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Research Article

Development of a Handheld System for Liquor Authenticity Detection Based on Laser Spectroscopy Technique

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In this paper, a handheld liquor authenticity detection system is demonstrated based on the laser spectroscopy technique, which consists of a handheld laser spectrometer and a mobile phone display terminal. In this system, the semiconductor laser is integrated into the spectrometer and the laser beam is further angled to the optical axis of the spectrometer to avoid interference of the fluorescence generated by the bottle wall. During the system operation, the laser excites the tested liquor to generate fluorescence and Raman spectroscopy signals, which are digitized and wirelessly transmitted by Wi-Fi to the Android mobile terminal. After the image processing by the mobile phone APP, the tested liquor spectrum curve is obtained. At the same time, based on the standard liquor spectrum curve stored in the database, the Pearson correlation coefficient is calculated and the matching similarity is given. In addition, this paper proposes a calibration method based on pure water Raman intensity to achieve accurate measurement of fluorescence intensity and minimize the influence of fluorescence intensity saturation on the measurement results. In the experiment, we measured the similarity of 12 brands of Chinese liquor by using our self-developed handheld laser spectrometer. Their authenticity of liquor could be given accurately and effectively.

1. Introduction

Nowadays, the global alcohol market is fast-growing; however, liquor counterfeit has also developed into one of the global economic criminal activities. The counterfeit liquor not only disturbs the order of the normal market management but also infringes the rights and interests of the regular manufacturers and consumers [1]. According to a survey on counterfeit liquor released by Chinese liquor industry media, a large number of consumers have bought counterfeit liquor (https://www.sohu.com/a/455700115_120133372). Because of the influence of human factors, the authenticity of many liquors is difficult to be judged by sensory identification alone. At present, researchers, liquor manufacturers, and consumers have adopted a variety of technical methods for anticounterfeit detection of liquor, including packaging anticounterfeit, electronic label anticounterfeit, and QR code label anticounterfeit [2]. However, these anticounterfeiting technologies have some shortcomings such as forged anticounterfeiting signs and reused outer packages.

Recently developed methods for detecting liquor also have significant disadvantages. For example, the electronic nose method [3, 4] which uses gas sensors and can only detect volatile substances in liquor. The sensor used in this method is also easily affected by the temperature of the detected objects and chemical substances, resulting in inaccurate experimental results. Though chromatographic methods have high accuracy, they need a complicated detection process, relatively harsh environmental requirements, and high cost of a single experiment and experimental results are susceptible to interference [5, 6]. The spectral detection system used for liquor evaluation is generally composed of a spectral acquisition system and spectral data processing software based on the PC platform. As the system requires a PC to process and display, it brings inconvenience for outdoor usage [7, 8]. Although its detection accuracy is high, its portability is poor. How to form an integrated, miniaturized, and intelligent portable detection system based on the combination of spectral acquisition and real-time processing has become a research hot spot in the field of liquor authenticity detection [9].

In this paper, a handheld liquor authenticity detection system based on laser spectrum technology is developed. The microlaser spectrometer independently developed by the laboratory is used to detect liquor, and the signal acquisition of laser spectrum is realized by the CPU chip. Then, the signals from the microspectrometer are wirelessly transmitted to the Android smartphone terminal by Wi-Fi. The mobile phone APP can automatically adjust the laser intensity to avoid the phenomenon of fluorescence saturation. Finally, we used the algorithm of Pearson correlation coefficient to match the test spectrum with the standard spectrum stored in the database and visualized its similarity on the APP interface. The experimental results show that the detection of the authenticity of liquor can be realized in real-time by our handheld microlaser spectrometer system.

2. Detection Principle

2.1. Laser-Excited Characteristic Spectra of Liquor. The laser has the advantages of high intensity, high brightness, and a good monochromator. When the laser beams are irradiated on the liquor, it can excite the water, ethanol, and soluble molecules to produce detectable laser spectroscopic signals such as Mie scattering [10], Raman scattering [11], fluorescence scattering [12], and so on [13, 14]. Because of the different components between liquors, the laser-excited spectra are also distinct. If the compositions or contents of the liquor change, the obvious differences should be observed in the spectrum, which provides an objective and reliable basis for the identification and evaluation of the liquor [15, 16]. Figure 1 is a typical spectrum curve of the liquor after laser (405 nm) excitation. As shown in Figure 1, it includes a laser scattering peak, a Raman scattering peak of ethanol, a Raman scattering peak of water, and the fluorescence spectrum of the liquor. The identification of liquor by the laser spectrum method needs to analyze fluorescence spectrum, laser scattering, and water Raman scattering simultaneously.

2.2. Matching Similarity Algorithms. To calculate the similarity of liquor matching, the spectral curve of the tested liquor should be compared with the liquor spectrum saved in the spectral database. The authenticity of the measured liquor can be achieved by the Pearson correlation coefficient [18]. First, by assuming the optical density values of the reference and measured spectra at the same sampling point λ_i to be x_i and y_i, respectively, the intensity of the reference spectrum and the measured spectrum can be expressed as follows:

$$\begin{aligned} x &= (x_1, x_2, x_3, \ldots, x_i), \\ y &= (y_1, y_2, y_3, \ldots, y_i), \end{aligned} \tag{1}$$

where i represents the position of the i^{th} wavelength point.

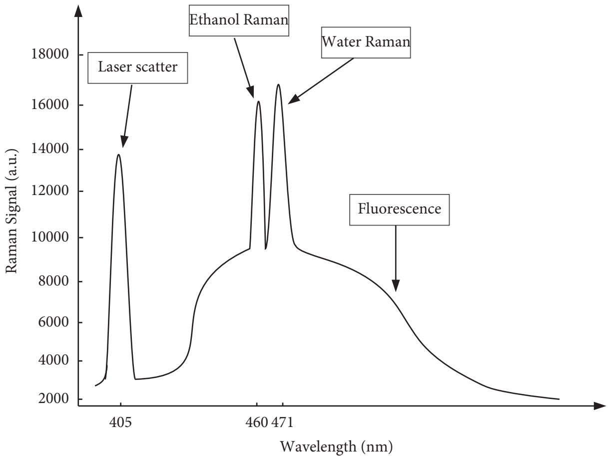


FIGURE 1: The typical spectrum of liquor is produced by laser (405 nm) excitation, which includes a laser scattering peak, a Raman scattering peak of ethanol, a Raman scattering peak of water, and the fluorescence spectrum of the liquor. By analyzing various peaks of the spectrum, the substance content of liquor can be detected, achieving the goal of authenticity detection of liquor [17]. a.u., arbitrary units; nm, nanometer.

The Pearson correlation coefficient is used to quantify the correlation between the two spectra within a value between −1 and 1. By averaging the variables, this coefficient reduces the influence of the individual point differences on the overall similarity between two variables. The Pearson correlation coefficient is defined as the quotient of the covariance and standard deviation of the two variables, which can be expressed as follows:

$$S_3 = \rho_{x,y} = \frac{\operatorname{cov}(x, y)}{\sigma_x \sigma_y} = \frac{\sum_{i=1}^{n} (x_i - \overline{x})(y_i - \overline{y})}{\sqrt{\sum_{i=1}^{n} (x_i - \overline{x})^2} \sqrt{\sum_{i=1}^{n} (y_i - \overline{y})^2}}, \tag{2}$$

where $\rho_{x,y}$ is the Pearson correlation coefficient of the two spectra, $\operatorname{cov}(x, y)$ is the covariance of the two spectra, σ_x is the standard deviation of spectral x, σ_y is the standard deviation of spectral y, $\overline{x}$ is the average value of spectral x, and $\overline{y}$ is the average value of spectral y. Based on this equation, it can be found that if the value s_3 is 0, it indicates that there is no relationship between the two variables. When s_3 is closer to 1, which means the more positive correlation between the two variables, the two spectra are more similar.

2.3. Laser Intensity Calibration. In principle, the Pearson correlation coefficient can give the similarity between the measured and the reference spectra, but in practice, the laser intensity should be considered since it has a great influence on the shape of the laser spectrum curve. If the laser intensity is too high, the phenomenon of fluorescence saturation or detector saturation may appear. If the laser intensity is too low, the spectral intensity will be relatively low and the noise will be relatively large, which will affect the accuracy of matching similarity. Therefore, it is necessary to set the appropriate laser intensity to calculate the similarity of the two spectral curves. In the calibration process, pure water or distilled water is selected as the standard sample. The spectra from pure water generated by laser (405 nm) excitation are shown in Figure 2. The spectrum curve includes laser scattering [19] and Raman scattering of water. There are no other organic or inorganic impurities in pure water, so no fluorescence spectra are generated. In our self-developed software (APP), it would automatically adjust the intensity of the laser source to make the excited water Raman intensity reach the preset value. For the curve shown in Figure 2, the laser intensity gear is set as the maximum. In addition, different liquors may excite curves with similar shapes under different laser intensities, which will affect the accuracy. In the actual measurement, if the measured liquor is close to fluorescence saturation, the gear of laser output will be automatically reduced to avoid fluorescence saturation. Only the spectral data near the laser intensity gear stored in the database is selected to calculate the matching similarity. Therefore, the laser intensity calibration method proposed here can greatly reduce the matching calculation amount and improve the accuracy of matching similarities.

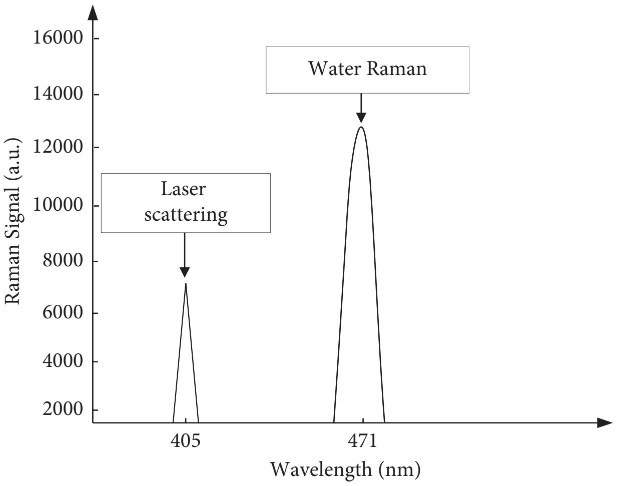


Figure 2: Spectra of purified water produced by laser (405 nm) excitation, which includes laser scattering and water Raman scattering. a.u., arbitrary units; nm, nanometer.

3. Structure of Liquor Authenticity Detection System

The liquor authenticity detection system based on laser spectrum technology is composed of two parts, as shown in Figure 3. The left part is the laser spectrum detector, and the other is the Android phone processing terminal. The laser spectrum detecting instrument includes a laser emission module, spectrometer module, CCD imaging module, charging battery module, wireless routing module, and CPU module. The CPU module is connected to the laser light source, CCD imaging, and wireless routing module, respectively.

To avoid the reflected light and fluorescence interference from the bottle wall of the sample, the laser emission should be directed at a certain angle with the optical axis of the spectrometer. As shown in Figure 3, the laser excitation point is located at the top region of the bottle. As mentioned, the angle of the laser beam ensures that the liquid in the bottle can be excited. On the other hand, the spectral receiving port is located in the lower region of the bottle. Therefore, the fluorescence from the bottle wall produced by laser excitation will not be detected directly by the spectrometer. Although the interference comes from the blurring of the bottle wall in front of the spectral receiver port, this is partially caused by the laser scattering. Considering the low optical power of the scattering, this interference should be extremely low and therefore can be ignored [20]. If this angle is too small, the spectrum received by the spectrometer will contain the reflected light and fluorescence interference from the bottle wall. If the angle is too large, the laser emission point will be too far from the laser excitation point, affecting the intensity of the received spectrum. After extensive experimental verification, for our system, this optimal angle is around 45 degrees.

The basic working process of the system can be described as follows: first, the pulse laser passes through the sample bottle wall and excites the measured liquor to produce the optical signals. Then, the received light through the spectrometer module is converged by the convex lens on the surface of the CCD and converted into an electrical signal which is then transmitted to the CPU module and converted into a digital image signal. Based on the OpenWrt system, the pixel distribution, brightness, and color information of the received image are processed and sent to the mobile phone in MJPG format by the wireless routing module. The spectral curves can be obtained by the processing of APP software. The tested spectrum is compared with spectral data in a standard database, and matching similarity can be given by calculating the Pearson correlation coefficient. If the similarity is greater than 96%, the tested liquor is considered to be true. To avoid the phenomenon of fluorescence saturation, the intensity of the emitting laser can be adjusted automatically by APP through laser intensity calibration.

4. Design of Software

The flowchart of the mobile phone software system is shown in Figure 4. First, the mobile phone terminal receives the laser and spectral image signals transmitted by the spectrometer, and then the spectral signal is filtered and denoised by the APP software to get a spectral curve. The spectral data of the current and adjacent gear of laser intensity in the database are selected and normalized to calculate the Pearson correlation coefficient with the received spectral curve. The software only selects the spectral data of the adjacent laser intensity gear, so it can reduce the number of comparisons with the spectral data in the database and improve the speed and accuracy of recognition.

The matching similarity obtained is displayed on the APP interface as a percentage, and the corresponding name with the highest similarity is displayed. The display interface of the APP software is shown in Figure 5, which includes the spectrum display area, the parameter adjustment area, and the measurement area. The spectrum display area can

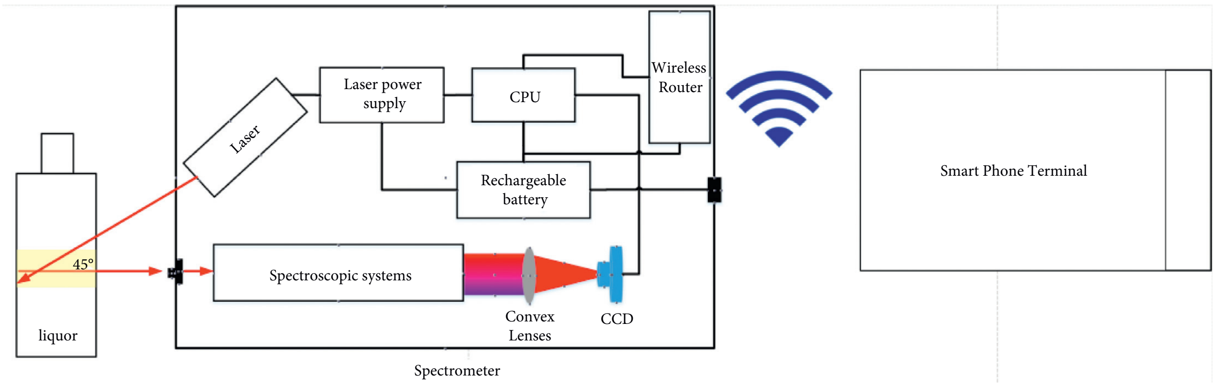


Figure 3: Structure of liquor authenticity detection system based on laser spectrum technology. The left part is the laser spectrum detector, and the other is the smartphone processing terminal. CPU, central processing unit; CCD, charge-coupled device.

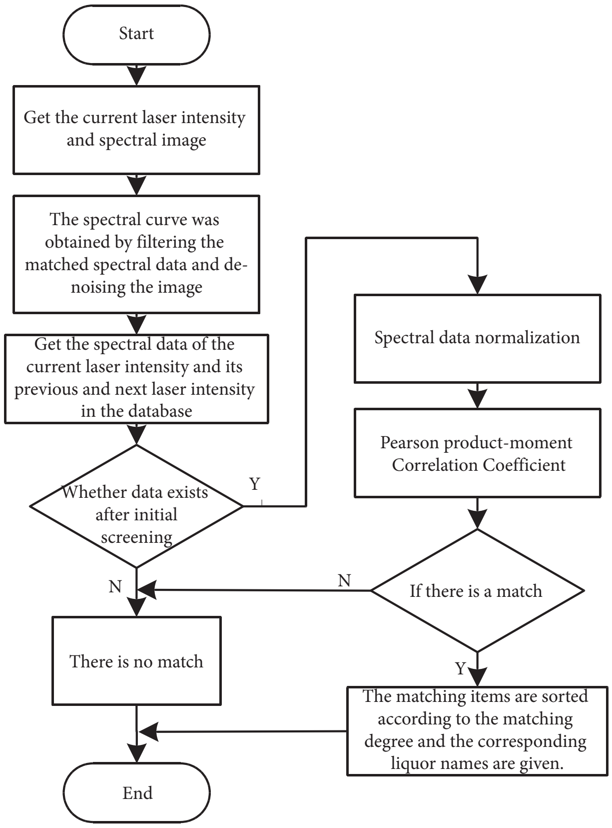


Figure 4: The flowchart of the system.

display the laser spectrum curve, the laser calibration curve, and the reference spectrum curve saved in the database. The parameter adjustment button includes the laser intensity adjustment button and the detector parameter adjustment button. The parameters of the instrument can be adjusted by these two buttons. The measuring button includes a manual measuring button, automatic measuring button, comparison button, pause button, and data saving button. In addition, the text box will be displayed when the spectrum match is finished, the similarity value and the liquor name will be displayed, and the liquor authenticity detection results are also given.

5. Test Results

The physical diagram of the experimental system is shown in Figure 6. The size of the self-developed microfluorescence spectrometer is 103 mm*58 mm*25 mm. In theory, the lower the wavelength of the laser, the better, but the price of the lower laser will rise sharply, and we need to reduce the cost to make the product cheap. At the same time, some current studies [20,21] in this field also show that 405 nm laser products are more commonly used and stable. Based on the above factors, we chose a laser with a wavelength of 405 nm. The CCD pixel is 1280*720, and the CPU chip is Atheros AR9331 SOC. The phone with the Android system and 2 GB memory is used for the test. The liquor sample is put into the glass sample bottle and inserted into the dark box for testing. The spectrum data are transmitted to the mobile phone terminal by the wireless circuit.

The tested liquors were selected from 12 brands in the Chinese market, including 3 Maotai-flavor liquors, 3 Luzhou-flavor liquors, and 6 Fen-flavor liquors. The names and numbers are Beijing Erguotou (${}^{\#}1$), Weihai Wei Shao Guo (${}^{\#}2$), clove love (${}^{\#}3$), the raw pulp (${}^{\#}4$), Beidacang (${}^{\#}5$), bitter mustard (${}^{\#}6$), Jing Zhi Bai Gan (${}^{\#}7$), Hengshui Lao Bai Gan (${}^{\#}8$), Lao Jiu Hu (${}^{\#}9$), Niu Lan Shan aged (${}^{\#}10$), du Er Jiu (${}^{\#}11$), and Xiao Lang Jiu (${}^{\#}12$). To guarantee the accuracy and consistency of measurement results, the laser intensity calibration of the instruments should be carried out first. The sample used for calibration is pure water. By clicking on the "intensity calibration" button in the software interface, the laser intensity is automatically adjusted. When the intensity of the Raman peak from the pure water excited by the laser reaches the set value, the corresponding laser gear is set to be the 10th, which is the maximum number. The APP then prompts successful calibration.

We used the system to measure the spectra of 12 Chinese liquor samples and pure water, as shown in Figure 7. It can be seen that the ethanol Raman and water Raman in Maotai-flavor liquor are submerged in the fluorescence spectrum because of the strong fluorescence from many compounds. The fluorescence intensity of Luzhou-flavor liquor is lower than that of Maotai-flavor liquor because there are fewer

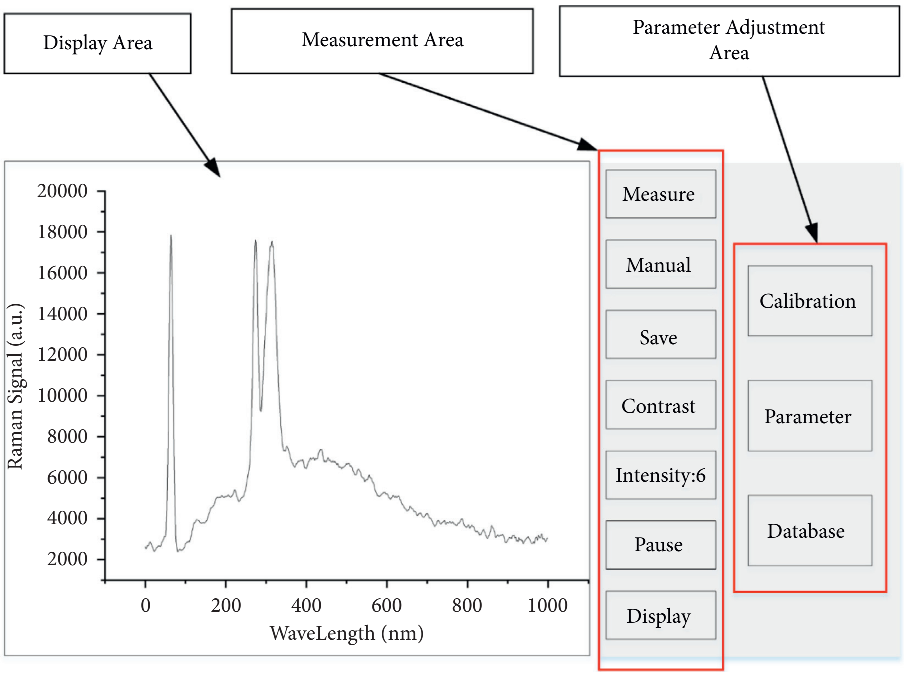


Figure 5: Software display interface, which includes the spectrum display area, the parameter adjustment area, and the measurement area.

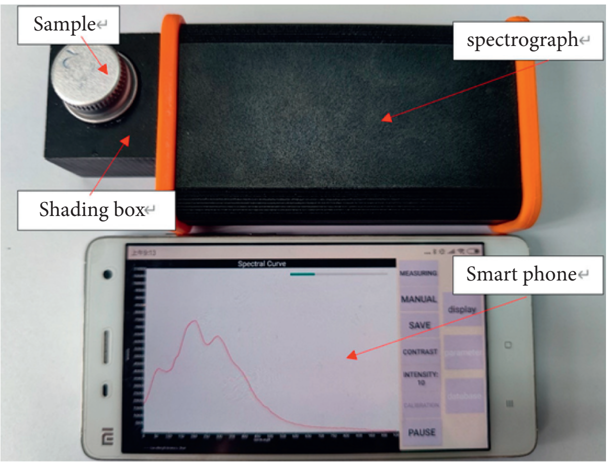
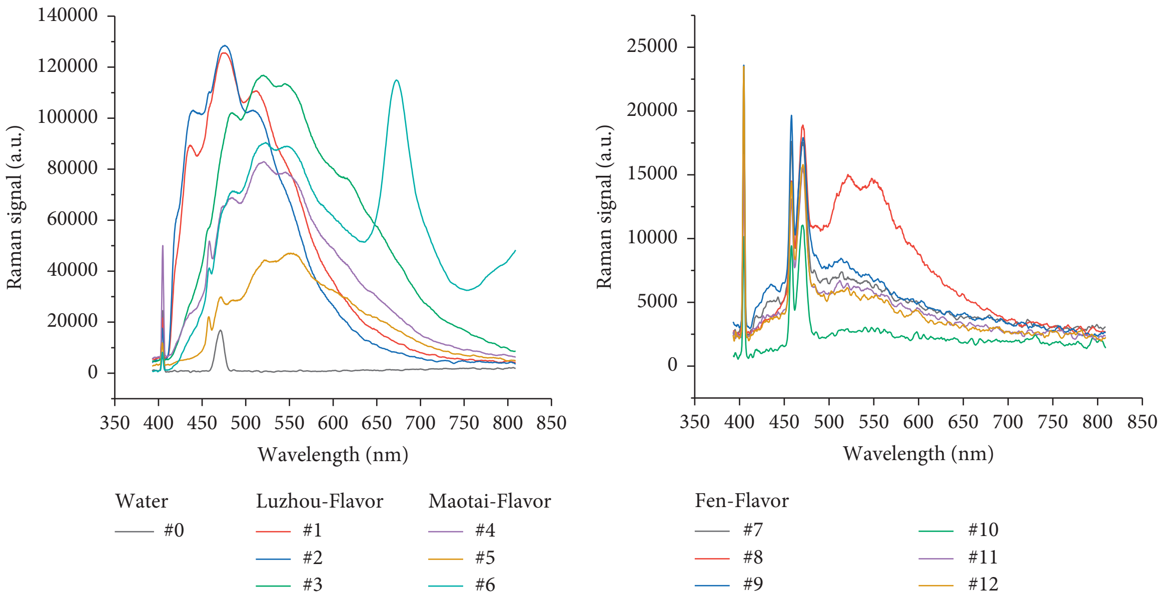


Figure 6: The physical diagram of the liquor authenticity detection system.

blending compounds in Luzhou-flavor liquor (see Figure 7(a)). The Raman signal intensity in Figure 7(a) is higher than that in Figure 7(b). This is because Luzhou-flavor liquor and Maotai-flavor liquor, compared to Fen-flavor, contain more organic matter, which can excite stronger fluorescence. From Figure 7, we can see that the spectral curve of Luzhou-flavor liquor contains ethanol Raman peak and water Raman peak with superimposed fluorescence background. The alcohol Raman peak and water Raman peak can be seen obviously in the Fen-flavor liquor due to less organic matter.

The similarity of 12 liquor samples and pure water is also calculated, respectively, and the results are shown in Table 1. The corresponding laser intensity of each sample is also marked in parentheses. In the table, "—" means that different spirits can be not matched with each other due to the noncorrelation of laser intensity; the self-comparing similarity is close to 100%, and the similarity of different liquors is between 30% and 95%. The similarity detection threshold is set to 96%, above which the liquor can be considered as the real one.

In addition, to verify the consistency of the measured results, we carried out the test on the matching degree stability. Each sample was measured 10 times, to achieve the matching degree error bar graph as shown in Figure 8. The maximum error of the system is kept within ±0.3%, which



(a) (b)

Figure 7: The spectral curves of the samples under their respective measured intensities. Different colors represent different samples, and different types of lines represent different categories, including Luzhou flavor, Maotai flavor, and Fen flavor.

Table 1: The similarity between liquor samples (%). The corresponding laser intensity of each sample is also marked in parentheses, and "—" means that different spirits can be not matched with each other due to the noncorrelation of laser intensity.

Num. (intensity)	Num. (intensity) Similarity												
	$^{\#}$0 (10)	$^{\#}$1 (6)	$^{\#}$2 (6)	$^{\#}$3 (4)	$^{\#}$4 (10)	$^{\#}$5 (10)	$^{\#}$6 (10)	$^{\#}$7 (10)	$^{\#}$8 (10)	$^{\#}$9 (10)	$^{\#}$10 (10)	$^{\#}$11 (10)	$^{\#}$12 (10)
$^{\#}$0 (10)	99.98	—	—	—	50.81	40.86	32.08	60.65	56.44	63.48	85.48	83.02	83.35
$^{\#}$1 (6)	—	99.95	94.32	—	—	—	—	—	—	—	—	—	—
$^{\#}$2 (6)	—	94.32	99.97	—	—	—	—	—	—	—	—	—	—
$^{\#}$3 (4)	—	—	—	99.98	—	—	—	—	—	—	—	—	—
$^{\#}$4 (10)	50.81	—	—	—	99.99	92.47	68.29	79.41	85.67	87.03	56.52	80.92	77.88
$^{\#}$5 (10)	40.86	—	—	—	92.47	99.96	75.01	78.69	91.66	85.33	52.18	75.51	71.30
$^{\#}$6 (10)	32.08	—	—	—	68.29	75.01	99.98	62.93	69.99	67.44	50.96	57.96	56.97
$^{\#}$7 (10)	60.65	—	—	—	79.41	78.69	62.93	99.51	78.13	94.40	63.11	76.10	73.81
$^{\#}$8 (10)	56.44	—	—	—	85.67	91.66	69.99	78.13	99.90	82.73	71.87	86.48	82.57
$^{\#}$9 (10)	63.48	—	—	—	87.03	85.33	67.44	94.40	82.73	99.62	65.88	81.31	78.80
$^{\#}$10 (10)	85.48	—	—	—	56.62	52.18	50.96	63.11	71.87	65.88	99.96	90.45	90.34
$^{\#}$11 (10)	83.02	—	—	—	80.92	75.51	57.96	76.10	86.48	81.31	90.45	99.55	93.15
$^{\#}$12 (10)	83.35	—	—	—	77.88	71.30	56.97	73.81	82.57	78.80	90.34	93.15	99.30

indicates good measurement stability. We can see that the stability of the Fen-flavor liquors ($^{\#}$7–12) in Figure 8 is relatively poor, and we think the reason is that its doping components are compared with the other two types of liquors ($^{\#}$1–3 and $^{\#}$4–6). Therefore, the fluorescence intensity is relatively weak (see Figure 7(b)), and the algorithm in this paper is a comprehensive measurement method of fluorescence and Raman. The signal-to-noise

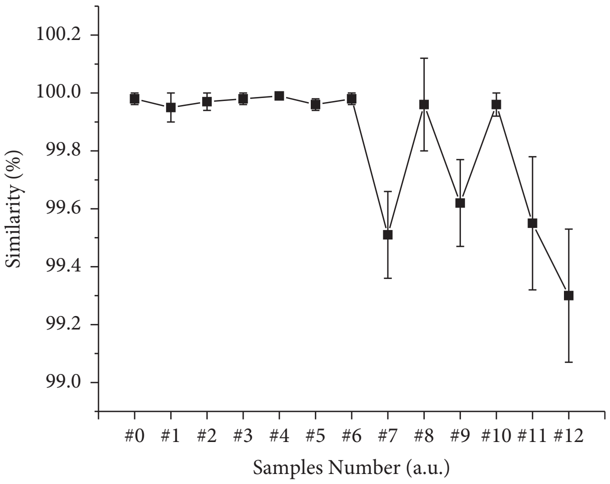


Figure 8: The error bar graph of sample matching error. Each sample was measured 10 times to achieve the matching degree error. %, percent; a.u., arbitrary units.

ratio of the fluorescence part is relatively low, resulting in relatively large errors. The error range is far less than the minimum discrimination interval of different samples in Table 1; therefore, the accuracy of the matching result can be guaranteed.

6. Conclusion

In this paper, the detection of liquor authenticity is studied by using laser spectrum technology. A handheld liquor authenticity detection system based on this technology is developed. In this structure design, the authenticity can be directly tested for the liquor in the transparent glass or bottle in the actual test, which greatly improves the convenience of use. In addition, a laser intensity calibration method based on pure water Raman is proposed in this paper, which can reduce the influence of different spectral instruments, instability of laser power, and changes in external environment. Therefore, the accuracy of spectrum matching between tested liquor and real liquor in the database is improved. The number of spectrum matching is reduced, and the matching speed is improved.

In the experiment, the system was used to test the matching similarity for different liquor samples. The experiment results showed that the result obtained from the system was accurate and stable. The system has the advantages of small volume, low cost, and convenient use. We provide a new method for rapid detection of liquor authenticity.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest to this work.

Acknowledgments

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