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Abstract. The use of laser-induced breakdown spectroscopy (LIBS) as a low-cost, nondestructive method for detecting counterfeit coins was examined. A pulsed laser was used to evaporate a minute amount of coin surface, and the emanating plasma was interrogated with an entry-level spectrometer. The spectra produced showed evidence of lead content in six of the eight counterfeits examined. Thus, LIBS could offer a viable low-cost technique for identifying a significant number of fake coins. © *2016 Society of Photo-Optical Instrumentation Engineers (SPIE)* [DOI: 10.1117/1.OE.55.4.044104]

1 Introduction

Laser-induced breakdown spectroscopy (LIBS) determines the atomic elements present in a target sample. A pulse of laser light is used to evaporate a small sample (typically less than a microgram) of the target to generate plasma of ionized atoms and free electrons. As this plasma cools and the free electrons recombine with ions, various spectral lines are emitted. The wavelengths and intensities of these lines identify the atomic elements in the original target. In addition, the percentage of those elements present in the target can also be deduced. With computer analysis of the spectral lines emitted, a measurement can be completed within a fraction of a second. Little or no sample preparation is required. The target may be any material that absorbs the chosen laser wavelength: solid, liquid, or gas.

LIBS is considered to have its roots in the paper by Brech and Cross.[1] The development of LIBS to its current state is now well-documented.[2–4] This simple, rapid, and versatile technique is widely applied in the laboratory and with *in situ* field measurements. The latter has been fueled by the advancements in laser and spectrometer technologies that have led to compact, portable LIBS systems.[5–7] Applications of LIBS now cover many sectors of physical and life sciences,[8–12] ranging from measurements in deep oceans[13,14] to the planet Mars.[15] This technique can be characterized as microdestructive (many applications consider it nondestructive), with applications even extending to precious art for identification of pigments in ancient painted works and treasures, such as examining ancient coins to determine their age and authenticity.[16–19]

Counterfeit coinage is a longstanding problem for currency manufacturers. Modern technology has made forgery of notes more difficult, but there are still significant rates of forgery of coins.[20,21] In the United Kingdom, the one-pound coin is particularly targeted for counterfeiting. Official estimates put the rate of one-pound counterfeits in circulation at 2.55% in 2015.[21] These counterfeits are typically manufactured using casting methods, whereby molten metal is poured into molds. A casting flaw line is clearly evident in one of the coins (Fig. 1) produced by this method. The genuine coins produced by the Royal Mint are made by the stamping method where two dies, one for each side of the coin, are pressed onto blank metals to deform them into the images of the dies.[22]

Counterfeit one-pound coins are currently identified by several methods that include visual inspection, physical measurements, conductivity measurement, and alloy composition measurement using techniques such as x-ray fluorescence.[23] For example, deviation from the standard weight (±0.5%), diameter and thickness are good indications of forgery, as are incorrect combination of faces, wrong relative orientation of the faces, and poor edge design. The features of counterfeit coins to look out for are clearly detailed on the Royal Mint website.[21] Some indications can be found by the naked eye: indistinct edges to the lettering (Fig. 2), poor quality of the edge milling and lettering (Fig. 3), and color of coin not matching that of genuine coins.

Visual identification of forgeries is not always conclusive. A general rise in the quality of forgeries in recent years, combined with the damage and discoloration sometimes present on genuine coins, can cast doubt.[24] The Royal Mint regularly takes coins out of circulation based on discrepancies in size and weight; these counterfeits are tested and then destroyed.

This paper proposes to investigate the use of LIBS to test one-pound coins for indications of forgery. Use of low-intensity, single-shot LIBS means that this method causes hardly any damage to the coins as compared to the wear and tear while in circulation. The aim is to identify either the difference in elemental composition or the presence of additional element(s) as compared to the specifications of genuine coins. For example, lead is not present in the manufacture of genuine one-pound coins, but is thought to be widely used in the production of fakes to achieve the correct conductivity for the alloy, thereby allowing it to pass as real in vending machines.[23] Vending machines have a number of sensors to authenticate and sort the coin denominations.

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Fig. 1 Counterfeit £1 coin with casting defect.

The authentication sensor normally uses a magnetic field to check the conductivity of the coins.

2 Methodology

2.1 Instrumentation

The LIBS setup is shown in Fig. 4. The Nd:YAG laser (Quanta-Ray DCR-2-10) was set to produce 6 mJ, 10-ns Q-switched pulses at the fundamental wavelength of 1064 nm. Although the flashlamp was running at 2 Hz, the Q-switch triggering was manually controlled to give either single or multiple pulses. The laser output was focused by a 250-mm focal length convex lens. At the focal point of the laser, a coin holder was placed for ease of interchange of coins without disturbing the optical setup, in particular the necessity of realigning the plasma collection optics. Although the coin holder maintained a fixed position for

Fig. 2 Counterfeit £1 coin with indistinct edges to the lettering.

Fig. 3 Counterfeit £1 coin (top), compared to a genuine coin (bottom), has poor quality edge milling and lettering.

the coin surface, the rotational position of the coin face was user-determined, which was random in our case. The 6-mJ laser pulse was just enough to evaporate a microscopic amount of material, thus producing sufficient plasma for analysis.

Plasma collection optics consisting of two 50-mm diameter fused silica (UV grade) biconvex lenses of 100- and 60-mm focal lengths were used in a cage setup to collect the plasma radiation and focus it onto a 400-μm fiber. A 1064-nm Rugate notch filter (25-mm diameter) was placed between the collection lens and fiber to block the laser radiation and thus protect the fiber and spectrometer from optical damage. The transmittance curve for the notch filter provided by the manufacturer showed >90% transmission from 400 to 1000 nm except for the region of 537 to 541 nm, where the value was from 80% to 90%.

The other end of the fiber was connected to an Ocean Optics USB2000+ spectrometer, a compact low-cost entry-level spectrometer. The light from the fiber that enters the spectrometer is dispersed via a fixed grating onto a 2048-pixel linear CCD array detector. The spectrometer was manufacturer-preconfigured for the 200 to 850 nm range with a 25-μm slit that resulted in a resolution of 1.5 nm. The spectrometer module was connected to a computer via a universal serial bus (USB) cable to enable the capture and display of spectra. The intensity levels of the recordings were in arbitrary units (AU) linked to the number of photons incident on the sensor during the image capture.

The plasma collection optics consisting of the two lenses and a notch filter were tested to check their combined transmission properties. A broad-spectrum halogen bulb covering the range of wavelengths from 400 to 850 nm and a mercury arc lamp for wavelength coverage below 400 nm were used for this purpose. It was found that there was no appreciable attenuation from 310 to 850 nm. The spectrum was completely cut off below 295 nm, mainly due to absorption by the notch filter.
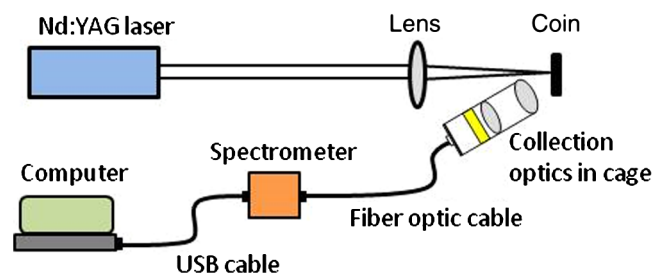

Fig. 4 Instrument setup for LIBS experiments on coins.

Fig. 5 The material removed by LIBS is less than or comparable to natural wear and tear while coins are in circulation.

2.2 Sample Coins

On its website,[25] the Royal Mint has published one-pound coin specifications as a nickel–brass alloy (nickel, copper, and zinc) with 22.5 mm diameter, 3.15 mm thickness, and 9.5 g weight. It also provides a guide for identifying genuine coins based on date compared to design, edge lettering, surface quality, and orientation between the "heads and tails" designs.

For this experiment, eight genuine and eight counterfeit one-pound coins were used. Of the forgeries, seven were kindly lent by a counterfeit coin collector and one was

found by the authors. The coins were all acquired in change. The distinction between genuine and counterfeit was made by visual inspection using the aforementioned guidelines by an expert in coin authentication.[26] The coins were labeled as genuine (G1 to G8) and counterfeit (C1 to C8).

2.3 Experimental Procedure

Each of the 16 coins was placed on the coin holder in random rotational orientation. Single-shot spectra were taken from the heads and tails side of the coin alternately, for a total of four spectra from each coin. The spectra were collected by the spectrometer, which was set to subtract the background noise. Although in some LIBS applications multiple shots are averaged for accuracy and improved signal-to-noise ratio (SNR), for this experiment, it was found that a single shot produced a sufficiently clear spectrum. Further, for practical reasons, we wanted to keep the ablation of the coins to a minimum. For that reason, after a single initial shot to clean the surface of contaminants, a second single shot was used to collect the plasma spectrum from the coin. Figure 5 shows the spots on a coin that were exposed to 2 and 10 laser pulses, respectively. It can be seen that the damage (ablation) is hardly obvious even after five times the number of pulses that the coins were exposed to.

Each individual one-shot spectrum is the image of a single plasma event. Variations in the chemical composition, laser energy, and, most importantly, topography of the sample surface meant that the overall intensity of the signal could vary from sample to sample. The relative intensities of different parts of the spectrum are consistent despite this, as they depend on the relative quantities of different elements present, whereas the overall intensity is affected by the overall quantity of material evaporated. When comparing signals, it is therefore necessary to scale the signals for comparison.
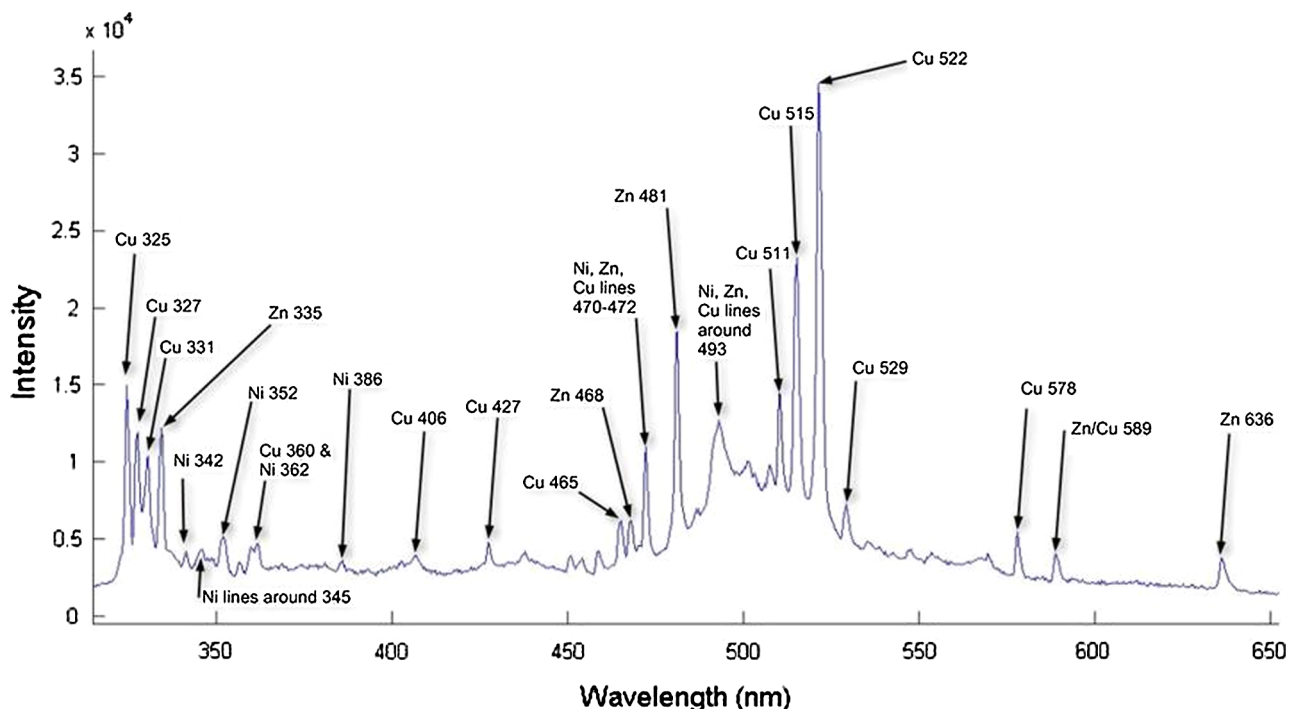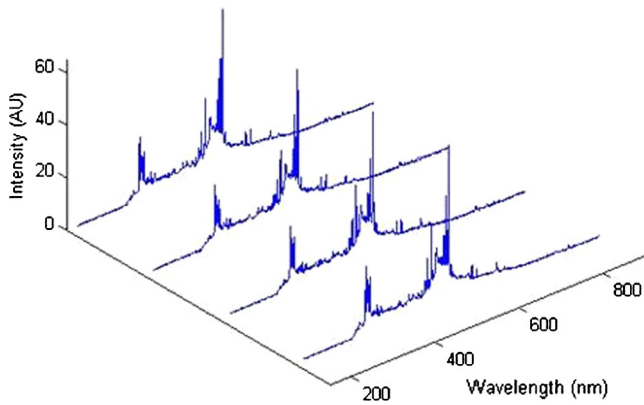

Fig. 6 LIBS spectrum of a genuine £1 coin.

Fig. 7 LIBS spectra of a genuine £1 coin from four random spots.

Fig. 9 LIBS spectra of a counterfeit £1 coin from four random spots.

Hence, all the spectra were normalized to the plasma background (mainly generated by Bremsstrahlung radiation) between 600 and 620 nm, a region where all the spectra had no detectable peak. As a result, the spectra maintained their true relative peak intensities but were now on a similar scale for ease of comparison. It is worth mentioning here that all the LIBS measurements were done under the same conditions without the interference of any ambient light.

3 Results and Discussion

3.1 *Laser-Induced Breakdown Spectroscopy Analysis of Genuine Coins*

In order to use LIBS for counterfeit identification, it is first necessary for genuine coins to be identified reliably. A typical spectrum of a genuine coin is shown in Fig. 6, which labels some of the elemental peaks of copper, nickel, and zinc. The respective elemental peaks were identified by comparing the coin spectrum with our element emission database. The database was created by collecting LIBS spectra of calibrated specimens and comparing them with the National Institute of Standards and Technology Atomic Spectra Database. In order to make comparisons between real and counterfeit coins, the data acquired from the real coins must be consistent. Figure 7 shows the spectra from four unscaled samples of the same coin. It can be seen that the same peaks are consistently present in similar magnitudes, with some variation in the height of each spectrum's set of peaks.
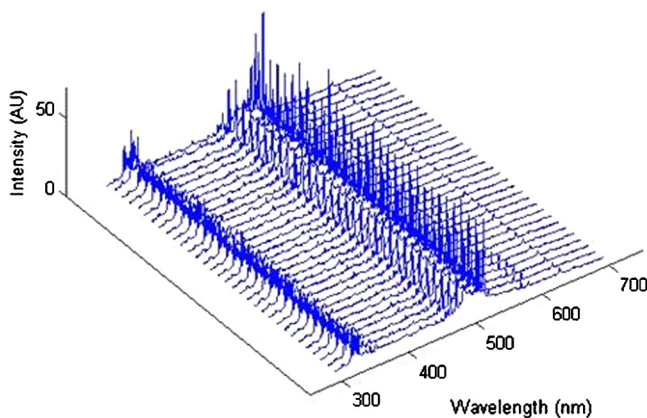
Comparing the entire range of 32 scaled spectra from the genuine coins (four samples from each of eight coins) in Fig. 8, there is evidence of consistency. Some slight variation is unavoidable because of the low laser intensity, which results in relatively low SNR levels: where one sample has a low overall intensity, its smaller peaks will be obscured by noise and will not appear when scaled up for comparison. However, the magnitude and location of the major peaks are consistent. The relative standard deviation (SD) of each of the major peak magnitudes was within the 6% to 10% range. Therefore, these spectra demonstrate, to a certain degree, the repeatability of data from genuine coins.

3.2 *Laser-Induced Breakdown Spectroscopy Analysis of Counterfeit Coins*

The first test was to see whether the elemental composition of the counterfeit coins is homogeneous, as nonhomogeneity can be used as a means of identifying counterfeits. Similar to the genuine coin test, four plasma spectra were recorded for each coin. Each of the eight sets of spectra showed consistency within it, implying that the counterfeit coins had homogeneous alloys, at least at the surface level. Figure 9 shows a set of spectra from a representative counterfeit coin.

Figure 10 shows the spectra of eight counterfeit coins, along with a spectrum of a genuine coin for comparison. It can be seen that the most obvious difference between the fake coins and the real is the presence of peaks in the 350- to 450-nm region of the spectrum. This is the region where lead lines are found, but because of the low relative

Fig. 8 Comparison of 32 LIBS spectra from eight genuine £1 coins (four spectra from each coin—two random spots from each face).

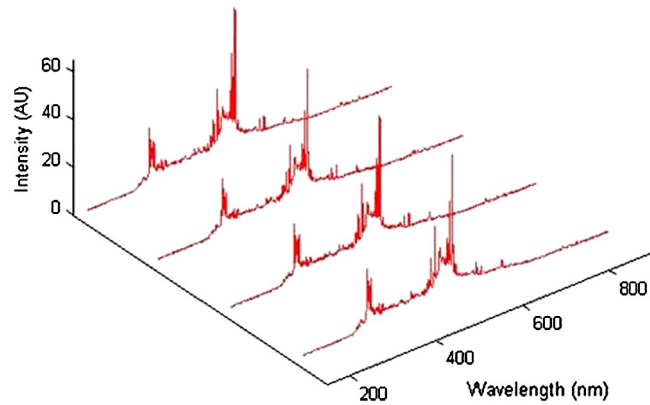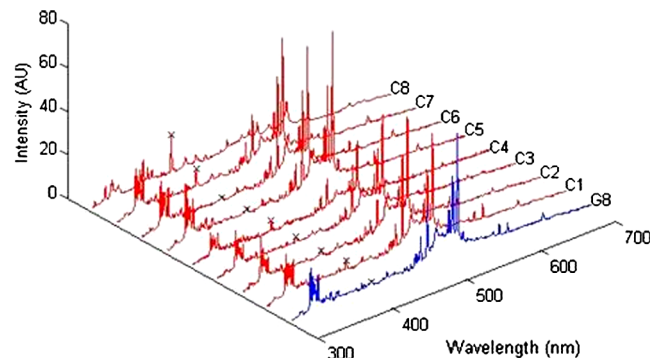**Fig. 10** Spectra from eight counterfeit £1 coins along with a spectrum of genuine coin for comparison.

intensity of the lead lines in comparison to the copper and the probable low concentration of lead in the alloy, the peaks are smaller than many nearby copper, nickel, and zinc lines. Figure 11 shows the region of interest for the genuine coins. Here, we can see the nickel lines at 356.6, 359.8, 361.9, and 385.8 nm and the copper line at 406.3 nm. The weak pair of peaks that sometimes appears at 393 and 397 nm is from ionized copper and zinc, respectively. These peaks are similar in size to the range of random noise; therefore, they are sometimes not present in noisy samples.

It can be seen in Fig. 12 that the peaks of interest in the counterfeits are at 406 and 368 nm. These are lead lines, an immediate indicator of forgery. It is important to distinguish between the 405.8-nm lead line and the line between 406 and 407, which is present in the real coins; this is the copper line at 406.3 nm. The counterfeit samples have the broad 406.3-nm peak because of their copper, nickel, and zinc content as well as the narrower, more intense 405.8 nm. There is also a weaker lead line at 374 nm clearly visible in C4, C7, and C8. C8 has very strong lead lines (including lead 357.3 nm, which is not present in the others) as well as a tin line at 380.1 nm and no copper, nickel, or zinc peaks. This fake is a simple Pb–Sn alloy painted gold to appear genuine. With the paint having worn off, the counterfeiting is visually obvious, but the LIBS result nevertheless confirms it. Two of the counterfeit pound coins' (C5 and C6) spectra are very similar to the genuine coin spectrum. It is likely that they are correct-alloy forgeries. Such counterfeits are not easily detectable with the LIBS technique.

The numerical values of the strongest lead line (405.8 nm) were analyzed. The average intensity for the real coins was 1.20 (AU) with an SD of 0.039 and a relative SD of 3.25%. For six (C1 to C4, C7, and C8) of the eight coins, there was a very strong indication of the presence of lead, with their intensity levels above 6 SDs as compared to genuine coins. The two samples previously mentioned (C5 and C6) had intensity values close to 1 SD of the real coins, suggesting that their lead content was very low or nonexistent.

LIBS will be a powerful tool to detect foreign elements (other than copper, nickel, and zinc) present in coins to classify them as counterfeits. The limit of detection (LOD) of lead for single-shot LIBS analysis using the same experimental setup as shown in Fig. 4 is about 1000 ppm (or 0.1%). Other low-cost metals such as iron, magnesium, and aluminum could also be used in manufacture of counterfeits. Their
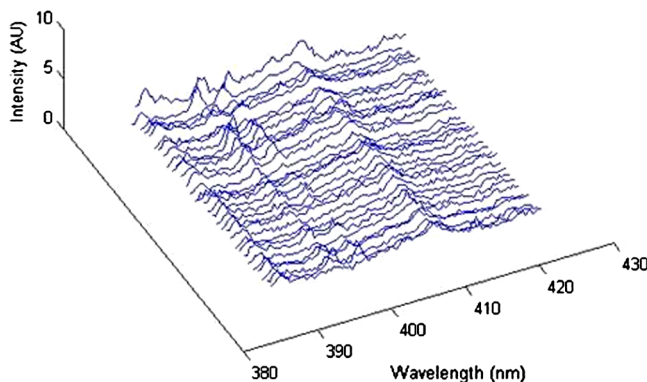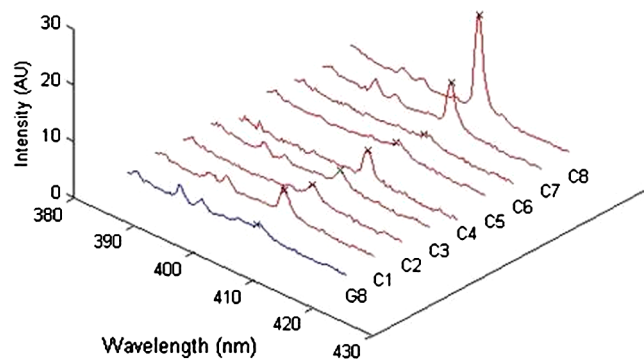

Fig. 12 Zoomed-in version of Fig. 10 shows the lead peaks (405.8 nm) in six of the counterfeit coins. For clarity, 405.8 nm points are marked by a cross in each spectrum.

LODs are found to be even lower than that of lead. However, in practical applications of counterfeit coin detection, detection of foreign element(s) down to a lower limit of 1000 ppm would more than suffice. Setting it any lower may cause ambiguity in the detection process. Counterfeiters use foreign metals for reasons such as cost and lowering the melting point. Therefore, the proportion of these foreign metals will be much higher than 1000 ppm to have any impact for their motives.

The second method considered for evaluation of the coins was an examination of the relative intensities of the peaks of the different elements for genuine and fake coins. The Cu:Ni, Ni:Zn, and Cu:Zn line ratios were studied. The Cu:Zn ratio yielded slightly more variation than the other two ratios. However, to our surprise, there was no clear distinction between the genuine and counterfeit coin sets to suggest using this technique as a universal identification method. Table 1 shows the comparison of ratios of the intensities of copper and zinc peaks in the genuine (average of the eight coins) and counterfeit coins. Counterfeit coins C2 and C5 (C5 had no detectable lead content) had Cu:Zn ratios outside 2 SDs of the genuine coins average. The C8, as previously mentioned, was found to be a purely lead–tin alloy.

Fig. 11 Zoomed-in version of Fig. 8 shows the smaller peaks in region of interest.

Table 1 Comparison of intensity ratios of selected copper and zinc peaks for genuine and counterfeit coins.

Coin type	Ratio of peaks Cu (522 nm):Zn (481 nm)	Ratio of peaks Cu (325 nm):Zn (335 nm)
G1 to G8	1.93 (SD − 0.24)	1.41 (SD − 0.16)
C1	1.84	1.39
C2	2.56	1.88
C3	2.03	1.42
C4	1.78	1.30
C5	2.49	1.67
C6	2.15	1.54
C7	1.65	1.29
C8	—	—

Without any peaks at the Cu, Zn, and Ni lines, it was easily identified as a counterfeit. The remaining counterfeit coins that had lead present in them had Cu:Zn:Ni ratios similar to the genuine coins. Although the detection rate is low for the intensity ratio method, it can be combined with lead detection to enhance the detection of counterfeit coins.

4 Conclusions

This experiment demonstrates the practical possibility of a compact, low-budget system for detecting incorrect-alloy counterfeit coins. It was able to clearly identify lead and tin present in the forgeries. The spectrometer can be reconfigured to cover only the 350- to 420-nm region, which would increase its resolution to about 0.2 nm. This would mean much improved accuracy of measurement, allowing detection of forgeries with a lower level of adulterating metals. As these elemental lines are clear and distinct in six of the eight coins tested, this technique, if automated, could be used in combination with other tests, such as measuring size, weight, and conductivity, to check large numbers of coins for counterfeits without damaging any genuine coins.

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