17]. This model has been extended to represent the entire echo train [18].

Although it has been used in the development of NQR detection algorithms, the presented echo signal model is not found in theory [19], nor in practical experiments [20]. Specifically, the sharp shape of the echoes does not resemble the response of a substance excited by the SLSE technique. For a more accurate simulation of the NQR response, an echo signal model obtained by applying the Kaiser window is proposed.

A category of noise often used in the study of algorithms applied in NQR spectroscopy is the white and additive Gaussian noise [21]. Often, noise is represented by a small-order autoregressive model estimated from the acquired data [16,17]. Also, radio frequency interference is modeled as sums of complex sinusoids.

4.2 Study of signal processing and analysis algorithms

As presented in the previous chapter, the recent research effort in this field has focused on signal analysis algorithms specific to machine learning. Thus, the following machine learning classification algorithms are proposed for analysis: the k-nearest neighbors, support vector machine, neural network and ensemble of classifiers. Also, statistical algorithms have provided good results in NQR detection and the RETAML and LSETAML algorithms [22] are proposed for this study. In order to give a higher degree of novelty to this comparative analysis, a multi-criteria algorithm (MFD) for detection is proposed and presented in three optimized variants [23-25].

4.3 Comparative analysis of signal processing and analysis algorithms

The purpose of this chapter is to compare the signal processing and analysis algorithms under the same conditions (data sets and signal-to-noise ratio). Taking into account the factors that influence the NQR response signal detection, but also the operational aspects, the following indicators are proposed in the comparative analysis: classification performance and processing time.

The analysis procedure, graphically illustrated in Fig. 4.1, aims to create its own reference regarding the performance indicators in order to compare the new solutions developed for NQR detection. In this sense, the procedure aims to generate the ROC curve in different simulation scenarios, focusing on evaluating the robustness of the algorithms at different noise levels (at SNRs -25, -30, -35, -40, -45 and -50 dB) and in the case of a resonant frequency offset due to the temperature variation of the substance (with the temperature of the substance 10 K lower than in previous cases).

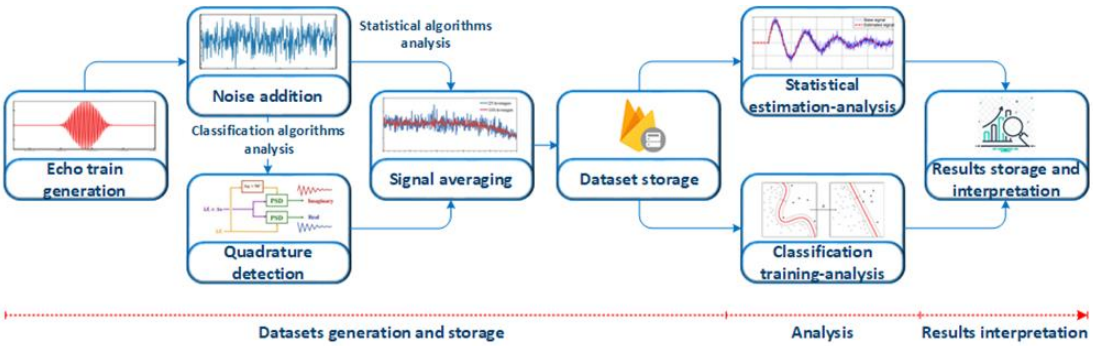

Fig. 4.1 *Comparative analysis procedure*

The analysis has shown that the classification algorithms are more robust to the influence of noise and are clearly superior to the statistical solutions, and can be used in scenarios where the SNR is about 15 dB lower than in the case of statistical algorithms.

Tab. 4.1 presents the detection accuracies for the investigated algorithms. They are determined at the optimal detection thresholds and are expressed as percentages. The table confirms the superiority of the ensemble of classifiers.

In terms of processing times, the results are obtained by averaging 100 measurements, in order to reduce the influence of the central processing unit's load. The final processing time is thousands of times longer for the statistical algorithms than for the classification solutions. Given that in a real application the final processing time is added to that of the signal acquisition (the actual scan), the classification algorithms offer a significant advantage in an operational context.

Tab. 4.1 *Detection accuracy for the investigated algorithms (T – temperature, LS – LSETAML, RE - RETAML)*

SNR [dB] / ACC	T [K]	LS	RE	MFD	MFD GA	MFD CS	k-NN	SVM	NN	EC
-50	297	48.7	48.4	49.0	51.0	50.0	57.0	44.4	56.6	62.0
-45		49.2	49.0	48.3	51.6	50.0	69.8	75.0	73.6	77.4
-40		52.4	53.3	47.5	51.6	50.0	93.6	97.2	95.0	98.2
-35		63.8	64.0	54.6	54.3	50.8	100	100	99.8	100
-30		81.8	87.8	80.1	79.6	81.8	100	100	100	100
-25		99.2	99.5	99.8	100	100	100	100	100	100
-50	287	49.9	49.8	47.7	50.2	50.0	38.4	47.6	52.0	47.4
-45		50.5	50.6	47.6	50.5	50.0	73.6	67.0	88.6	96.0
-40		52.1	53.3	46.3	51.7	50.0	92.8	91.8	93.0	96.4
-35		57.7	63.5	54.4	54.2	50.9	100	100	99.8	100
-30		76.0	82.4	79.9	79.4	82.3	100	100	100	100
-25		96.7	97.6	99.9	100	100	100	100	100	100

The comparative study's results have showed the better performance of the classification algorithms compared to the statistical ones regarding the detection of the NQR signal. Therefore, future research will focus on classification algorithms specific

to the field of machine learning in order to achieve an acceptable detection performance in real scenarios.

Chapter 5. Development of signal analysis and detection optimization algorithms

This chapter aims to present contributions in the development of signal analysis algorithms based on machine learning and the investigation of optimization techniques in order to improve the detection of substances using nuclear quadrupole resonance.

5.1 Analysis of NQR response signals

In order to obtain the most relevant results, the algorithms were developed based on real data, which were acquired using a custom-designed spectrometer [26]. The data were acquired for the following substances: 130 g paracetamol (precursor found in narcotics combinations) and 150 g sodium nitrite (toxic substance in large quantities). Also, the acquisitions were performed in various scanning conditions (scenarios), as follows:

1. At different distances between substances and the excitation/detection coil;
2. In the presence of sand;
3. With a substance temperature offset;
4. With a detuned resonant circuit;
5. Using a small quantity of substance;
6. In the substance's absence.

The feature is a measurable individual property or an observed phenomenon and is used by the signal analysis algorithm to make the prediction (classification). Given that discrimination in the presence of the substance's response is performed by humans primarily by evaluating the spectral diagram, then this is proposed as a feature.

In order to increase the probability of detection and reduce false alarms, some additional features resulting from the visual analysis of the spectra obtained in each scan scenario are proposed. Thus, the following characteristics are proposed: maximum spectral peak frequency (SMF), the ratio between the maximum peak amplitude and the spectral average (SAMP), the Pearson correlation coefficient with a reference spectrum (PCC), the ratio between the central bandwidth's average and that of the sidebands (SRAR) and 11 signal features described in the literature (C1-C11) [27].

An exploratory analysis of the data set was performed in terms of the signal features defined above. The distributions of the feature's values were visualized in the form of histograms and the symmetrical, unimodal distribution of SMF was observed, explained by the fact that the response frequency is close to the reference, and in the case of acquisitions in the substance's absence it is determined by random noise. SAMP and SRAR present bimodal distributions and indicate that most substances in the substance's absence give approximately equal values for these features, while acquisitions in the substance's presence give higher but more scattered values. The PCC has a multimodal distribution, noting that acquisitions in the substance's absence are grouped at lower values and the others at higher values.

An alternative method of observing the correlations between the features is to use the Heatmap matrix. Fig. 5.1 illustrates the correlation matrix for samples 384-640, representing the first 128 samples to the left and right of the reference frequency. The latter is located in sample 512, in the middle of the matrix. As expected, the samples in the center band are poorly correlated with the most distant ones, due to the fact that the response signal is located in a narrow band around the reference frequency.

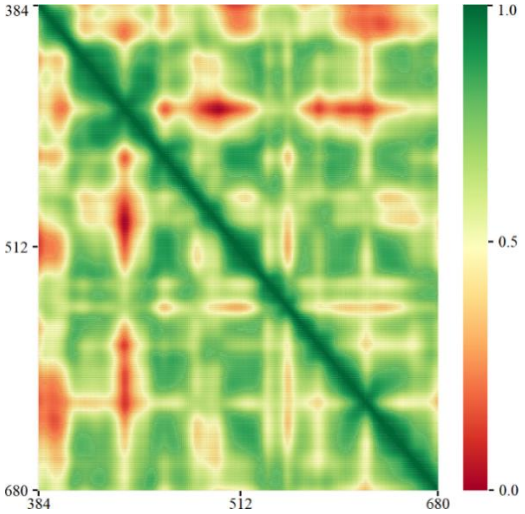

Fig. 5.1 *Heatmap matrix showing the correlation of the spectral diagram's samples 384-640*

5.2 Development of signal analysis algorithms

The development starts from existing signal analysis algorithms and aims to modify them for the detection of NQR response signals and to comparatively evaluate them in order to choose the most performant solution. The following deep learning solutions are chosen for this study due to the high performance reported and in order to cover several categories of algorithms: DNN, CNN, LeNet-5, AlexNet, VGG-16, ResNet-34, LSTM, CNN LSTM and ConvLSTM [28]. Some of them have been developed for image recognition and will be modified for 1D signal classification.

The implementation is performed in the integrated development environment Spyder 3.3.6, using Python 3.7.3, running in the docker virtual environment Anaconda 4.7.10. The workflow followed for the development and evaluation of the signal analysis algorithms is illustrated in Fig. 5.2.

The training and evaluation stage aims to train the algorithms on real data and analyze the results obtained in order to choose the most performant solution. After training the models, they are evaluated on the test set by calculating the ROC curve and measuring the accuracy of the model. Following the determination of the optimal detection thresholds for all algorithms, their evaluation on the test set has provided the results presented in Tab. 5.1.

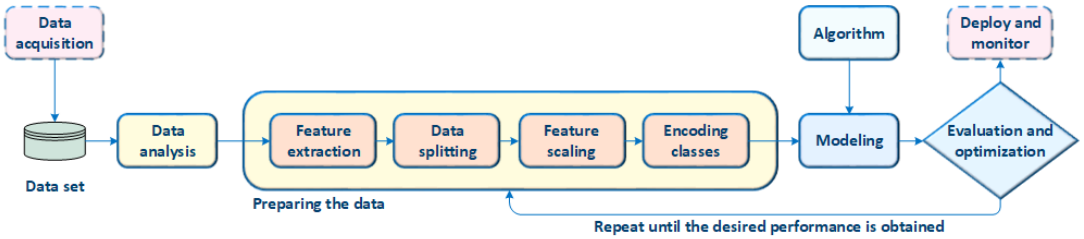

Fig. 5.2 *The workflow followed for the development and evaluation of the signal analysis algorithms*

All algorithms have high and close accuracy values. Despite the overfitting, ResNet-34 has achieved good performance, but, although it is the most complex algorithm developed, its accuracy is the lowest. DNN also provided the second highest accuracy and is a simpler algorithm than the others. This indicates that a simpler algorithm is recommended for this type of detection. In general, the more complex the signal, the more complex the algorithm used and the larger the data set required. The very good results provided by the recurrent networks is noted.

Tab. 5.1 *Detection accuracy obtained with developed algorithms*

Algorithm	**Accuracy [%]**
DNN	99.6
CNN	94.7
LeNet-5	94.9
AlexNet	**99.8**
VGG-16	99.3
ResNet-34	93.7
LSTM	99.3
CNN LSTM	99.6
ConvLSTM	99.6

The analysis performed at this stage has shown that the AlexNet algorithm achieves the highest detection accuracy of 99.8%, and is therefore proposed for optimization. The use of a more complex algorithm, such as AlexNet, for the analysis of a relatively simple signal can ensure robustness to variations in noise or useful signal compared to the data set used.

The first optimization step is the analysis of errors (wrong classifications) in order to identify their causes. The percentage of errors in the case of the AlexNet model is 0.2%, representing 4 false negatives and 4 false positives. The analysis of the errors has shown that they are caused by the detection limit of the spectrometer. The optimization of the algorithm is proposed to reduce the number of misclassifications. Thus, the model is retrained for different combinations of the following parameters: number of training epochs, batch size, optimization algorithm, learning rate, weight initialization function, activation function, number of neurons in the last layers, filter size, pool size used by the MaxPooling1D function.

Finally, the value that provides the highest score was identified. Each result was obtained by 2 cross-validations of the data set, which use randomly selected training/test

sets and average the results. However, the model compiled with these values reached an accuracy of 99.7%, lower than the initial one. This can be explained by the fact that these parameters were analyzed individually, and not correlated. This optimization approach did not provide a better model. However, by successively training the initial network, a better model was obtained. This is explained by the use of better initialization values, which are chosen randomly and are not under the control of the simulation program. The model reached an accuracy of 99.9%, having only 4 false positives and no false negatives which is also significant from an operational point of view. Hereinafter, it will be called the optimized model.

In order to obtain a more realistic result, 10 cross-validations were performed. The values obtained are located in the range of 99.2-99.4%, with two outliers and an average of 99.31%, a good value that allows the use of the model in the operational (real) environment [28].

The model was compared with the ensemble of classifiers, identified in the previous chapter as the most performant algorithm. In the case of simulated data, the ensemble of classifiers has an accuracy of about 4% higher than AlexNet. But in the case of real data AlexNet is superior. The weaker result in simulated data can be explained by the fact that this set is more predictable and AlexNet is suitable for more complex signals (containing nonlinearities due to the spectrometer, as well as internal and external interferences, which were not simulated).

5.3 Solutions to improve NQR detection

This section seeks to investigate solutions to improve NQR detection based on the optimized AlexNet algorithm.

Transfer learning can allow the generalization of the algorithm developed in the previous section to different detection equipment. The need for this approach is imposed by the fact that existing NQR spectrometers may have different transfer characteristics and noise conditions, due to the components, electronic components and their construction (shielding materials, modules locations). Thus, the received signal may contain spectral peaks due to the hardware components, which differ from one equipment to another and may cause misclassifications.

The AlexNet network is trained on the source data set and the knowledge obtained is transferred using the data acquired from the target spectrometer, on which the model will run. This requires the existence of at least two different spectrometers to acquire the source data and improve the initial model with the data from the target equipment. Since there is only one spectrometer available, the simulated data set will be used to train the network and the real one to improve it using transfer learning.

Taking into account that the simulated and real data sets are similar and aiming to study the influence the number of trained layers has on the detection performance, the accuracy obtained in case of training different numbers of layers is investigated. The analysis indicated that the model learns better the target data set when fewer layers are frozen. The best model is obtained by freezing the first 3 layers and reaches an accuracy of 99.8%, providing the lowest number of false negatives. It was evaluated on the actual test set, reaching an accuracy of 99.85%, approximating that of the optimized model

developed in the previous section. When reevaluating the solution on the simulated test set, it reached an accuracy of 85.08%, about one percentage point lower than the initial one.

NQR detection is affected by disturbing signals represented by noise and radio frequency interferences. Thus, a way to improve detection is to eliminate these signals. One of the machine learning solutions that can be applied in this situation is represented by autoencoders (AE).

The study aims to perform signal denoising using AE. Two different architectures are investigated: (1) autoencoder based on artificial neurons (DNN-AE) and (2) convolutional autoencoder (ConvAE). The spectral diagrams of the acquired signals are used for training the autoencoders and the resulting models are applied before the AlexNet network for denoising, as shown in Fig. 5.3.

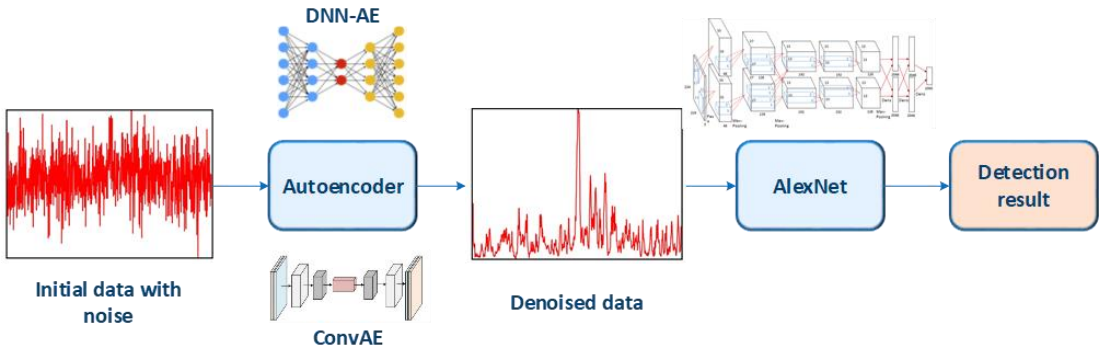

Fig. 5.3 *The structure of the network composed of the autoencoder and AlexNet*

In order to perform the training, an additive Gaussian noise with zero mean and unit variance is added to the initial signals. A signal-to-noise ratio study was performed to visualize the model's performance in different noise conditions (-30, -25, -20, -15, -10, -5, 0, 5, 10, 15 and 20 dB). Fig. 5.4 illustrates the average accuracy obtained at each SNR after 10 cross-validations.

The performance of the AlexNet model is very good for SNRs over 10 dB, but decreases quickly to approx. 60% at -30 dB. As seen, the autoencoders significantly improve the detection, increasing the accuracy by approx. 10% and 20%, respectively, at -30 dB. The growth is maintained throughout the investigated range.

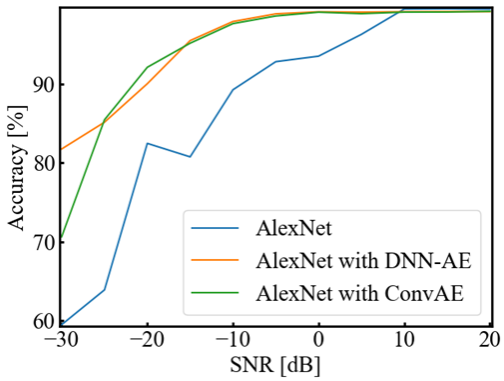

Fig. 5.4 *Average accuracy of the denoising AlexNet model after 10-fold cross-validation*

5.4 Optimization of excitation sequences by using black-box techniques

NQR detection is highly dependent on the excitation sequence's parameters and any misadjusted values can severely impact the SNR and even prohibit detection.

The challenge of finding the optimal excitation sequence parameters can be viewed as an optimization problem. In order to avoid using a mathematical model that may not take into account all the variables that influence the results (e.g., due to different substances and measurement conditions), black-box optimization is considered. The following methods are used in this study: Gaussian processes, random trees, extra trees, gradient boosted trees and tree Parzen estimators. In addition to the previous methods, this study also uses an evolutionary algorithm called the covariance matrix adaptation evolution strategy, respectively the simulated annealing algorithm. The random search technique is also applied for comparison.

The techniques were evaluated comparatively by performing mono- and multi-parametric optimizations on sodium nitrite. Finally, the optimization provides a SNR increase of 8 dB, as seen in Fig. 5.5.

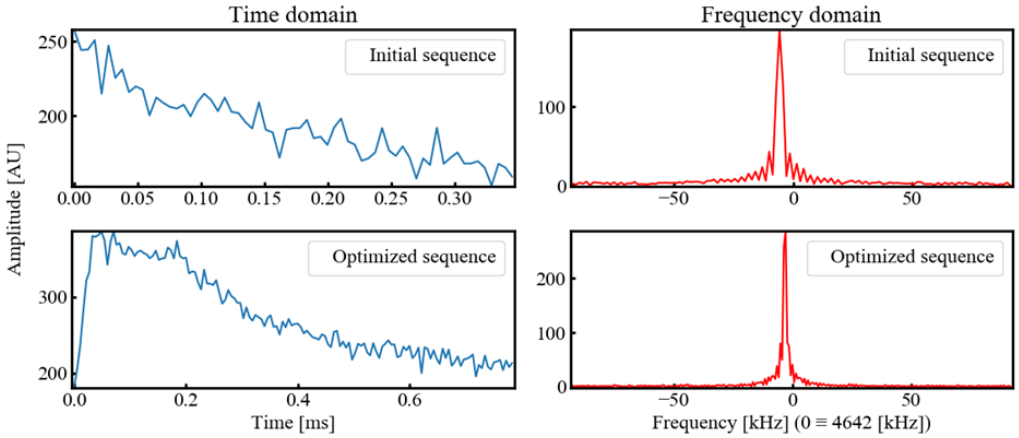

Fig. 5.5 *NQR response signal with the initial (top) and optimized (bottom) excitation sequences (AU – arbitrary units)*

Chapter 6. Implementation of a signal pre-processing, processing and analysis system used in nuclear quadrupole resonance

This chapter aims to present contributions in the implementation of a detection system based on the nuclear quadrupole resonance technique.

6.1 Hardware implementation of the detection system

The developed system represents an NQR spectrometer based on multi-pulse excitation in the frequency band 0.4-6 MHz and designed to scan envelopes and small parcels for prohibited substances. It integrates several solutions to compensate for factors influencing NQR detection and to reduce noise, such as fiber optic transmission, during data acquisition, of the control signals to the modules located near the analog circuits, and powering the low-noise amplifiers from batteries that are electrically isolated from the rest of the system. The general block diagram of the spectrometer is illustrated in Fig. 6.1.

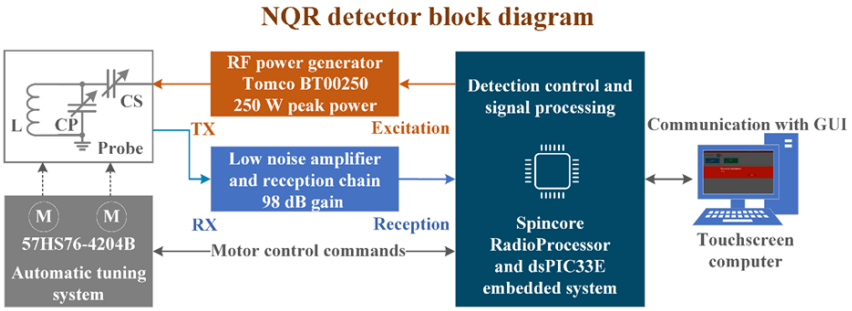

Fig. 6.1 *NQR detector general block diagram*

6.2 Software implementation of the detection system

The general software architecture is illustrated in Fig. 6.2.

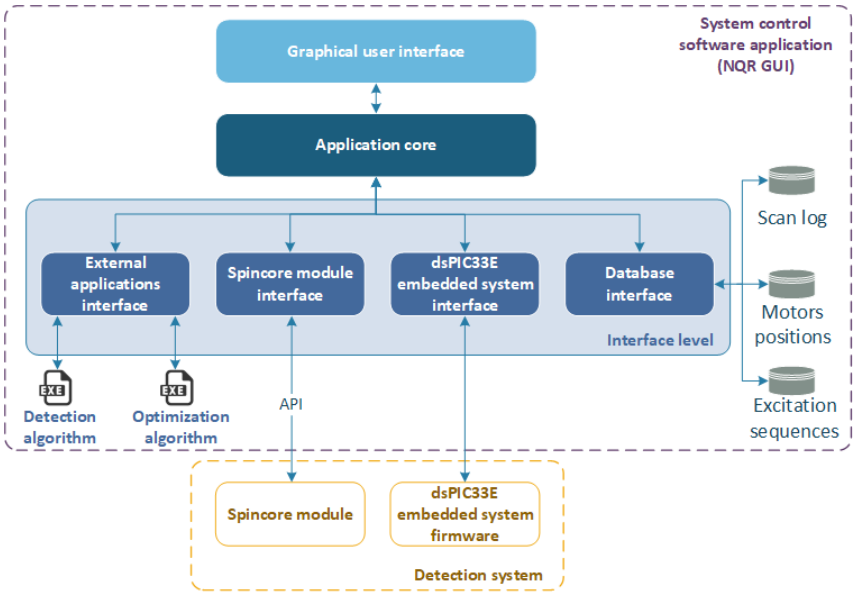

Fig. 6.2 *Detection system software architecture*

The software component of the NQR detection system is responsible for controlling its operation, monitoring it and allowing the user to operate the detector. It consists of two software sub-components: the system control software application (also called NQR GUI) and the embedded system firmware.

6.3 Evaluation of the detection system

The RF switch's capability to block the reception of the excitation signal by the reception chain was evaluated and a good noise rejection factor was obtained, which varies between 46-50 dB in the investigated frequency band.

The system was evaluated on sodium nitrite and paracetamol by measuring the distance and substance quantity in correlation to the excitation power. It was shown that at an excitation power of 7 W, the maximum detection distance was 2 cm for paracetamol and 6 cm for sodium nitrite. The minimum limit of detection was 1 g of sodium nitrite and 15 g of paracetamol at the same power. Also, the solutions implemented to improve the detection have increased the signal-to-noise ratio by approximately 31 dB compared to a prototype detector that does not integrate such solutions.

The detection system was presented nationally and internationally, being awarded a gold medal at the exhibitions in Geneva and Barcelona in 2017 [29]; several patent applications were filed, and in 2019 a Romanian patent was granted [30].

Chapter 7. Conclusions

This paper aimed to propose hardware and software solutions to improve the detection of prohibited substances using the nuclear quadrupole resonance technique. The paper brings original contributions and presents interdisciplinary research covering the following areas: electronics, physics, computer science.

7.1 Results

In **Chap. 1**, I made an introduction to the issues specific to this paper's field and I highlighted both its purpose and its content.

In **Chap. 2,** I made a review of the methods and equipment for the acquisition and analysis of signals for the detection of prohibited substances.

Chap. 3 aimed to perform a literature review of the signal processing and analysis techniques used in substance detection applications based on nuclear quadrupole resonance.

In **Chap. 4**, I performed a comparative study of the signal processing and analysis algorithms used in NQR detection applications, in order to verify and create my own benchmark for the performance indicators. I showed that the classification algorithms are superior to the statistical solutions and MFD, achieving the best detection performance in all simulated scenarios and regarding all the evaluated indicators.

Chap. 5 aimed to present contributions in the development of signal analysis algorithms based on machine learning in order to improve the NQR detection of substances. I selected and implemented in a dedicated programming environment 9 deep learning algorithms and I identified the AlexNet algorithm as the most performant, reaching an accuracy of 99.8% on real data consisting of 20,000 signal acquisitions. The model obtained after optimizing the algorithm achieved a significant performance from an operational point of view, with an accuracy of 99.9%, having only 4 false positives and no false negatives. I proposed the generalization of the algorithm using transfer learning and the application of autoencoders for signal denoising. Finally, I proposed several black-box optimization techniques for adjusting the excitation sequence.

Chap. 6 aimed to present contributions in the implementation of a system for signal pre-processing, processing and analysis based on the nuclear quadrupole resonance technique. Thus, I presented the developed system both in terms of hardware and software. I proposed and implemented several noise reduction solutions during signal acquisition, and finally evaluated the detection system on sodium nitrite and paracetamol.

7.2 Successful and unsuccessful research activities

This section summarizes the research activities carried out during the doctoral period for creating this work and also mentions their results.

7.3 Original contributions

From the point of view of the research carried out during the doctoral period, the original contributions (of a scientific and technical nature) are the following:

Bibliographic studies and scientific and technical reviews

1. I made a review of the methods and equipment for the acquisition and analysis of signals for the detection of prohibited substances, presenting their advantages and disadvantages [13,16];
2. I made a review of nuclear quadrupole resonance spectroscopy, presenting its applications, as well as the laboratory and commercial equipment [6,16];
3. I made a review of the signal processing and analysis techniques applied in nuclear quadrupole resonance spectroscopy [12,2,17,26,27];
4. I proposed a new classification of the signal processing and analysis techniques applied in nuclear quadrupole resonance spectroscopy, presenting the relationships between them and the chronology of their development, respectively [2,17];

Proposals, developments, generalizations or revisions of models and performance indicators, algorithms and techniques for optimized signal processing

5. I performed a comparative study of the signal models that can be used to represent a real echo [9];
6. I proposed a new signal model that better approximates the real shape of an echo [18];
7. I proposed a new multi-criteria algorithm (MFD) for performing NQR detection [11,18,28];
8. I proposed 1040 signal features for the application of artificial intelligence algorithms in order to perform NQR detection [4,19];
9. I proposed the application of deep learning techniques to perform NQR detection [4,19];
10. I proposed the generalization of the AlexNet algorithm using transfer learning [5,19];
11. I proposed the improvement of the AlexNet algorithm by applying denoising autoencoders [5,19];
12. I proposed the application of black-box optimization techniques to identify the optimal excitation sequence [3,19];
13. I proposed two new indicators to assess the system's detection limits [1,20];
14. I proposed hardware solutions to improve the signal-to-noise ratio [1,20];

Implementations proposed and validated by simulation

15. I evaluated the influence the number of trained layers has on the detection performance when generalizing the AlexNet algorithm using transfer learning [5,19];
16. I evaluated the detection performance at several signal-to-noise ratios when

applying the denoising autoencoders [5,19];

17. I performed a comparative study of the main NQR signal processing and analysis techniques and I highlighted the superiority of the ensemble of classifiers [2,18];

Implementations proposed and validated experimentally

18. I performed the acquisition and statistical analysis of a data set for the development of NQR detection algorithms [19];
19. I performed the optimization of the MFD solution using a genetic algorithm [7];
20. I performed the optimization of the MFD solution using a clonal selection algorithm [8];
21. I performed a comparative analysis of several deep learning techniques and highlighted the AlexNet algorithm as the most performant [4,19];
22. I optimized the AlexNet algorithm, reaching the detection accuracy of 99.9% and allowing its application in the operational environment [4,19];
23. I performed mono- and multi-parametric optimizations of the excitation sequence using black-box techniques [3,19];
24. I implemented a system for signal pre-processing, processing and analysis used in nuclear quadrupolar resonance and I have evaluated its performance [1,20,22,23,24,25,28];
25. I developed a software application for performing and optimizing the NQR detection [1,20];
26. I have implemented several hardware solutions to improve the signal-to-noise ratio [1,20];
27. I performed a study regarding the improvement of the signal-to-noise ratio brought by the pre-processing techniques [10,20].

7.4 Original publications list

Papers published during the doctoral research program or in the process of publication

Papers published in Clarivate Analytics WoS indexed journals

[1] **C. Monea**, G.V. Iana, S. Ionita, L.M. Ionescu, S.A. Zaharia, S. Ilie, N. Bizon, *An optimized NQR spectrometer for detection of prohibited substances*, **Measurement**, vol. 151, p. 107158, 2019, ISSN 0263-2241, IF 3,364 (2019), Q1, WOS:000500942200092, https://doi.org/10.1016/j.measurement.2019.107158.

[2] **C. Monea**, *A review of NQR signal processing and analysis techniques*, **Journal of The Franklin Institute**, vol. 357, no. 17, pp. 13085-13124, 2020, ISSN 0016-0032, IF 4,036 (2019), Q1, WOS:000586803400050, https://doi.org/10.1016/j.jfranklin.2020.09.013.

[3] **C. Monea**, *Optimization of NQR excitation sequences using black-box techniques*, **Journal of Magnetic Resonance**, vol. 321, p. 106858, 2020, ISSN 1090-7807, IF 2,624 (2019), Q2, WOS:000596612900011, https://doi.org/10.1016/j.jmr.2020.106858.

[4] **C. Monea**, *Nuclear quadrupole resonance response detection using deep neural networks*, **Expert Systems with Applications**, vol. 182, p. 115227, 2021, ISSN 0957-4174, IF 5,452 (2019), Q1, https://doi.org/10.1016/j.eswa.2021.115227.

Papers in the review phase for publication in Clarivate Analytics WoS indexed journals

[5] **C. Monea**, *Enhancing deep learning nuclear quadrupole resonance detection using transfer learning and autoencoders*, **Expert Systems with Applications**, submitted in May 2021, ISSN 0957-4174, IF 5,452 (2019), Q1.

Papers published in conferences indexed in international databases (Clarivate Analytics WoS, IEEE Xplore)

[6] **C. Monea**, N. Bizon, *The use of nuclear quadrupole resonance spectroscopy for detection of prohibited substances: Techniques and equipment*, **11th International Conference on Electronics, Computers and Artificial Intelligence (ECAI)**, Romania, pp. 1-6, 2019, WOS:000569985400175.

[7] M. Oproescu, V.G. Iana, **C. Monea**, *Application of genetic algorithm for optimization of NQR signal detection*, **11th International Conference on Electronics, Computers and Artificial Intelligence (ECAI)**, Romania, pp. 1-4, 2019, WOS:000569985400010.

[8] V.G. Iana, **C. Monea**, M. Oproescu, *The use of the clonal selection algorithm for NQR signal detection optimization*, **11th International Conference on Electronics, Computers and Artificial Intelligence (ECAI)**, Romania, pp. 1-4, 2019, WOS:000569985400111.

[9] **C. Monea**, *Improved signal models for nuclear quadrupole resonance spectroscopy*, **12th International Conference on Electronics, Computers and Artificial Intelligence (ECAI)**, Romania, 2020, WOS:000627393500063.

[10] **C. Monea**, *Study of signal pre-processing techniques in nuclear quadrupole resonance spectroscopy*, **12th International Conference on Electronics, Computers and Artificial Intelligence (ECAI)**, Romania, 2020, WOS:000627393500114.

Papers published in conferences in the process of indexing in international databases (Clarivate Analytics WoS, IEEE Xplore)

[11] **C. Monea**, G.V. Iana, S. Ionita, M. Oproescu, *Multi-criteria decision algorithm for NQR signal detection*, **25th International Conference on Applied Electronics (AE)**, Czech Republic, 2020.

Papers published in journals indexed in other international databases (Copernicus, Google Scholar)

[12] **C. Monea**, *Signal processing and analysis methods in nuclear quadrupole resonance spectroscopy*, **Journal of Electrical Engineering, Electronics, Control and Computer Science**, vol. 4, no. 2, pp. 1-8, 2018, ISSN 2457-7812, https://jeeeccs.net/index.php/journal/article/view/98/84.

[13] **C. Monea**, *Techniques and equipment for detection of prohibited substances: A brief overview*, **Journal of Electrical Engineering, Electronics, Control and Computer Science**, vol. 4, no. 4, pp. 7-16, 2018, ISSN 2457-7812, https://jeeeccs.net/index.php/journal/article/view/109/91.

Papers published before doctoral admission

[14] G.V. Iana, **C. Monea**, *Coin recognition system based on a neural network*, **Proceedings of the 2014 6th International Conference on Electronics, Computers and Artificial Intelligence (ECAI)**, Romania, pp. 13-18, 2014, WOS:000380489500061, https://doi.org/10.1109/ecai.2014.7090172.

[15] **C. Monea**, G. Iana, *Coin signal recognition based on neural networks*, **Scientific Bulletin of University of Pitesti**, vol. 14, no. 1, pp. 19-24, 2014, ISSN 2344-2158, www.shorturl.at/uzCK5.

Research reports

[16] **C. Monea**, *Metode și echipamente de achiziție și analiză a semnalelor pentru detecția substanțelor interzise*, Doctoral research report no. 1, Dec. 2018.

[17] **C. Monea**, *Tehnici de procesare și analiză de semnal utilizate în rezonanța nucleară cuadrupolară*, Doctoral research report no. 2, Jul. 2019.

[18] **C. Monea**, *Modelarea și simularea algoritmilor de procesare și analiză de semnal utilizați în rezonanța nucleară cuadrupolară*, Doctoral research report no. 3, Dec. 2019.

[19] **C. Monea**, *Contribuții în dezvoltarea algoritmilor de analiză de semnal bazați pe machine learning*, Doctoral research report no. 4, Jun. 2020.

[20] **C. Monea**, *Contribuții la implementarea unui sistem de pre-procesare, procesare și analiză de semnal utilizat în rezonanța nucleară cuadrupolară*, Doctoral research report no. 5, Sept. 2020.

Research contracts

[21] National research project, *Utilizarea metodei rezonanței quadripolare nucleare la producerea de echipamente pentru detecția explozivilor și drogurilor – DESMTEH*, POS CCE program, Priority axis 2, Operation 2.3.3.

Patents

[22] S. Zaharia, M. Apostol, S. Ioniță, V. Iana, **C. Monea**, L.M. Ionescu, D. Anghel, M. Ilie, A. Varga, *Detector mobil și metodă de detecție a substanțelor cu risc exploziv, a explozivilor și a drogurilor pe baza efectului rezonanței nucleare cuadrupolare (NQR)*, Romanian patent, no. RO131585 (B1), 30.04.2019.

[23] S. Zaharia, M. Apostol, S. Ioniță, V. Iana, **C. Monea**, L.M. Ionescu, D. Anghel, M. Ilie, A. Varga, *Detector fix pentru descoperirea substanțelor cu risc exploziv, a explozivilor și a drogurilor din bagaje pe baza efectului rezonanței nucleare cuadrupolare (NQR)*, Romanian patent application, no. RO131586 (A0), 30.12.2016.

[24] S. Zaharia, M. Apostol, S. Ioniță, V. Iana, **C. Monea**, L.M. Ionescu, D. Anghel, M. Ilie, A. Varga, *Mobile detector and method for detecting potentially explosive*

substances, explosives and drugs by nuclear quadrupole resonance (NQR), European patent application, no. WO2018124905 (A1), 05.07.2018.

[25] S. Zaharia, M. Apostol, S. Ioniță, V. Iana, **C. Monea**, L.M. Ionescu, D. Anghel, M. Ilie, A. Varga, *Mobile detector and method for detecting potentially explosive substances, explosives and drugs by nuclear quadrupole resonance (NQR)*, USA patent, no. US20200182810 (B2), 16.02.2021.

Unpublished scientific papers and presentations

[26] **C. Monea**, *Metode de procesare și analiză de semnal utilizate în spectroscopia cu rezonanță nucleară cuadrupolară*, **Communications Session of the Students' Scientific Circles in Electrical, Electronics, Control Engineering and Computer Science – SCCSS 2018**, national phase, 5th edition, Valahia University of Târgoviște, Jul. 12-13, 2018.

[27] **C. Monea**, *Tehnici de procesare și analiză de semnal utilizate în rezonanța nucleară cuadrupolară*, **Communications Session of the Students' Scientific Circles in Electrical, Electronics, Control Engineering and Computer Science – SCCSS 2019**, national phase, 6th edition, Lucian Blaga University of Sibiu, Jun. 5-6, 2019.

[28] **C. Monea**, *An optimized NQR spectrometer for detection of prohibited substances*, **Annual Symposium of Doctoral School of Electronics, Telecommunications and Information Technology – SAD-ETTI**, 2nd edition, Jul. 4, 2019 (diploma of merit for the best unpublished work).

7.5 Perspectives for future development

From a hardware point of view, future research will focus on the implementation of coherent noise cancellation techniques, such as piezoelectric and magnetoacoustic noise. In the long term, a dual detector will be developed containing an X-ray scanner to identify suspicious objects and an NQR spectrometer to confirm substances.

From a software point of view, future research will focus on adding an application that centralizes the detection results from multiple systems. It can run on a local server or in the cloud and will give the user an overview of the operation of all equipment in a security checkpoint.

Selected references

[1] Global Terrorism Database, *Incidents over time*. Internet: www.start.umd.edu/gtd/search/Results.aspx?start_yearonly=1970&end_yearonly=2017 [10 Aug. 2020].

[2] European Monitoring Centre for Drugs and Drug Addiction, *European Drug Report: Trends and Developments*, 2019.

[3] H. Chihara, Ed., *Nuclear Quadrupole Resonance Spectroscopy Data*, Landolt-Börnstein - Group III Condensed Matter, 1997.

[4] R.D. Hippenstiel, *Detection theory: applications and digital signal processing*, CRC Press, 2002.

[5] **C. Monea**, *Techniques and equipment for detection of prohibited substances: A brief overview*, **J. of Electrical Eng., Electronics, Control and Computer Science**, vol. 4, no. 4, pp. 7-16, 2018.

[6] **C. Monea**, *The use of nuclear quadrupole resonance spectroscopy for detection of prohibited substances: Techniques and equipment*, **11th Int. Conf. on Electronics, Computers and Artificial Intelligence**, pp. 1-6, 2019.

[7] SpinCore Technologies, *RadioProcessor*. Internet: http://spincore.com/products/RadioProcessor/ [12 Aug. 2020].

[8] Tecmag, *Redstone*. Internet: http://www.tecmag.com/redstone/ [12 Aug. 2020].

[9] J. Yinon, *Counterterrorist Detection Techniques of Explosives*, Elsevier, 2007.

[10] L. Cardona, Y. Miyato, H. Itozaki, J. Jiménez, N. Vanegas, H. Sato-Akaba, *Remote Detection of Ammonium Nitrate by Nuclear Quadrupole Resonance using a Portable System*, **Appl. Magn. Reson.**, vol. 46, no. 3, pp. 295-307, 2015.

[11] A. Kaspar, CONPHIRMER – Project final report, Grant Agreement Number 261670, King's College London, 2014.

[12] J. Barras, K. Althoefer, M.D. Rowe, I.J. Poplett, J.A.S. Smith, *The Emerging Field of Medicines Authentication by Nuclear Quadrupole Resonance Spectroscopy*, **Appl. Magn. Reson.**, vol. 43, no. 4, pp. 511-529, 2012.

[13] X. Zhang, N. Schemm, S. Balkir, M.W. Hoffman, *A Low-Power Compact NQR Based Explosive Detection System*, **IEEE Sensors J.**, vol. 14, no. 2, pp. 497-507, 2014.

[14] P. Tagare, *Signal Averaging*, Biomedical digital signal processing, pp. 184-192, Prentice-Hall, 1993.

[15] **C. Monea**, *Tehnici de procesare și analiză de semnal utilizate în rezonanța nucleară cuadrupolară*, Doctoral research report no. 2, Jul. 2019.

[16] S.D. Somasundaram, A. Jakobsson, E. Gudmundson, *Exploiting spin echo decay in the detection of nuclear quadrupole resonance signals*, **IEEE Trans. on Geosci. and Remote Sensing**, vol. 45, no. 4, pp. 925-933, 2007.

[17] A. Jakobsson, M. Mossberg, M.D. Rowe, J.A.S. Smith, *Exploiting temperature dependency in the detection of NQR signals*, **IEEE Trans. on Signal Process.**, vol. 54, no. 5, pp. 1610-1616, 2006.

[18] W. Shao, J. Barras, K. Althoefer, P. Kosmas, *Detecting NQR signals severely polluted by interference*, **Signal Proces.**, vol. 138, pp. 256-264, 2017.

[19] A. Coy, P.T. Callaghan, *Pulsed gradient spin echo nuclear magnetic resonance for molecules diffusing between partially reflecting rectangular barriers*, **The J. of Chem. Phys.**, vol. 101, no. 6, pp. 4599-4609, 1994.

[20] S. Beguš, V. Jazbinšek, J. Pirnat, Z. Trontelj, *A miniaturized NQR spectrometer for a multi-channel NQR-based detection device*, **J. of Magn. Reson.**, vol. 247, pp. 22-30, 2014.

[21] S.D. Somasundaram, et al., *Detecting stochastic nuclear quadrupole resonance signals in the presence of strong radio frequency interference*, **IEEE Int. Conf. on Acoustics, Speech and Signal Process.**, 2008.

[22] S.D. Somasundaram, A. Jakobsson, E. Gudmundson, *Robust nuclear quadrupole resonance signal detection allowing for amplitude uncertainties*, **IEEE Trans. on Signal Process.**, vol. 56, no. 3, pp. 887-894, 2008.

[23] **C. Monea**, *Modelarea și simularea algoritmilor de procesare și analiză de semnal utilizați în rezonanța nucleară cuadrupolară*, Doctoral research report no. 3, Dec. 2019.

[24] M. Oproescu, G.V. Iana, **C. Monea**, *Application of genetic algorithm for optimization of NQR signal detection*, **11th Int. Conf. on Electronics, Computers and Artificial Intelligence**, pp. 1-4, 2019.

[25] G.V. Iana, **C. Monea**, M. Oproescu, *The use of the clonal selection algorithm for NQR signal detection optimization*, **11th Int. Conf. on Electronics, Computers and Artificial Intelligence**, pp. 1-4, 2019.

[26] **C. Monea**, G.V. Iana, S. Ionita, L.M. Ionescu, S.A. Zaharia, S. Ilie, N. Bizon, *An optimized NQR spectrometer for detection of prohibited substances*, **Meas.**, vol. 151, p. 107158, 2019.

[27] L. Cardona, H. Itozaki, J. Jiménez, N. Vanegas, H. Sato-Akaba, *Spectral descriptors and supervised classifier for ammonium nitrate detection in landmines by nuclear quadrupole resonance*, **J. of Magn. Reson.**, vol. 305, pp. 104-111, 2019.

[28] **C. Monea**, *Nuclear quadrupole resonance response detection using deep neural networks*, **Expert Syst. with Applications**, vol. 182, p. 115227, 2021.

[29] Mira Technologies Group, *Premii*. Internet: http://www.miratechnologies.ro/premii/ [24 Aug. 2020].

[30] S. Zaharia, M. Apostol, S. Ioniță, V. Iana, **C. Monea**, L.M. Ionescu, D. Anghel, M. Ilie, A. Varga, *Detector mobil și metodă de detecție a substanțelor cu risc exploziv, a explozivilor și a drogurilor pe baza efectului rezonanței nucleare cuadrupolare (NQR)*, Romanian patent, no. RO131585 (B1), 30.04.2019.