Detection of counterfeit U.S. paper money using intrinsic fluorescence lifetime

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Abstract: Genuine U.S. Federal Reserve Notes have a consistent, two-component intrinsic fluorescence lifetime. This allows for detection of counterfeit paper money because of its significant differences in fluorescence lifetime when compared to genuine paper money. We used scanning two-photon laser excitation and the time-correlated single photon counting (TCSPC) method to sample a ~4 mm^2 region. Three types of counterfeit samples were tested. Four out of the nine counterfeit samples fit to a one-component decay. Five out of nine counterfeit samples fit to a two-component model, but are identified as counterfeit due to significant deviations in the longer lifetime component compared to genuine bills.

OCIS codes: (190.0190) Nonlinear optics; (300.2530) Fluorescence, laser-induced; (170.3650) Lifetime-based sensing.

References and links

1. W. Becker, The bh TCSPC Handbook (Becker & Hickl, 2008).
2. K. Suhling, P. M. French, and D. Phillips, "Time-resolved fluorescence microscopy," Photochem. Photobiol. Sci. **4**(1), 13–22 (2005).
3. T. H. Chia, A. Williamson, D. D. Spencer, and M. J. Levene, "Multiphoton fluorescence lifetime imaging of intrinsic fluorescence in human and rat brain tissue reveals spatially distinct NADH binding," Opt. Express **16**(6), 4237–4249 (2008).
4. R. Cubeddu, D. Comelli, C. D'Andrea, P. Taroni, and G. Valentini, "Time-resolved fluorescence imaging in biology and medicine," J. Phys. D Appl. Phys. **35**(9), R61–R76 (2002).
5. T. Q. Ni, and L. A. Melton, "Two-dimensional gas-phase temperature measurements using fluorescence lifetime imaging," Appl. Spectrosc. **50**(9), 1112–1116 (1996).
6. D. Comelli, G. Valentini, R. Cubeddu, and L. Toniolo, "Fluorescence lifetime imaging and Fourier transform infrared spectroscopy of Michelangelo's David," Appl. Spectrosc. **59**(9), 1174–1181 (2005).
7. M. Suzuki, "Development of a simple and non-destructive examination for counterfeit coins using acoustic characteristics," Forensic Sci. Int. **177**(1), e5–e8 (2008).
8. V. Rusanov, K. Chakarova, and T. Madolev, "Mössbauer spectroscopy investigation of the properties and stability of dollar bank note pigments," Appl. Spectrosc. **56**(9), 1228–1236 (2002).
9. W. Denk, J. H. Strickler, and W. W. Webb, "Two-photon laser scanning fluorescence microscopy," Science **248**(4951), 73–76 (1990).
10. W. R. Zipfel, R. M. Williams, and W. W. Webb, "Nonlinear magic: multiphoton microscopy in the biosciences," Nat. Biotechnol. **21**(11), 1369–1377 (2003).
11. W. Becker, A. Bergmann, M. A. Hink, K. König, K. Benndorf, and C. Biskup, "Fluorescence lifetime imaging by time-correlated single-photon counting," Microsc. Res. Tech. **63**(1), 58–66 (2004).
12. R. Judson, and R. Porter, "Estimating the worldwide volume of counterfeit U.S. currency: data and extrapolation," FEDs Working Paper No. 2003–52 (2003).
13. D. Comelli, C. D'Andrea, G. Valentini, R. Cubeddu, C. Colombo, and L. Toniolo, "Fluorescence lifetime imaging and spectroscopy as tools for nondestructive analysis of works of art," Appl. Opt. **43**(10), 2175–2183 (2004).
14. J. Cameron, J. Marengo, R. Judson, and J. Pruiksma, "The use and counterfeiting of United States currency abroad, part 3," presented to United States Congress by the Secretary of the Treasury, Sept. 2006.
15. Q. S. Hanley, D. J. Arndt-Jovin, and T. M. Jovin, "Spectrally resolved fluorescence lifetime imaging microscopy," Appl. Spectrosc. **56**(2), 155–166 (2002).
16. D. K. Bird, K. W. Eliceiri, C. H. Fan, and J. G. White, "Simultaneous two-photon spectral and lifetime fluorescence microscopy," Appl. Opt. **43**(27), 5173–5182 (2004).

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1. Introduction

The United States currency is influential and important to both domestic and international economies. Despite the anti-counterfeiting measures designed into U.S. currency, attempts to forge U.S. Federal Reserve Notes are ever-present and can pose a serious threat to the public's confidence in their money while providing financial support to organized crime groups. As a result, there is strong demand for a better understanding of the U.S. paper currency's material properties and the development of reliable methods to detect counterfeit bank notes. Many advanced counterfeiters focus on the production of counterfeit U.S. Federal Reserve Notes that can pass the "look-and-feel" test and are able to mimic salient anti-counterfeiting measures such as inscribed security threads and watermarks. We present a new method of reliably discerning genuine paper money from counterfeit banknotes by taking advantage of intrinsic fluorescence properties from the paper substrate. The approach reported in this paper is unique because we measure an inherent physical property of genuine U.S. Federal Reserve Notes with a non-obvious method of duplication. Beyond simple counterfeit detection, this technique may serve the forensic science community by helping to identify groups of counterfeit notes that all originate from a single source based on identical fluorescence lifetime and amplitude signatures.

United States paper money is predominately a blend of cotton and linen fibers. These banknotes, like many textiles and papers, produce intrinsic fluorescence when exposed to high-intensity light (e.g. a laser beam). The focus of this study was to determine the differences in fluorescence lifetime of genuine versus counterfeit paper money. The fluorescence lifetime (τ) is a measurement of the time a fluorescent molecule is in its excited state before relaxing back to its ground state and emitting a photon [1]. A specific molecule's lifetime is constant and generally on the order of hundreds of picoseconds (10^{-12} seconds) to several nanoseconds (10^{-9} seconds). However, a molecule's lifetime can vary depending on a number of microenvironmental conditions such as local viscosity, pH, or temperature [2]. In the case of paper money, these factors are unlikely to affect the intrinsic fluorescence lifetimes from the currency. Techniques to measure fluorescence lifetime are highly sensitive and have been applied to biological [3,4] and non-biological systems [5,6]. Other approaches to counterfeit recognition include examining acoustical characteristics in genuine and counterfeit coins [7] and Mössbauer spectroscopy for the analysis of paper money [8]. To our knowledge this is the first time fluorescence lifetime has been applied to the counterfeit detection of paper currency.

We used a two-photon microscope [9,10] capable of measuring fluorescence lifetime to show a very consistent fluorescence lifetime "signature" that is substantially different from known counterfeits and other basic textiles and papers. In addition, this method requires only a small sample area (~4 mm^2), is non-destructive, and does not leave any marks or changes in coloration to the currency. We believe that this technique can accurately discern the differences between genuine and counterfeit currency and could have important applications in fields of numismatics and forensic science.

2. Material and methods

2.1 Microscope Apparatus

We used a custom-built two-photon microscope based on an Olympus BX51 WI upright fluorescence microscope (Olympus America, Center Valley, PA). The excitation source was an 80 MHz pulsed Ti:Sapphire (Ti:S) laser (Mai Tai, Spectra-Physics, Mountain View, CA) tunable between 710 nm and 990 nm. The excitation wavelength was set to 735 nm with a 100 fs pulsewidth. The microscope objective was a 4x, 0.28 NA air objective (Olympus, XLFLUOR 4x/340, Center Valley, PA). Samples were held flat on a motorized 3-axis microscope stage (ASI Imaging, Eugene, OR). Power at the sample was ~15 mW.

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The intrinsic fluorescence spectra of samples were obtained using a fiber optic spectrometer (USB2000, Ocean Optics, Dunedin, FL) connected to the two-photon microscope. Fluorescence generated at the sample was reflected by a dichroic mirror (680 long-pass, Chroma Technologies, Rockingham, VT) and focused with a plano-convex lens into a 1 mm core fiber optic to maximize collection efficiency.

Fluorescence lifetime capabilities were made possible through the addition of a multi-channel plate PMT (R3809U-52, Hamamatsu, Bridgewater, NJ) and a time-correlated single photon counting (TCSPC) card (SPC-150, Becker & Hickl, Berlin, Germany) [11]. The fluorescence of the paper money was filtered through a 555 nm short-pass filter (Chroma Technologies, Rockingham, VT). A fluorescence lifetime decay curve was produced by raster scanning the laser beam over a 4 mm^2 area (or frame) and summing the emission photons for 60 seconds (~0.8 seconds / frame). Figure 1 shows the location of the 4 mm^2 on the bill. Each fluorescence decay curve was processed using SPCImage software (Becker & Hickl). The software (Fig. 2) was able to determine the best-fit parameters and account for the instrument response function (IRF). The full width at half maximum (FWHM) of the IRF was 62 ps as measured from the second-harmonic generation of rat-tail collagen.

Fig. 1. The red square between the Federal Reserve Bank seal and the serial number on the U.S. $100 bill indicates the ~4 mm^2 region where fluorescence lifetime was collected.

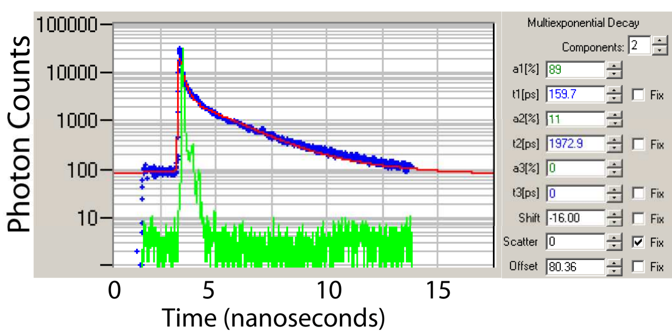

Fig. 2. The blue points represent a typical fluorescence lifetime decay curve taken from a one hundred-dollar bill. Blue points correspond to the photon counts for a given time interval after the excitation laser pulse. The red line is a two-component fit based on a minimization of the χ^2 value. The green line is the instrument response function (IRF). The measured lifetimes (τ_1, τ_2) and amplitudes (a_1, a_2) of the two-component fit are on the right.

Typical counts following each 60 second scan was 600,000 - 1,000,000 photons. Calculations from prior literature indicate an accurate two-component fit requires a minimum of 10,000 to several 100,000 photons [11]. Fluorescence lifetime decays, F(t), for genuine Federal Reserve Notes were fit to the two-component lifetime model (Eq. (1)),

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$$F(t) = a_1 e^{-\frac{t}{\tau_1}} + a_2 e^{-\frac{t}{\tau_2}} \quad (1)$$

by obtaining the best χ^2 fit value. Four parameters are generated: lifetime of components one and two (τ_1, τ_2) and the amplitudes of each lifetime (a_1, a_2). It is this combination of lifetimes and amplitudes from genuine paper notes that serves as its unique signature different from all other samples measured. Counterfeit samples were fit to either a one-component or two-component model depending on its χ^2 fit value.

2.2 Samples

The U.S. one hundred-dollar bill was chosen as the primary focus of this study because it is not only the largest denomination U.S. Federal Reserve Note in circulation, but it also represents the largest value on the counterfeit market. As of 2002, it is estimated that total value of counterfeit $100 notes worldwide could be as high as $98 million USD [12]. Therefore, one control group included genuine $100 Federal Reserve Notes from the printing series 1996 and later (n = 10).

Additional control groups included $50 Federal Reserve Notes (n = 5, Series 1996 – 2004), $20 Federal Reserve Notes (n = 5, Series 1996 – 2006), and $1 Federal Reserve Notes (n = 54, Series 1999 – 2006) in order to test for fluorescence lifetime variations between banknote denominations. The 54, $1 Federal Reserve Notes were distributed throughout the past 10 years of printing to specifically test whether increased circulation time, and in theory increased wear and tear, affected the fluorescence lifetime measurement. All control samples were genuine Federal Reserve Notes, in circulation, non-sequential, and untreated prior to lifetime measurements. The location of intrinsic lifetime collection for all denominations tested was the unmarked space to the left of the portrait, below the serial number, and above the Federal Reserve Bank seal, as seen in Fig. 1.

Three types of known counterfeits were tested in these experiments: 1. Copies made by digitally scanning a bill into a computer followed by printing on both sides using a consumer-grade color inkjet or laser printer (herein referred to as "digital"). 2. Traditional counterfeits made with a cotton and linen blend and printed using more sophisticated methods. These bills are often produced by foreign organized crime groups (herein referred to as "traditional"). 3. Bleached (or "washed") bills made by removing the ink from a lower denomination bill and then reprinting a larger denomination over the ink-less paper (herein referred to as "bleached"). Three counterfeits bills of each type were tested. All bills were confirmed as counterfeit by an authorized government agency. Fluorescence lifetime measurements were also obtained from several control materials. These include printer paper made from wood pulp, 100% cotton stationary paper, and swatches of 100% linen cloth.

3. Results and discussion

The fluorescence spectrum of a genuine $1 U.S. Federal Reserve Note and a $100 U.S. Federal Reserve Note is shown in Fig. 3. All genuine notes tested ($1, $5, $10, $50, $100 denominations) have a similar broadband fluorescence spectra spanning 400 nm to 650 nm with greater emission in the 460 - 640 nm bandwidth. Several swatches of cotton paper, wood pulp paper, and linen cloth were also tested and shown to emit broadband fluorescence. Similar broadband fluorescence from textiles has also been reported [13]. The 555 nm short-pass emission filter used in the lifetime measurements allowed us to collect a substantial amount of intrinsic fluorescence spectrum from all papers, cloths, genuine and counterfeit U.S. Federal Bank Notes.

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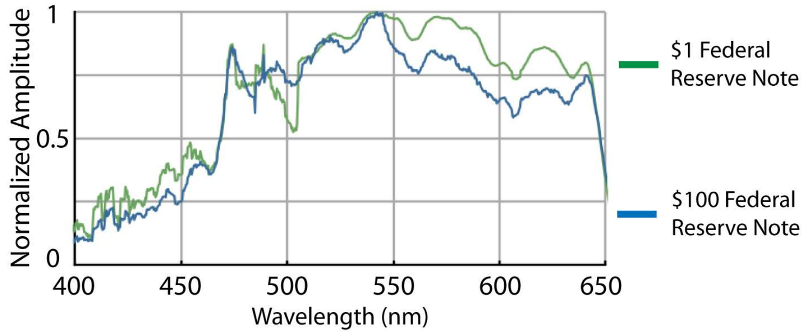

Fig. 3. The intrinsic fluorescence spectra of a $1 and a $100 U.S. Federal Reserve Note. All genuine notes tested possessed similar broadband fluorescence from 400 - 650 nm.

Table 1 shows the calculated lifetimes and amplitudes for all the samples measured.

Table 1. Intrinsic fluorescence lifetimes for four denominations of genuine U.S. Federal Reserve Notes, three types of counterfeits, and three types of basic materials. In samples fit to a two-component decay, the longer lifetime (τ_2) is the salient value in determining genuine versus counterfeit Federal Reserve Notes. Values are given as mean ± SD.

TYPE	Average Lifetime 1	Average Lifetime 2	Average Amplitude 1	Average Amplitude 2
Bleached Counterfeits (n = 3)	160 ± 0.6 ps	1552 ± 118 ps	86.0 ± 4.9%	14.0 ± 4.9%
Traditional Counterfeits (n = 3)	174 ± 5.7 ps (n = 2) 918 ps (n = 1)	1725 ± 46 ps N/A	79.4 + 2.2% 100%	20.6 + 2.2% N/A
Digital Counterfeits (n = 3)	802 ± 170 ps	N/A	100%	N/A
Genuine $100s (n = 10)	162 ± 4.6 ps	2010 ± 64 ps	84.8 ± 3.3%	15.2 ± 3.3%
Genuine $50s (n = 5)	161 ± 1.7 ps	2070 ± 60.1 ps	84.2 ± 0.9%	15.8 ± 0.9%
Genuine $20s (n = 5)	161 ± 4.1 ps	2023 ± 53.3 ps	86.2 ± 4.6%	13.8 ± 4.6%
Genuine $1s (n = 54)	166 ± 10.3 ps	2001 ± 60.3 ps	85.0 ± 3.3%	15.0 ± 3.3%
Cotton Paper (n = 3)	1060 ± 84 ps	N/A	100%	N/A
Linen Cloth (n = 3)	1374 ± 57 ps	N/A	100%	N/A
Wood Pulp Paper (n = 3)	919 ± 56 ps	N/A	100%	N/A

The two consistent fluorescence lifetimes found in genuine U.S. one hundred-dollar bills are: a short lifetime component with an average τ_1 = 162 ± 4.6 ps and an average amplitude of 84.8 ± 3.3%. A long lifetime component with an average τ_2 = 2010 ± 64 ps and an average amplitude of amplitude of 15.2 ± 3.3%. Genuine $50s, $20s, and $1s exhibit similar two-component fluorescence lifetimes and amplitudes. One advantage of our technique is that no pre-treatment of the paper money samples is required. All genuine Federal Reserve Notes were randomly obtained from public circulation, with some notes dated as far back as the 1996 printing series. There was no significant change in the fluorescence lifetimes and

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amplitudes as a function of circulation time (as approximated by the serial number and series year on each Federal Reserve Note). These data suggest the paper substrate's lifetime properties are highly consistent within each denomination, between denominations, and are minimally influenced by wear-and-tear and/or residual skin oils from regular handling. Counterfeit bills were also left untreated and were subject to some level of public circulation and handling prior to seizure by authorities.

All bleached and most traditional counterfeits fit to a two-component model. All digital counterfeits and one traditional counterfeit fit to a one-component model. In addition, cotton papers, wood pulp papers, and linen cloths all fit to a one-component model. Samples displaying one-component fluorescence decay are readily identified by simple visual inspection of the decay curve (Fig. 4).

Counterfeit currencies made by digital copying methods are the easiest to detect by the fluorescence lifetime method. All digital counterfeit samples tested with this method show a fluorescence lifetime decay curve that fits best to a single component. We were informed by an authorized government agency that these types of counterfeits are typically printed with consumer-grade color printers on standard wood pulp or cotton paper. A single-component lifetime data from these samples may arise from a single constituent (wood pulp or cotton). However, there are a multitude of paper substrates available, each one undergoing a different manufacture and treatment process. Furthermore, fluorescence lifetimes involve complex energy relaxation processes that can be difficult to fully elucidate in a heterogeneous material such as wood pulp or cotton paper. Therefore it is not surprising the lifetime values obtained from wood pulp paper and cotton paper (Table 1) are not identical to the lifetimes from the digital counterfeits.

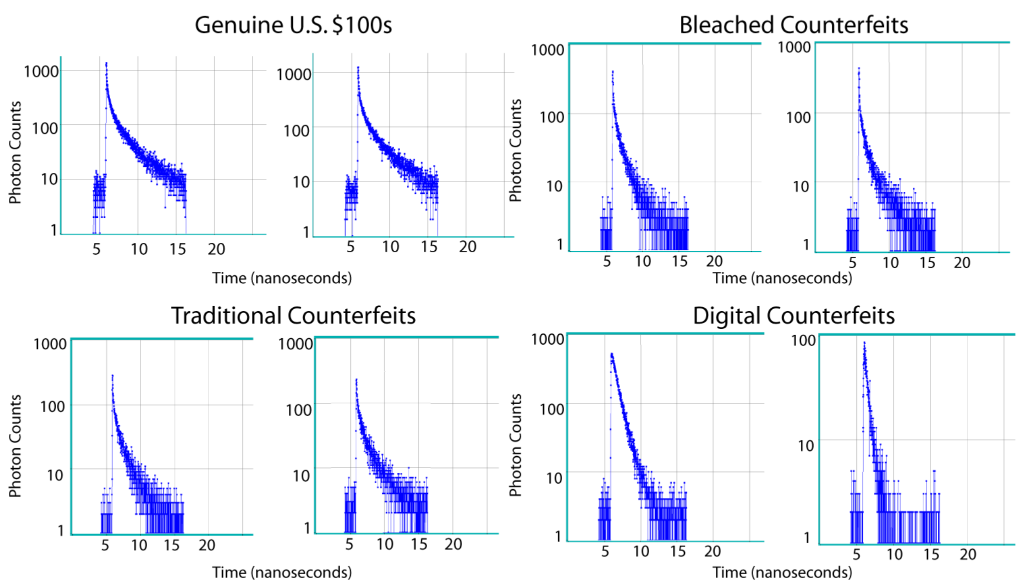

Fig. 4. Representative fluorescence lifetime decay curves from genuine paper money and three different types of counterfeit currency. Digital counterfeits immediately appear different from genuine $100 Federal Reserve Notes because of the linear (one-component) decay. Washed counterfeits and most traditional counterfeits fit to a two-component decay. However, the long lifetime component (τ_2) is measured to be significantly shorter in these samples than the long lifetime component in genuine $100 bills. This difference is also apparent when examining the shape of the fluorescence lifetime decay curves.

Given that this technique is analyzing the fluorescence properties of the paper, it was initially thought that bleached counterfeits would dupe our method since these counterfeits use the same cotton and linen paper blend as genuine notes. However, this is certainly not the

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case when looking at the data for these two groups. Once again, the longer lifetime component in bleached counterfeits has a significantly shorter lifetime (1552 ± 118 ps) compared to genuine $100, $50, $20, and $1 Federal Reserve Notes. Several reasons could cause this difference in lifetimes. First, the harsh chemicals used to remove the ink from the bills could alter the shape and / or binding of the molecules within the paper, giving rise to a change in their fluorescence lifetime. Another possible explanation is that there are residual bleaching chemicals impregnated inside the bill providing some contribution of intrinsic fluorescence different from the lifetime of the native paper. However, we find this unlikely to be the sole explanation, given that the magnitude of the contribution from the residual chemicals required to influence the lifetime fits would likely be discernible as an additional component.

Our data also shows that this method can clearly distinguish genuine notes from traditionally printed counterfeit notes. These counterfeiting groups may create or modify their own cotton and linen paper for printing forged notes [14]. In any case, it is unlikely these groups can replicate the U.S. paper used to print authentic notes, resulting in obvious lifetime measurements differences. These variations in lifetime may come from an incorrect proportion of components, such as cotton and linen, used to make the paper. Another difference with traditional counterfeits could come from the method of synthesizing paper from the raw materials. As these synthesis methods differ, the resulting paper may have different molecular properties or have lingering chemicals imbedded within as mentioned previously with the bleached notes. Finally, variations in fluorescence lifetime for these notes may come from differences in the species or strain of the raw materials. Since many traditionally counterfeited notes come from outside the United States, the cotton and flax plants used in these different parts of the world may be genetically different from what is found in the U.S.

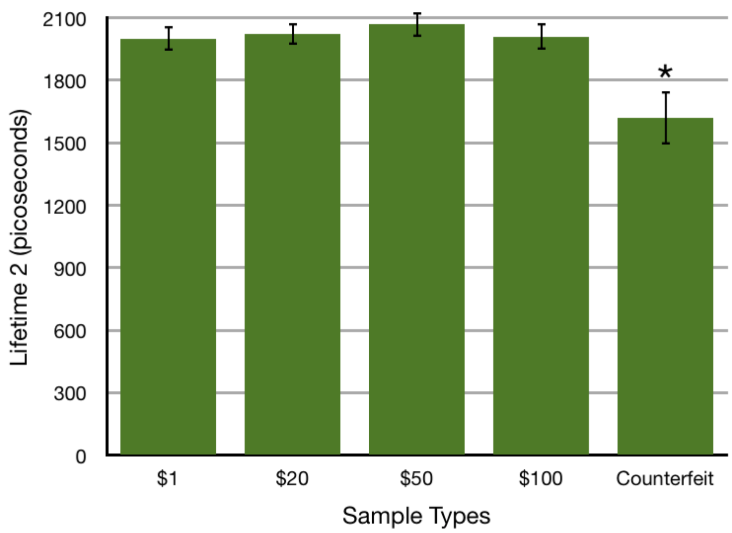

Fig. 5. Comparisons between longer lifetime values (τ_2) of different denominations of genuine Federal Reserve Notes and counterfeit notes fit to a two-component model. Only counterfeit samples fit to two-components (three bleached and two traditional) were included in order to provide a better comparison to bills with a more advanced level of counterfeiting. The longer lifetime component is significantly different in genuine samples when compared to the counterfeits ($p < 0.001$). Values are given as mean ± SD.

Figure 5 compares the intrinsic fluorescence lifetime measurements from genuine Federal Reserve Notes to counterfeit bills fit to a two-component model (three washed notes and two traditional counterfeits). The shorter lifetime measurement between these two groups is not appreciably different. However, the lifetime 2 component of counterfeit notes consistently

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have a shorter lifetime. ANOVA analysis revealed a statistically significant difference in the lifetime 2 value of all genuine U.S. banknotes compared to counterfeit bank notes fit to two-components ($p < 0.001$). There is no significant difference in lifetime 2 among genuine U.S. Federal Reserve Notes. The digital counterfeits and one of the traditional counterfeits were not included into this calculation because these samples can be excluded on the basis of fitting to a one-component model.

Many counterfeiters focus on duplicating colors, textures, watermarks, and other anti-counterfeiting technologies built into the note. However, counterfeiters would find it very difficult to mimic the fluorescence lifetime of genuine paper money. One reason is the equipment required for these measurements are not readily available to the general public. Even if lifetime measurements can be obtained, it is non-obvious how one could tweak counterfeit banknotes to match the two-component fluorescence lifetime of genuine paper money. Incorporation of the sample's fluorescence spectrum may offer an additional means of differentiating a counterfeit bank note from a genuine note. A more advanced approach could utilize a spectrally-resolved fluorescence lifetime imaging system to ascertain multiple fluorescence lifetimes from the sample across various spectral windows [15,16].

4. Conclusion

We have demonstrated a promising technique for the discernment of genuine U.S. paper money from counterfeits. The excellent repeatability within denominations and between different denominations is likely due to a consistent supply of paper from specific manufactures. As a result, this paper substrate provides us with a repeatable and unique fluorescence lifetime "signature" that can set genuine U.S. Federal Reserve Notes apart from all other materials and counterfeits tested. With the assistance of an authorized government agency, we were also able to test known counterfeits made using different techniques and show unique lifetime characteristics between some common counterfeiting methods.

This study contributes to the overall body of knowledge available for the study of U.S. paper money and fluorescence analysis of textiles and papers. The size, costs, and training required for our experiments currently precludes this technique from practical implementation in government institutions, banks, or businesses. However, this technique may find utility in forensic science laboratories and in the analysis of extremely high-quality counterfeits for critical investigations. Affordable detection systems for simple identification of genuine versus counterfeit notes can be achieved by using less demanding electronics to reduce the temporal resolution and switching to single-photon excitation instead of two-photon excitation. Currently, we collect fluorescence from the sample for one minute to gather a more than sufficient number of photons for an accurate calculation of the lifetime(s) and amplitude(s). Future work on determining the minimal sampling time and minimal temporal resolution necessary may make this technique more feasible for rapid, large-scale analysis of paper money. In conclusion, our results provide a new technique for identifying counterfeit paper money, as well advance the concept that intrinsic fluorescence lifetime measurements can provide important information on a material's properties.

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