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Latent Fingerprint Development by Frequency-Doubled Neodymium:Yttrium Aluminum Garnet (Nd:YAG) Laser: Benzo(f)ninhydrin

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ABSTRACT: The ninhydrin/zinc chloride method for laser latent fingerprint development is not well suited to the use of frequency-doubled neodymium:yttrium aluminum garnet (Nd:YAG) lasers. We have examined several ninhydrin analogues and find that benzo(f)ninhydrin is very well matched to the use of this laser.

KEYWORDS: criminalistics, fingerprints, ninhydrin, zinc chloride, benzo(f)ninhydrin, lasers, frequency-doubled Nd:YAG laser

Since the late 1960s, ninhydrin has been the most commonly used reagent for chemical development of latent fingerprints. Even the advent of latent fingerprint development by laser in 1976 has not diminished the use of ninhydrin. Indeed, zinc chloride ($ZnCl_2$) treatment of latent fingerprints subsequent to ninhydrin application converts the customary purple-blue ninhydrin-aminoacid reaction product to an orange species that is highly fluorescent under argon-ion laser illumination [1]. Laser examination of latent fingerprints can thus be very effectively combined with the ninhydrin method. For safety, the use of fume hoods and gloves is generally recommended for chemical treatments of latent prints and this is pertinent here as well since $ZnCl_2$ is poisonous.

Until about two years ago, the argon-ion laser (Ar-laser) was the only type of laser used for latent fingerprint development. Since then, two additional kinds of laser have found use in fingerprint work, first the copper-vapor laser (Cu-vapor laser), and most recently the frequency-doubled neodymium:yttrium aluminum garnet (Nd:YAG) laser. For fingerprint detection, the Ar-laser is normally operated all lines blue-green (457.9, 465.8, 472.7, 476.5, 488.0, 496.5, 501.7, and 514.5 nm, the 488- and 514.5-nm lines being the strongest). At times it is more advantageous to operate the Ar-laser at a single line to enhance contrast when strong background fluorescence tends to overwhelm the latent fingerprint fluorescence. One generally uses either the 488- or 514.5-nm line, depending on the situation at hand. This will be considered further below. Ar-lasers can also operate in the near ultraviolet [UV] (between about 350 and 365 nm). Although ultraviolet laser illumination for latent fingerprint detection can be effective, [2,3] it is rarely used at present. This may change in the near future, however, since latent print treat-

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ments designed for the ultraviolet have been developed for surfaces, such as brown paper and wood, which are usually not amenable to blue-green laser illumination because of excessive background luminescence [4].

The Cu-vapor laser delivers lines at 510 and 578 nm. The latter is of no value at present for fingerprint development and is quenched by an intracavity dichroic filter. The Nd:YAG laser operates at 532 nm. The Ar-lasers used in law enforcement at this time deliver either 20-W blue-green (and about 2.5-W UV) or 5-W blue-green (and over 100-mW UV, which is adequate for fingerprint work), and are continuous (CW) lasers. The Cu-vapor and Nd:YAG lasers, on the other hand, are pulsed lasers. The Cu-vapor laser operates at about 5 kHz, with pulse energies of about 1.4 mJ, and with pulse duration of about 30 ns. The laser's average power (averaged over many pulses) is about 7 W. While pulsed laser illumination may on occasion be advantageous over CW illumination (the converse occurs as well), the average power generally dictates fingerprint detectability when Cu-vapor and Ar-laser are compared. For fingerprint work, the Cu-vapor laser thus essentially acts much like an Ar-laser operated at 514.5 nm. The Nd:YAG laser (Laser Printfinder, Laser Photonics, Inc.) has pulse energies of about 7 mJ, with pulse duration of about 10 ns. The pulse repetition rate is low, 20 Hz, so that the laser's average power is only about 140 mW. Nonetheless, the Nd:YAG laser is quite effective for fingerprint detection because it "fools" the eye. The eye cannot resolve times shorter than about 0.025 s. Thus, the Nd:YAG laser light and fingerprint fluorescence appear to the eye to be nearly CW, with only a slight flicker, and the Nd:YAG laser light appears quite bright to the eye because each pulse is very intense and the eye's response is logarithmic.

Latent Fingerprint Development by Nd:YAG Laser

While the Nd:YAG laser, because of its portability, is uniquely suited to crime scene work, it should be competitive in the laboratory as well. We have examined the presently most widely used laser fingerprint development procedures for compatibility with the Nd:YAG laser. We have also made a comparison with the Ar-laser in terms of general fingerprint detectability. This comparison, which shows that the Nd:YAG laser is effective for fingerprint work, will be reported in a separate paper [5].

Inherent Fingerprint Fluorescence

The results one obtains with Nd:YAG lasers are similar to those obtained with Ar-lasers or Cu-vapor lasers. We note at this juncture that the orange long-wavelength-pass filter with nominal cutoff wavelength (50% of maximum transmission) of 550 nm one customarily uses with Ar-lasers for latent fingerprint detection transmits too much light at 532 nm and is therefore not useful together with the Nd:YAG laser. Instead, an orange long-wavelength-pass filter with cutoff wavelength of 570 nm is used. Band-pass interference filters can be used in addition to enhance contrast [6]. For the Nd:YAG laser, the generally most useful band-pass interference filter has its maximum transmission at about 580 nm and has a bandwidth of about 40 nm.

Dusting with Fluorescent Powders

Commercially available dusting powders designed for UV illumination can be used with the Ar-laser but not the Nd:YAG laser. Commercial powders, such as Mars Red (Criminal Research Products), that respond to green illumination can readily be used with the Nd:YAG laser. Most laser users, however, apply powders containing the fluorescent dye rhodamine 6G (Rh 6G). Such powders can be prepared as described elsewhere [6, Chap. 3, 7]. An effective

magnetic powder, for instance, blends Rh 6G with fine iron filings in 10-mg to 5-g ratio. Rh 6G powders are very well suited to the Nd:YAG laser because the Rh 6G absorption is very well matched to the laser's wavelength [8].

Solution Dye Staining

On nonporous items, latent fingerprints can be developed effectively by Rh 6G solution staining, particularly after cyanoacrylate ester treatment [9]. Again, the Rh 6G absorption is well suited to Nd:YAG laser use. Rhodamine B (Rh B) is a very good alternative to rhodamine 6G for solution (as well as evaporative) staining. Although the Rh B absorption is not as well matched to the Nd:YAG laser as the Rh 6G absorption, the Rh B fluorescence is better matched to the Nd:YAG laser safety filter than the Rh 6G fluorescence [8].

For latent fingerprint development on adhesive tapes, Rh 6G and Rh B can be effective. However, some adhesive tapes show strong yellow fluorescence competitive with the rhodamine 6G or B fluorescence. In these instances, 3,3'-diethyloxadicarbocyanine iodide (DODC) can be an effective substitute [5]. DODC, incidentally, is also well suited to solution staining, but not evaporative staining.

Evaporative Dye Staining

On porous items, evaporative dye staining can be very effective [10], particularly after cyanoacrylate ester treatment [9]. For use with Ar-, Cu-vapor, and Nd:YAG lasers, Rh 6G is the dye of choice. For Nd:YAG laser use, Rh B is comparably effective. For processing of items such as brown paper, wood, and leather, dyes that respond to UV illumination are more effective [4], but are useful only together with Ar-lasers or UV lamps.

Ninhydrin/Zinc Chloride

This chemical method useful for porous items, paper in particular, works very well with the Ar-laser, particularly when the laser is operated at 488 nm where the match with the absorption of the aminoacid/ninhydrin/ZnCl₂ reaction product is at its best [11]. The match with the Nd:YAG laser line is not as good, however, and the ninhydrin/ZnCl₂ treatment does not work nearly as well with that laser as with the Ar-laser. Because the Nd:YAG laser works well with all other current routine laser procedures, and because of its unique portability, hence utility in crime scene work, it is certainly a viable alternative to the Ar- or Cu-vapor lasers. At the same time, a viable laser should be effective in processing paper items, which, after all, occur rather frequently in criminal investigation. Accordingly, we have investigated modifications to the ninhydrin/ZnCl₂ method tailored to the Nd:YAG laser.

Benzo(*f*)ninhydrin

Solution Spectra

One of us (JA) has for some time been engaged in the study of ninhydrin analogues for the development of latent fingerprints [11]. Such analogues, which chemically act like ninhydrin, but which spectroscopically differ significantly from ninhydrin, appeared to us to be logical candidates for use together with the Nd:YAG laser. Accordingly we have examined 2,2-dihydroxy-5-chloro-6-methoxyindane-1,3-dione, 2,2-dihydroxybenz(*e*)indane-1,3-dione (benzo(*e*)ninhydrin), and 2,2-dihydroxybenz(*f*)indane-1,3-dione (benzo(*f*)ninhydrin), namely Com-

pounds II, III, and IV of Ref 11. Solution absorption spectra were measured after reacting these compounds with the amino acid leucine and then with $ZnCl_2$ to determine which, if any, of these compounds might be compatible with the Nd:YAG laser. Spectroscopic measurements used techniques and instrumentation described previously [6, Chap. 2]. The first two of the above listed compounds showed spectra very similar to that which one obtains when reacting ninhydrin itself with leucine and then $ZnCl_2$. Thus, we did not expect that these two ninhydrin analogues would offer any advantage over ninhydrin. Later treatment with them of latent fingerprints, described below, confirmed this expectation. Our results here are quite analogous to those obtained by Almog's group [11] in investigation of these compounds for room light, rather than laser, development of latent fingerprints.

Figure 1 shows the solution absorption spectrum for the benzo(*f*)ninhydrin case. The absorption maximum at 530 nm in Fig. 1 indicates that the 532-nm Nd:YAG laser excitation should be ideally suited to benzo(*f*)ninhydrin/ $ZnCl_2$ treatment of latent prints, whereas the absorption maximum for ninhydrin/ $ZnCl_2$ at 490 nm [1] indicates that the 488-nm Ar-laser line should be best suited to this treatment.

The absorption spectrum in Fig. 1 involved a solution prepared by reacting benzo(*f*)ninhydrin and leucine in acetone with gentle heating until the solution turned greenish brown, resembling the color of the reaction product of benzo(*f*)ninhydrin with fingerprint residue, which is dark green [11]. For measurement of fluorescence spectra, $ZnCl_2$ was added to benzo(*f*)ninhydrin/leucine solutions until they turned reddish purple, resembling, as we shall see, the color of latent prints after benzo(*f*)ninhydrin and $ZnCl_2$ treatment. The concentration of the reagents were not fixed for measurement of absorption as well as fluorescence spectra. Since thus prepared solutions may contain not only the desired reaction product, but also

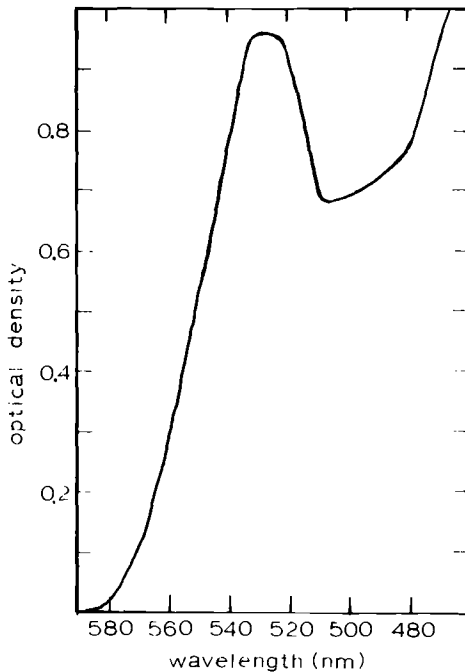


FIG. 1—Solution absorption spectrum of leucine/benzo(*f*)ninhydrin/ $ZnCl_2$ in acetone. See text for discussion.

intermediates, by-products, or aggregates, we measured fluorescence spectra of solutions under 488-, 514.5-, and 532-nm excitation. The resulting fluorescence spectra were virtually indistinguishable from the corresponding ones using a latent print treated with benzo(*f*)ninhydrin-ZnCl₂, which we will present shortly. Solutions, before and after ZnCl₂, will change color and show precipitates if left standing. Similar behavior is found also with ninhydrin.

We investigated some 50 metal salts as potential alternatives to ZnCl₂ as a follow-up to latent print treatment with ninhydrin or its analogues. Not too surprisingly, cadmium and mercury salts converted the ninhydrin-amino acid reaction product to a fluorescent form (zinc, cadmium, and mercury are Group IIB transition metals), but not nearly as well as ZnCl₂, and we found no competitive salts of other metals.

Fingerprint Development

In terms of room light development of latent prints, benzo(*f*)ninhydrin has previously been found to be superior to the other two above mentioned ninhydrin analogues [11]. ZnCl₂ treatment and Ar- or Nd:YAG laser examination revealed that it is also by far the best ninhydrin analogue of the three for laser fingerprint development. The other two analogues were found not to be nearly as good as ninhydrin for purposes of laser examination either with the Ar- or the Nd:YAG laser. The comparison of benzo(*f*)ninhydrin and ninhydrin is shown in Figs. 2 to 7. Figure 2 shows the room light photograph of a latent print on white paper. The left half of the print was treated with benzo(*f*)ninhydrin. The right half was treated with ninhydrin. The left half developed a dark green, the right half the customary purple blue. The developments were comparable, with the left half showing slightly more intense color, however.

All lines blue-green Ar-laser examination after ZnCl₂ showed an orange fluorescence on the left half and a greenish-yellow fluorescence on the right half. Note here that weak prints developed by ninhydrin-ZnCl₂ tend to show greenish-yellow fluorescence under the Ar-laser, whereas

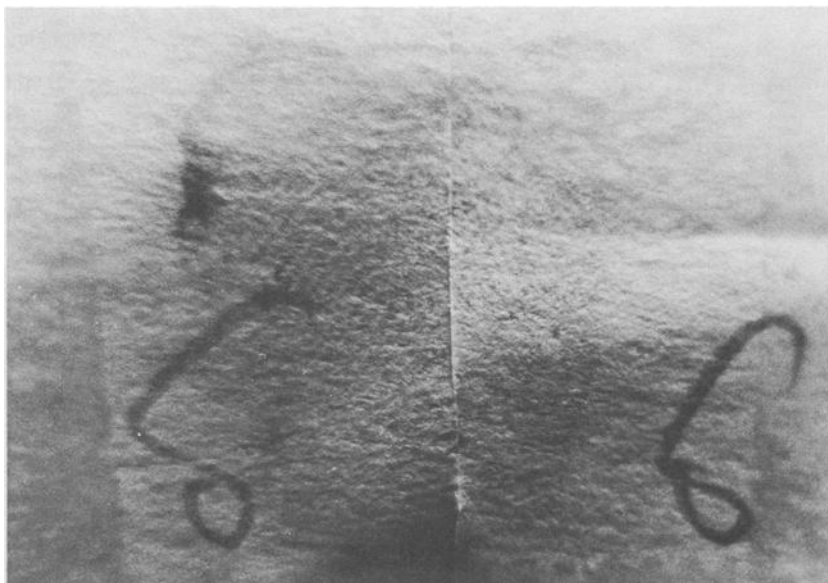


FIG. 2—Room light photograph of latent print on paper developed with benzo(*f*)ninhydrin (left half) and ninhydrin (right half).

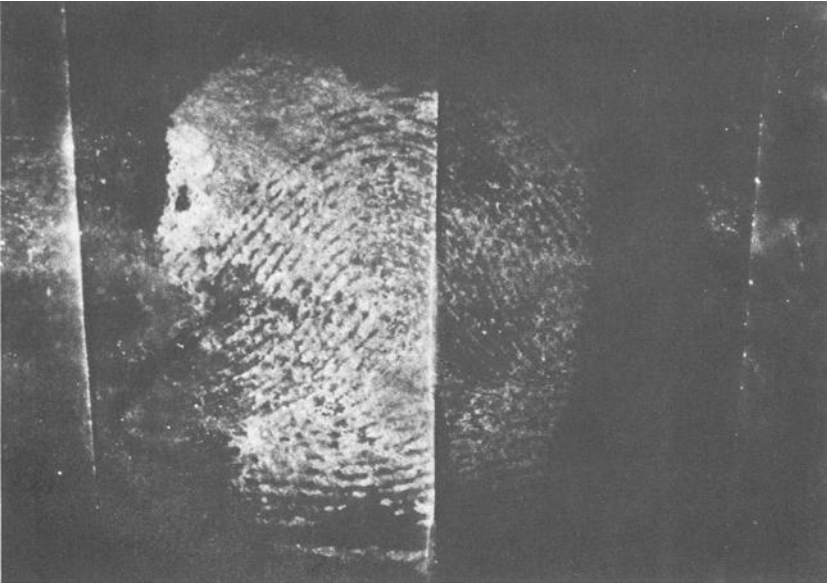


FIG. 3—Print of Fig. 1 treated with $ZnCl_2$ and developed under all lines blue-green Ar-laser excitation.

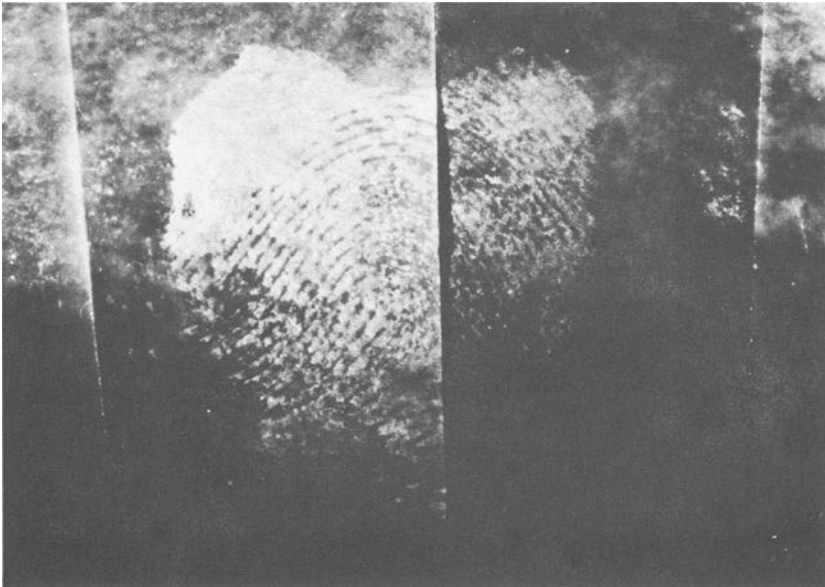


FIG. 4—Print of Fig. 2 developed under 514.5-nm Ar-laser excitation.

stronger prints show orange fluorescence. Prints developed by benzo(*f*)minhydrin $ZnCl_2$ show orange fluorescence regardless of the strength of the latent print. The fluorescence (all lines Ar-laser excitation) of the left half of the print of Fig. 2 was somewhat brighter than of the right half, as shown in Fig. 3. Figures 4 to 6 show the relative fluorescence intensities under 514.5-, 488-, and 457.9-nm Ar-laser excitation, respectively. With 514.5-nm excitation, the

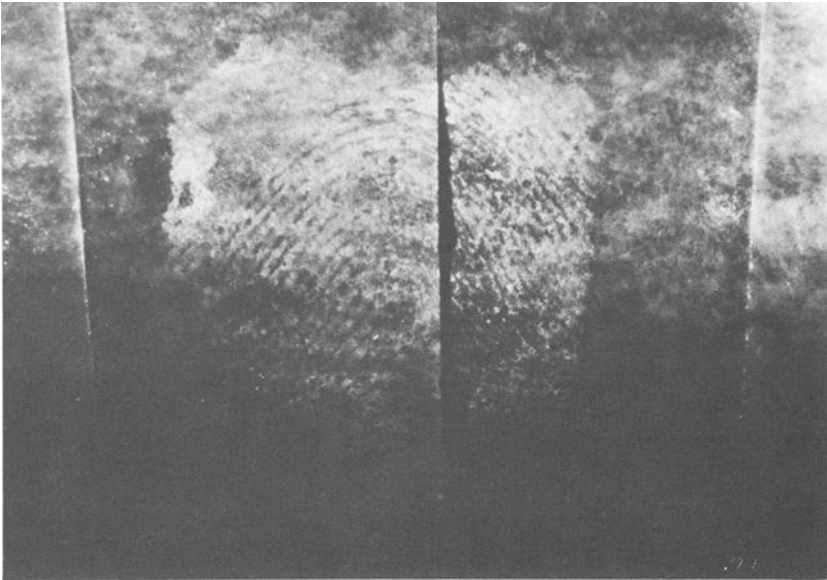


FIG. 5—Print of Fig. 2 developed under 488-nm Ar-laser excitation.

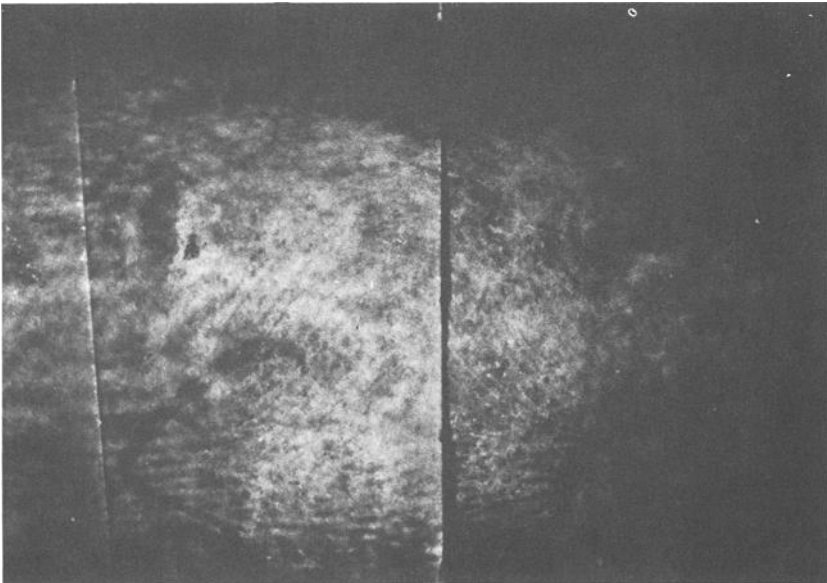


FIG. 6—Print of Fig. 2 developed under 457.9-nm Ar-laser excitation.

benzo(*f*)ninhydrin half of the print developed considerably better than the ninhydrin half as shown in Fig. 4. With 488-nm excitation, the ninhydrin half developed slightly stronger than the benzo(*f*)ninhydrin half, as seen in Fig. 5. Comparable, but faint, developments were obtained with 457.9-nm excitation as seen in Fig. 6.

Finally, Fig. 7 shows the relative fluorescence strengths under the Nd:YAG laser's excita-



FIG. 7—Print of Fig. 2 developed under 532-nm frequency-doubled Nd:YAG laser excitation.

tion at 532 nm. Under this excitation, benzo(*f*)ninhydrin is seen to be far superior to ninhydrin. Benzo(*f*)ninhydrin is, according to Figs. 3 and 4, also a promising alternative to ninhydrin for Ar- and Cu-vapor laser use.

The salient features of Figs. 2 to 7 are as follows:

For room light development of latent prints, benzo(*f*)ninhydrin is as good as ninhydrin, perhaps slightly better. After ZnCl₂, 488-nm excitation is very well suited to ninhydrin. The 514.5-nm excitation is better suited to benzo(*f*)ninhydrin.

Benzo(*f*)ninhydrin is much superior to ninhydrin in concert with the Nd:YAG laser's 532-nm light.

Strong laser exposure will cause photodecomposition, hence fingerprint fluorescence degradation. Fingerprint fluorescence degradation occurs as well within one to several days if treated prints are left in ambient conditions. Finally, increased background fluorescence may also occur in the same time span.

Fingerprint Spectra

To corroborate the compatibility of benzo(*f*)ninhydrin with Nd:YAG laser excitation, we measured the excitation spectrum of the left half of the print of Figs. 3 to 7. An excitation maximum about 540 nm was obtained, closely corresponding to the absorption maximum in Fig. 1. The slight red shift between the solution absorption and fingerprint excitation spectra is reminiscent of red shifts one often encounters when going from solutions to solids generally.

Measurement of fingerprint fluorescence spectra involving both the left and right halves of the latent print of Figs. 2 to 7 served a two-fold purpose, namely to get detailed insight into the fluorescence resulting from the benzo(*f*)ninhydrin treatment to allow selection of filters for optimum fingerprint detectability, and to determine whether the fluorescence efficiency for this ninhydrin analogue is in fact as high as one can reasonably expect.

Measured fluorescence peaks ranged from 550 to 585 nm, depending on excitation, and are shown in Fig. 8. All spectra in this figure are normalized to have the same peak height to facilitate comparison of peak wavelengths and spectral widths. All fluorescences were broad and

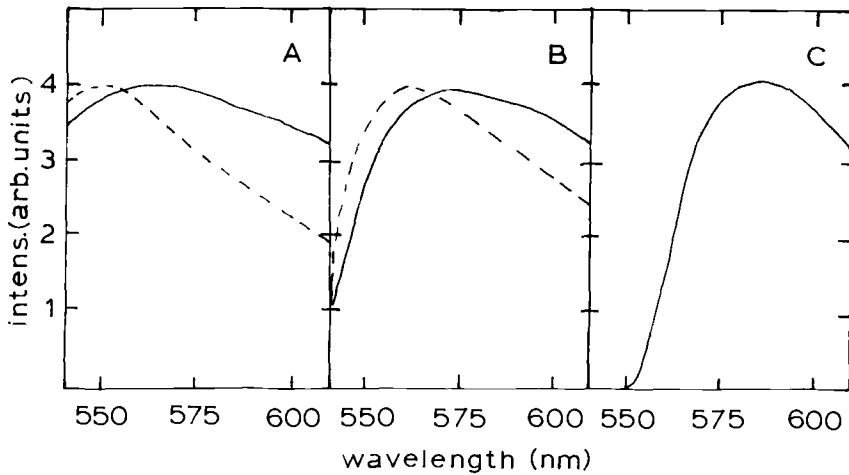


FIG. 8—Fluorescence spectra of print of Fig. 2 (left half = solid line, right half = dashed line) under 488 (A), 514.5 (B), and 532 (C) nm excitation. Peak intensities are normalized to be equal.

featureless, with the fluorescence corresponding to the benzo(*f*)ninhydrin treatment broader, extending substantially further into the red.

For 488-, 514.5-, and 531-nm excitation, the fluorescences were prefiltered with long-wavelength-pass filters with cut-off wavelengths of 530, 550, and 570 nm, respectively. The red shifts in going from A to B to C in Fig. 8 are primarily a matter of this prefiltering, needed to eliminate laser light scattering into the $\frac{1}{3}$ -m monochromator with which we measured the fluorescence spectra. However, the red shift of the fluorescence resulting from the benzo(*f*)ninhydrin treatment (solid lines in Fig. 8) with respect to that from the ninhydrin treatment (dashed lines) is a real effect, and amounts to about 10 nm. At first thought, this seems a rather small red shift, considering the very obvious color difference between the fluorescences produced by the benzoninhydrin/ ZnCl_2 and ninhydrin/ ZnCl_2 treatments, namely orange versus greenish yellow. Note, however, that a comparison of monochromatic greenish yellow and orange reveals only a 10-nm difference, namely 570 to 580 versus 580 to 590 nm, consistent with the "average" colors corresponding to the spectra in Fig. 8 and the measured 10-nm red shift.

Comparison of the peak intensities of the fluorescences of the left and right halves of the print shown in Figs. 2 to 7 gave the following data:

With 488-nm excitation the left/right ratio was 0.8, with 514.5-nm excitation it was 1.2. For the 514.5- versus the 488-nm excitation, the left fluorescence peak intensity remained unchanged. These results, together with absorption and excitation spectral data, indicate that the aminoacid/benzo(*f*)ninhydrin/ ZnCl_2 reaction product produces stronger fluorescence than the corresponding product with ninhydrin under optimized illumination wavelengths.

Interpretation of Spectral Features

If one considers the main spectral features of the reaction products of an aminoacid with ninhydrin and benzo(*f*)ninhydrin and of the reaction product after ZnCl_2 reaction, the following salient features emerge.

1. The green product seen in room light after fingerprint treatment with benzo(*f*)ninhydrin is slightly more intensely colored than the corresponding purple product obtained with ninhydrin, as shown in Fig. 2.

2. Stronger fluorescence is obtained with benzo(*f*)ninhydrin than with ninhydrin under wavelength optimized excitation.

3. Comparison of the aminoacid/ninhydrin and the aminoacid/benzoninhydrin reaction products reveals a red shift between the lowest energy absorption bands of roughly 40 nm.

4. Following the $ZnCl_2$ reaction, a blue shift of some 100 nm occurs in both cases, so that the 40-nm wavelength difference is preserved.

5. In the fluorescence spectra, this wavelength difference is reduced to about 10 nm. The Stokes shift for the aminoacid/benzo(*f*)ninhydrin/ $ZnCl_2$ reaction product is thus rather smaller than for the corresponding ninhydrin product.

We present a qualitative interpretation that provides a consistent picture not only in terms of the involved chemical reactions, but also in terms of the observed spectroscopic behaviour.

Ninhydrin, Compound I of Fig. 9, and benzo(*f*)ninhydrin, Compound II of Fig. 9, react with aminoacids to form Compounds III and IV of Fig. 9, respectively. In the subsequent $ZnCl_2$ reaction, two Molecules III or IV chelate with one Zn^{+2} ion to form a coordination compound involving the charged oxygen and the nitrogen of each of the two Molecules III or IV. This coordination compound is shown as V in Fig. 9. Zinc has the right charge (+2) as an ion, and is well known to chelate with coordination number four. Similar behavior is expected for cadmium and mercury (less for the latter), and we find fluorescent products with cadmium

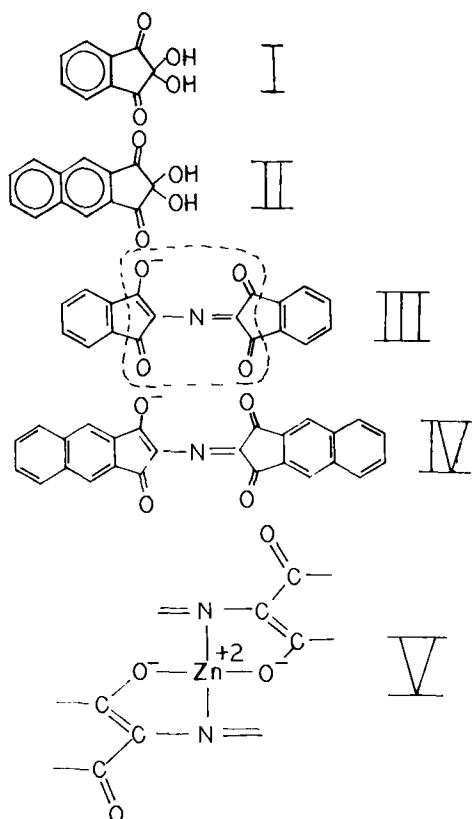


FIG. 9—Ninhydrin (I), benzo(*f*)ninhydrin (II), respective reaction products (III and IV), formed with aminoacids, and coordination compounds formed by III and IV with Zn^{+2} (V). The dashed line in III shows the chromophore of the molecule.

and mercury salts (albeit with fluorescence that is not as good as that obtained with ZnCl_2). Few other metal salts will simultaneously display the proper ionic charge, coordination number, and tendency to chelate. Also, there must be no low-lying metal states that can quench the ligand-centered luminescence. Thus, the unique nature of the ZnCl_2 treatment is perhaps not too surprising.

The increased aromaticity in going from ninhydrin to benzo(*f*)ninhydrin is expected to increase the molar extinction coefficient. This is consistent with 1, and also with 2 because greater absorbance means greater fluorescence, all else being equal. The increased aromaticity, hence increased electron delocalization, is also expected to cause a red shift in absorption, consistent with 3.

The chelation with zinc causes a tight binding of electrons to the zinc, and this is expected to cause a spectral blue shift, consistent with 4. Stokes shifts are essentially a solvent effect. The chromophore in III and IV of Fig. 9 is in the region shown by the dashed line in that figure. The added ring in II compared to I means that solvent molecules should exert a lesser effect in IV than in III. This should also be the situation in V, consistent with 5.

Summary

Our study of fingerprint development with the Nd:YAG laser shows that this is a good laser for fingerprint development. At the same time, it is clear that its most efficient use is achieved when latent print treatments are tailored to its wavelength.

The present routine laser latent print procedures are development by inherent fingerprint fluorescence, dusting or staining with rhodamine 6G (particularly in concert with cyanoacrylate fuming) and ninhydrin/ ZnCl_2 [12]. For dusting with rhodamine 6G based powder or rhodamine 6G staining (vapor or solution), the 532-nm wavelength of the Nd:YAG laser is better matched to the rhodamine 6G absorption [8] than those of the Ar- or Cu-vapor laser. However, some of the rhodamine 6G fluorescence is blocked by the 570-nm cut-off filter needed to block scattered laser light. For Ar- or Cu-vapor lasers, the 550-nm cut-off filter blocks less of the fluorescence. Use of Rhodamine B or DODC may at times be more advantageous for Nd:YAG laser excitation.

The fluorescence and excitation spectra for inherent fingerprint fluorescence [6] show that there the situation is very similar. Thus, in terms of inherent fingerprint fluorescence, dusting and staining, Ar-, Cu-vapor, and Nd:YAG lasers are essentially equivalent. The value of tailoring procedures to the laser on hand becomes very obvious, however, when the benzo(*f*)ninhydrin treatment, the main subject of this paper, is considered. Whereas the ninhydrin/ ZnCl_2 treatment is excellently suited, in terms of its absorption, to the Ar-laser (particularly when operated at the 488-nm line), it is less well suited to the Nd:YAG laser. On the other hand, the benzo(*f*)ninhydrin substitution is best matched to the Nd:YAG laser. The fluorescence of the aminoacid/benzo(*f*)ninhydrin/ ZnCl_2 reaction product is very broad, so that laser line filtering does not present a problem in terms of fluorescence blocking. The fluorescence efficiency obtained with the benzo(*f*)ninhydrin/ ZnCl_2 treatment is good, certainly comparable to that of the ninhydrin/ ZnCl_2 treatment. Thus, it will likely be difficult to find ninhydrin analogues superior to benzo(*f*)ninhydrin. We plan, however, to extend the present study to vicinal triketones that are not ninhydrin analogues.

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