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^{13}C-NMR Experimental Methods for Determination of Resonance Multiplicities: *Strychnos* Alkaloids

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Received January 21, 1980, from the *Department of Medicinal Chemistry and Pharmacognosy, College of Pharmacy, University of Houston, Houston, TX 77004.* Accepted for publication July 16, 1980.

Abstract □ ^{13}C-NMR spectral assignments of the *Strychnos* alkaloids brucine and strychnine have been reported by numerous investigators. One recent report contained several disparities in the assignments that were attributable to incorrect determinations of spin multiplicities. The source of the inaccuracies in the spin-multiplicity determinations of very complex molecules is discussed, and several additional techniques for the determination of these multiplicities are described that are less subject to interpretational errors than the conventional and routinely employed single-frequency off-resonance decoupling methods. These procedures are applied using brucine as a representative example.

Keyphrases □ ^{13}C-NMR spectroscopy—determination of resonance multiplicities, brucine □ Brucine—determination of resonance multiplicities □ Resonance multiplicities—brucine

Several recent reports dealt with the ^{13}C-NMR spectral assignments of strychnine, brucine, and related *Strychnos* alkaloids (1–9). The assignments in one report (8) for several resonances in both strychnine and brucine are in direct conflict to those reported previously. It is possible that errors occurred in this work that were due to the incorrect determination of spin multiplicities for the resonances in question.

The results obtained from two complementary methods that can provide unambiguous determination of the resonance multiplicities in even the most complex ^{13}C-NMR spectra are presented using brucine as an example. These methods are alternatives to conventional single-frequency off-resonance decoupling techniques and can be executed on modern Fourier transform spectrometers with only minor modification.

RESULTS AND DISCUSSION

A spectral parameter that is readily accessible for the pulsed Fourier transform technique and that provides an alternative method for the determination of spin multiplicities is the spin-lattice relaxation time, T_1 (10–12). Experimental evidence strongly supports the premise that in molecules of moderate size, the ^{1}H–^{13}C dipolar mechanism is predominantly responsible for ^{13}C-NMR relaxation. On this basis, for molecules that tumble at rates in excess of the motional narrowing limit (Larmor resonance frequency), the relaxation rate may be expressed as:

$$1/T_1^{DD} = N\hbar^2\gamma_C^2\gamma_H^2 r_{CH}^{-6}\tau_c \qquad \text{(Eq. 1)}$$

where γ_C and γ_H represent the magnetogyric ratios of the ^{13}C and ^{1}H nuclei, respectively, and $\hbar^2$ is Planck's constant. Thus, relaxation is determined by the terms Nr_{CH}^{-6} and τ_c, where the former term is modulated by the number of protons at specific internuclear distances (typically, relaxation is mediated by the directly attached protons, if any), while the latter term is the reorientational correlation time, typically 10^{-12}–10^{-10} sec for natural products of moderate molecular weight (*i.e.*, mol. wt. 250–1000) (13).

A further requirement is that the molecule tumble in a random fashion (isotropic), which can strictly be true only for spherical molecules or close approximations such as adamantane (14). In practice, however, the relaxation of a relatively large number of molecules has been found to conform to this simple requirement, thereby allowing the use of Eq. 1 rather than the more complex equations necessary to describe ordered (anisotropic) reorientation. This behavior leads to the very useful result that the relaxation time of a given carbon is inversely proportional to the number of directly attached protons. Thus, in relatively rigid cyclic compounds, methylene resonances are expected to undergo relaxation at a rate twice as fast as methine carbons. In contrast, methyl groups are capable of an internal reorientation in addition to the overall isotropic reorientation, which imparts a different reorientational correlation time, τ_i (referred to as τ_μ by some investigators), typically on the order of 10^{-13}–10^{-11} sec, resulting in longer relaxation times. In the extreme case, these relaxation times can be up to three times as long as those observed for methine resonances.

In complex molecules in which large numbers of carbon resonances have substantially similar chemical shifts, thereby preventing the unambiguous interpretation of single-frequency off-resonance decoupling spin multiplicities, relaxation considerations present a viable alternative for the demonstration of this information. This consideration is especially useful where a methylene carbon bears nonequivalent (anisochronous)

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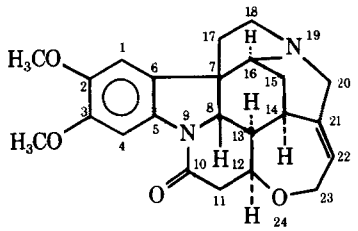

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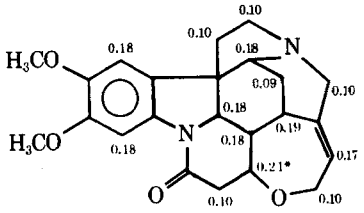
protons that can lead to the observation of non-first-order spin multiplets (15), which may account for the incorrect signal multiplicity of at least one resonance misassigned by Singh *et al.* (8). Circumvention of these interpretational problems by the application of relaxation phenomena has been used for various natural products, including steroids (16), terpenoids (17–21), macrolide antibiotics (22, 23), and lincomycin and its analogs (24). Furthermore, although partial relaxation data have been reported for brucine (I) (1, 2), the main application of these data was in the assignment of the quaternary carbon resonances, with no discussion of its application to spin-multiplicity determination in this compound.



The T_1 relaxation data for brucine (I) are shown here for the protonated resonances. (No effort was made to incorporate the measurement of the nonprotonated carbon relaxation times.) The spin-lattice relaxation times (T_1) (expressed in seconds) were measured by the inversion recovery method (22) using the three-parameter nonlinear fit program (23). Examination of the average T_1CH/T_1TH_2 ratio gave an observed ratio of 1.82 and a calculated reorientational correlation time (τ_c) of 2.5×10^{-10} sec. While it is not always convenient or practical to determine T_1 relaxation times rigorously, the same multiplicity data can be inferred effectively and conveniently from a partial relaxation experiment (Fig. 1). This latter technique is particularly powerful when used in conjunction with single-frequency off-resonance decoupling, as described by Nakanishi and coworkers (18, 19). The techniques, when used in conjunction with one another, also hold promise for extending applications of relaxation behavior to include nonisotropically reorienting systems such as the retinoids (25, 26).

An additional pair of alternative techniques for the determination of spin multiplicities is the use of selective excitation, as described by Freeman and coworkers (27, 28), in conjunction with either gated decoupling (29) or single-frequency off-resonance decoupling, which was described recently by Martin *et al.* (20). In both techniques, a train of soft pulses, typically 0.5–1.0°, is applied to the sample at the precession interval of the resonance to be selectively excited. This procedure has

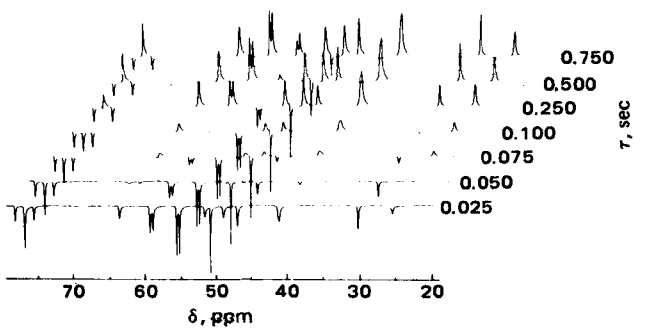


Figure 1—*Stack plot of a partial inversion recovery experiment showing the high-field region of brucine (I). Since relaxation is inversely proportional to the number of directly bonded protons, methylene carbons are expected to exhibit a positive inflection first (τ = 0.075 sec) followed by methine carbons (τ = 0.25 sec) and then methyl carbons. The nonprotonated C-7 resonances never become positive in this experiment.*

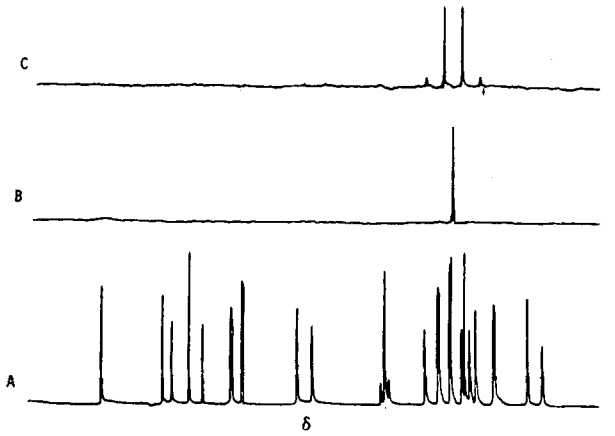
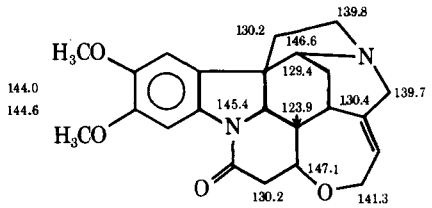


Figure 2—^{13}C-*NMR spectra of brucine. Key: A, fully decoupled* ^{13}C-*NMR spectrum; B, selective excitation with full decoupling of the* OCH_3 *resonance at 55.3°; and C, selective excitation with gated decoupling of the* OCH_3 *resonance showing full spin–spin coupling,* J = 144.6 *Hz.*

the effect of producing a cumulative tip of the desired resonance 90° into the x–y plane for observation. In contrast, the remaining resonances, which have different precession intervals, receive relatively little benefit from the pulse train application and thus remain as essentially unperturbed Z-magnetization. The net result of these two events is the acquisition of a subspectrum containing only information from the desired resonance.

By the application of selective excitation with gated decoupling, ^1H–^{13}C coupled spin multiplets were obtained for all resonances in the high-field region of the ^{13}C-NMR spectrum of brucine (I) (Fig. 2). In all cases, the multiplicities determined by T_1 relaxation methods and selective excitation were in complete agreement. However, the latter method has the further advantage of providing easy experimental access to the primary ^1H–^{13}C spin-coupling constants ($^1J_{CH}$), which are otherwise inaccessible, despite their considerable diagnostic utility. These data are summarized in the structure presented here and in Table I.



The primary coupling constants ($^1J_{CH}$) for brucine were measured by selective excitation with gated decoupling (27–30). It is certain that the resonances for C-11, C-18, and C-20, which Singh *et al.* (8) referred to as being not clear, were reported accurately by other investigators (1–7, 9).

A further point that is clarified by examination of the selectively excited ^1H–^{13}C spin-coupled subspectra is the contention by Wehrli and Nishida (13) that the assignments for C-11, C-18, and C-20 are insufficiently substantiated. Unequivocal assignment of the resonance for C-11 thus is made to the resonance at δ 41.6, based on the primary coupling constant $^1J_{C_{11}H_{11}}$ = 130.2 Hz, while C-18 and C-20 exhibit somewhat larger couplings of 139.8 and 139.7 Hz, respectively. Further discrimination between C-18 and C-20 also is possible by selective excitation, the former exhibiting considerable broadening of its spin-coupled resonance, probably as a result of couplings to the anisochronous C-17 methylene protons. This contention is supported by the observation that the two-bond coupling constants of heterocyclic systems frequently are as large as the three-bond couplings, which normally are the largest of the long-range couplings seen in ^1H–^{13}C coupled spectra. In contrast, C-20, which is positioned between N-19 and the vinyl carbon, C-21, would be expected to exhibit significantly less long-range coupling. Thus, assignment of C-18 is made to the resonance at δ 49.3, while C-20 is assigned to the resonance at δ 51.8; these assignments also are in agreement with previous reports (5–7, 9).

While considerable utility has been demonstrated for selective excitation with gated decoupling, some information is lost to the researcher

Table I—25.158-MHz ^{13}C-NMR Shift Assignments, Multiplicities, Relaxation Times, and Coupling Constants for Brucine

Carbon	^{13}C, δ	Multiplicity	T_1, sec	$^1J_{CH}$, Hz
10	168.0	s	—	—
3	148.4	s	—	—
2	145.4	s	—	—
21	139.7	s	—	—
5	135.3	s	—	—
22	126.3	d	0.17	—
6	122.9	s	—	—
1	105.1	d	0.18	—
4	100.3	d	0.18	—
12	76.9	d	0.21[a]	147.1
23	63.7	t	0.10	141.3
8	59.9	d	0.18	145.4
16	59.1	d	0.18	146.6
OCH_3	55.7	q	0.84	144.0
OCH_3	55.3	q	0.68	144.6
20	51.8	t	0.10	139.7
7	51.0	s	—	—
18	49.3	t	0.10	139.8
13	47.4	d	0.18	123.9
11	41.6	t	0.10	130.2
17	41.5	t	0.10	130.2
14	30.8	d	0.19	130.4
15	26.0	t	0.09	129.4

[a] Degenerate with center line of deuterochloroform triplet; relaxation time may be suspect.

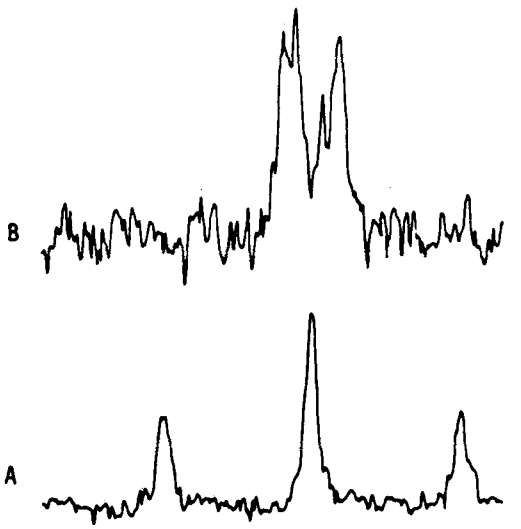


Figure 3—*Selective excitation in conjunction with gated decoupling of the C-15 methylene resonance at δ 26.0 (A) and single-frequency off-resonance decoupling subspectrum showing the anisochronicity of the methylene protons attached to the same carbon atom (B).*

using this technique which can be especially valuable when dealing with a molecule of unknown structure. Specifically, the lost information is that provided by the anisochronicity of methylene protons under certain circumstances (15), but it is available by conventional single-frequency off-resonance decoupling techniques. However, the advantages of single-frequency off-resonance decoupling observation with the unambiguous character of selectively excited and gated decoupled subspectra are combined in selective excitation with single-frequency off-resonance decoupling (20). The technique, in principle, is quite similar to selective excitation in the preparation of the spin system for observation. The two techniques differ in that the former employs conventional gated decoupling while the latter utilizes the collapse of the decoupling field from broad-band noise irradiation of the proton spectral window to a preselected single frequency. The net result of selective excitation method is to provide a selectively excited subspectrum with single-frequency off-resonance decoupling spectral characteristics. An illustrative example is shown in Fig. 3 for the C-15 methylene resonances, which, because of the anisochronous nature of the attached protons, may exhibit a doublet of doublets under certain decoupler conditions rather than the more conventional triplet (9, 15).

Several techniques have been described that provide the means of determining resonance spin multiplicities unambiguously. Considerable useful information in the form of primary coupling constants also is obtained, which can be of tremendous assistance in the assemblage of structural fragments into larger contiguous blocks. The errors in signal assignments such as those in the report of Singh *et al.* (8) can be completely avoided by the use of techniques that are capable of completely removing the ambiguities inherent in single-frequency off-resonance decoupling spectra of very large molecules.

EXPERIMENTAL[1]

All ^{13}C-NMR experiments were performed on a sample prepared by dissolving 500 mg of brucine[2] in 3.0 ml of deuterochloroform[3] and the solution was degassed with zero-grade argon[4] for 15 min followed by transfer to a 12-mm NMR tube fitted with a polytef vortex plug. All chemical shifts were referenced relative to the center line of the deuterochloroform triplet, which was taken as being 76.9 ppm downfield of tetramethylsilane. Typical instrument acquisition parameters were 5-kHz spectral observation with 8K data points (4K after transformation) and sufficient acquisitions to provide usable signal-to-noise ratios and an acquisition time of 0.8192 sec. Typical 90° pulses were obtained with a 20-sec pulse, with no effort made to optimize this pulse to the sample.

The T_1 relaxation studies were conducted using the inversion recovery pulse sequence (31, 32), and data reduction was accomplished with the three-parameter nonlinear fitting program (33). Selective excitation experiments were conducted using previously reported procedures and instrument modifications (20, 30).

[1] Varian Associates model XL-100-15 spectrometer system, operating at 25.158 MHz in the Fourier transform mode, equipped with a Nicolet TT-100 data system, a model TT-760 decoupler, and an NT-440 Multi-Observe Nuclei Accessory.
[2] Sigma Chemical Co., St. Louis, Mo.
[3] Aldrich Chemical Co., Milwaukee, Wis.
[4] Iweco, Houston, Tex.

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NOTES

Synthesis and Antibacterial and Anticancer Evaluations of α-Methylene-γ-butyrolactones

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Abstract □ Nine new α-methylene-γ-butyrolactones were synthesized by the Reformatsky condensation of ethyl α-bromomethylacrylate with ketones, aldehydes, and an epoxide. A unique spirobutyrolactone class was prepared by reaction of the zinc alkyl derivative and *N*-methylisatins. The compounds were evaluated against L-1210 and P-388 leukemia and the 9KB carcinoma of the nasopharynx. They also were screened in a microbiocidal and an antifungal assay. The spiro methylene lactone of 5-iodo-*N*-methylisatin displayed activity in the P-388, 9KB, and antifungal screens.

Keyphrases □ α-Methylene-γ-butyrolactones—synthesis and evaluation for antibacterial and antineoplastic activity □ Antibacterial activity, potential—evaluation of α-methylene-γ-butyrolactones □ Antineoplastic activity, potential—evaluation of α-methylene-γ-butyrolactones

Recognized as the active moiety in a wide variety of antineoplastic sesquiterpene lactones (1, 2), the α-methylene-γ-butyrolactone structure has been synthetically incorporated in numerous drug candidates in the search for anticancer activity (3, 4). Many synthetic approaches have been used (5), and new methods are reported regularly (6, 7) since many of the model substances are active.

The application of the Reformatsky method by Öhler *et al.* (8) is a facile, one-step conversion of aldehydes and ketones to the requisite lactones with the greatest potential of altering the functionality at C-5 (Scheme I). This condensation of the zinc alkyl derivative of ethyl α-bromomethylacrylate with *N*-methylisatins (II*a* and II*b*) also yields a unique class of spirolactones (V*a* and V*b*) (Scheme II). Nine lactones were synthesized in 25–59% yields using this procedure (Table I).

EXPERIMENTAL[1]

***p*-(Pyrrolidinosulfonamido)benzaldehyde (I*d*)**—Compound I*d* was obtained by the dropwise addition of 2.78 g (39.2 mmoles) of anhydrous pyrrolidine in 10 ml of acetone to 6.00 g (19.5 mmoles) of *p*-chlorosulfonylbenzaldehyde diacetate (9) in 25 ml of acetone. The medium was stirred for 1 hr, the acetone was removed *in vacuo*, and the residue was dissolved in methylene chloride and washed with 20 ml of 10% aqueous HCl. The solvent was removed by distillation, and the crystalline diacetate was hydrolyzed by refluxing for 1 hr in a solution of 25 ml of ethanol, 25 ml of water, and 3 ml of concentrated sulfuric acid. The solution was chilled and it deposited the aldehyde as white crystals, which were filtered, recrystallized twice from ethanol, and dried *in vacuo* to yield 3.35 g (50%) of I*d*, mp 109–111°; IR (mineral oil): 1710 (C=O) cm^{-1}; NMR (deuterochloroform): δ 1.60–1.95 (m, 4H, CH_2CH_2), 3.16–3.50 (m, 4H, CH_2NCH_2), 8.1 (broad s, 4H, aromatic H), and 10.17 (s, 1H, CHO) ppm.

Anal.—Calc. for $C_{11}H_{13}NOS_2$: C, 55.19; H, 5.47; N, 5.85. Found: C, 55.16; H, 5.68; N, 5.71.

***p*-(Pyrrolidinosulfonamido)acetophenone (I*f*)**—Compound I*f* was prepared by the addition of 3.50 g (49.3 mmoles) of anhydrous pyrrolidine in 10 ml of acetone to 5.00 g (22.9 mmoles) of *p*-chlorosulfonylacetophenone (10) in 25 ml of acetone. After 2 hr of refluxing and stirring, the solution was poured into 75 ml of cold water. The resulting precipitate was collected, washed with cold water, dried *in vacuo*, and recrystallized twice from ethanol to yield 4.30 g (74%) of I*f* as pale-yellow needles, mp 142–144°; IR (mineral oil): 1690 (C=O) cm^{-1}; NMR (deuterochloroform): δ 1.65–2.00 (m, 4H, CH_2CH_2), 2.70 (s, 3H, $COCH_3$), 3.20–3.50 (m, 4H, CH_2NCH_2), and 7.85–8.30 (m, 4H, aromatic H) ppm.

Anal.—Calc. for $C_{12}H_{15}NO_3S$: C, 56.95; H, 5.97; N, 5.53. Found: C, 57.13; H, 6.16; N, 5.67.

[1] Analyses were performed by Dr. G. I. Robertson, Florham Park, N.J. Melting points were determined between glass disks on a Fisher-Johns apparatus and are uncorrected. NMR spectra were obtained on a Perkin-Elmer Hitachi R20A spectrometer and were calibrated against tetramethylsilane. IR spectra were obtained on a Perkin-Elmer 257 spectrophotometer as petroleum oil mulls.

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