to faster vibrational relaxation and reduced internal conversion rate in the matrix.[5] In conclusion, quenching of internal energy by the matrix probably plays a major role in sharpening cation absorptions in the matrix environment.

The presence of excess internal energy in the **4** cation formed in the PDS experiment is demonstrated by its essentially complete rearrangement to the **3** cation.[1] On the other hand, the matrix photoionization experiment with **4** quenches many **4** cations quickly enough to trap at least comparable amounts of **3** and **4** cations, based upon the assumption that the absorption cross sections for the $\pi \rightarrow \pi$ transitions of **3** and **4** cations are similar.

Conclusions

The matrix photoionization technique produces phenylalkene molecular cations with absorption spectra slightly red shifted from PDS observations. In several cases, the matrix bands are sharp enough to resolve vibrational structure. Rearrangements of several molecular ions were observed during sample condensation and upon photolysis of the cold sample. Of particular interest, the allylbenzene cation was rearranged to the β-methylstyrene cation and the indan cation by 290 nm radiation. The matrix absorptions are substantially sharper than PDS bands, which suggests that the PDS bandwidth may, in part, be due to excess internal energy in the molecular ions produced by electron impact. It is perhaps noteworthy that the allylbenzene cation rearranges completely to the β-methylstyrene cation in PDS experiments, but a substantial fraction of the allylbenzene cation formed in matrix photoionization experiments is quenched and trapped in the matrix.

Acknowledgment. The authors gratefully aknowledge financial support for this reasearch by the National Science Foundation under Grant No. CHE 79-10966 and a glassy matrix spectrum of β-methylstyrene cation from Dr. T. Shida.

Perfluoroacetylmethylmethylene and Its Quintet Radical Pair: Isolation and ESR Detection

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Received September 5, 1980

Abstract: Perfluoroacetylmethylmethylene (**1**) was produced by the photolysis of 3-diazohexafluoro-2-butanone at $T < 30$ K. Two triplet ESR spectra were observed with relative intensities of ca. 9:1 and assigned to the cis and trans geometrical conformers of **1**, respectively. These triplets are most likely the ground electronic state of **1**. The relative yields of the conformers appear to be governed by the geometry of the source compound. *cis*-**1** is kinetically more stable than *trans*-**1** and decays at a higher temperature featuring first-order kinetics with $E_a = 5.0$ kcal/mol, probably corresponding to triplet → singlet intersystem crossing prior to or concertedly with a Wolff rearrangement to the ketone structure. An additional weak ESR spectrum attributed to a quintet state radical pair arising from the interaction of two triplet **1** was also observed.

The existence of geometrical isomerism in triplet carbenes having an α double bond and frozen in low-temperature matrices was first demonstrated for the case of naphthylmethylenes with the use of ESR spectroscopy by Trozzolo, Wasserman, and Yager.[1] Since then cis/trans conformer pairs have been detected in the triplet state of several other α-unsaturated carbenes including vinylmethylene[2] (**2**), carbomethoxymethylene[3] (**3**), carboethoxymethylene[3] (**4**), iminocyclohexadienylidene,[4] benzoylphenylmethylene[5,6] (**5**), and dibenzoylmethylene.[7] Thiobenzoylphenylmethylene[8] prepared recently by the photolysis of 4,5-diphenyl-1,2,3-thiadiazole appeared to be present only as the cis isomer: this was attributed to the controlling effect of the geometry of the precursor molecule on the product carbene geometry.

In previous studies on triplet benzoylphenylmethylene[5,6] and triplet 9-oxo-10-anthrylene[9] we have observed in each case a new quintet state ESR spectrum, in addition to the triplet spectrum. These we assigned to a stable, quintet state radical pair formed by the interaction of two close neighbor triplet carbenes. Prior to these studies, radical pairs had not been detected in higher than triplet multiplicity although contemporaneously the existence of a short-lived quintet state radical pair has been reported.[10]

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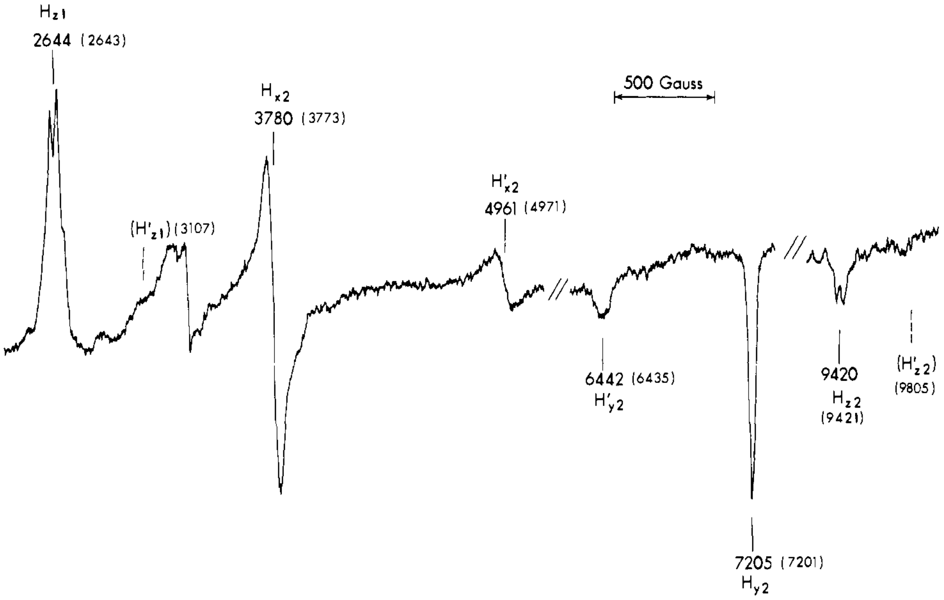

Figure 1. ESR spectrum of **1a** (H'_{x2} and H'_{y2}) and **1b** (H_{z1}, H_{x2}, H_{y2}, and H_{z2}) generated in the ($\lambda > 300$ nm) photolysis of 3-diazohexafluoro-2-butanone polycrystal at 11 K. The klystron frequency was 9.369 GHz. The values in brackets are the calculated ones obtained from eq 3 and 5–7.

Multiplicities higher than triplet are scarce even among stablized organic radicals. Trozzolo, Wasserman, and co-workers[11] observed complex ESR spectra in the low-temperature matrix photolysate of 1,3-bis(α-diazobenzyl)benzene and 1,3-diazidobenzene and assigned them to the quintet ground states of *m*-dicarbene and *m*-dinitrene, respectively. Itoh[12] also succeeded in stabilizing the quintet state of *m*-dicarbene in the precursor single crystal. There are also reports on the detection of septet-state organic radicals produced in the photolysis of 1,3,5-triazido-2,4,6-tricyanobenzene[13] and 1,3,5-tris(α-diazobenzyl)benzene.[14] More recently, Weissman and Kothe[15] reported the quartet ESR spectrum of the stable molecule 1,3,5-(2,4,6-tricyanobenzenetriyl)tris(N^1,N^1-diphenylhydrazyl). Using an L-band spectrometer, these authors were able to observe the $\Delta m = 3$ transition, making the assignment of the spectrum unambiguous.

The purpose of the present study was to attempt the synthesis of a new ketocarbene, perfluoroacetylmethylmethylene (**1**), and to explore its ESR spectroscopy.

Experimental Section

The ESR spectra were recorded with a Bruker ER-420 X-band spectrometer equipped with 100 kHz modulation in which the magnetic field was controlled by a Varian Fieldial V-FR 2503 and monitored by an Alpha digital NMR gaussmeter Model 3093. The microwave frequency was determined with a Hewlett Packard Model X532B frequency meter.

3-Diazohexafluoro-2-butanone was prepared according to the procedure given by Dyatkin and Mochalina.[16] The neat sample was degassed under vacuum and sealed in a suprasil ESR tube with a diameter of 3 mm. By quenching this sample at 77 K a glass was obtained, which on repeated warming and cooling turned into a polycrystal.

The sample prepared this way was inserted into a previously cooled cryogenic Dewar, Model LTD-3-110A Heli Tran Transfer System provided by Air Product and Chemicals, installed inside the ESR cavity. A variable temperature Dewar (Bruker) was used to study the decay kinetics. The sample was photolyzed by a medium pressure mercury lamp through a Pyrex glass filter ($\lambda > 300$ nm) and the sample temperature was monitored at the upper position of the sample inside the cryogenic Dewar by a thermocouple of chromel vs. gold, 0.07 atom % iron.

Results and Discussion

Ketocarbene **1** was prepared by the $\lambda > 300$ nm photolysis of 3-diazohexafluoro-2-butanone either in the glass or polycrystal phase. An illustrative example of the triplet portion of the spectra obtained is shown in Figure 1. Two sets of triplet absorptions can be distinguished, an intense one, H_{z1}, H_{x2}, H_{y2}, and H_{z2}, and a weak one, (H'_{z1}), H'_{x2}, H'_{y2}, and (H'_{z2}).

The spectra were analyzed with the aid of the theoretically derived resonance field equations for ESR absorption reported in the literature,[17] which were derived by the exact solution of the Hamiltonian,

$$\mathcal{H} = g\beta\vec{H}\cdot\vec{S} + D\left\{S_z^2 - \frac{S(S+1)}{3}\right\} + E(S_x^2 - S_y^2) \quad (1)$$

where $S = 1$ for a triplet. If the g tensor is assumed to be isotropic and $g = g_e$, as was used for the best fit of this work, then the values of D and E can be obtained from the experimentally observed absorptions by the equations:

$$H_{x1} = (g_e/g)[(H_0 - D' + E')(H_0 + 2E')]^{1/2} \quad (2)$$

$$H_{x2} = (g_e/g)[(H_0 + D' - E')(H_0 - 2E')]^{1/2} \quad (3)$$

$$H_{y1} = (g_e/g)[(H_0 - D' - E')(H_0 - 2E')]^{1/2} \quad (4)$$

$$H_{y2} = (g_e/g)[(H_0 + D' + E')(H_0 + 2E')]^{1/2} \quad (5)$$

$$H_{z1} = (g_e/g)[(H_0 - D')^2 - E'^2]^{1/2} \quad (6)$$

$$H_{z2} = (g_e/g)[(H_0 + D')^2 - E'^2]^{1/2} \quad (7)$$

where $D' = D/(g_e\beta)$, $E' = E/(g_e\beta)$, and $H_0 = h\nu/(g_e\beta)$.

On the basis of apparent similarities in the appearance, kinetic behavior, and values of computer zero-field splitting parameters, the two spectra are assigned to the geometrical isomers of **1**: the set of strong signals—H_{z1}, H_{x2}, H_{y2}, and H_{z2}—to isomer **1b** and the other set of weak signals—H'_{x2} and H'_{y2}, the lowest and highest absorptions of which, (H'_{z1}) and H'_{z2}), are not clearly discernible from the spectrum—to isomer **1a**.

As seen from Figure 1, only four absorptions are apparent for isomers **1b** and two for **1a** while resonance field eq 2–7 predict

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Table I. Zero Field Splitting Parameters[a]

species	D, cm^{-1}	E, cm^{-1}	ref
1a	0.605	0.034	this work
1b	0.572	0.079	this work
2a	0.4578	0.0193	2
2b	0.4093	0.0224	2
3a	0.663	0.030	3
3b	0.617	0.053	3
4a	0.661	0.031	3
4b	0.616	0.053	3
5a	0.3916	0.0518	6
	0.3815	0.0489	23
5b	0.3123	0.0519	6

[a] $g = 2.002$ was used as the best fit of 1a and 1b.

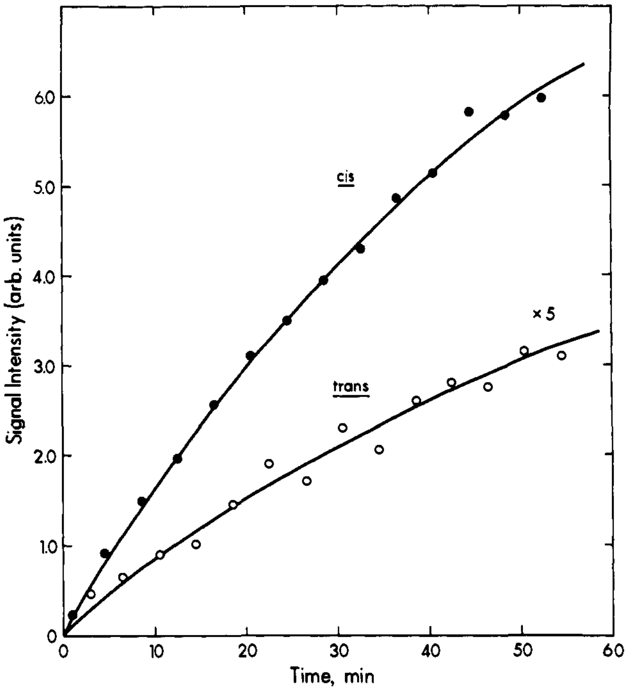


Figure 2. Growth of ESR signals of **1a** (open circle) and **1b** (closed circle) during photolysis of 3-diazohexafluoro-2-butanone polycrystal at 10 K.

the existence of six absorptions for each isomer. The reason for the absence of some absorptions becomes immediately clear on substituting the numerical values into the equation. Thus, being the values of H_0 and D' and 3342 G and 6470 G, respectively, eq 2 and 4 give imaginary values for H. In other words, when the triplet species have D values higher in energy than H_0 then H_{x1} and H_{y1} (or H_{x2} and H_{y2} depending on the absolute value of D which cannot be determined from conventional absorption measurements) will become imaginary. The calculated and experimental values of H and H' are given in Figure 1. The intensity distribution of the two spectra is probably quite similar and therefore the nonappearance of the H'_{z2} transition is due to its inherently low intensity while the nonappearance of the H'_{z1} transition is more likely due to its overlap with the color center absorption of quartz.

The H_{z1} and H_{z2} absorptions of **1b** each clearly show a doublet splitting of 33 G with indications of possible further splittings. The H_{x2} absorption may also feature some hyperfine structure but the H_{y2} transition is clearly structureless. Undoubtedly the cause of hyperfine structure is the presence of fluorine atoms.

The zero-field splitting parameters of these triplets as determined from the spectra are tabulated in Table I along with the literature values of other related conformer pairs reported to date.

Both the triplet signals were also observed in the neat glass of the precursor and the D and E values derived from the glass phase spectra were nearly identical with those obtained in the polycrystal phase. However, the line width of **1b** was considerably affected;

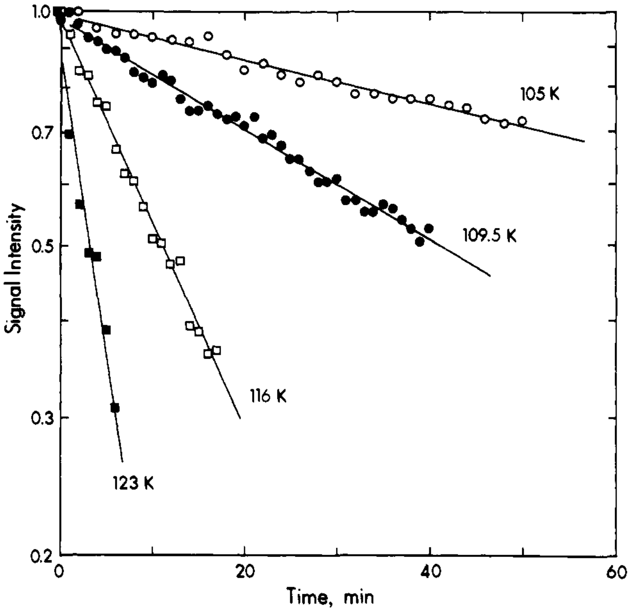


Figure 3. Typical first-order decay of **1b**.

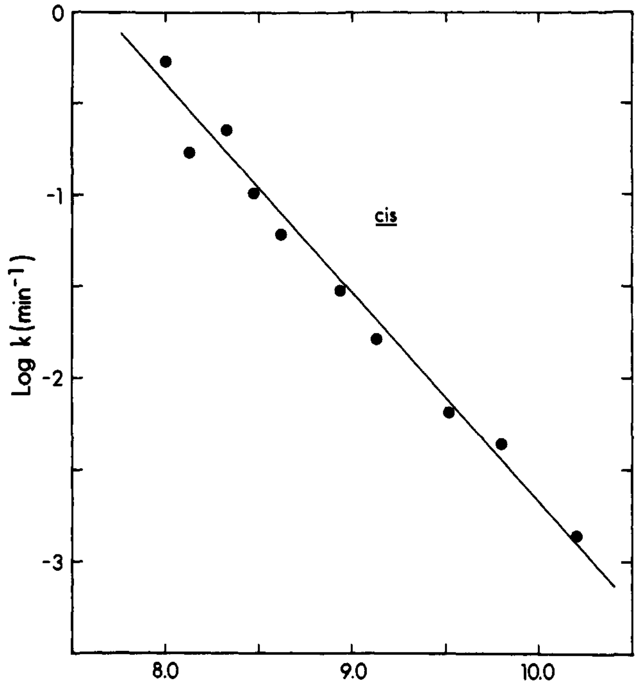


Figure 4. Arrhenius plot of the first-order rate constant for the decay of **1b**.

it was broadened and the signal intensities weakened. For example, the peak-to-peak width of the H_{x2} line was ~230 G as compared to ~75 G in polycrystals and the hfs of the H_{z1} and H_{z2} absorptions became indiscernible. All subsequent experiments were then done in polycrystals.

In order to determine whether **1a** and **1b** are primary products of the photolysis, the variation of triplet signal intensities with exposure time was examined. As seen from Figure 2, both plots give smooth curves extrapolating to zero intensity at zero exposure time and it is concluded that both **1a** and **1b** are primary products of the decomposition of the precursor molecule, 3-diazohexafluoro-2-butanone.

Below 30 K both **1a** and **1b** appear to be stable for at least several hours. **1a** decays between 50 and 70 K but **1b** is stable up to 100 K, above which it decays with good first-order kinetics, Figure 3. The Arrhenius plot for the decay, Figure 4, is also a good straight line giving E_a = 5.0 ± 0.5 kcal mol^{-1} and log A (s^{-1})

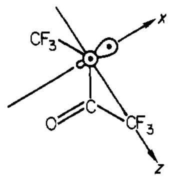
= 6.5 ± 0.9. Analogous data on **1a** could not be obtained owing to the weakness of signal intensities, ca. 10% of **1b**.

The values of the zero-field splitting parameters, D and E, are given by the sum of one-center and two-center interactions. The main contribution comes from the one-center term of the divalent carbon with a smaller contribution from the two-center term involving π-spin distributions on the carbonyl carbon and the carbonyl oxygen. These are both governed by the distance r between the two spins and by the components of r along the principal x, y, z axes and are given by the formulas:

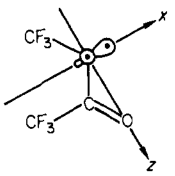
$$D \propto \langle (r^2 - 3z^2)/r^5 \rangle \qquad (8)$$

$$E \propto \langle (y^2 - x^2)/r^5 \rangle \qquad (9)$$

Assuming that the principal axes are close to those in other triplet carbenes,[2,3,6-8] the carbonyl oxygen should make a larger negative contribution to the D value in the cis isomer, because the oxygen atom, on which relatively high π-spin density could be expected, lies almost on the z axis. Concerning the E value, a negative contribution of the two-center term could be expected in the trans isomer in a like manner. Based on these considerations, we assign **1a** to the trans conformer and **1b** to the *cis* conformer, respectively.

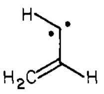


trans-1a

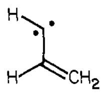


cis-1b

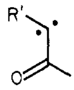
The existence of these correlations was first pointed out and applied to the interpretation of the ESR spectra of rotameric isomers of vinylmethylene (**2**), by Hutton, Manion, Roth, and Wasserman[2] and later for carbalkoxymethylenes (**3** and **4**) by Hutton and Roth.[3] More recently, we have been able to isolate the two rotameric isomers of benzoylphenylmethylene (**5**) and assign them on the basis of this principle.[6] The zero-field splitting parameters of these carbenes are also listed in Table I for comparison. Assuming that the geometry of the carbene is determined by the geometry of the precursor molecule, the preponderance of **1b** over **1a** is taken as an indication that the 3-diazohexafluoro-2-butanone molecule is present largely in the cis geometry in the polycrystal.



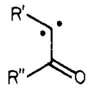
2a



2b



3a
4a
5a



3b, R' = H; R'' = OMe
4b, R' = H, R'' = OEt
5b, R' = R'' = Ph

The π-spin density of the divalent carbon is estimated to be 0.8 from the D value of the methylene triplet,[18] and the whole π-spin distribution in **1** is probably very close to that in the carbalkoxymethylenes **3** and **4**.[3]

It is of interest to note that in the case of benzoylphenylmethylene (**5**) the signal intensities of the cis/trans conformers are inverted and the trans conformer is the preponderant product from the photolysis of azibenzil. This can be attributed to steric repulsion between the two bulk phenyl groups causing destabilization in the cis compared to the trans conformer of azibenzil.[19] With alkyl α-diazoketones in general the rotational equilibrium may be shifted toward the cis or the trans conformer depending on the direction and magnitude of the electronic and steric effects of the substituent.[20] Thiobenzoylphenylmethylene on the other hand, as was mentioned above, is present only in the cis form when produced by the photolysis of 4,5-diphenyl-1,2,3-thiadiazole.[8] In this case the cyclic structure of the precursor molecule clearly preimposes the cis geometry on the carbene product.

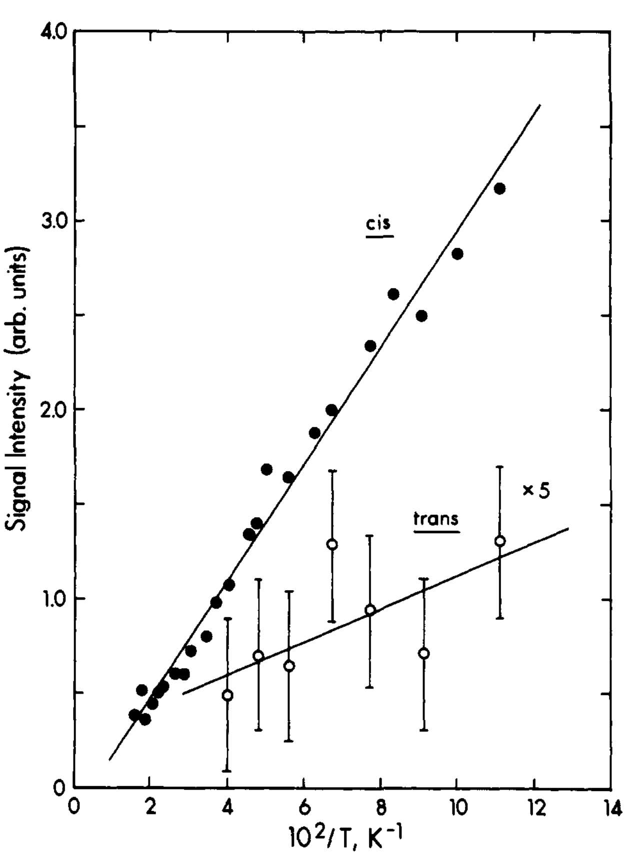


Figure 5. Signal intensities of the H_{x2} line of **1b** (closed circle) and the H'_{x2} line of **1a** (open circle) vs. T^{-1}. The lines represent the best linear least-squares fit of the data. The error bars of **1a** show the uncertainty by noise.

The trend in the kinetic behavior of the two ketocarbenes **1** and **5**, the only pair for which comparative data are available, is similar in that the trans conformers are kinetically less stable and decay at lower temperatures than the cis conformers. In neither case are the conformers interconvertible thermally or photolytically which in turn suggests that the rotational energy barriers are considerably higher than the activation energies measured for the decay of the cis conformer, which for **1** is 5 kcal mol^{-1}. This suggests that a Wolff rearrangement to ketenes from the ground triplet state of ketocarbenes is more facile in the trans than in the cis conformer, although the opposite has been claimed in the solution phase[21] in which the lowest singlet potential surface is involved. On the basis of the low value of the preexponential factor measured, $10^{6.5}$ s^{-1}, it is proposed that the decay of **1b** is via intersystem crossing either to the S_1 state, followed by a Wolff rearrangement to the ketone,

$$cis\text{-}CF_3CCOCF_3(T_0) \xrightarrow{\Delta} cis\text{-}CF_3CCOCF_3(S_1) \qquad (10)$$

$$cis\text{-}CF_3CCOCF_3(S_1) \rightarrow (CF_3)_2C{=}C{=}O(S_0) \qquad (11)$$

or concertedly to the S_0 surface of the ketone,

$$cis\text{-}CF_3CCOCF_3(T_0) \xrightarrow{\Delta} (CF_3)_2C{=}C{=}O(S_0) \qquad (12)$$

Parallel IR studies of the 10 K photolysis of argon matrix isolated 3-diazohexafluoro-2-butanone showed that the IR detectable end product of the photolysis is the ketene. Most of the ketene appears to form via a stable intermediate, most likely the T_0 state keto-

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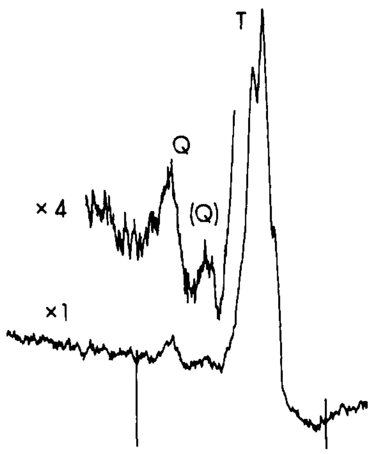

Figure 6. ESR spectrum of quintet radical pair (Q) and the H_{z1} line of triplet **1b** (T) generated in the photolysis of 3-diazohexafluoro-2-butanone polycrystal at 18 K. The klystron frequency was 9.371 GHz.

carbene, indicating that at 10 K intersystem crossing competes favorably with the Wolff rearrangement (eq 11).

$$cis\text{-}CF_3CN_2COCF_3 + h\nu \xrightarrow{-N_2} cis\text{-}CF_3CCOCF_3(S_1) \quad (13)$$

$$cis\text{-}CF_3CCOCF_3(S_1) \xrightarrow{ICS} cis\text{-}CF_3CCOCF_3(T_0) \quad (14)$$

The temperature dependence of signal intensities was also examined in the 8–50 K range and the results are plotted in Figure 5. The signal intensity changes are reversible and the intensity of the cis conformer clearly obeys the Curie law. The data on the trans conformer are scattered but also appear to follow the Curie law. The linearity of the plots in Figure 5 is consistent with a triplet ground state or with a near degenerate triplet–singlet state. The former is likely.

In addition to the triplet spectra discussed above, the other weak but well-reproducible transitions were observed in the 2300–2500 G range, Figure 6. These appeared in the polycrystal but not in the neat glass photolysis and only at $T \leq 40$ K. From previous studies on triplet benzoylphenylmethylene,[5] 9-oxo-10-anthrylene,[9] and pivaloyl-*tert*-butylmethylene[22] we known that the quintet state radical pairs which have been shown to form in these systems from the interaction of two close neighbor triplet carbenes exhibit weak absorptions in this spectral range corresponding to $m = 0 \rightarrow |m| = 1$ transitions. These occur at H_z, H_y, and H_x of about 2360, 2620, and 2670 G for benzoylphenylmethylene, 2300, 2490, and 2530 G for 9-oxo-10-anthrylene, and 2300 and 2450 G for pivaloyl-*tert*-butylmethylene, respectively. On the basis of the close similarity among these spectra in regard to location and intensity, nonappearance in glassy matrix photolysis, low thermal stability, and persistent good reproducibility we assign these absorptions to the quintet state radical pairs formed from two neighbor **1** molecules.

Acknowledgment. We thank the Natural Science and Engineering Research Council of Canada for financial support.

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On the Equilibrium [10]Annulene⇌Bisnorcaradiene. X-ray Study of the β Form of 11-Methyltricyclo[4.4.1.0^{1,6}]undeca-2,4,7,9-tetraene-11-carbonitrile at Two Temperatures

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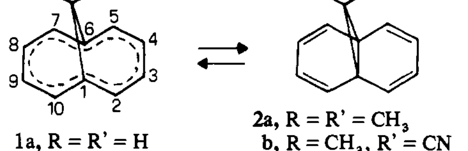
Contribution from the Istituto di Chimica-Fisica e Centro C.N.R., Università, 20133 Milano, Italy. *Received February 3, 1981. Revised Manuscript Received June 9, 1981*

Abstract: A new crystal phase (β form) of 11-methyltricyclo[4.4.1.0^{1,6}]undeca-2,4,7,9-tetraene-11-carbonitrile was studied by X-ray diffractometry at room temperature and at -100 °C. The unusually long C–C bond (mean value 1.817 Å), found in the previously studied phase (α form), is strongly reduced in the β form at room temperature (1.712 Å) and even more at -100 °C (1.622 Å). All other geometrical parameters vary in such a way as to emphasize the bisnorcaradiene character of the molecule. The remarkable dependence of the molecular geometry on crystal field and temperature suggests that the molecule is better described as a "fluxional system".

The geometry of a number of 1,6-methano[10]annulene derivatives has been determined in our laboratory in connection with the study of the equilibrium [10]annulene (**1**) ⇌ bisnorcaradiene (**2**).



1a, R = R' = H
b, R = R' = F

2a, R = R' = CH_3
b, R = CH_3, R' = CN
c, R = R' = CN

It is known[1,2] that the introduction of π-donating groups at carbon C(1) in the cyclopropane ring lengthens all C–C bonds in the ring, while the introduction of π-acceptor substituents shortens the C(2)–C(3) bond and lengthens the other two. Hence, it can be argued that the 1,6 distance should decrease in the series **1b**, **1a**, **2a**, **2b**, **2c**. In fact Günther and Schmickler[3] showed that the ^{13}C NMR chemical shift of C(1) and C(6) changes system-

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(1) R. Hoffman, *Proc. Int. Congr. Pure Appl. Chem.*, **23** (1971).

(2) R. Hoffman and R. B. Davidson, *J. Am. Chem. Soc.*, **93**, 5699 (1971).

(3) H. Günther and H. Schmickler, paper presented at the Second International Symposium on Non-benzenoid Aromatic Compounds, Lindau, Sept 23-27, 1974, ISNA II.

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