with excess methyl iodide. An exothermic reaction occurred giving a semicrystalline mass. This was dissolved in water, the aqueous solution was extracted with ether, and then an aqueous solution of sodium perchlorate and perchloric acid was added. The crystals that separated from the aqueous solution were collected and recrystallized from water to give 1.4 g. of N,N,N-tri-methyl-*cis*-hydrindane-1-ammonium perchlorate as white crystals, m.p. 218–220°.

Anal. Calcd. for $C_{12}H_{24}NO_4Cl$: C, 51.14; H, 8.58; N, 4.97. Found: C, 50.49; H, 8.29; N, 4.89.

A solution of 1.2 g. of the perchlorate in 20 ml. of a 20% aqueous potassium hydroxide solution was heated at 120° for 2 hr. The cold solution was extracted with ether and the ether extract was carefully concentrated. The residual oil was dissolved in ethanol, 100 mg. of a 5% palladium-on-charcoal catalyst[11] was added, and the mixture was subjected to hydrogenation at room temperature and atmospheric pressure. There was a rapid up-take of 61 ml. of hydrogen indicating the presence of 320 mg. (61%) of olefin. After removal of the catalyst, the solution was carefully concentrated. Vapor phase charomatography of the residue using a Carbowax column was carried out and the main component was collected and examined in the infrared using carbon tetrachloride as solvent. The infrared spectrum thus obtained agreed very closely with that given for *cis*-hydrindane[13] and was different from that of *trans*-hydrindane.[14]

(13) American Petroleum Institute Tables of Infrared Spectra, No. 1647.
(14) American Petroleum Institute Tables of Infrared Spectra, No. 1648.

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Conjugation Effects in Phenylcyclopropanes

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The ultraviolet absorption spectrum of phenylcyclopropane has previously been interpreted as evidence that the cyclopropane ring electrons interact with those of the benzene ring to form a conjugated system. It has now been shown, using rigid model compounds, that the steric relationship between the cyclopropane and benzene rings is of little consequence spectroscopically.

The ability of the cyclopropane ring to enter into conjugation with a neighboring unsaturated system has been the subject of a number of recent investigations.[2] However, few of these have been concerned with ascertaining the steric requirement necessary for such interaction to occur.

Walsh[3a] predicted for interaction that the plane of the cyclopropane ring should be perpendicular to the plane of the unsaturated side chain; his model was used by Music and Matsen[3b] to predict the ultraviolet absorption maximum of phenylcyclopropane from molecular orbital calculations. Experimental evidence was given by Cromwell[4] in the case of the *cis* and *trans* forms of arylaroylethylenimines and ethylene oxides to the effect that a rather critical orientation of the three-ring atoms and the adjacent p-orbital bearing carbon atom must be met for appreciable conjugation to occur. Fuchs and Bloomfield[5a] have provided kinetic evidence for a pronounced geometrical bias in the transmission of electronic effects by the cyclopropane ring, whereas the equilibrium studies of Trachtenberg and Odian[5b] showed the cyclopropane system to be inferior to a two-carbon saturated chain in such transmission.

Kosower and Ito,[6a] in the cases of the two cyclopropyl ketones spiro[4.2]heptan-1-one and bicyclo[3.1.0]hexan-2-one, predicted and found the spiroketone to absorb at longer wave lengths than the fused ring ketone. They interpreted their data as supporting the importance of the geometrical factor for which evidence had earlier been reported by Cromwell.[2,4] The ultraviolet spectra of compounds derived from natural products where the cyclopropyl ring is a γ-substituent on an α,β-unsaturated ketone have also been interpreted to correspond to changes in the overlapping of the electrons of the ring and the double bond in the excited state.[6a,b]

Striking differences in the position of maximum absorption were observed in a series of substituted *trans*-2-phenylcyclopropanecarboxamides by Perold.[7] According to Walsh's theory,[3] for maximum conjugative interaction the hydrogen atom on that carbon atom of the cyclopropane ring which is bonded to the aromatic nucleus must lie in the plane of the benzene ring. If bulky groups such as methyl are substituted in the position *ortho* to the cyclopropyl substituent, the rings might be expected to twist out of conjugation. Perold[7] reports that 2-*o*-tolylcyclopropanecarboxamide possesses no measurable band near 220 mµ, while the corresponding *m*- and *p*-tolyl compounds have absorption maxima at 225 mµ (log ε 3.96) and 226 mµ (log ε 4.12). On the other hand, *trans*-1-methyl-2-phenylcyclopropanecarboxamide has its absorption maximum at 220 mµ (log ε 4.0), and the corresponding *trans*-2-phenyl-3,3-dimethyl compound has a maximum at 218 mµ (log ε 4.01). The bathochromic shifts relative to the *o*-tolyl compound were taken by Perold[7] to correspond to the appearance of a complete chain of conjugation from the phenyl group through the cyclopropane ring to the amide. It seemed likely that it was the phenylcyclopropane chromophore that was being affected.[8]

The relative ease of shifting the position of the absorption maximum in Perold's amide series suggested that a possibility existed for semiquantitative measure-
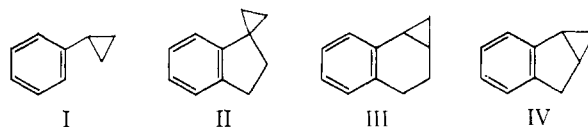
(1) This material is taken from the Dissertation of Alan L. Goodman offered in partial fulfillment of the requirements for the degree of Doctor of Philosophy at Stanford University. National Science Foundation Predoctoral Fellow, 1959–1960; Du Pont Fellow, 1961–1962.

(2) For leading references see: M. T. Rogers and J. D. Roberts, *J. Am. Chem. Soc.*, **68**, 843 (1946); R. H. Eastman and S. K. Freeman, *ibid.*, **77**, 6642 (1955), and earlier papers; R. S. Mohrbacher and N. H. Cromwell, *ibid.*, **79**, 401 (1957); G. W. Cannon, A. A. Santilli, and P. Shenian, *ibid.*, **81**, 1660 (1959); R. Fuchs, C. A. Kaplan, J. J. Bloomfield, and L. F. Hatch, *J. Org. Chem.*, **27**, 733 (1962).

(3) (a) A. D. Walsh, *Trans. Faraday Soc.*, **45**, 179 (1949); (b) J. F. Music and F. A. Matsen, *J. Am. Chem. Soc.*, **72**, 5256 (1950).

(4) See N. H. Cromwell, F. H. Schumacher, and J. L. Adelfang, *ibid.*, **83**, 974 (1961), for a bibliography of the extensive work in this field.

(5) (a) R. Fuchs and J. J. Bloomfield, *J. Org. Chem.*, **28**, 910 (1963); (b) E. N. Trachtenberg and G. Odian, *J. Am. Chem. Soc.*, **80**, 4081 (1958).

(6) (a) E. M. Kosower and M. Ito, *Proc. Chem. Soc.*, 25 (1962); (b) G. Buchi and H. J. E. Loewenthal, *ibid.*, 280 (1962).

I   II   III   IV

(7) G. W. Perold, *J. S. African Chem. Inst.*, **6**, 22 (1953); **8**, 1 (1955); **10**, 11 (1957); *Chem. Abstr.*, **48**, 4314c (1954); **50**, 6326d (1956); **52**, 1074i (1958).

(8) M. T. Rogers, *J. Am. Chem. Soc.*, **69**, 2544 (1947).

Figure 1.   Figure 2.

Figure 3.   Figure 4.
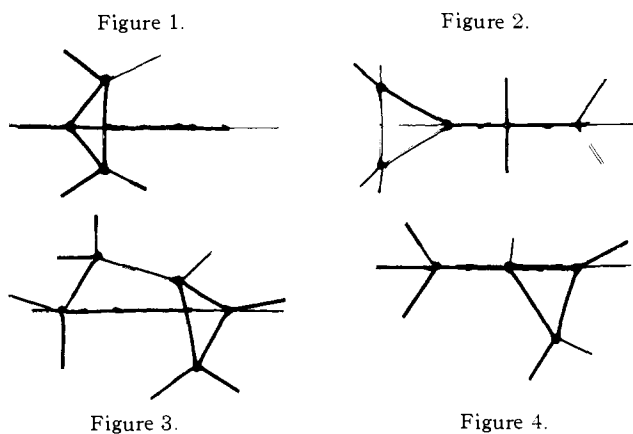
ment of the angular relationship necessary for conjugation between the cyclopropane and benzene rings.

In the present study, model compounds were prepared such that there would be a variation in the spatial relationships between the electron clouds of the cyclopropane and benzene rings. The steric relationships can be seen by inspection of the photographs of the molecular models (Fig. 1–4). The models are to be compared with that of phenylcyclopropane (I) which is taken as the standard for this study. The small ring in phenylcyclopropane is not fixed, but is free to rotate about the bond joining the two rings. In the figure of phenylcyclopropane (Fig. 1), the methine carbon–hydrogen bond is arbitrarily placed in the plane of the aromatic nucleus, and θ is equal to $0°$.[9] This conformation was used by Music and Matsen[3b] in their molecular orbital calculation of the ultraviolet absorption spectrum of the compound, and was considered to provide maximum overlap of the benzene and cyclopropane electron orbitals. Obviously, because of the free rotation about the bond joining the two rings, the conformer in Fig. 1 is present only a fraction of the time in any sample of phenylcyclopropane.

Spiro-[cyclopropan-1,1'-indan](II, Fig. 2) is the rigid analog of the phenylcyclopropane conformer in Fig. 1. The cyclopropane ring is now held so that its plane is perpendicular to the plane of the benzene ring. The methine carbon–hydrogen bond described above for phenylcyclopropane has been incorporated into the five-membered ring to effect coplanarity with the benzene ring ($\theta = 0°$).

On the basis of the theoretical treatment of Music and Matsen,[3b] the spiro compound would be expected to absorb at longer wave lengths and to exhibit a larger molar extinction coefficient than phenylcyclopropane because the desired conformer now exists exclusive of the other forms where θ is not equal to $0°$.

Two values of θ may be proposed for tetrahydrocyclopropanaphthalene[10] (III) since this compound might exist in two conformations. In the structure shown (Fig. 3), the carbon–hydrogen bonds of the methylene group adjacent to the small ring are staggered with respect to the methine carbon–hydrogen bond and the carbon–carbon bond of the cyclopropane ring. In the other conformation, these bonds are eclipsed, contributing to a destabilization of the conformer by bond opposition.[11] In the conformer which is proposed to be the more stable one (Fig. 3), the methine carbon–hydrogen bond under discussion is not in the plane of the benzene ring, but is about $10°$ from

(9) θ is defined as the angle made by the methine carbon–hydrogen bond on the cyclopropane ring with the plane of the benzene ring.

(10) 1a,2,3,7b-Tetrahydro-1H-cyclopropa[a]naphthalene.

(11) E. L. Eliel, "Stereochemistry of Carbon Compounds," McGraw–Hill Book Co., Inc., New York, N. Y., 1962, p. 205.
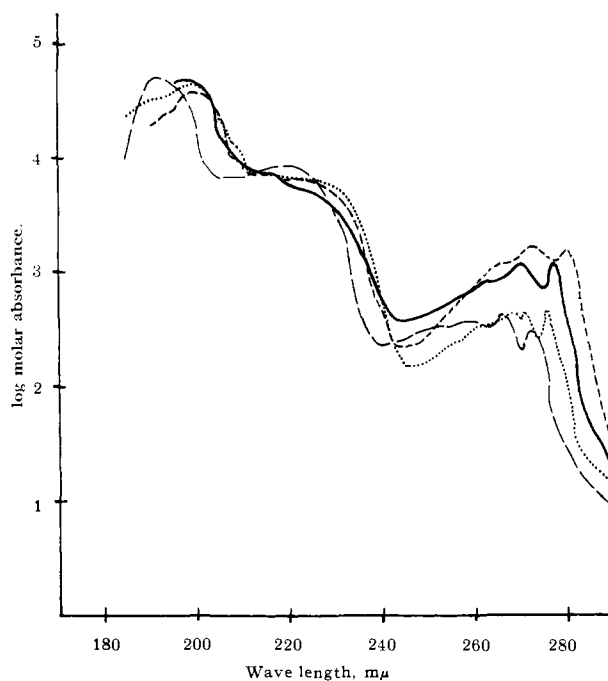

Fig. 5.—Ultraviolet absorption spectra: —— 1,1a,6,6a-tetrahydrocycloprop[a]indene (IV); ···· 1a,2,3,7b-tetrahydro-1H-cyclopropa[a]naphthalene (III); - - - - spiro[cyclopropane-1,1'-indan] (II); — — phenylcyclopropane (I).

coplanarity. This angle (θ) is enlarged to approximately $25°$ in tetrahydrocyclopropindene[12] (IV, Fig. 4).

These larger values for the angle θ lead to the prediction, on the theory of Music and Matsen,[3b] that tetrahydrocyclopropanaphthalene and tetrahydrocyclopropindene should absorb radiation at shorter wave lengths and with smaller extinction coefficients than the spiro compound.[13]

However, the predictions do not agree with the observed ultraviolet absorption spectra of the compounds. The three spectra (Fig. 5) are almost superimposable in the region of the primary band[14] at 210–240 mμ which is also the region where the maximum for the primary band of phenylcyclopropane itself occurs (Fig. 5).[8] The small differences in intensity in the maxima near 270 mμ (secondary band[14]) are attributed to a perturbation in the electronic states of the benzene nucleus by increasing strain in the molecule. The slight shift to longer wave lengths is attributed to the increase in strain in the ground state. The fact that phenylcyclopropane itself absorbs at slightly shorter wave lengths than the three compounds in all three bands is thought to be due to the lack of strain in this molecule as compared to the more highly condensed compounds. There is a 3 mμ bathochromic shift in the spectra of the cyclopropanes from the spectra of the hydrocarbons (indan, Fig. 6) in the 200 mμ region (second primary band[14]), but whether this is due to the interaction of the electrons of the small ring or a strain effect introduced by the cyclopropane ring cannot be answered.

In an effort to get more information as to the character of the excited state, the spectra of tetrahydrocyclopropindene and tetrahydrocyclopropanaphthalene

(12) 1,1a,6,6a-Tetrahydrocycloprop[a]indene.

(13) Application of resonance theory (cf. G. W. Wheland, "Resonance in Organic Chemistry," John Wiley and Sons, Inc., New York, N. Y., 1955, p. 256) also leads to the prediction of a dependence of spectral properties on the steric disposition of the benzene and cyclopropane rings.

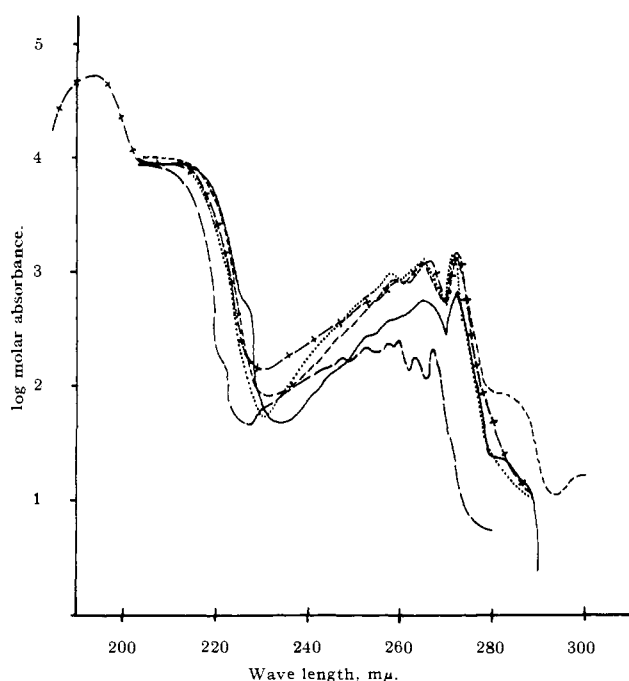(14) L. Doub and J. M. Vandenbelt, J. Am. Chem. Soc., 69, 2714 (1947); 71, 2414 (1949).

Fig. 6.—Ultraviolet absorption spectra: - - - - 1-methylindan; . . . . 1,1-dimethylindan; —— tetrahydronaphthalene; — — ethylbenzene; — + indan.

Fig. 7.—Ultraviolet absorption spectra: —— 1,2-dihydronaphthalene; — — styrene; - - - - 1-methyleneindan: . . . . indene.

were obtained in cyclohexane solution. The only difference in the spectra of hydrocarbon solutions over those obtained with ethanol solutions is in the intensity of the primary band, which is higher by several thousand units in cyclohexane solution. It was hoped the shift to a less polar solvent would allow some of the vibrational structure of the spectra to appear, but such was not the case. The vapor phase spectrum of tetrahydrocyclopropindene also exhibits continually increasing absorption and no fine structure as shorter wave lengths are approached. This result is in agreement with other information on the spectra of complex aromatic hydrocarbons.[15]

The cyclopropanes under consideration absorb at wave lengths intermediate to those of the analogous hydrocarbons and olefins (Fig. 7), as does the original compound studied, phenylcyclopropane.[8] The essential identity of the spectra of the cyclopropa compounds III and IV, the spiro compound II, and phenylcyclopropane (I) is taken as evidence that there is no preferred geometry for conjugation of the cyclopropane ring with a benzene ring, as has been shown necessary in the case of more electronegative substituents bearing p-electrons, such as the carbonyl group,[16] and in the polar systems cited earlier.[5,6] The bathochromic shift of about 20 mμ in the primary band of benzene occasioned by cyclopropyl substitution[8] would thus seem to be interpretable as a result of purely inductive effects, akin in nature to the similar, though somewhat smaller (5–10 mμ), effects of simple alkyl group substitution.[15]

An alkyl group substituted on a dienoid system causes a bathochromic shift of 5 mμ.[17] Alkyl substitution at the carbon atoms of α,β-unsaturated ketones cause bathochromic shifts of about 11 mμ.[18] Simple considerations of this type illustrate the point that induc-
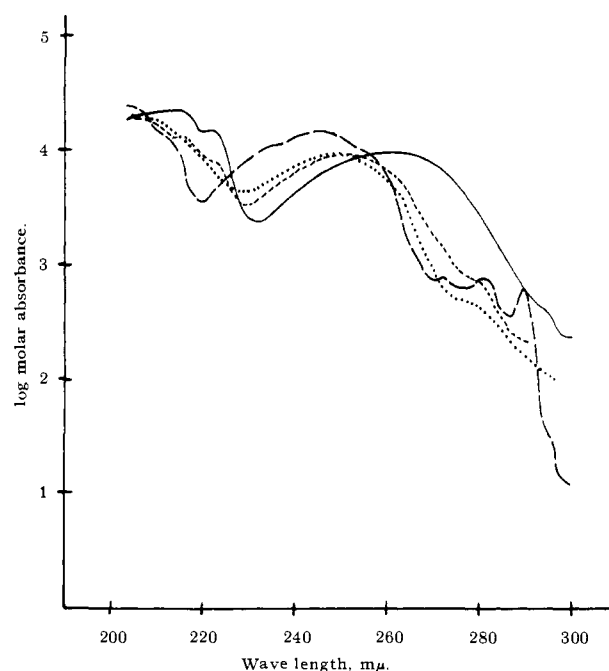
tive effects are enhanced appreciably in systems which possess polar character. However, the intimate relation between inductive and resonance effects in the spectra of benzene derivatives has not been completely explained as yet.[15] It has been established that an inductive effect, in the absence of a resonance effect, does not modify the spectrum of benzene appreciably. Thus, the cause of the large bathochromic shift occasioned by the attachment of a cyclopropane ring to benzene remains obscure. Our results are explicable on the postulate that the cyclopropane ring is a seat of high electron density whose polarizability is devoid of stereochemical bias.

Experimental

1,1a,6,6a-Tetrahydrocycloprop[a]indene (IV).—Diiodomethane (107.2 g., 0.4 mole), iodine (0.15 g.), and zinc–copper couple[19] (32.6 g., 0.5 mole) were added to 300 ml. of anhydrous ether, and the mixture was heated at reflux with stirring for 0.5 hr. under anhydrous conditions. Indene (93.0 g., 0.8 mole) in 100 ml. of anhydrous ether was added dropwise to the mixture during 1 hr. After being heated for 40 hr., the mixture was cooled and filtered through Supercel. The residue was washed with ether, and the combined filtrate and washings were shaken successively with 5% hydrochloric acid, 10% sodium bicarbonate solution, and finally, saturated salt water. The aqueous layers were shaken with ether, and the ether layers were combined with the original ether solution. After the solution was dried over anhydrous sodium sulfate, the ether was removed at reduced pressure to leave an oil that was fractionally distilled on a 13 theoretical plate, glass helices packed column to give 15.4 g. (30% yield) of 1,1a,6,6a-tetrahydrocycloprop[a]indene, b.p. 104° (40 mm.), n^{26}D 1.5545.

Anal. Calcd. for $C_{10}H_{10}$: C, 92.26; H, 7.74; mol. wt., 130.1. Found: C, 92.40; H, 7.58; mol. wt., 130 (mass spec.).

1a,2,3,7b-Tetrahydro-1H-cyclopropa[a]naphthalene (III).—Diiodomethane (200 g., 0.75 mole), zinc–copper couple[19] (55.6 g., 0.85 mole), and a few crytals of iodine were added to 400 ml. of anhydrous ether. Upon heating the mixture to reflux, an exothermic reaction occurred which was moderated by cooling with ice water. When the reaction had moderated, 1,2-dihydronaphthalene[20] (86.4 g., 0.66 mole) in 100 ml. of anhydrous ether was added dropwise. Heating and stirring were continued for 30 hr. after the addition was completed.

After being cooled, the mixture was filtered through Supercel. The residue was washed with ether, and the washings and filtrate

(15) H. H. Jaffé and M. Orchin, "Theory and Applications of Ultraviolet Spectroscopy," John Wiley and Sons, Inc., New York, N. Y., 1962, pp. 248–254.

(16) G. D. Heddon and W. G. Brown, *J. Am. Chem. Soc.*, **75**, 3744 (1953); R. Huisgen and W. Rapp, *Ber.*, **85**, 826 (1952).

(17) Ref. 15, p. 200.

(18) Ref. 15, p. 207.

(19) R. S. Shank and H. Shechter, *J. Org. Chem.*, **24**, 1825 (1959).

(20) Prepared by the method of C. O. Hurd and L. H. Juel, *J. Am. Chem. Soc.*, **77**, 6013 (1955).

were combined. The ether solution was shaken successively with saturated ammonium chloride solution, 10% ammonium hydroxide solution, and saturated salt water. The ethereal solution was distilled at reduced pressure to give an oil, b.p. 106–125° (30 mm.), which was purified by preparative vapor phase chromatography using columns 6 ft. in length and 1 in. in diameter to 20% Ukon Polar on firebrick to give 22.5 g. (23.5% yield) of 1a,2,3,7b-tetrahydro-1H-cyclopropa[a]naphthalene, b.p. 99–102° (15 mm.), n^{24}D 1.5642. The reported[21] constants are b.p. 60° (2 mm.), n^{20}D 1.5688.

Anal. Calcd. for $C_{11}H_{12}$: C, 91.61; H, 8.39; mol. wt., 144.2. Found: C, 91.35; H, 8.37; mol. wt., 144 (mass spec.).

Spiro[cyclopropane-1,1′-indan] (II). 1-Indancarboxylic Acid. —Indene (80 g., 0.68 mole) was converted to 1-indenecarboxylic acid (43 g.) according to Wunderlich's procedure,[22] which was then hydrogenated in ethanol solution at atmospheric pressure using 1.5 g. of platinum oxide as catalyst to give 42 g. (38% yield) of 1-indancarboxylic acid, m.p. 55–57°. The reported[22] value is 55–56°.

1-Hydroxymethylindan.—1-Indancarboxylic acid (25 g., 0.15 mole) in 100 ml. of anhydrous ether was added slowly to a filtered solution of lithium aluminum hydride (11.4 g., 0.3 mole) in 200 ml. of anhydrous ether. After being stirred for 1 hr., the mixture was heated at reflux for 1 hr., and then was cooled by an ice bath before decomposition by dilute hydrochloric acid. After separation of the layers, the water layer was extracted with ether. The ether layers were combined, shaken with dilute sodium bicarbonate solution, and dried over anhydrous magnesium sulfate. 1-Hydroxymethylindan (19.6 g., 86% yield) was obtained as a clear, colorless liquid, b.p. 97–99° (3 mm.), n^{24}D 1.5558; infrared absorption spectrum, 3325 cm.$^{-1}$ (O–H). The phenyl urethan of the alcohol on crystallization from hexane melted at 112.5–113°.

Anal. Calcd. for $C_{17}H_{17}O_2N$: C, 76.38; H, 6.41. Found: C, 76.61; H, 6.47.

1-Chloromethylindan.—1-Hydroxymethylindan (5 g., 34 mmoles) in 10 ml. of anhydrous benzene was slowly added to a stirred solution of thionyl chloride (5.3 g., 45 mmoles) and 1 drop of pyridine in 10 ml. of benzene.[23] After the addition was completed, the solution was heated on a steam bath for 2 hr., and then was poured into 50 ml. of water. The mixture was shaken vigorously. The benzene layer was dried over anhydrous magnesium sulfate before distillation to give 4.8 g. (84% yield) of 1-chloromethylindan, b.p. 101–104° (8 mm.), n^{23}D 1.5535. The compound was not purified[24] for analysis but was used directly in the next step of the reaction sequence.

1-Methyleneindan.—1-Chloromethylindan (4.05 g., 24 mmoles), potassium hydroxide (8.1 g., 145 mmoles), 1.5 ml. of water, and 35 ml. of methanol were heated at reflux for 45 min., and then the solution was poured into 200 ml. of water. The oily layer was taken up in pentane. The pentane solution was dried over anhydrous magnesium sulfate. After removal of the solvent, the residual oil was distilled to give 2.5 g. (80% yield) of 1-methyleneindan, b.p. 99.5–101.5° (29 mm.), n^{23}D 1.5759; infrared absorption spectrum, 1635 (Ph—C=C), 860 cm.$^{-1}$ (=CH₂).

(21) M. S. Newman, W. C. Sagar, and C. C. Cochrane, *J. Org. Chem.*, **23**, 1832 (1958).

(22) W. Wunderlich, *Arch. Pharm.*, **286**, 512 (1953).

(23) C. R. Noller and R. A. Bannerot, *J. Am. Chem. Soc.*, **56**, 1563 (1934).

(24) An increase in the scale of the reaction led to large amounts of 1,2-dihydronaphthalene, which, was formed, presumably, by a carbonium ion ring enlargement mechanism. Separation was effected using preparative vapor phase chromatography.

Anal. Calcd. for $C_{10}H_{10}$: C, 92.26; H, 7.74. Found: C, 91.89; H, 8.10.

Spiro[cyclopropane-1,1′-indan] (II).—Diiodomethane (8.25 g., 30.8 mmoles), zinc–copper couple[14] (2.62 g., 40 mmoles), a crystal of iodine, and 25 ml. of anhydrous ether were heated at reflux with stirring for 45 min. under anhydrous conditions. 1-Methyleneindan (1 g., 7.7 mmoles) in 10 ml. of ether was added, and the mixture was heated for an additional 18 hr. Zinc–copper couple (2.6 g., 40 mmoles) a crystal of iodine, and 2-methyl-2-butene (2.1 g., 30 mmoles) were added to remove the excess diiodomethane,[25] and the mixture was heated for a further 8 hr.

After being cooled and filtered through Celite, the ether solution was shaken with three 75-ml. portions of ammonium chloride solution, three 75-ml. portions of ammonium hydroxide solution, and three 75-ml. portions of water. The aqueous layers were extracted with ether, and the combined ether extracts were dried over sodium sulfate before removal of the solvent. After distillation, 0.7 g. of an oil was obtained, which was further purified by vapor phase chromatography using columns 6 ft. in length and 0.5 in. in diameter packed with 20% Carbowax 20M on firebrick to give spiro[cyclopropane-1,1′-indan], n^{23}D 1.5389.

Anal. Calcd. for $C_{11}H_{12}$: C, 91.61; H, 8.39; mol. wt., 144.2. Found: C, 91.28; H, 8.71; mol. wt., 144 (mass spec.).

Phenylcyclopropane (I).—Phenylcyclopropane, n^{24}D 1.5310, was prepared from styrene according to the procedure of Simmons and Smith[26] using the zinc–copper couple described by Shank and Shechter.[19] The reported[26] value is n^{25}D 1.5309.

Anal. Calcd. for C_9H_{10}: C, 91.47; H, 8.53. Found: C, 91.71; H, 8.50.

1-Methylindan.—1-Methyleneindan was hydrogenated at atmospheric pressure using platinum oxide as catalyst to give 1-methylindane, n^{24}D 1.5222.

Anal. Calcd. for $C_{10}H_{12}$: C, 90.85; H, 9.15. Found: C, 90.66; H, 9.15.

1,1-Dimethylindan was obtained through the kind generosity of Professor J. W. Wilt,[27] and was purified by preparative vapor phase chromatography using a column 20 ft. in length and ³/₈ in. in diameter of 20% silicone oil (SE-30) on firebrick.

Anal. Calcd. for $C_{11}H_{14}$: C, 90.35; H, 9.65. Found: C, 90.32; H, 9.71.

Spectra.—Infrared absorption spectra were obtained on films using a Perkin–Elmer Model 421 spectrophotometer. Ultraviolet absorption spectra were obtained on 95% ethanol solutions in fused quartz cells, 1-cm. path length, using a Cary Model 14M spectrophotometer; spectra below 205 mμ were obtained in cells, 0.01-cm. path length, using a Cary Model 15 spectrophotometer whose optical path was flushed with pure nitrogen. The ultraviolet spectrophotometers were calibrated on benzene–water solutions at 203.5 mμ. Spectra run on both instruments were identical.

(25) S. D. Koch, R. M. Kliss, D. V. Lopiekes, and R. J. Wineman, *J. Org. Chem.*, **26**, 3122 (1961).

(26) H. E. Simmons and R. D. Smith, *J. Am. Chem. Soc.*, **81**, 4256 (1959).

(27) J. W. Wilt and C. A. Schneider, *J. Org. Chem.*, **26**, 4196 (1961).