Anal. Calcd for $C_{22}H_{18}O_2S$: C, 76.27; H, 5.24. Found: C, 75.99; H, 5.34.

The pmr spectrum of **7** in carbon tetrachloride shows a singlet (1 H) at τ 5.43, a pair of doublets (2 H each, J_{gem} = 16.4 Hz) at 6.62 and 6.83, and a complex multiplet (13 H) from 2.5 to 3.1.

Anal. Calcd for $C_{22}H_{18}S$: C, 84.03; H, 5.77. Found: C, 84.00; H, 5.97.

Oxidation of **7** under the same conditions as that described for **5** gave the sulfone of **7** (95%), mp 178–179°.

The pmr spectrum of the sulfone in chloroform-*d* shows a singlet (1 H) at τ 4.69, a pair of doublets (2 H each, J_{gem} = 17.6 Hz) at 6.10 and 6.78, and a complex multiplet from 2.0 to 3.0.

Anal. Calcd for $C_{22}H_{18}O_2S$: C, 76.27; H, 5.24. Found: C, 76.40; H, 5.29.

When **1** is treated with thiophenol at 150° for 30 hr, **5**, **6**, **7**, and **8** were isolated in high yield in the ratio of 7.5:1:0.5:1, respectively. The presence of oxygen, benzoyl peroxide, or benzoic acid had no observable effect on the rate or product distribution of the reaction. The reaction of **1** with thiophenol at 110° catalyzed by a trace of *p*-toluenesulfonic acid proceeded very rapidly to give 85% **5** and 15% **6**; no **7** or **8** was observed. Treatment of **1** (500 mg) with 10 ml of thiophenol in which 60 mg of potassium metal had been dissolved at 170° for 18 hr gave essentially identical results with that observed in the absence of potassium thiophenoxide. In all these cases, products **5**–**8** were stable to the conditions of the reactions. Attempts to photoinitiate the addition of thiophenol to **1** with either medium or low pressure mercury uv lamps failed to give any 1:1 adducts.

Treatment of thio ethers **5**–**7** with a 20-fold excess (by weight) of Raney nickel W-2[38] in refluxing ethanol for 14 hr gave an 80–85% yield of hydrocarbon **8** in each case.

Attempted Addition of *n*-Butanethiol to 1.—Treatment of **1** with *n*-butanethiol in the presence of either medium or low pressure mercury uv lamps did not result in any observable addition products. When **1** was treated in refluxing *n*-butanethiol with benzoyl peroxide, **1** was recovered unchanged after several days. Chromatography over silica gel did result in the isolation of *n*-butyl phenyl sulfide (eluted with 10% benzene in Skellysolve B) which proved to be identical (pmr and ir spectra) with an authentic sample.[39] This sulfide could be isolated from a solution of *n*-butanethiol treated with benzoyl peroxide in the absence of **1**.

Treatments of 1 with Fluorene and Dihydroanthracene.—A mixture of 0.50 g of **1** and 5.0 g of fluorene was sealed in a glass tube under nitrogen. The tube was heated at 195–200° in an oil bath for 2 days. The majority of the fluorene was removed by crystallization from methanol, and a pmr spectrum of the mother liquor showed only **1** and fluorene to be present. No **8** could be observed.

This same procedure was employed for 9,10-dihydroanthracene, and a pmr spectrum of the resulting mixture indicated that *ca.* 10% of **1** had been hydrogenated to **8**. This mixture was chromatographed over 60 g of silica gel packed in Skellysolve B. Elution with 3% benzene in Skellysolve B gave 950 mg of 9,10-dihydroanthracene, 45 mg of **8**, 400 mg of **1**, and 70 mg of 9,9′,-10,10′-tetrahydro-9,10-bianthryl, mp 256–258° (lit.[40] mp 255°).

Registry No.—**1**, 2199-28-2; **2**, 23367-54-6; **5**, 23265-33-0; **5** sulfone, 23265-34-1; **6**, 23265-35-2; **6** sulfone, 23265-36-3; **7**, 23288-66-6; **7** sulfone, 23265-37-4.

Acknowledgment.—Financial support from the donors of the Petroleum Research Fund of the American Chemical Society is gratefully acknowledged.

(38) R. Mozingo, "Organic Syntheses," Coll. Vol. III, John Wiley & Sons, Inc., New York, N. Y., 1955, p 181.

(39) G. Modena, *Gazz. Chim. Ital.*, **89**, 834 (1959).

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Conformational Studies of Perfluoro-2-halo-1,2-oxazetidines Using Nuclear Magnetic Resonance Spectroscopy[1]

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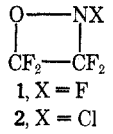
The high-resolution nmr spectra of perfluoro-2-fluoro-1,2-oxazetidine (**1**) and perfluoro-2-chloro-1,2-oxazetidine (**2**) were obtained over the temperature range 85 to −120°. The observed nonequivalence of geminal fluorines was attributed to restricted nitrogen inversion. The temperature dependence of the geminal fluorine–fluorine chemical-shift differences indicated equilibrating nonplanar conformers. The conformational free-energy differences for **1** and **2** were determined to be 900 and 1000 cal/mol, respectively.

The use of nmr spectroscopy to demonstrate the nonplanarity of cyclobutane rings has been reported by Lambert and Roberts.[2] These authors observed that the chemical-shift differences of geminal fluorines in certain substituted cyclobutanes showed temperature dependence. This was attributed to an equilibrium between the two possible puckered-ring conformations. We wish to present nmr evidence indicating similar nonplanarity in a perfluorooxazetidine ring system.

The room-temperature ^{19}F nmr spectra of perfluoro-2-fluoro-1,2-oxazetidine (**1**) and perfluoro-2-chloro-1,2-oxazetidine (**2**) showed AB quartets which were assigned to the CF_2O and CF_2N fluorines. The spectrum of **1** also contained a broad peak owing to the NF fluorine. The chemical shifts and geminal coupling constants are given in Table I. With the temperature varied from 85 to −120°, the same general pattern was obtained in the spectra of **1** and **2** with the geminal coupling constants remaining essentially unchanged. The volatility of the N-halooxazetidines precluded nmr studies above 85°. However, even at this temperature the quartet structures were clearly visible. The NF signal in the spectrum of **1** was detectably sharper at lower temperatures.[3]

The nonequivalence of the geminal fluorines in **1** and **2** results either from restricted oxazetidine ring inversion or from restricted nitrogen inversion. However, it seems very unlikely that the barrier to ring inversion would be sufficient to slow the ring-intercon-



1, X = F
2, X = Cl

(1) This investigation was performed under Contract No. N00019-67-c-0454 for the Naval Air Systems Command, Department of the Navy, Washington, D. C. 20360, with Mr. John Gurtowski as Project Officer.

(2) J. B. Lambert and J. D. Roberts, *J. Amer. Chem. Soc.*, **85**, 3710 (1963); **87**, 3884 (1965).

(3) Measurements of $W_{1/2}$ (signal width at half-height) indicate a change from 47 Hz at −120° to 108 Hz at 24° with further broadening to 135 Hz at 85°.

TABLE I
CHEMICAL SHIFTS AND GEMINAL COUPLING CONSTANTS FROM THE ^{19}F NMR SPECTRA OF 1 AND 2[a]

1

Assignment	24°	−73°	−115°
CF_2O	78.4, 80.1	77.7, 79.0	77.4, 78.5
J_{F-F}, Hz	89	89	90
CF_2N	105.0, 106.9	103.4, 107.0	102.7, 106.9
J_{F-F}, Hz	140	139	139
NF	−25.3	−25.6	−25.8

2

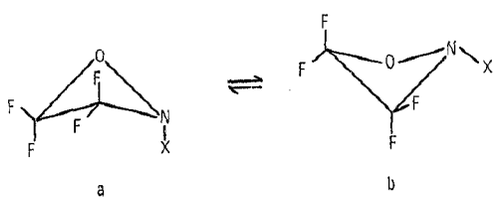
Assignment	24°	−73°	−115°
CF_2O	78.0, 81.6	77.4, 81.2	76.6, 80.6
J_{F-F}, Hz	89	90	90
CF_2N	95.5, 100.2	95.0, 99.1	94.4, 98.2
J_{F-F}, Hz	122	123	122

[a] Determined chemical shift of each fluorine in parts per million with $CFCl_3$ as internal standard.

version process at 85°. At this temperature a barrier of *ca.* 17 kcal/mol would be required.[4] The barrier to the ring inversion of cyclobutane has been estimated to be 0.47 kcal/mol.[5] Consideration of the effects of replacing hydrogens by fluorines and substitution of NF and O in the cyclobutane ring leads to the conclusion that the barrier to ring inversion of a perfluorooxazetidine ring should not be appreciably different.[6] Hence the observed nonequivalence must arise as a consequence of restricted nitrogen inversion.

Consistent with these results are the recent nmr studies of N-haloaziridines,[7,8] which demonstrate that nitrogen inversion is remarkably restricted in the N-chloro and N-bromo compounds. Although there apparently are no examples of similar behavior by an NF substituent,[9] equally effective retardation of nitrogen inversion by fluorine in small-ring compounds would not be unreasonable. Lee and Orrell[10] reported that nitrogen inversion in the related perfluoro-2-methyl-1,2-oxazetidine is essentially frozen at −74°. The higher barrier observed for **1** is thus consistent with this fact, since substitution of F for CF_3 would on steric and electrostatic grounds[11] lead to a higher barrier to nitrogen inversion.

The chemical-shift difference, δ,[12] for the geminal fluorines of **1** and **2** showed temperature dependence and was determined over the range 85 to −120°. These values of δ are given in Tables II and III. The fact that the δ values for the pairs of geminal fluorines in both compounds respond in different and opposite manners to change in temperature indicates that a direct temperature effect is not involved. Instead the change of δ with temperature suggests the presence of two conformers which are equilibrating at a rate such that only one AB pattern is observed for each set of geminal fluorines. This equilibrium involves the nonplanar conformations, a and b, and is represented as follows.



1a, 1b, X = F
2a, 2b, X = Cl

As a result of the 1,3 fluorine–fluorine and 1,3 fluorine–chlorine interactions in the a conformers, it might be expected that the b conformers would be the more stable. The fraction of molecules, p, in this latter conformation is related to the conformational free-energy difference, ΔG, by the expression

$$p/(1 - p) = K = e^{-\Delta G/RT} \quad (1)$$

This fraction, p, of molecules in conformer b is likewise related to δ, since the observed chemical-shift difference is simply the weighted average of the chemical-shift differences, δ_a and δ_b, of the individual conformers.

$$\delta = p\delta_b + (1 - p)\,\delta_a \text{ or } \delta = \delta_a + p(\delta_b - \delta_a) \quad (2)$$

Values of p for those temperatures utilized in the nmr study were calculated using expression 1 with ΔG varied in units of 100 cal/mol from −1600 to −400 cal/mol. These values of p[13] were then plotted *vs.* the

TABLE II
FLUORINE–FLUORINE CHEMICAL-SHIFT DIFFERENCES FOR PERFLUORO-2-FLUORO-1,2-OXAZETIDINE (1)

Temp, °C	δ_{CF_2N}, Hz	δ_{CF_2O}, Hz
85	68[a]	108
24	112	95
1	134	91
−20	153	84
−35	167	80
−55	184	74
−76	206	68
−85	215	66
−98	230	60[a]
−120	255	52[a]

[a] Center peaks of quartet not resolved. $(d_3 - d_1)$ calculated from $(d_4 - d_1) - J$.

TABLE III
FLUORINE–FLUORINE CHEMICAL-SHIFT DIFFERENCES FOR PERFLUORO-2-CHLORO-1,2-OXAZETIDINE (2)

Temp, °C	δ_{CF_2N}, Hz	δ_{CF_2O}, Hz
75	277	194
24	262	202
−10	254	207
−37	247	211
−73	232	216
−91	226	219
−115	214	222

(4) Assuming a coalescence temperature (T_c) of 85° and δ 100 Hz, ΔG^* = 17 kcal/mol is calculated using the expression $\Delta G^* = 4.57\ T_c\ (9.97 + \log T_c/\delta)$.

(5) H. E. Simmons and J. K. Williams, *J. Amer. Chem. Soc.*, **86**, 3222 (1964).

(6) (a) G. V. D. Tiers, *Proc. Chem. Soc.*, 389 (1960); (b) R. K. Harris and R. A. Spragg, *J. Chem. Soc.*, 864 (1968); (c) J. Lee and K. G. Orrell, *Trans. Faraday Soc.*, **63**, 16 (1967).

(7) S. J. Brois, *J. Amer. Chem. Soc.*, **90**, 506, 508 (1968).

(8) J. M. Lehn and J. Wagner, *Chem. Commun.*, 148 (1968).

(9) (a) Rapid inversion at nitrogen in perfluoro-N-fluoropiperidine was reported to be occurring even at −74°.[6c] (b) Restricted nitrogen inversion in an NF_2 group has been reported: F. A. Johnson, C. Haney, and T. E. Stevens, *J. Org. Chem.*, **32**, 466 (1967).

(10) J. Lee and K. G. Orrell, *Trans. Faraday Soc.*, **61**, 2342 (1965).

(11) F. A. L. Anet, R. D. Trepka, and D. J. Cram, *J. Amer. Chem. Soc.*, **89**, 357 (1967).

(12) $\delta = [(d_3 - d_1)^2 - J^2]^{1/2}$, where d_3 and d_1 are the chemical shifts of peaks 3 and 1 of an AB quartet.

(13) With $\Delta G = -900$ cal/mol, p values at different temperatures are, at 85°, 0.780; 24°, 0.821; 1°, 0.839; −20°, 0.857; −35°, 0.870; −55°, 0.889; −76°, 0.909; −85°, 0.912; −98°, 0.930; −120°, 0.953. With $\Delta G = -1000$ cal/mol, p values at different temperatures are, at 75°, 0.809; 24°, 0.845; −10°, 0.871; −37°, 0.894; −73°, 0.925; −91°, 0.941; −115°, 0.960.

corresponding values of δ to give a series of curves. This procedure was followed for both the CF_2N and CF_2O fluorines of **1** and **2**. In each case the best linear relationship between δ and p was obtained when $\Delta G = -900 \pm 100$ cal/mol (compound with NF) and $\Delta G = -1000 \pm 100$ cal/mol (compound with NCl).[14] From the slopes of the best straight lines, $\delta_b - \delta_a$ and subsequently values of δ_a and δ_b for all pairs of fluorines were determined. These values are given in Table IV. The relationship between p and δ is indicated in Figure 1.

TABLE IV
CONFORMATIONAL DATA

	δ_{1a}, Hz	δ_{1b}, Hz	δ_{2a}, Hz	δ_{2b}, Hz
CF_2N	−798	+311	+604	+202
CF_2O	+387	+35	+50	+229
ΔG, cal/mol	−900 ± 100		−1000 ± 100	

The determined conformational free-energy differences lend support to the initial assignment of greater stability to conformer b. Such destabilization of conformer a as a consequence of the 1,3-halogen interactions is not unreasonable[15] with the lesser destabilization in **1** consistent with the smaller size of the fluorine.

The chemical shifts of the geminal fluorines of **1** and **2** obtained from spectra at several temperatures are given in Table I. It may be concluded that both fluorines of the CF_2O of **1** and **2** are less shielded in conformer b, since all signals move downfield as the temperature is lowered and the proportion of the more stable conformer increases. The upfield fluorine of the CF_2O group of **1** appears to undergo the greater change in chemical shift, while the opposite is the case with the related fluorines of **2**. The chemical shift of the upfield fluorine of the CF_2N group of **1** is essentially unchanged throughout the temperature range, indicating nearly the same value in both conformers. Relative to this stationary fluorine, the other CF_2N fluorine is less shielded in conformer **1b** and more shielded in conformer **1a**. The negative value of δ_a for these fluorines reflects this reversal in the relative signal positions. Both the CF_2N fluorines of **2** appear to be less shielded in **2b** than in **2a** with the upfield fluorine undergoing the greater change in chemical shift.

High-resolution spectra of the CF_2O and CF_2N fluorines of **1** and **2** were obtained. The outer members of the AB quartet observed for the CF_2O fluorines of **1** showed an eight-peak pattern, indicating that these fluorines are coupled not only with the CF_2N fluorines, but also with NF. One of the CF_2N fluorines of **1** in its spectrum at 24° shows apparent coupling with all vicinal fluorines (eight-peak patterns) whereas the upfield members of this quartet are very broad, unresolved peaks. However, at low temperature (−100°), each member of the CF_2N quartet is clearly resolved into eight peaks. The spectrum of the N-chloro compound **2** similarly reveals that both of the CF_2O and one of the CF_2N fluorines undergo coupling with the adjacent fluorines and are well resolved peaks (doublet of doublets). In this case, the downfield

(14) It is assumed in this treatment that ΔG is constant over the temperature range studied; hence $\Delta S = 0$.

(15) Lambert and Roberts[2] determined $\Delta G = -750$ cal/mol as the energy difference for conformers of 1,1-difluoro-2,2,3-trichlorocyclobutane.

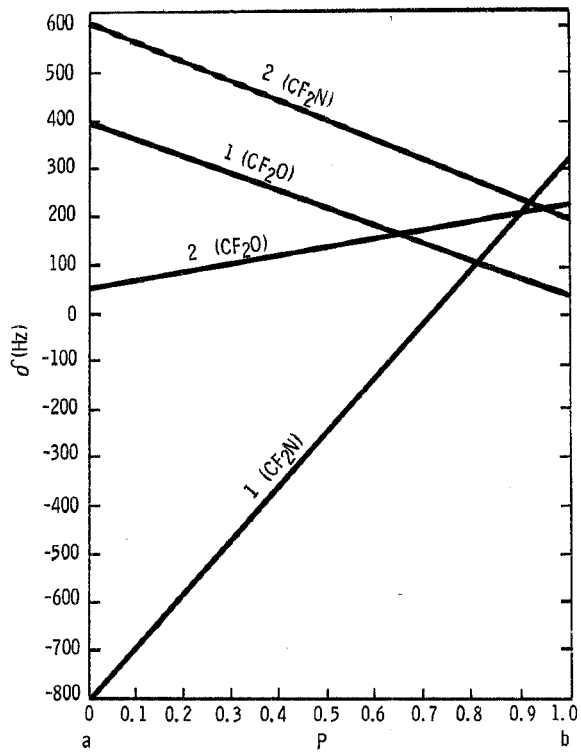


Figure 1.—Chemical-shift difference *vs.* conformer population.

members of the CF_2N quartet are those which are broad and essentially structureless. These members are likewise resolved in −100° spectra (doublet of doublets). Neither the complex ABMXY pattern of **1** nor the ABXY pattern of **2** were analyzed; thus the vicinal coupling constants are not given.

The assignment of nmr peaks to the CF_2N fluorines in **1** can be made if one assumes that a significant change in the nuclear shielding will be experienced by the fluorine *trans* and axial to the free electron pair of nitrogen. This assumption is based on the observation that the chemical shift of a proton located on a carbon bonded to nitrogen depends on its orientation relative to the unshared electrons of nitrogen, the *trans*-axial relationship giving rise to a pronounced upfield shift. The equatorial proton *cis* to the electron pair is essentially unaffected.[16] Since the CF_2N fluorine *cis* to NF would become axial and coplanar with the unbonded nitrogen electrons in conformer **1b**, one might expect that the chemical shift of this fluorine would be significantly different in conformer **1b** than in **1a**. Only one fluorine of the CF_2N group undergoes any detectable change in chemical shift in going from conformer **1a** to **1b**, that being the fluorine which appears as the resolved downfield portion of the AB quartet. This half is thus assigned to the fluorine *cis* to NF, while the broad upfield absorptions are attributed to the *trans* fluorine. Although the downfield shift of the *cis* fluorine in **1b** is contrary to the results referenced above for a similarly substituted hydrogen, the absence of change in chemical shift observed for the upfield fluorine makes the alternative assignment much less attractive.

The apparent relationship between configuration and peak broadening is the basis for the assignment of the

(16) H. P. Hamlow, S. Okuda, and N. Nakagawa, *Tetrahedron Lett.*, 2553 (1964); J. B. Lambert, R. G. Keske, R. E. Carhart, and A. P. Jovanovich, *J. Amer. Chem. Soc.*, **89**, 3761 (1967).

unresolved peaks in the spectrum of the CF_2N fluorines of **2** to the fluorine *trans* to NCl. The downfield shift of the *cis* CF_2N fluorine on going from **2a** to **2b** is consistent with that observed for the related fluorine in **1**. Lee and Orrell[10] have made the same assignment to the CF_2N fluorines of perfluoro-2-methyl-1,2-oxazetidine (spectrum obtained at −74°). The fluorine showing coupling with the CF_3 group (upfield half of AB quartet) was assigned *cis* to NCF_3, while the downfield fluorine was observed as a broad, structureless absorption.

The assignment of nmr peaks to the CF_2O fluorines is somewhat more difficult. However, it may be argued that the fluorine of this group, which is *cis* to the N-halo group, will experience a greater environmental change and consequently a more pronounced variation in chemical shift as a result of the 1,3-diaxial interaction in conformer a. On this basis, then, the *cis* CF_2O fluorine is assigned to the upfield half of the AB pattern in the spectrum of **1** and to the lower field half in the spectrum of **2**.

A small chemical-shift change was detected for the NF of **1** when the sample was cooled from 24 to −120°. The downfield shift amounted to *ca.* 0.5 ppm (ϕ^* −25.8 ppm at −120°). The broadness of the signal made the exact measurements of peak position difficult. Since the fraction of conformer **1b** would increase by 0.13 over this temperature range, the change in the NF chemical-shift in going from **1a** to **1b** would represent *ca.* 217 Hz.

Experimental Section

The ^{19}F nmr spectra were obtained with a Varian Model V-4302B spectrometer operating at 56.4 MHz. The spectra were calibrated by the sideband modulation technique using a Hewlett-Packard wide-range oscillator. Chemical shifts and coupling constants represent the average of at least eight measurements. Errors of ±0.1 ppm and ±1 Hz, respectively, were estimated.

For both low- and high-temperature studies, the variable-temperature accessory supplied by Varian was used. Temperature measurements were made both before and after recording spectra by means of a copper–constantan thermocouple immersed in a tube filled with a Kel-F oil. The temperature measurements are believed to be accurate to ±1°.

The chemical-shift differences (Tables II and III) obtained from nmr spectra of $CFCl_3$ solutions of **1** and **2** were essentially unchanged with the weight per cent of **1** and **2** varied from 25 to 50. However, the chemical-shift values were affected significantly by traces of acetone.

Perfluoro-2-fluoro-1,2-oxazetidine (**1**) and **perfluoro-2-chloro-1,2-oxazetidine** (**2**) were prepared by fluorination and chlorination, respectively, of perfluoro-1,2-oxazetidine as described previously.[17] Both compounds are low-boiling materials, with boiling points below −30°.

Registry No.—**1**, 21720-81-0; **2**, 21720-80-9.

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(17) R. A. Falk and J. D. Readio, *J. Org. Chem.*, **34**, 4088 (1969).

Polyfluoroaryl β-Dicarbonyl Compounds[1]

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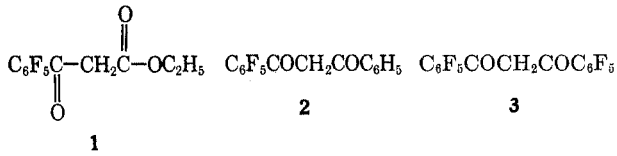
Ethyl pentafluorobenzoylacetate (**1**) is prepared by oxidation of ethyl 3-hydroxy-3-pentafluorophenylpropionate with Jones reagent, or, better, by reaction of pentafluorobenzoyl chloride (**4**) with diethyl malonate in the presence of magnesium ethoxide. Compound **1** exhibits 54% enolic character as the neat liquid, whereas ethyl benzoylacetate possesses 22% enol. The unsymmetrical 1,3 diketone pentafluorodibenzoylmethane (**2**) is prepared by reaction of the morpholine enamine of acetophenone with (**4**) or from pentafluoroacetophenone and methyl benzoate in the presence of sodium hydride. The symmetrical 1,3 diketone bis(pentafluorobenzoyl)-methane (**3**) has been obtained by three methods, the preferred route being the reaction of **4** with vinyl acetate.

As part of studies aimed at evaluating the effect of pentafluorophenyl substitution on the properties and chemical behavior of neighboring functional groups in organic molecules, we have examined several polyfluoroaryl β-dicarbonyl compounds. In this paper, we report the preparation and some properties of ethyl pentafluorobenzoylacetate (**1**) and the 1,3 diketones pentafluorodibenzoylmethane (**2**) and bis(pentafluorobenzoyl)methane (**3**).



1

2

3

(1) Presented, in part, at the Southeastern Regional Meeting of the American Chemical Society, Louisville, Ky., Oct 1966, and at the 156th National Meeting of the American Chemical Society, Atlantic City, N. J., Sept. 1968.

(2) To whom inquiries should be sent.

(3) Abstracted, in part, from the M.S. thesis of V. D. B., Jan 1966, and the Ph.D. thesis of F. N. M., Jan 1967.

Ethyl Pentafluorobenzoylacetate (**1**).—In our initial approach to compound **1**, pentafluorobenzoyl chloride (**4**) was treated with ethyl acetoacetate in alkaline medium, according to an established procedure for the preparation of ethyl benzoylacetate.[4] Instead of the desired β-keto ester, the sole product isolated was a substance whose elemental composition and infrared and proton magnetic resonance spectra were consistent with compound **5**, a substituted chromone (eq 1).

Compound **5** is formed by intramolecular displacement of *ortho* fluorine by the intermediate enolate anion. Such nucleophilic substitution cannot occur on a non-halogenated aromatic ring, and the reaction proceeds by an alternate course, *i.e.*, cleavage of the acetyl group to give the β-keto ester.

Shortly after completion of this work, our attention was drawn to similar observations by Soviet workers,[5]

(4) J. M. Straley and A. C. Adams, "Organic Syntheses," Coll. Vol. IV, John Wiley & Sons, Inc., New York, N. Y., 1963, p 415.

(5) N. N. Vorozhtsov, Jr., V. A. Barkhash, A. T. Prudchenko, and T. I. Khomenko, *Zh. Obshch. Khim.*, **35**, 1501 (1965).