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The Fluorescence and Phosphorescence Spectra and Phosphorescence Decay Time of Harmine, Harmaline, Harmalol, Harmane, and Norharman in Aqueous Solutions and EPA at 77 K

The plant *Paganum Harmala* is found in India, Mongolia, northern China, and in the dry range lands of the American Southwest. The seeds of the plant contain the hallucinogenic alkaloids harmine, harmaline, harmol, harmalol, and harmane [*1–4*]. These alkaloids are also found in the South American plant species *Banisteriopis*. The hallucinogenic properties of the juice from *Banisteriopis* plants are employed by several Amazonian Indian tribes to produce visions for ceremonial purposes [*1–4*]. In addition a similar alkaloid is found in the pineal body [*5*]. As hallucinogens these alkaloids are reportedly more active than mescaline or LSD [*4*].

The nonhallucinogenic parent compound of these alkaloids is norharman (β-carboline). Harmane is 1-methyl-β-carboline. Harmine is 7-methoxy-1-methyl-β-carboline. Harmol is 7-hydroxy-1-methyl-β-carboline. These four compounds retain the full π orbital characteristics of the skeletal ring structure. Harmaline and harmalol substitute additional hydrogens onto the ring. Harmaline is 3,4-dihydro-7-methoxy-1-methyl-β-carboline. Harmalol is 3,4-dihydro-7-hydroxy-1-methyl-β-carboline. The exact mechanism of the hallucinogenic action of these alkaloids is not known. It is known that harmine is a monoamine oxidase inhibitor [*6*]. Also it is obvious that substituent groups must be added to norharman's structure for hallucinogenic activity.

A number of papers have now been published on the physiological effects of harmine and harmaline. It is not known at this time whether these drugs are sold on the illicit drug market [*7*]. The primary aim of this research was to further establish reliable quantitative means of detection of the drugs. Slotkin and co-workers successfully demonstrated the feasibility of harmine detection in blood and urine of man and rats [*8*]. The authors injected harmine hydrochloride into the blood of rats, and human volunteers then took blood and urine samples at appropriate time intervals. Sample separations including paper chromatography were used before quantitative detection by fluorometry. Thus, analysis by emission spectroscopy as demonstrated by Slotkin and co-workers has been shown to be very sensitive even in the dilution range of nanograms/millilitre. Therefore,

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we were interested in characterizing the emission spectra of these alkaloids utilizing techniques readily suitable for routine analysis.

The absorption spectra for all these commercially available compounds are known. Absorption takes place in the near ultraviolet with the first absorption maximum typically near 340 nm with an extinction coefficient of $10^{4.6}$ [9]. Harmaline and harmalol are yellow-green solids forming yellow solutions in water. The other compounds are essentially colorless. S. Udenfriend et al [9,10] examined the fluorescence spectrum of harmine. However, only the emission maximum near 400 nm was reported. Even though the blue and/or green emission is known for these compounds, apparently these are the only references to a characterized emission spectrum.

It was our purpose to, first, characterize the fluorescence emission spectra of the six compounds in order to determine the band origins and band shapes. Second, we wished to establish limits of detectability for aqueous solutions with increased sample dilution. Third, we were curious to know the effect of pH on emission band shape and origin, as these compounds are amines and in the case of harmalol and harmol have hydroxy groups that are acidic. A second aspect of this research was to search for phosphorescence emission in EPA (ethyl ether, isopentane, ethanol mixed by volume 5:5:2) glass matrices at 77 K. Measurement of phosphorescence band origin and band shape, relative fluorescence to phosphorescence quantum yield, and phosphorescence decay times would provide additional means for detection and identification of these compounds.

Experimental

Harmine, harmaline, harmalol, harmane, harmol, and norharman were purchased from Aldrich Chemical Co. As purchased the compounds were 98 to 99 percent pure and were used without further purification. For fluorescence studies in aqueous solution, stock solutions of the six compounds were made at exact concentrations of 1×10^5 ng/ml which is approximately 5×10^{-4} *M*. Harmine, harmaline, harmalol, and harmol were purchased as HCl salts. Thus, the pH of these solutions was 5.75 to 6.70. Norharman and harmane dissolved in water only with the addition of a few drops of 1 *N* HCl, which give a pH of 2.75 to 3.00. These stock solutions at these pH values were used for the entire aqueous study.

Fluorescence excitation was accomplished using a PEK 75-W high pressure xenon arc filtered with a 313-nm interference filter (Corion Instrument Corp.). The filtered light was focused onto a 1-cm square UV cuvet (Beckman) with a 1-in. diameter and 1-in. focal length quartz lens. The fluorescence was measured 90 deg to the excitation light. To eliminate stray light a sample holder was constructed which attached directly to a plate covering the slit of the monochromator. Thus, no stray light entered the slits of the monochromator and the experiments were carried out in room light. A modified Czerny-Turner 0.5-m McPherson model 216.5 f/8.7 scanning monochromator with a 1200 lines/mm replica grating blazed for 500 nm in the first order was used. Typical slit widths were 800 μm as the emission was quite broad. The emission was photoelectrically detected using an EMI 9785B S-20 extended range photomultiplier, typically operated at 800 to 1000 V with a Fluke 415B voltage power supply. The signal was amplified with a Keithly Instruments model 610 picoammeter and recorded on a Sargeant-Welch SRG strip chart recorder.

Measurements using EPA mixed solvent matrices (Matheson, Coleman, Bell) at 77 K, incorporated a quartz tube filled with approximately 20 ml of solution immersed in liquid nitrogen contained in a quartz dewar. The dewar was placed in a sheet metal container

which attached to the monochromator. Again the emission was measured at 90 deg to the exciting light, which was filtered as previously described. Typical solution concentrations were 2×10^{-4} g of sample dissolved in 20 ml EPA mixture, giving a molarity of about 10^{-4} *M*.

For pure phosphorescence studies a chopper was constructed which was simply a rotating can with three 0.5-in.-wide slits arranged 120 deg from each other. The dewar and sheet metal container were held inside the can. The chopper operated at about 250 rpm, giving an effective time between excitation and emission detection of 60 milliseconds. With adequate radiation shielding to eliminate stray light, no filtering was necessary. However, a Corning blue 7-54 band pass filter was used to ensure that no stray light was recorded in the region of phosphorescence. Solvent blanks showed no long-lived emission.

The phosphorescence emission was usually intense and could be seen up to 45 s after excitation cut off. Therefore, measurement of phosphorescence decay versus time was easily performed by chopping the exciting light by hand and following the emission decay on a strip chart recorder. In all cases the decay was first order. The phosphorescence decay time was obtained from the slope of the log of intensity versus time plots.

Results in Aqueous Solutions

The emission spectrum of each of the six compounds in aqueous solution was scanned from 300 to 600 nm. The fluorescence emission spectra of the six solutions are shown in Figs. 1 through 6. Also shown in the same figures are the emissions as a function of pH. In all cases the fluorescence was very intense blue or blue-green emission. As expected, no phosphorescence was observed in aqueous solutions at room temperature.

As shown in Fig. 1 the fluorescence origin of the parent compound, norharman, at pH of 2.75, is at 395 nm (25.4×10^3 cm^{-1}). The fluorescence shows two main features: a broad peak at 430 nm and a second broad peak of equal intensity at 463 nm. There is no change in the spectrum in the acid region. However, addition of two drops of 1 *N* NaOH to 50 ml of norharman solution produces a pH of 12.15 and a significantly different spectrum. The basic solution spectrum is very weak, broad, and featureless. The emission origin is shifted to about 357 nm. Of course the change in emission intensity may in part be due to the change in the absorbance in the region of the 313-nm exciting light. With the optical arrangement previously described, the band shape and origin of the acid solution were readily observable at a dilution of 10^3 ng/ml. An additional dilution to 10^2 ng/ml produced an observable emission, but the features of the spectrum were not clear.

Fig. 2 shows the spectrum of harmane in acid and base solutions. In acid the spectrum's origin is similar to that of norharman. The fluorescence origin is at 387 nm (25.9×10^3 cm^{-1}). The spectrum shows a maximum at 420 nm and a shoulder at 460 nm. The spectrum is unchanged in the acid region, but addition of base produces the second spectrum shown in Fig. 2. The origin of the fluorescence is shifted to 355 nm and a new shoulder is observed at 370 nm. The other features of the spectrum remain the same but with an overall loss of intensity. The emission spectrum in acid is still discernible, even at 10^2 ng/ml.

The fluorescence emission spectrum of harmine as a function of pH is shown in Fig. 3. At a pH of 6.35 the emission origin is at 369 nm (25.1×10^3 cm^{-1}). The spectrum shows a maximum at 410 nm with a second peak of nearly equal intensity at 428 nm. In addition, there is a shoulder located at 458 nm which is half as intense as the maximum. The emission spectrum is the same at a pH of 2.50 but with some loss in intensity. However, in

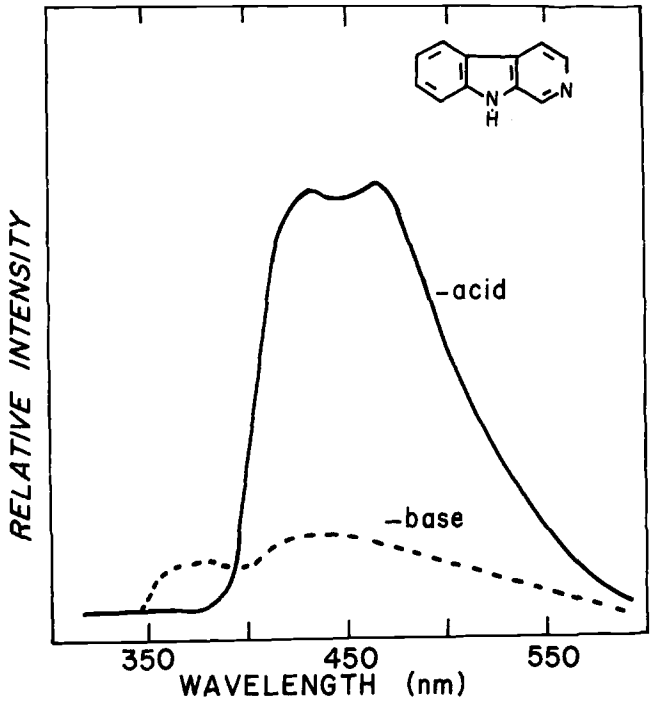

FIG. 1—*Fluorescence spectrum of aqueous norharman.*

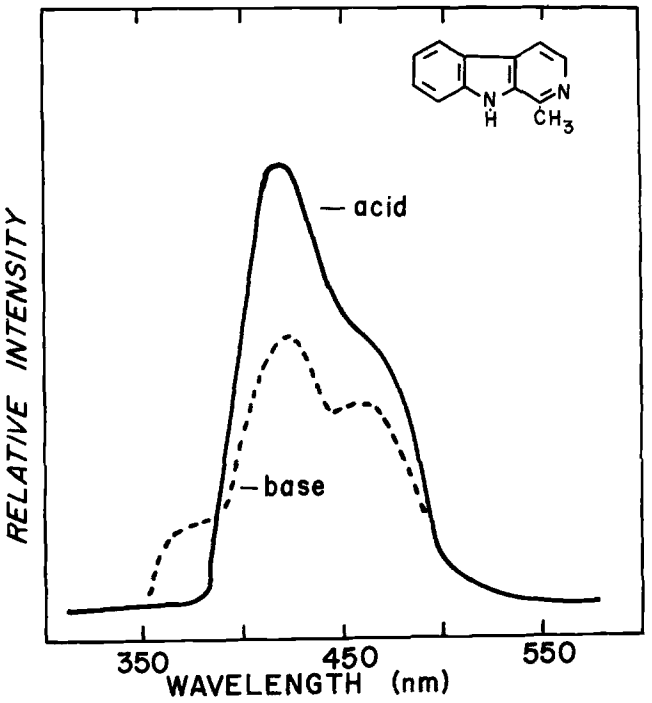

FIG. 2—*Fluorescence spectrum of aqueous harmane.*

base at pH of 11.20 the emission spectrum is stronger than in acid. The emission origin is shifted to 342 nm, the shoulder at 458 nm develops into a separate peak, and another shoulder develops at 360 nm. The emission of harmine is in any case very intense and is readily observable at 10^2 ng/ml.

The emission of harmol, shown in Fig. 4, originates at 370 nm (27.0×10^3 cm^{-1}) with maxima at 410 and 428 nm and a shoulder at 455 nm. The spectrum is unchanged in acid, but in base at pH of 11.00 the emission is a broad weak continuum originating at 410 nm, with a broad maximum centered about 460 nm. The emission is strong and may be observed at 10^2 ng/ml.

The fluorescence of aqueous harmaline is shown in Fig. 5. The emission originates at 401 nm (24.9×10^3 cm^{-1}) with a shoulder or peak at 425 nm and a maximum at 470 nm. Apparently there is no change in these spectral features in either acid or base. However, the emission has its maximum intensity at pH of 3.00. As shown in Fig. 5, the emission in basic solutions is considerably less intense than in acid. Dilution of the stock harmaline solution at pH of 5.75 reduces the fluorescence intensity to the extent that with our optical arrangement, the emission could be detected with certainty at only 10^3 ng/ml. In addition, we noted that after a few months of storage the original yellow solution becomes yellow green, perhaps indicating photolytic decomposition. This change is also reflected in a slight change in the emission spectrum.

The fluorescence emission of aqueous harmalol is shown in Fig. 6. At pH of 6.40 the emission originates at 376 nm (26.6×10^3 cm^{-1}), with a peak at 420 nm and another of equal intensity at 465 nm. In acid the spectrum does not change, even down to pH of 1.50. In base at pH of 11.00, the spectrum is a broad continuum originating at 450 nm with a maximum at 520 nm. Addition of more NaOH to bring the pH to 12.70 shifts the spectrum back to an origin at 400 nm, with a broad maximum at 465 nm. The fluorescence emission of harmalol is similar in intensity to harmaline's. It too can only be observed with certainty down to 10^3 ng/ml.

Results in EPA Matrices

Examination of the emission spectra of the six alkaloids in EPA matrices at 77 K revealed that, except for slight solvent shifts, the emissions were similar in location and intensity, as were those at room temperature in aqueous solution. Usually no separate emission to the red of the fluorescence could be positively identified with the expected phosphorescence. However, a long-lived blue or green phosphorescence was easily observed by chopping the exciting light. Therefore, we concluded that the phosphorescence was weaker than the fluorescence and hidden under the red tail of the fluorescence. By examination of the emission spectra of the alkaloids using a phosphoroscope, the pure phosphorescence was observed in the same region as the fluorescence. Figures 7 through 12 show both the mixed fluorescence and phosphorescence and the pure phosphorescence.

As shown in Fig. 7, when excited by the 313-nm line, norharman in EPA at 77 K emits strongly in the blue region. The mixed fluorescence-phosphorescence originates at 385 nm (26.7×10^3 cm^{-1}), with a broad peak at 415 nm and a slight shoulder at 460 nm. As observed with the phosphoroscope the pure phosphorescence is 20 times weaker. However, much of the intensity loss is due to the use of the phosphoroscope technique. The origin of the triplet system is located at 412 nm (24.3×10^3 cm^{-1}), which is only 2400 cm^{-1} further to the red of the fluorescence system. The phosphorescence has three peaks located at 422, 436, and 465 nm. The phosphorescence decay time was measured in at least two different experiments and at different wavelengths over the entire region of the spectrum. The decay time of norharman in EPA is 4.27 ± 0.14 s.

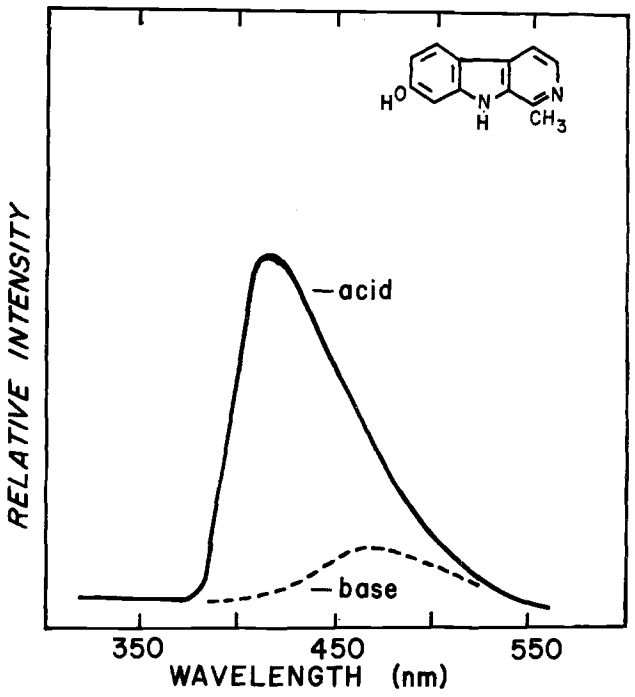

FIG. 3—*Fluorescence spectrum of aqueous harmine.*

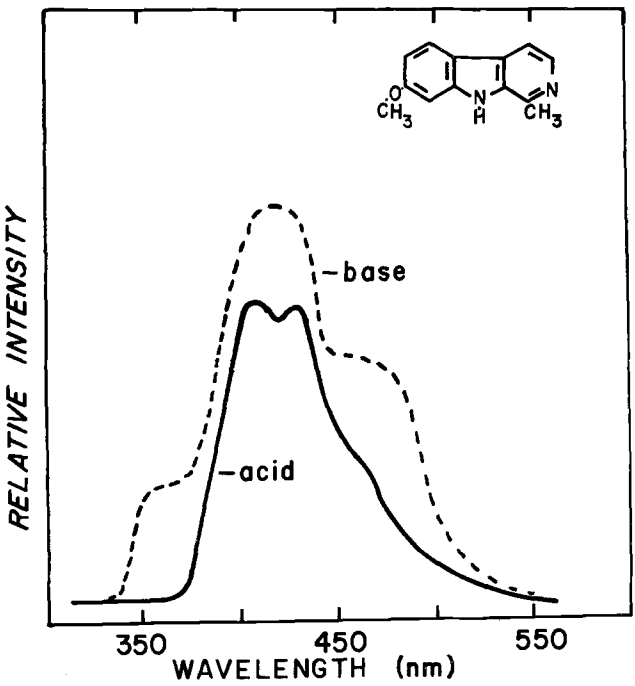

FIG. 4—*Fluorescence spectrum of aqueous harmol.*

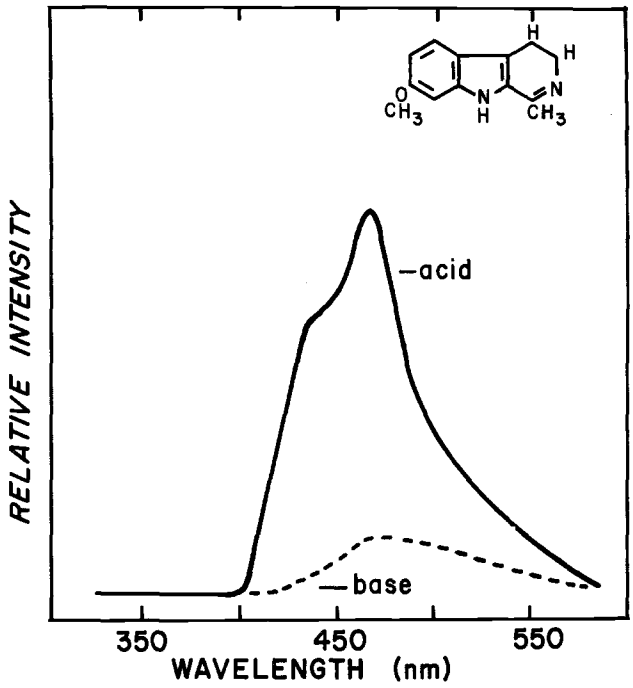

FIG. 5—*Fluorescence spectrum of aqueous harmaline.*

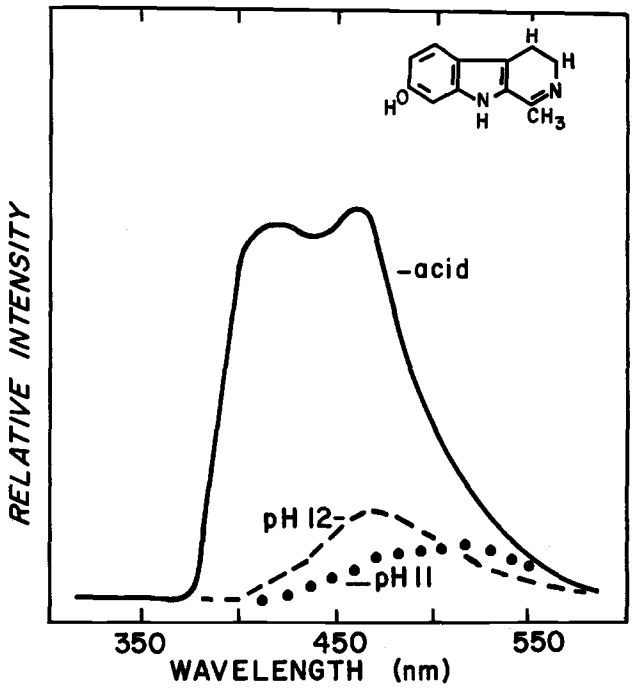

FIG. 6—*Fluorescence spectrum of aqueous harmalol.*

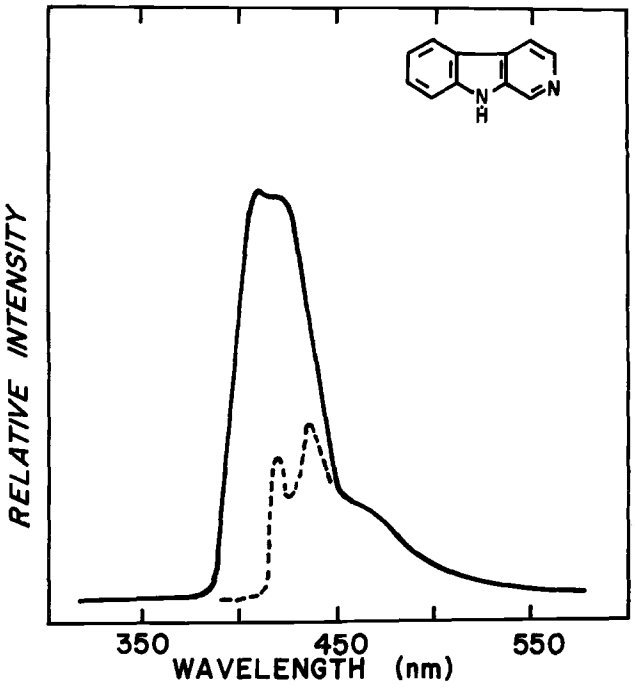

FIG. 7—*Fluorescence and phosphorescence spectrum of norharman in EPA matrix at 77 K.*

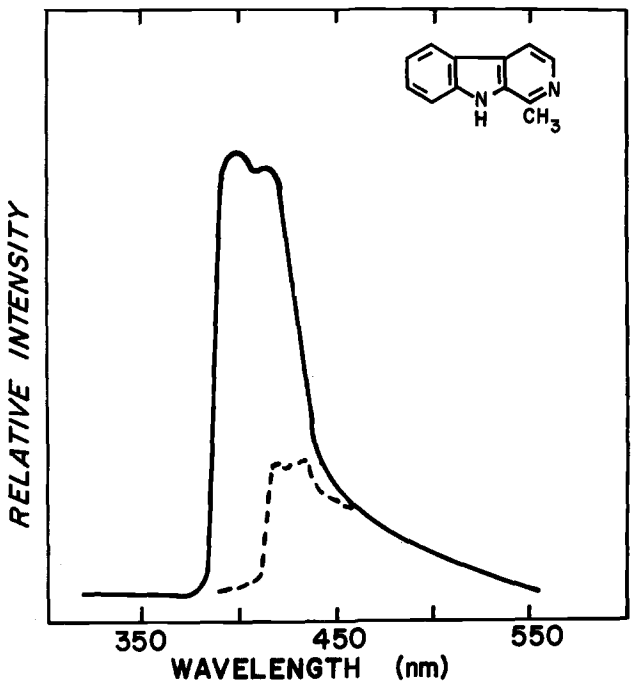

FIG. 8—*Fluorescence and phosphorescence spectrum of harmane in EPA matrix at 77 K.*

Figure 8 shows the mixed fluorescence-phosphorescence spectrum of harmane in EPA matrix. The spectrum originates at 377 nm (26.5×10^3 cm^{-1}). Two peaks of nearly equal intensity are observed at 395 and 412 nm. Also shown in Fig. 8 is the pure phosphorescence spectrum, which is approximately 10 times weaker. The spectrum is very similar to that of norharman. It originates at 412 nm (24.3×10^3 cm^{-1}) and has three peaks at 422, 433, and 462 nm. Thus, the observed triplet state emission is only 2200 cm^{-1} below the emitting singlet. The decay time of harmane phosphorescence is quite long. As measured over several wavelengths the average decay is 5.70 ± 0.20 s.

The emission spectrum of harmine in EPA is shown in Fig. 9. The emission has two distinct regions. First the fluorescence emission origin is near 368 nm (27.2×10^3 cm^{-1}). In this region two intense peaks are observed at 381 and 400 nm. To the red of this system is a second broad band centered around 460 nm, which is approximately five times weaker than the blue system. As shown in Fig. 9, the system to the red is identified as phosphorescence. As observed with the phosphoroscope the origin of this system is at 403 nm (24.8×10^3 cm^{-1}). Two small peaks at 408 and 420 nm are observed to the blue of the main peak at 465 nm. Again the emitting triplet state is 2400 cm^{-1} below the emitting singlet state. The phosphorescence nature of the system was confirmed by decay time measurement. The observed decay time in the entire region is 3.20 ± 0.16 s.

The emission spectrum of harmol in EPA is shown in Fig. 10. Here again both the fluorescence and phosphorescence are resolved. Using the phosphoroscope the pure phosphorescence origin is observed. In EPA the phosphorescence is approximately five times weaker than the fluorescence. The fluorescence origin is near 369 nm (27.1×10^3 cm^{-1}), with two peaks of equal intensity at 383 and 400 nm. The green phosphorescence origin is located at 445 nm (22.5×10^3 cm^{-1}), or 4600 cm^{-1} to the red of the fluorescence origin. The phosphorescence has one main peak located at 475 nm, but in addition there is some structure evident in this system. The observed phosphorescence decay time in EPA at 77 K is 3.35 ± 0.02 s.

The mixed emission spectrum of harmaline in EPA matrix is shown in Fig. 11. The mixed spectrum shows some structure, but since the phosphorescence is only three times weaker some of this structure must be due to the phosphorescence. The fluorescence origin is located near 404 nm (24.8×10^3 cm^{-1}), with peaks at 425 and 455 nm. The phosphorescence originates at 408 nm (24.5×10^3 cm^{-1}), with maxima located at 422, 432, and 463 nm. Thus, the emitting triplet is within 300 cm^{-1} of the excited singlet state. The measured decay time of the phosphorescence in EPA is 3.46 ± 0.13 s.

Figure 12 shows the emission spectrum of harmalol in EPA at liquid nitrogen temperature. Both the fluorescence spectrum and the phosphorescence spectrum are resolved. In EPA the fluorescence originates at 369 nm (27.1×10^3 cm^{-1}), with maxima of nearly equal intensity at 385 and 402 nm. The green phosphorescence origin is near 447 nm (22.4×10^3 cm^{-1}), or 4700 cm^{-1} lower in energy than the fluorescence. The phosphorescence shows one broad peak centered around 471 nm. The decay time of the phosphorescence in EPA at 77 K is 3.61 ± 0.06 s. Both harmaline and harmalol form light yellow solutions at room temperature in EPA. Upon cooling to 77 K the yellow color disappears. Presumably the loss of color is due to lowered population of the ground state hot bands. A similar phenomenon is observed in pure sulfur upon cooling to liquid nitrogen temperature [*11*].

Discussion

As expected, the fluorescence emission spectra of the six β-carbolines are mirror images of the absorption into the first excited singlet state. From the strength of both the absorp-

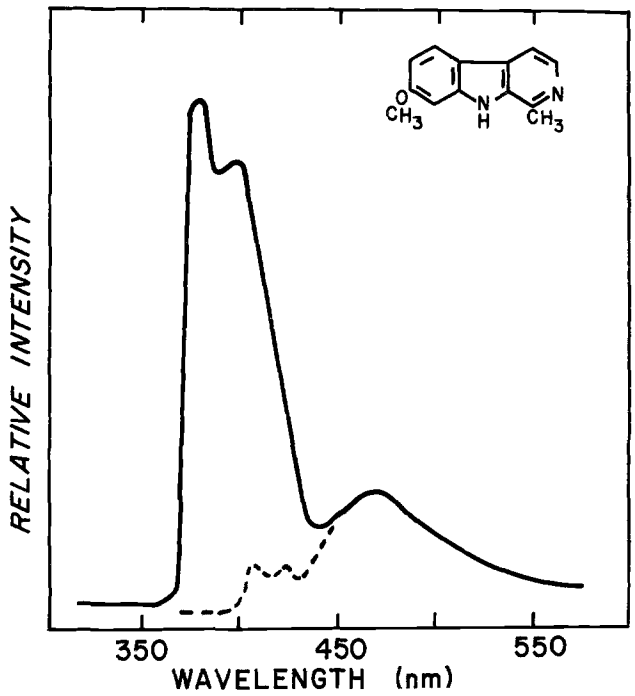

FIG. 9—*Fluorescence and phosphorescence spectrum of harmine in EPA matrix at 77 K.*

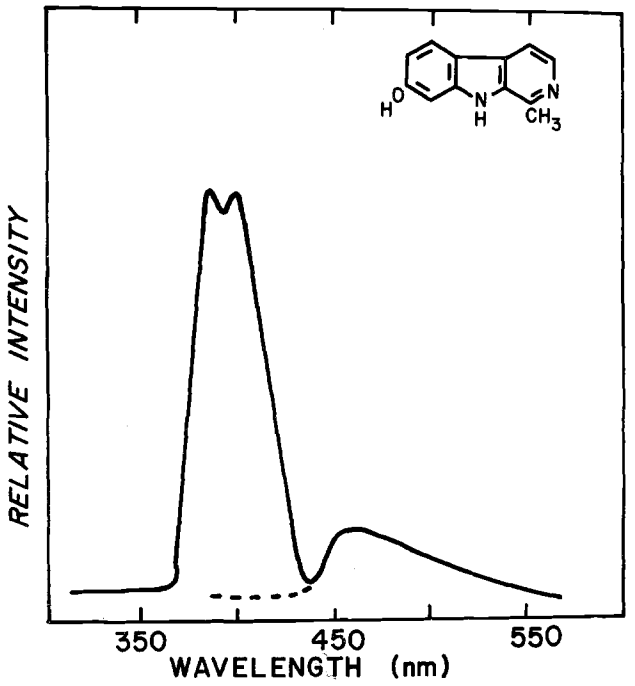

FIG. 10—*Fluorescence and phosphorescence spectrum of harmol in EPA matrix at 77 K.*

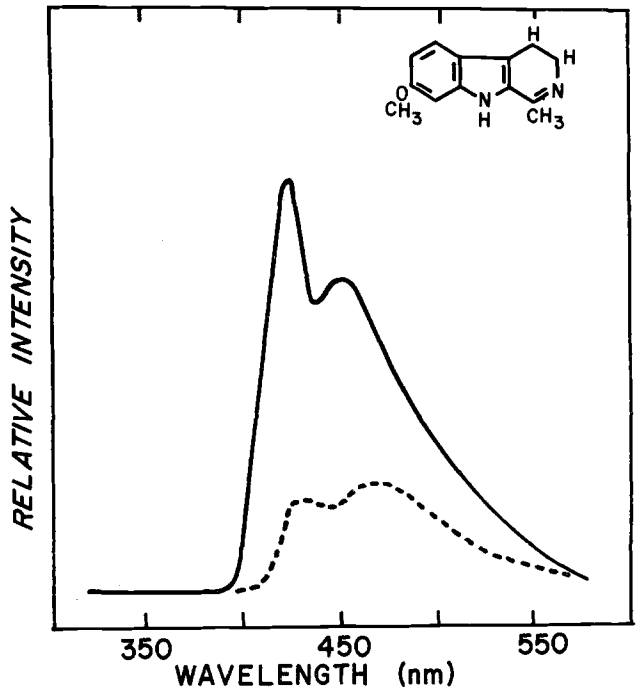

FIG. 11—*Fluorescence and phosphorescence spectrum of harmaline in EPA matrix at 77 K.*

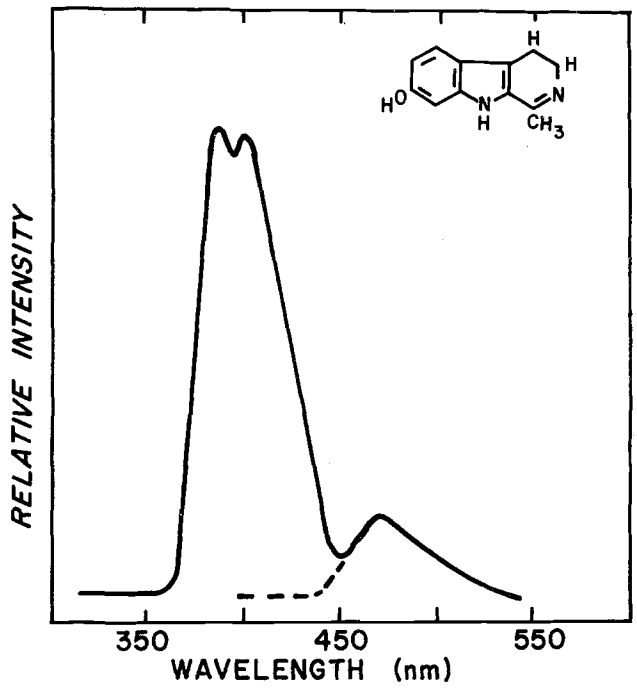

FIG. 12—*Fluorescence and phosphorescence spectrum of harmalol in EPA matrix at 77 K.*

tion and the emission the transition is assumed to be a $\pi^* \leftrightarrow \pi$ transition. Addition of the β-nitrogen into the ring shifts the absorption and emission to the red of that observed in carbazole [*12*]. Apparently nitrogen's nonbonding electrons play no part in the transition observed. Substitution of other functional groups onto the skeleton does not change the location of the emission to any great extent. Addition of hydrogens into the skeletal ring, as in harmalol and harmaline, invalidates any analogy to carbazole. The fluorescence emission of harmaline is red, shifted by 500 cm^{-1} with respect to norharman. The emission of harmalol is blue, shifted by 1200 cm^{-1} with respect to norharman.

The six β-carbolines studied here have a central amine group capable of acting as a Lewis base. Therefore, it is not surprising that the cation formed in acid has a different emission spectrum than the nonprotonated basic form. In addition, harmol and harmalol have phenolic hydroxy groups. The acidic hydrogen may form in equilibria a zwitterion with the amine group. Three different emission spectra observed in harmalol reflect the different forms available. In acid presumably both the amine and phenolic groups are protonated. With added base the neutral species is formed, which is probably a zwitterion. Addition of excess base deprotonizes the phenolic group giving the anion. In all six alkaloid solutions the emission spectra are of equilibria systems, with one form or another in greater abundance depending upon the pH.

It is not clear what form predominates in EPA matrices. Norharman and harmane were purchased as free bases. However, the fluorescence emission of these two molecules is similar to the spectra observed in acid. The other four molecules were purchased as acid salts. As expected, the fluorescence in EPA matrices is also similar to the aqueous solution spectra in acid. The fluorescence in EPA is blue, shifted by 1300 cm^{-1} with respect to the acidic aqueous solution.

As with fluorescence, the phosphorescence spectra of the six β-carbolines in EPA at 77 K are red, shifted with respect to carbazole in EPA [*13*]. The strong phosphorescence observed for carbazole and the alkaloids studied here, indicates the transition is $\pi^* \rightarrow \pi$. It is interesting to note that the $S_1 - T_1$ split of carbazole is 4600 cm^{-1}, but only 2400 cm^{-1} for norharman and harmine, 2200 cm^{-1} for harmane, only 300 cm^{-1} for harmaline, 4600 cm^{-1} for harmol, and 4700 cm^{-1} for harmalol. Kasha and others have proposed for carbazole that the transition borrows some of its intensity from intramolecular charge transfer states involving the nonbonding electrons of the amine group and the π^* orbitals [*14*].

The phosphorescence decay times observed in this study are only slightly shorter than those reported for indole and carbazole [*14*]. The times of 3 to 6 s are similar to other aromatic amines in EPA as well. The relative quantum yield of phosphorescence to fluorescence is smaller in the six β-carbolines than in carbazole. We estimate the relative yields of phosphorescence to fluorescence at 0.2 to 0.05, whereas carbazole's relative quantum yield is 0.55 and indole's is 0.36. Since the fluorescence overlapped the phosphorescence in these six compounds, the yields reported here are only approximate.

Conclusion

In this study we have identified the origin and band shape of the intense blue fluorescence of norharman, harmane, harmine, harmol, harmalol, and harmaline. The fluorescence is the mirror image of the absorption. The emission origin and shape changes with the addition of base to the aqueous solution. In addition, we have discovered the previously unreported lowest triplet state of these alkaloids. The phosphorescence is weak with respect to the fluorescence in EPA at 77 K.

The emissions observed are in all respects similar to those of indole and carbazole. Addition of functional groups which do not change the skeletal π structure, do not change the location of the emission greatly. Harmaline and harmalol form separate classes from the other four molecules due to the loss of π structure. The relative quantum yield and decay times are similar to other aromatic amines.

Our primary goal in this study was to establish the reliability of emission spectroscopy as an aid to detection of these alkaloids. The fluorescence is easily detected at 0.1 μg/ml. With improved optics the detection limit would surely be smaller.

Acknowledgment

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