Forensic Advantages and Disadvantages of Raman Spectroscopy Methods in Various Banknotes Analysis and the Observed Discordant Results

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Abstract:- This work discussed and highlighted advantages and disadvantages of Raman spectroscopy methods in the analysis of various security areas of banknote around the world, trending in the current literature. Various methods applied on the three main security areas of banknotes investigation were considered - ink, prints and substrate security areas. Specifically, the advantages and disadvantages envisage from the various methods focuses on laser focal spot size used, Raman signal detection on the selected security areas and the samples detection efficiency of the various methods reported. The research also explains some discordant results observed in the various works conducted on similar security areas of different banknotes. Overall, Ink and print security analysis are the most widely analyzed security areas employed in the various researches, while the spectral analysis of substrate security area was the least explored area. The Raman forensic advantages of banknotes investigation outweighed the envisage disadvantages, and the most common disadvantage notice in the various methods are the fluorescent effect and single note detection (per analysis) limitation in the various methods.

Keywords:- Banknote, Raman Spectroscopy, Advantages, Disadvantages, Discordant, Ink Security, Print Security, Substrate Security.

I. INTRODUCTION

Banknotes are part and parcel of our daily life. Even with the advent of digital banking system we still require hard copies of banknotes in our daily transactions and record keeping. These banknotes are systematically produced to guarantee their security when in circulation, since they are subject to counterfeiting by criminals. Despite these measures, criminals still uses advance technology at their disposal to produce similar copies of these banknotes (Brandão et al., 2016) within a shortest possible time (Guedes et al., 2013; Katarina et al., 2018). To tackle this criminality, financial institution uses various scanners and visual inspection techniques to infer the authenticity of suspected notes in their financial operations. This may have some limitations with the current advancement in banknote counterfeiting production and deployment. Meanwhile, it is still difficult for forgers to produce banknotes with exact molecular constituent of ink, print or substrate security constituent of genuine banknote (Sonnex et al., 2014). Recent literature therefore introduced molecular photonic finger print methods of Raman spectroscopy to detect various counterfeiting in banknotes around the world specifically on the ink, print or substrate security areas (Fig. 1). These methods have recorded various advantages and disadvantages in the project of Raman spectroscopy analysis of banknote around the globe.



Fig 1 Some Selected Points of Analysis on ₩500 Banknote Plain Surfaces

Since 1928 when Indian Physicist Sir Chandrasekhar Venkata Raman was awarded with novel price in Physics for discovering Raman spectroscopy phenomenon, Raman has gained several momenta and become a valuable tool for molecular photonic finger prints detection of specimen in molecular level with little or no sample preparation. It provides unique molecular vibration information (photonic finger print) of molecules within a specimen in either solid, liquid or gas phase and also in either cold or hot state without destruction. Various techniques were developed as an improvement to suit a specific investigation procedure in sample analysis. For instance, in 1955 Marvin Minsky developed a confocal technique of Raman spectroscopy (Giridhar et al., 2017). In contrast to the traditional Raman micro spectroscopy, the procedure uses an additional attached microscope in the machine configuration to focus the laser light onto the sample through a microscope objective. This technique possesses fluorescent rejection system which allowed only some desire Raman spectra reaches the charge couple device (CCD) detector through a pinhole (Fig. 2), which is in confocal with the illuminating focal spot. The detected light is restricted to a small area within the focal point so that light above or below the focal point is rejected (defocus). Still the techniques use traditional micrometer focal spot of light source to illuminate sample and collected Raman spectra, and this affect only localize small volume area of the sample, and may result in sub-sample problem in heterogeneous samples analysis (Shin & Chung, 2013) such as paper. Moreover, when this technique is applied on low optical absorption samples (such as paper), the intensity of the source laser has to be reduced to avoid damage on the sample point of analysis and this will result in low Raman intensity generation and detection. As the generated Raman intensity is proportional to the area from which light is collected (Downes, 2019; Duy et al., 2017; Shin & Chung, 2013) and therefore on the density (number) of active Raman molecules involve (Pelletier M. J., 2003). Meanwhile, the micro focal spot laser beam use in the Raman spectroscopy setting will allow for the selection of specific region of the sample analysis area (Novais et al., 2019). Recently, a diameter of the focal spot was increase so that wider area of the sample point of analysis can be cover to allowed for more Raman signal generation and collection from deep inside the sample. This will also reduce cost and time spent in some Raman spectra acquisition schemes used in some Raman spectroscopy method, requiring averaging Raman spectra acquired over different points (Novais et al., 2019) and the 2-D compressive sensing scheme used by Zhang et al., 2020 team, which required multiple incident laser spots. Therefore, the wider area Raman spectroscopy will have vrious advantages which may not be achieved with the traditional confocal micro-Raman spectroscopy.

In either way, the aspect of Raman spectroscopy scheme that comprises laser focal spot configuration, spectral detection and sample detection capacity of Raman methods may have intrinsic advantages and disadvantages associated with them. For instance, emitted light with longer wavelength (fluorescent) may be observed by the CCD in the analysis of substrate security, which may be influence by

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the uneven surfaces of the paper (Causin et al., 2010), optical brightening agents (OBAs) or fluorescent whitening agents (FWAs) added to the substrates paper during it manufacturing stage etc. Fluorescent additives (such as quantum dots) used in the print ink can also emit light with longer wavelength in the Raman spectroscopy analysis. Environment exposure such as handling of notes may contaminate the sample banknote (Lawandy & Smuk, 2014) which can also course fluorescent in Raman spectroscopy analysis of banknotes. In addition, other ambient factors such as cosmic rays and signals could also contribute to Raman spectra impurities (fluorescent).

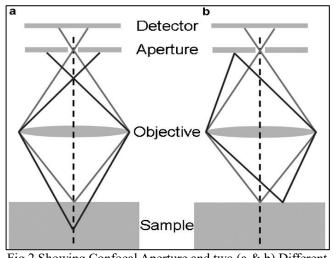


Fig 2 Showing Confocal Aperture and two (a & b) Different Signal Defocusing (Gordon & McGoverin, 2011)

With these improvements, constraint, advantages and disadvantages, various researches were conducted with Raman spectroscopy in the literature to authenticate various genuine and counterfeited banknote around the world. Therefore, in this work we review relevant researches conducted with Raman spectroscopy alone or in combination with other procedure to analyze both or solely ink, prints and substrate security areas of banknotes aiming to envisage some of the advantages and disadvantages associated with the various laser focal spot size used, Raman signal detection on the selected security areas and the number of banknotes detection efficiency in the various methods. We also discourse some discordant observations therein. When research was conducted in combination with other techniques as the case may be, we only consider the Raman spectroscopy aspect in the research work.

II. REVIEW OF RAMAN ANALYSIS IN VARIOUS BANKNOTES' SECURITY-AREAS

A. Raman Spectral Investigation on Ink Security of Banknotes

Raman photonic method to analyze same security ink made with complex ink recipe, ultra violet ink and colourshifting ink (Chambers et al., n.d.) on genuine and fake banknote has been used in the literature to established status of banknotes as genuine or counterfeit without the prior knowledge of the ink composition (Hana et al., 2019). Different spectral peaks indicate specific spectral finger Volume 9, Issue 4, April – 2024

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prints of molecules in the document. Generally, Raman analysis of banknote were conducted with low power micrometer focal spot laser to avoid damage on the point of analysis. And the analysis is usually conducted on a small sample area of the security ink printed on the banknotes which tend to be difficult due to reduce laser power use (Novais et al., 2019). To characterizes R\$50 and R\$100 Real banknotes with portable Raman spectroscopy, Novais et al in 2019 conducted a qualitative Raman analysis on genuine and counterfeit bills of Brazilian R\$100 and R\$50 banknotes. Due to the difficulty of low power requirements in the procedure not all samples were analyzed. Meanwhile, the team were able to detect the Raman characteristic bands (1528, 1340, 748, 680 & 487 cm⁻¹,) for phthalocyanine pigment (also detected by (Jauković, 2019) and bands characteristic of diarillyde (1596, 1398, 1256 & 953 cm⁻¹) from the brown and orange security ink areas of genuine R\$50 banknote. But the direct comparison of genuine and counterfeited note spectra shows strong similarities, thus making it difficult for the team to discriminate between the notes. Therefore, the team deployed partial least squares discriminant analysis (PLS-DA) and principal component analysis (PCA) for the accurate sample classification. Still the result show that most Raman band of inks on the R\$100 genuine and fake notes coincide with each other which

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could be an indication that the security ink has been reproduce successfully by the counterfeiters (Novais et al., 2019). Despite the R\$100 samples discrimination dificulty, the discrimination of R\$50 notes with PCA and PLS-DA was found to be effective.

Micro-Raman spectroscopy approach was also used (Guedes et al., 2013) to analyze 15 samples of genuine and counterfeit of €10 and €20 banknotes from various European countries, aiming to established their status from the spectra of their security inks. They discovered that micro-Raman spectroscopy results cannot reliably differentiate counterfeited note from genuine Euro notes, since ink spectra detected by the team from both sample notes show similar Raman spectra. Meanwhile, (Božičević, Gajović and Zjakić., 2012) used Micro-Raman spectroscopy to identify the origing of different countefreited banknote printed with the same toner on the same machine. Spectra from magenta, cyan and yellow toners were considered on various commercial print papers. Their findings revealed that only yellow toner on the banknote specimen can reliably link the common origin of fake banknotes (Fig. 3) The observed number of sample detection efficiency of all the researches was a single banknote in each analysis.

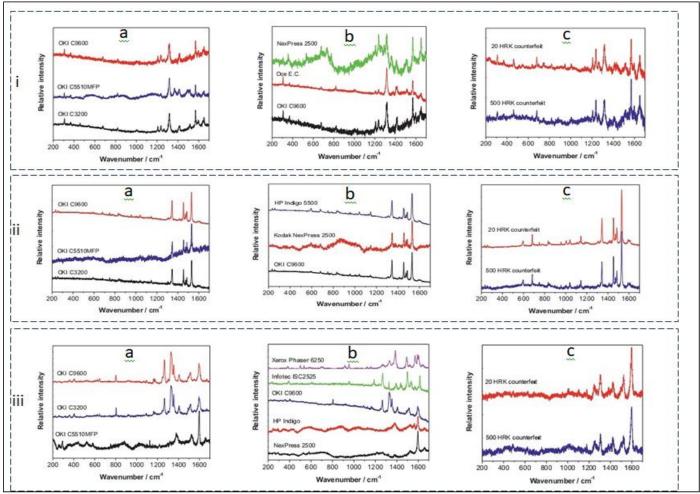


Fig 3 (i) Raman spectra of Magenta toners, (ii) Cyan toners and (iii) Yellow toners for a) the same manufacturer, different printers and cartridges results b) one liquid and two powder toners from different manufacturers results c) 20 and 500 Kuna counterfeits results respectively (Božičević *et al.*, 2012).

The advantage and disadvantage of testing paper banknote with Raman spectroscopy was study (Jauković, 2019). In the second case study of this research, the circulated genuine and fakes 1000 dinars note investigated give clear Raman spectra on each of the selected security areas (Jauković, 2019). Spectrum of phthalocyanic for green and blue pigment were observed. Although, fluorescent disadvantage was reported in the first case study, but in the second case study Jauković was able to directly authenticate genuine and fake 1000 dinars note through the characteristic molecular photonic finger prints recorded. However, the method is also limited to single note analysis and authentication. Johnson et al., 2017 analyzed 319 ink and print security areas (76 toner, 79 offset, 78 inkjets, and 86 intaglio) to demonstrate the capabilities of Raman spectroscopy in the discrimination of print inks. The capability success rate was more than 70% for inkjet, toner, and offset print. Some of the aforementioned researches we conducted in combine with other spectral techniques such as X-ray fluorescence (pXRF) (Novais et al., 2019) in solely or both security areas analysis for additional banknote authentication.

When conducting Raman analysis of ink security printed on a substrate paper, researchers often check if the substrate's characteristics spectra affected the detected ink spectra. Various studies, such as those by Božičević et al., (2012) and Jauković, (2019), have noted this potential interference. According to research team led by Novais, Luiz & Neris (2019), the substrate spectra may or may not influence the ink spectra detected on the substrate paper. While research finding by (Božičević et al., 2012) and (Jauković, 2019) found that the substrate paper does indeed affect the Raman spectra results of ink security within the analysis area, which they later subtracted from the detected composite spectra. In contrast, (Braz, López-lópez and Montalvo, 2014) and (Johnson et al., 2017) did not observed this effect in their ink spectral results. This observed discordance suggests that the influence of substrate paper on ink spectra in Raman analysis may vary depending on chemical composition of the ink, interaction of ink with the substrate paper occur during the halftone prints or carbon printing, the properties of the substrate paper it self and the experimental conditions in the spectral detection.

> Chemical Composition of the Ink

The chemical composition of the ink plays a crucial role in its interaction with the substrate paper and the Raman spectroscopy analysis. Different types of inks contain various pigments, dyes, solvents, and additives, each of which can exhibit unique Raman spectra (Božičević *et al.*, 2012) and could have various penetration range which is also influenced by chroma and hue of the ink (). Additionally, some ink compositions may have molecules that are more prone to Raman fluorescence (Takalo et al., 2015) When the analyzed samples are circulated types of notes, they can be conterminant with sebum (Lawandy & Smuk, 2014) at the point of analysis can also alter the Raman signals, affecting the observed ink signal.

> Properties of the Substrate Paper

The substrate paper's characteristics, including its chemical composition, surface roughness (Causin et al., 2010), thickness, and porosity, can influence the interaction between the ink and the paper surface depending on the ink penetration in the substrate. Paper substrates with rougher surfaces or higher porosity may allow for deeper penetration of the ink into the paper matrix, potentially affecting the Raman signals obtained from the ink. The presence of impurities or additives in the paper, such as fillers, coatings, or sizing agents, can also contribute to spectral variations (Bitla, 2002) and interference in the Raman analysis.

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> Experimental Conditions

The conditions under which the Raman spectroscopy analysis is performed can significantly impact the observed spectra. Factors such as laser power, wavelength, exposure time and focal spot size can influence the intensity and quality of the Raman signals obtained from ink printed on the substrate. Variation in experimental parameters across different studies can lead to discrepancies in results thereby influencing the interpretations.

Considering these factors collectively is essential for understanding the complexities involved in Raman spectroscopy analysis of ink security on substrate paper and for interpreting the observed spectral data accurately. Addressing these factors in experimental design and data analysis is crucial for obtaining reliable and reproducible results in forensic and security applications of banknote authentication. Therefore, further research is needed to better understand and address these discrepancies for accurate and reliable Raman analysis of ink security printed on substrate of banknotes.

B. Raman Method on Banknote Print Security

Numerous research has shown that print security of banknotes is also a unique component in Raman photonic discrimination of genuine and counterfeited banknotes (Imperio et al., 2015; Takalo et al., 2015). The print security areas constitute serial numbers, Intaglio printing, offset lithographic printing and design complexity (Chambers et al., n.d.) printed on the banknotes. We haven't come across any Raman imaging of print-security area analysis intended for banknotes discrimination. However, banknote print imaging from other techniques were used for banknote Doush & AL-Btoush, authentication (Abu 2017: Muhammad, 2017). Therefore, in the alternative, we review those literatures that analyzes constituent spectra of print security areas on banknotes. One of the related works was carried out on the blue pigment of the Euro-flag print area on $\in 5$, $\in 10$, $\in 20$, $\in 50$, $\in 100$ and $\in 200$ notes with time resolve Raman spectroscopy technique to discriminates genuine and fake notes. Despite the fluorescent effect of the blue pigment from the flag area of euro-note (especially of the old euro note), the team claimed that time resolve spectra from the blue pigment was a good parameter for identifying genuine and counterfeit euro banknotes (Takalo et al., 2015). The spectra of the blue pigment from old and new notes (Fig. 6a & b) printed by various European print companies were successfully detected.

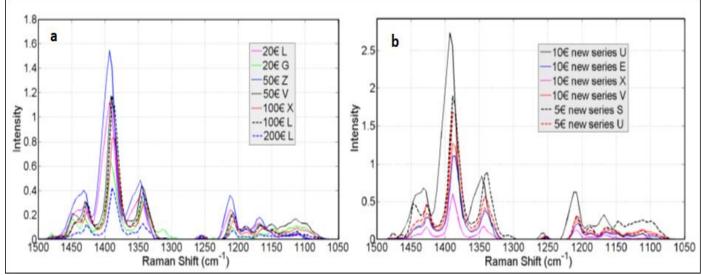


Fig 4 Time resolve Raman spectra of blue pigment on Euro flag print area of some genuine and counterfeited Euro banknotes from various european print companies a) Old euro series b) New euro series (Takalo et al., 2015)

A document spectroscopy was carried out with atomic force microscopy (AFM) coupled with Raman micro spectroscopy (Brandão *et al.*, 2016) on the serial number and chalcography print of Brazilian R\$100 banknote. Clear Raman spectra was obtained on the authentic R\$100 note (Fig. 7a). Meanwhile fluorescence was recorded from the counterfeited Brazilian R\$100 banknotes due to its significant surface difference (see section 1 for other possible reason). Since confocal techniques only recorded the spectra of the focal surface (Gordon & McGoverin, 2011) and obliterate all other defocus part (Fig. 2). The obliterated spectra contributed to high fluorescence in the counterfeited spectra (Fig. 7b). On that criterion the two security documents were discriminated. Another team of researchers (Imperio et al., 2015) conducted Raman spectroscopy analyses on Italian banknotes (1000 lira) used within 1947 to 2001, aiming to authenticate fake 1000 lira withing the time-lime. Serial number, watermark and security mark were considered. Direct spectral comparison has successfully discriminated genuine and counterfeited 1000 lira notes. Other team have also succeeded in banknote discrimination with Raman spectroscopy measurement on the print chalcography, orange and red areas (de Almeida et al., 2013) and other print features (Manukyan et al., 2023) We give summary of these various study methods including the advantages and disadvantages envisage in the various methods (Table 1).

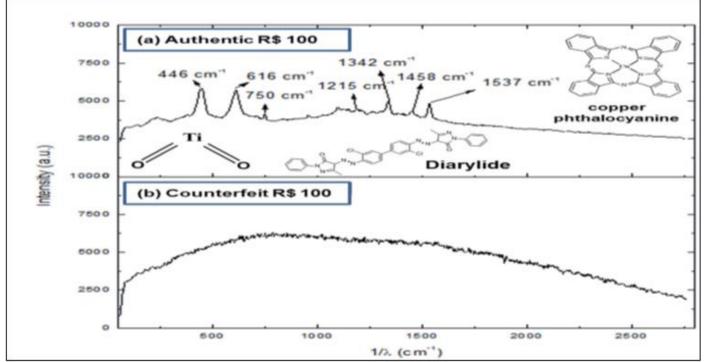


Fig 5 Raman Spectra for (a) Authentic and (b) Counterfeit R\$100 Banknotes (Brandão et al., 2016)

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Table 1 Summary on Advantages and Disadvantages of Raman Spectroscopy Methods in the Analysis of Ink Security of Various Banknotes

	Analysis of Ink Security of Various Banknotes									
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Authenti	Ink	Authenticat	Authentic	Micro	Floresce	The	The	Method	The	Brand
c US	security	ion of	banknote	focal	nt was	method	micro	may not	method	ão et
dollar,	(Serial	banknote	has	spot	successf	allowed	focal spot	be	is	al.,
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starling.	and	atomic	inks and	enables	identify	banknote	only	for	to single	
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ited and		Raman	Raman	composit	paramete	nation in	represent	exhibit	each	
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ited		machine			in colour			toner mk		
banknote		machine			toners					
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and	various	the	Raman		notes			method		s et
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20-euro	areas	Raman	obtained		fluoresce			suitable		
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s from	different	oscopy in	genuine		the			analysis		
various	colors	identifying	and fake		genuine			of old		
European		fake	banknote		notes			note		
country		banknotes	s, making		which			reported		
of origin.		by	it		help in			to have		
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banknote s used in Italy from 1947 to 2001, including both authentic specimen s and certified fakes.	features of the banknote s such as serial number, waterma rk, and security mark were examine d	infrared spectra analysis of Italian 1000 lira banknotes issued between 1947 and 2001 and discriminati on between their authentic and counterfeit specimens.	e in the various paper compositi on was revealed (Red colour pigment was used for security marks and black printing ink for serial numbers among different specimen s of banknote s)		fluoresce nt effect was reported in the method					o et al., 2015
Original and counterfe it Real (Brazilia n currency) banknote s, specifical ly R\$ 50	Ink security from the print chalcogr aphic, orange and red areas	To characteriz e inks used on authentic and counterfeit Real banknotes for their discriminati on.	s) The procedur e correctly classified all fake and genuine R\$ 50 denomina tions and was found to be suitable for use as a complem entary method to classical forensic inspectio n		✓	✓ The procedur e offered a fast analysis method that can be used by non- specialis t operator s	×	✓		de Almei da et al., 2013
Pre- Federal America n paper money	ink pigments , paper composit ion (Substrat e), and other print security features.	To analyze and compare the chemical compositio ns of paper fibers, inks, and fillers in pre- Federal American	n. The research identified Franklin used natural graphite pigments, vermilion , and Prussian blue for	×	No fluoresce nt was reported	The method allowed single banknote analysis and discrimi nation in each analysis	Ý	Not applicab le.	×	Manu kyan et al., 2023

		paper	ink							
		money	pigments							
Kościusz ko banknote s	Paper (substrat e), ink, and dyes	To establish the provenance of materials used in manufactur ing Kościuszko banknotes and materials used in the dyeing and printing processes	Raman spectrosc opy identified smalt an optical brightene r typically used in paper productio n	✓	✓.	The method allowed single banknote analysis	•	✓.	✓	Klisi et al., 2019
Genuine and counterfe its Czech banknote s.	Paper and inks used on Czech banknote s.	To distinguish genuine from counterfeits Czech banknotes through their characterist ic Raman spectra	Various substance s, were identified and a spectral library of inks measured on Czech banknote s was created.	~	~	~	×	The method may not be suitable for the analysis of old note reported to have shown stronger fluoresc ent	×	Hana et al., 2019
Genuine, counterfe ited and dummy 1000- and 500- Naira banknote s in a bunch	Substrate security area on the shorter side of the notes	simultaneo usly analysis and detection of multiple genuine and counterfeite d banknote	36 sample notes were simultane ously analyzed and authentic ate as genuine or counterfe it note	Wider focal spot laser enables simultan eous analysis of security substrate paper composit ion in a bunch	V	The method allowed simultan eous detection of multiple banknote s in a bunch in situ	Sample in the center of the focal spot are more represent ed than those samples at the extreme end of the bunch in the composit e spectra	No fluoresc ent was reported in the analysis	V	Murta la & Mukht ar 2023

C. Raman Method on Banknote Substrate-Security

The substrate paper types we considered here consist of cellulose fibres made from wood and plant (cotton), coating made with specific print figments and other additives for required print property. The substrate security of genuine banknote comprises complex substrate recipe (made with specialize cellulose fibre (cotton) and coating made with specific print figment), security fibres, windowed security thread, watermark and see through windows (Chambers et al., n.d.). When Jauković tested paper banknote base on two case studies. One of the analyses conducted in the first case study was on the clear paper surface of 1- and 5-Dinar notes. Pronounce Raman fluorescent originating from the paper was reported. According to them, the yellowish impregnation in the paper is to blame for the fluorescent. Part of the work presented by (Takalo et al., 2015) has also study the use of substrate security to discrimination between euro banknotes. Their technique has detected unique time resolve spectra from the substrate security (paper) of genuine euro banknote different from the substrate of their corresponding fake copies (Fig. 7)

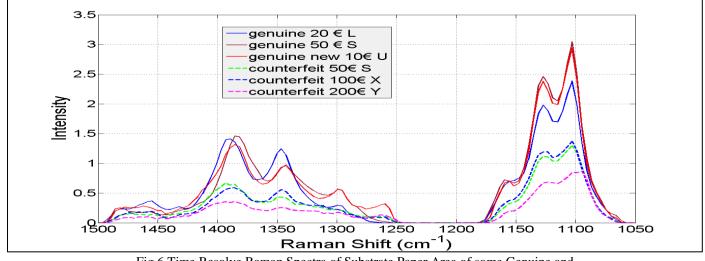


Fig 6 Time Resolve Raman Spectra of Substrate Paper Area of some Genuine and Counterfeited Euro Banknotes (Takalo et al., 2015)

III. STRATEGY FOR SPEEDY AND RELIABLE RAMAN SPECTRA ACQUISITION

Zhang et al., in 2020 have introduce a 2-D parallel acquisition strategy to increase acquisition speed of conventional confocal technique to more than two-fold magnitude in single acquisition. Their technique uses 2-D multiple (8) focal spots to simultaneously acquire multiple sample spectra with reduce acquisition time. The reconstruction of the individual original spectra was achieved through compressive sensing algorithm. (Shin & Chung, 2013), present a review on the 3 recently improve schemes for a good sample representation in Raman measurement. According to this review the most efficient scheme to acquire a representative Raman spectra collection strategy was the use of wide area simultaneous illumination scheme which can overcome problems such as sample overheating that can course sample damage, inefficiency in sample collection, lack of sample spectra representation due

to sample inhomogeneity, non requirement of multiple focal spot in the analysis of multiple sample which may be cost intensive and it does not require repeated Raman measurement on a single analysis area.

Recently, we used same scheme to published a paper (Murtala & Mukhtar, 2023) which is a breakthrough from the current single banknote authentication limitation used in the present literature. The work was able to simultaneously detect multiple genuine and counterfeited banknotes in a bunch in situ with 36 sample banknotes. It utilizes wider area focal spot illumination technique to simultaneously target each banknote substrate security surfaces on the longer side in the bunch (Fig. 8). To the best of our knowledge, increase in the detection number of banknote (detection efficiency) per analysis was not debated in the current literature and the number of counterfeited banknotes produced and deployed in the world economy has been increasing yearly.

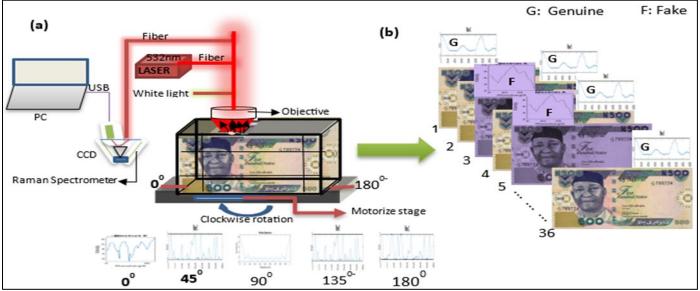


Fig 7 Simultaneous Multiple Banknote Analysis and Discrimination in a Bunch on the Substrates Longer Side (Ahmed & Mutari, 2023)

IV. CONCLUSION

Raman spectroscopy methods on Ink, print and substrate security analysis for the discrimination of various banknote around the world was review and discussed. Some specific advantages and disadvantages relating to laser focal spot size used, spectral impurities (fluorescent) associated with the detection of Raman molecules in the analyzed security areas and the sample detection capacity of the various analysis method were covered. The Raman photonic finger prints obtained from both security areas of sample banknotes can be used solely or in combination with other security areas to authenticate individual genuine and counterfeited banknotes. It is obvious, various factors resulted in the observed discordance need to be address in the feature experiment, for a better understanding of light, ink and substrate interaction in banknote photonic finger prints analysis. It is also obvious that Raman molecular photonic advantages of banknotes investigation outweighed envisage disadvantages. The most noticeable the disadvantage in the various methods was the spectral impurities effect and single note detection limitation in the various methods. while only photonic finger prints detected from the substrate security alone have been shown to archived simultaneous detection of multiple genuine and counterfeited banknotes.

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