

Table I. Relative Reactivity and Orientation of Substituted Benzenes in the Gas-Phase Nitration by Radiolytically Formed MeO<sup>+</sup>(H)NO<sub>2</sub> Ions

substrate PhX	rel reactivity <sup>a</sup> $k_{\text{PhX}}:k_{\text{PhH}}$	orientation <sup>b</sup>			partial rate factors <sup>c</sup>		
		% ortho	% meta	% para	$f_o$	$f_m$	$f_p$
toluene	2.9	52	≈4	44	4.5		7.7
fluorobenzene	0.31	26	≈3	71	0.24		1.3
chlorobenzene	0.25	43	11	46	0.32	0.082	0.69
α,α,α-trifluorotoluene	$3.7 \times 10^{-3}$	d	100	d		0.011	
anisole	3.2	27	d	73	2.6		14
mesitylene	5.6		e				

<sup>a</sup>Standard deviation ca. 20%, except in the case of trifluorotoluene, ca. 30%. <sup>b</sup>Standard deviation ca. 10%, except for *m*-nitrotoluene and *m*-fluoronitrobenzene, whose reported percentages are crude estimates, owing to their low abundance among isomers. <sup>c</sup>Partial rate factors are omitted when the corresponding products could not be detected, or the yields were so low as to confer a largely approximate character to the data, e.g., in the case of *m*-F and *m*-Me factors. <sup>d</sup>Below detection limit, ca. 2%. <sup>e</sup>Only 2,4,6-trimethylnitrobenzene formed.

PhMe and other aromatics, while double-resonance ICR experiments confirm that MeO<sup>+</sup>(H)NO<sub>2</sub> is indeed their precursor, consistent with the results of Bursey and co-workers.<sup>2</sup>

The radiolytic experiments involve irradiation of mixtures containing CH<sub>4</sub> (720 torr), MeNO<sub>3</sub> (10–15 torr), the aromatic substrate(s) (≤1 torr), and a radical scavenger (O<sub>2</sub>, 10 torr), at 37.5 °C in a 220 Gammacell (Nuclear Canada Ltd.) to a dose of 3 Mrad.<sup>10</sup> Analysis of the irradiated samples by GLC and GLC/MS<sup>11</sup> demonstrates significant yields of the expected nitrobenzenes, e.g., G<sub>+PhNO<sub>2</sub></sub> 1.2 ± 0.2 from PhH. The mass spectrometric data make interpretation of the radiolytic results straightforward. CH<sub>3</sub><sup>+</sup> and C<sub>2</sub>H<sub>5</sub><sup>+</sup>, formed from the radiolysis of CH<sub>4</sub> with a combined G<sub>+M</sub> = 2.8,<sup>12</sup> promote nitration according to eq 1 and 2. Deprotonation of the nitrated adducts, which obviously contain a C–N bond, by any gaseous base, including the substrate itself, eventually yields neutral nitrobenzenes. The ionic character of the nitration is independently demonstrated by the dramatic depression of the yields caused by additives (e.g., NH<sub>3</sub>) which intercept C<sub>n</sub>H<sub>5</sub><sup>+</sup><sup>13</sup> and MeO<sup>+</sup>(H)NO<sub>2</sub>. The role of the latter as the nitrating agent emerges from the observation that replacing CH<sub>4</sub> with *i*-C<sub>4</sub>H<sub>10</sub> nearly suppresses the formation of PhNO<sub>2</sub> (G<sub>+M</sub> ≤ 0.015), owing to the inability of *t*-C<sub>4</sub>H<sub>9</sub><sup>+</sup> to protonate MeNO<sub>3</sub>.<sup>14</sup>

The radiolytic results are summarized in Table I. With the exception of those concerning anisole, the partial rate factors fit a reasonably linear (correlation coefficient 0.983) Hammett's plot,<sup>15</sup> whose ρ<sup>+</sup> value, –3.3, characterizes the gas-phase nitration as a typically electrophilic substitution. As a comparison, typical ρ<sup>+</sup> values of aromatic nitration are ca. –9 (HNO<sub>3</sub> in H<sub>2</sub>SO<sub>4</sub>) and –6 (AcONO<sub>2</sub> in Ac<sub>2</sub>O).<sup>16</sup> The lower than expected reactivity of anisole is explained by the incursion of the proton transfer PhOMe + MeO<sup>+</sup>(H)NO<sub>2</sub> → PhO<sup>+</sup>(H)Me + MeNO<sub>3</sub> (3)

Indeed, anisole is the only substrate investigated containing a n-type site more basic than MeNO<sub>3</sub>.<sup>17</sup> Since exothermic proton transfer processes involving n-type bases generally proceed at

nearly unit collision efficiency in the gas phase,<sup>18</sup> competition by reaction 3 provides a neat explanation of the abnormally low nitration rate of anisole.

In conclusion, the present preliminary results show that the combination of mass spectrometry with radiolytic techniques and use of a suitable electrophile allow the study of aromatic nitration in the gas phase without incurring into earlier, disturbing inconsistencies with solution-chemistry trends.

Much scope exists for exploiting the approach outlined in this report, since the unique properties of the gaseous reaction environment allow direct comparison with theoretical results and construction of crucial, quantitative arguments bearing on fundamental aspects of aromatic reactivity, long debated in solution chemistry.<sup>19</sup>

**Warning:** Methyl nitrate may explode if overheated in closed vessels. Attempts to shatter fragile Pyrex ampules containing liquid MeNO<sub>3</sub> may result in explosions.

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(18) Su, T.; Bowers, M. T., ref 7b, Vol. 1, pp 83–118.

(19) For a discussion concerning aromatic nitration at encounter rate, and the question of π-type vs σ-type transition states, see ref 1 and: (a) Olah, G. A. *Acc. Chem. Res.* 1971, 4, 240–248. (b) Ridd, J. H. *Acc. Chem. Res.* 1971, 48 248–253.

## Footballene: A Theoretical Prediction for the Stable, Truncated Icosahedral Molecule C<sub>60</sub>

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This paper presents preliminary theoretical evidence for the stable molecule C<sub>60</sub>. In the proposed molecule, each carbon atom is sp<sup>2</sup> hybridized and bonded to three other carbon atoms. Hence C<sub>60</sub> is an "alkene" containing no hydrogen and approximates a spherical shell of graphite.

Any structure with 12 pentagonal rings and 20 hexagonal rings satisfies the requirements of chemical bonding and constitutes a roughly spherical molecule, as shown below. The pentagonal rings may be viewed as "defects" compared to the unstrained hexagonal rings. The most symmetrical possible structure is the truncated icosahedron (Figure 1a). The pentagonal rings sit as far as possible from each other, at the vertices of an icosahedron (Figure 1b). For convenience this molecule is called here "footballene" ("soccerballene" in the US). Each carbon atom is equivalent to every other carbon atom, being the vertex joining a pentagon and

(10) For details, cf.: Cacace, F. *Radiat. Phys. Chem.* 1982, 20, 99–110 and references therein.

(11) The following columns were used: (i) A 3.3-m-long, 3-mm-i.d. glass column, packed with SP-2100 (20% w/w) and Carbowax 1500 (1%) on 100–120 mesh Supelcoport, operated at 110–130 °C. (ii) A 25-m-long, 0.25-mm-i.d. silica column, coated with methylsilicone fluid, operated from 80 to 120 °C. (iii) A 25-m-long, 0.53-mm-i.d. silica column, coated with OV-101, operated from 60 to 120 °C.

(12) Ausloos, P.; Lias, S. G.; Gorden, R., Jr. *J. Chem. Phys.* 1963, 39, 3341–3348. From this value, the PhNO<sub>2</sub> yield can be estimated to be ca. 40%.

(13) (a) Meot-Ner, M. *J. Am. Chem. Soc.* 1979, 101, 2589–2595. (b) Attinà, M.; Cacace, F.; Giacomello, P.; Speranza, M. *J. Am. Chem. Soc.* 1980, 102, 6896–6898.

(14) From the data of ref 8, the PA of MeNO<sub>3</sub> can be calculated to be ca. 185 kcal mol<sup>-1</sup>, vs. 197 kcal mol<sup>-1</sup> of isobutene, ref 7b.

(15) While it is questionable whether ρ<sup>+</sup> values from solution-chemistry reactions can be extended with any degree of accuracy to the gas phase, previous attempts have revealed no conspicuous discrepancies, cf.: Lau, Y. Y.; Kebarle, P. *J. Am. Chem. Soc.* 1976, 98, 7452–7453.

(16) Coombes, R. G.; Crout, D. H. G.; Hoggett, J. G.; Moodie, R. R.; Schofield, K. *J. Chem. Soc. B* 1970, 347 and references therein.

(17) From the calculated PA of the oxygen atom of PhOH (cf.: De Frees, D. J.; Mc Iver, R. T., Jr.; Hehre, W. J. *J. Am. Chem. Soc.* 1977, 99, 3853–3854 and the basicity increase due to Me substitution, the oxygen atom of PhOMe can be expected to be assuredly more basic than MeNO<sub>3</sub>.

<sup>†</sup>U.S. Presidential Young Investigator, 1985–1990.

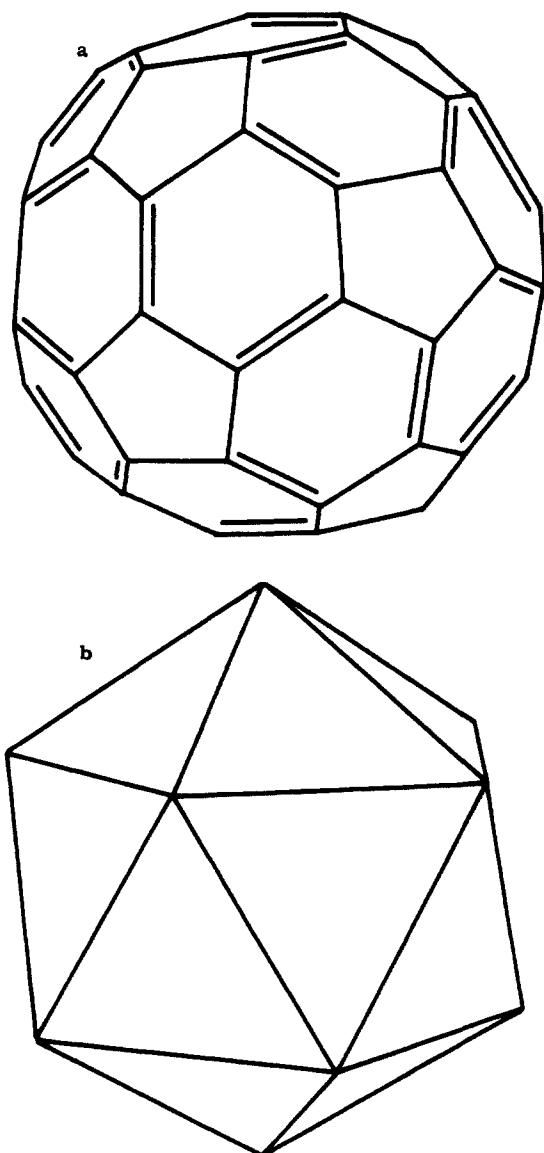


Figure 1. (a) Proposed  $C_{60}$  molecule "Footballene". (b) An Icosahedron.

two hexagons. Footballene has icosahedral point group symmetry.<sup>1</sup>

The evidence for the stability of footballene is simply stated. There is a dramatic stabilization energy from the delocalization of  $\pi$  electrons. The only destabilizing factor is that one of the C-C-C bond angles at each atom is  $108^\circ$  rather than  $120^\circ$ . Each atom must lie not in the plane defined by its three neighbors but a distance  $0.2r$  above the plane, where  $r$  is the C-C bond length (approximately 1.4 Å). This degree of nonplanarity apparently exists in [5.6.1]corrannulene (Figure 2).

A simple Hückel calculation<sup>2</sup> shows the delocalization energy of footballene. Energy is measured here in units of  $\beta$ , the usual nearest-neighbor orbital overlap parameter. In these units, the delocalization energy of benzene is  $2\beta$ , or  $0.3333\beta$  per carbon atom. The Hückel theory estimate for the delocalization energy of footballene is  $33.1616\beta$ , or  $0.5527\beta$  per carbon atom. Although Hückel theory does not account for the nonplanar strain at each carbon atom, it is a suggestive argument for the stability of footballene.

The evidence for  $C_{60}$  is strengthened by the existence of [5.6.1]corrannulene.<sup>3,4</sup> This molecule is believed to be nonplanar

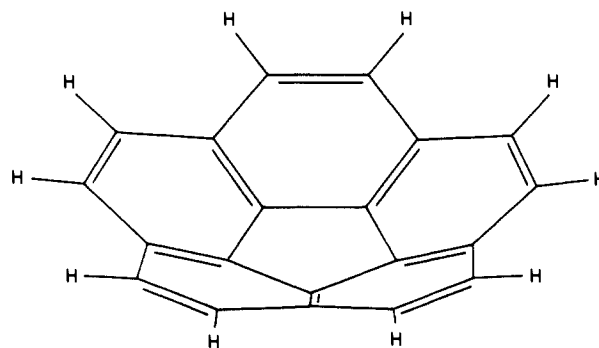


Figure 2. [5.6.1]Corrannulene.

and its atomic positions represent, roughly speaking, one half of footballene. For this molecule Hückel theory predicts a delocalization energy of  $0.4369\beta$  per carbon atom. Again, some of this stabilization energy is balanced by the strain at the nonplanar  $sp^2$  carbon atoms of the pentagonal ring. Molecular mechanics calculations<sup>5</sup> using the MMP2 program of Allinger,<sup>6</sup> which incorporate the effects of  $\pi$  electron delocalization via PPP theory, predict that [5.6.1]corrannulene has  $C_{5v}$  symmetry with a degree of nonplanarity at the pentagonal ring equal to that required in footballene.

If footballene exists and can be synthesized, it will have remarkable properties. Although a (macroscopic) football or soccerball has just one "bond length", there are two characteristic lengths in the molecule footballene, the C-C bond separating two hexagonal rings and the C-C bond separating a pentagon from a hexagon. These are likely to have similar but not necessarily identical lengths. Assuming equal C-C bond lengths of 1.4 Å, footballene will be a little spherical shell of graphite about 7 Å in diameter, with an electron-rich core due to the inward pointing lobes of the p-orbitals participating in the delocalized  $\pi$ -bonds. Small metal ions may sit comfortably inside footballene, and although not chemically bonded to the carbon shell the ion would be trapped inside, unable to tunnel through a hexagonal or pentagonal carbon ring.

The possible structure of a spherical shell of  $sp^2$  hybridized carbon atoms can be deduced mathematically. Consider a molecule of " $v$ "  $sp^2$  hybridized carbon atoms. The number of C-C bonds or edges " $e$ " is just  $3v/2$ . Let " $f$ " be the total number of faces,  $f = m + n$  where " $m$ " and " $n$ " are the number of pentagonal and hexagonal rings. Furthermore,

$$e = (5m + 6n)/2 = 3v/2$$

since each edge joins two faces. Finally Euler's famous theorem proves that

$$v + f = e + 2$$

and hence the only possible molecules have  $m = 12 =$  number of pentagonal rings and  $n = v/2 - 10 =$  number of hexagonal rings. Only molecules with an even number " $v$ " of carbon atoms are possible. The case  $n = 0$  corresponds to the highly strained dodecahedrene molecule which is unlikely to be stable. Footballene is one possible structure of the case  $n = 20$  ( $v = 60$ ). If the nonplanar strain makes footballene unstable (which it is argued above will not happen) then larger graphite shells with less strain are available.

After these calculations were completed Smalley and collaborators announced<sup>7</sup> an experiment on carbon clusters which displays a large peak in a mass spectrum at 60 carbon atoms. Independently they advance the possibility that footballene is responsible for the observed peak. However, it should be noted that any  $C_{60}$  molecule with 12 pentagonal and 20 hexagonal rings could be responsible. In particular, the following structure derived more closely from graphite should be considered. This structure,

(5) Haymet, A. D. J., Unpublished results.

(6) Allinger, N. L.; Flanagan, H. L. *J. Comput. Chem.* **1983**, *4*, 399.

(7) Kroto, H. W.; Heath, J. R.; O'Brien, S. C.; Curl, R. F.; Smalley, R. E. *Nature (London)*, submitted for publication.

(1) See also: Ternansky, R. J.; Balogh, D. W.; Paquette, L. A. *J. Am. Chem. Soc.* **1982**, *104*, 4503.

(2) See, for example: Karplus, M.; Porter, R. N. "Atoms and Molecules"; W. A. Benjamin: New York, 1970.

(3) Barth, W. E.; Lawton, R. C. *J. Am. Chem. Soc.* **1971**, *93*, 1730.

(4) Randic, M.; Trinajstić, N. *J. Am. Chem. Soc.* **1984**, *106*, 4428.

denoted for convenience "graphitene", consists of two sheets of seven hexagonal rings, six arranged symmetrically about one, linked by six pairs of carbon atoms connecting pairs of pentagonal rings. The remaining six hexagonal rings are found between the pairs of pentagons. This structure has a marginally lower Hückel stabilization energy of  $0.5418 \beta$  per carbon atom but is possibly destabilized by the extra strain of connected pentagonal rings. Vibrational analysis of  $C_{60}$  molecules will resolve the structure.

Due to the high symmetry of footballene, ab initio molecular orbital calculations can be used to calculate more accurately the properties of the molecule. More importantly, a synthetic route to footballene is eagerly awaited.

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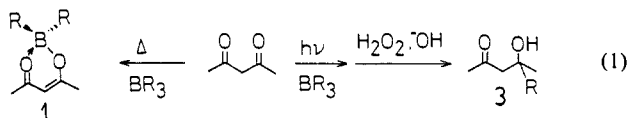
## Novel Photochemical Alkyl Migrations of Dialkylboryl Acetylacetonate Complexes

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Although much attention has been focused on the ground-state chemistry of organoboranes, relatively little is known about their excited states.<sup>1,2</sup> Hawthorne and Reintjes reported that thermal reactions of trialkylboranes with acetylacetonate yield dialkylboryl acetylacetonate complexes **1**.<sup>3,4</sup> Several years later, Nozaki and co-workers reported a sharp contrast between photochemical and thermal reactions of trialkylboranes with acetylacetonate (eq 1).<sup>5</sup>



In their study, photochemically induced conjugate addition<sup>2a</sup> of trialkylboranes to the enolized acetylacetonate was speculated to be responsible for the aldol formation. The participation of **1** in

(1) Notable examples of photoreactions of organoboranes: (a) Glogowsky, M. E.; Zumbulyadis, N.; Williams, J. L. R. *J. Organomet. Chem.* **1982**, *231*, 97; and a series of their papers. (b) Hancock, K. G.; Uriarte, A. K. *J. Am. Chem. Soc.* **1970**, *92*, 6374. (c) Hancock, K. G.; Dickinson, D. A. *J. Chem. Soc., Chem. Commun.* **1972**, 962. (d) Hancock, K. G.; Dickinson, D. A. *J. Am. Chem. Soc.* **1972**, *94*, 4396. (e) Hancock, K. G.; Dickinson, D. A. *Ibid.* **1973**, *95*, 280. (f) Hancock, K. G.; Kramer, J. D. *Ibid.* **1973**, *95*, 3425. (g) Hancock, K. G.; Uriarte, A. K.; Dickinson, D. A. *Ibid.* **1973**, *95*, 6980. (h) Trubini, L. J.; Poter, R. F. *Inorg. Chem.* **1975**, *14*, 1252. (i) Calhoun, G. C.; Schuster, G. B. *J. Org. Chem.* **1984**, *49*, 1925 and references cited therein. (j) Pachaly, B.; West, R. *Angew. Chem., Int. Ed. Engl.* **1984**, *23*, 454. (k) Pachaly, B.; West, R. *J. Am. Chem. Soc.* **1985**, *107*, 2987.

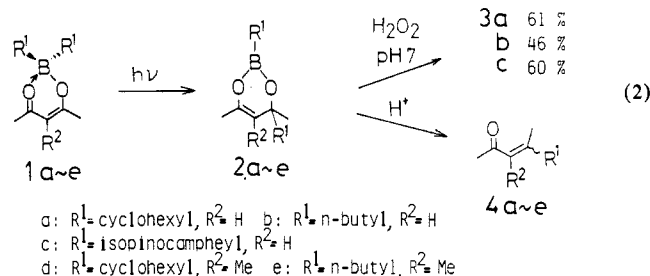
(2) Reactions of organoboranes with excited states of organic molecules: (a) Brown, H. C.; Kabalka, G. W. *J. Am. Chem. Soc.* **1970**, *92*, 714. (b) Miyamoto, N.; Isiyama, S.; Utimoto, K.; Nozaki, H. *Tetrahedron Lett.* **1971**, 4597. (c) Davies, A. G.; Griller, D.; Roberts, B. P.; Scaiano, J. C. *J. Chem. Soc., Chem. Commun.* **1971**, 196. (d) Scaiano, J. C.; Ingold, K. U. *Ibid.* **1975**, 878. (e) Lane, C. F.; Brown, H. C. *J. Am. Chem. Soc.* **1971**, *93*, 1025. (f) Yamamoto, Y.; Brown, H. C. *J. Chem. Soc., Chem. Commun.* **1973**, 801. (g) Brown, H. C.; De Lue, N. R. *Tetrahedron Lett.* **1977**, 3007.

(3) (a) Hawthorne, M. F.; Reintjes, M. *J. Am. Chem. Soc.* **1964**, *86*, 5016. (b) Hawthorne, M. F.; Reintjes, M. *J. Org. Chem.* **1965**, *30*, 3851.

(4) Other derivatives of this type of compound: (a) Roth, H. J.; Miller, B. *Arch. Pharm. (Weinheim, Ger.)* **1964**, *297*, 617. (b) Umland, F.; Schleyerbach, C. *Angew. Chem., Int. Ed. Engl.* **1965**, *4*, 432. (c) Balaban, A. T.; Rentea, C. N.; Mocanu-Paraschiv, M.; Romas, E. *Rev. Roum. Chim.* **1965**, *10*, 849. (d) Balaban, A. T.; Rentea, C. N.; Bacescu-Roman, M. *Ibid.* **1965**, *10*, 865. (e) Toporcer, L. H.; Dessy, R. E.; Green, S. I. E. *Inorg. Chem.* **1965**, *4*, 1649. (f) Köster, R.; Rotermund, G. W. *Liebigs Ann. Chem.* **1965**, *689*, 40. (g) Umland, F.; Hohaus, E.; Brodte, K. *Chem. Ber.* **1973**, *106*, 2427.

(5) Utimoto, K.; Tanaka, T.; Nozaki, H. *Tetrahedron Lett.* **1972**, 1167. The absence of alkane in the reaction mixture in Nozaki's study might suggest the occurrence of conjugate additions of trialkylboranes for the aldol formation. However, since the complex formation of acetylacetonate with trialkylboranes proceeds even at room temperature in a suitable concentration region and can be easily recognized by yellow color development, the borane complex **1** is at least one of the key intermediates of the photochemical reactions of acetylacetonate with trialkylboranes.

the photochemical reactions was not considered. From our interest in the borane complexes **1**, particularly in their unique absorption spectra (**1a**,  $\lambda_{\max}$  348 nm in  $C_6H_{12}$ ),<sup>6</sup> we have been led to study the photochemical reactions of **1**. We now report a new photoalkylation reaction where an alkyl group migrates from boron to the acetylacetonate carbonyl carbon of **1** (eq 2).



The borane complexes **1** are thermally stable and in fact heating a toluene solution of **1a** at 150 °C for 10 h in a sealed tube results in complete recovery of **1a**. However, when a benzene solution of **1a** is irradiated with a 500-W xenon lamp through a UV filter (>330 nm), the yellow color due to **1a** rapidly disappeared. Evaporation of the solvent and measurement of the <sup>1</sup>H NMR spectrum ( $CDCl_3$ ) of the residual transparent oil showed a narrow quartet (1 H) at 4.48, a narrow doublet (3 H) at 1.76, a singlet (3 H) at 1.20 ppm, and a multiplet due to cyclohexyl protons, suggesting the existence of an olefinic enol proton, a vinyl methyl group, and a methyl group on a saturated tertiary carbon, respectively. An IR spectrum of the oil showed an enol borate absorption ( $C=C$  stretching) at  $1695 \text{ cm}^{-1}$ .<sup>7</sup> These data and the following chemical reactions establish the structure of this oil as **2**. Oxidative hydrolysis with a solution of pH 7 phosphate buffer, MeOH, THF, and aqueous 30%  $H_2O_2$  (5:8:10:1 v/v) gave an aldol **3a** in 61% yield. Acidic hydrolysis gave the corresponding  $\alpha,\beta$ -unsaturated carbonyl compound **4a** in 60% yield.<sup>8</sup>

The reaction proceeded in moderate quantum yield ( $\Phi = 0.08$ ) and was not quenched by oxygen ( $2 \times 10^{-3} \text{ M}$ ) or piperylene (2.0 M), suggesting that the singlet is the reactive excited state. Product analysis of the volatile components obtained after distillation from the reaction mixtures of benzene-*d*<sub>6</sub> or decalin solutions revealed the formation of cyclohexane and cyclohexene<sup>9</sup> in a 6:1 ratio amounting to ca. 10% of the total yield. Bicyclohexyl and phenylcyclohexane, which are typical products from cyclohexyl radicals, were also detected by GC analysis as minor products (<1%).<sup>10</sup>

In order to clarify whether the reaction proceeds intramolecularly or intermolecularly, crossover experiments were performed using **1a** and **1e** (or **1b** and **1d**). Photolysis of an equimolar mixture of **1a** and **1e** in benzene followed by GC analysis of the hydrolyzed mixture of  $\alpha,\beta$ -unsaturated carbonyl compounds showed that the alkyl migration is essentially intramolecular (95%) with 5% of the crossed products **4b,d**: similar results were obtained for **1b,d**. The participation of radical species, indicated by the formation of the hydrocarbons and the production of small amounts of crossed products, suggests a radical pair mechanism (eq 3).

(6) The unusual spectrum is probably to be attributed to the red-shifted  $\pi-\pi^*$  absorption by the intramolecular coordination of the borane group to the carbonyl group. The fact that the absorption maximum is slightly red-shifted in nonpolar solvents (**1a**,  $\lambda_{\max}$  348 nm in  $C_6H_{12}$ , 345 nm in  $CH_2Cl_2$ , 344 nm in  $CH_3CN$ ) may suggest that the excited state has a smaller CT contribution than the ground state. Experiments on this unusual absorption including the substituent effect are in progress.

(7) For  $C=C$  stretching vibrations of enol borates, see, for instance: (a) Fenzl, W.; Kosfeld, H.; Koster, R. *Liebigs Ann. Chem.* **1976**, *1370*. (b) Koster, R.; Zimmermann, H.-J.; Fenzl, W. *Ibid.* **1976**, 1116.

(8)  $\alpha,\beta$ -Unsaturated carbonyl compounds were obtained as a mixture of cis and trans isomers (1:8 for **4a**, 1:1 for **4b**).

(9) (a) Gordon, A. S.; Smith, S. R. *J. Chem. Phys.* **1961**, *34*, 331. (b) Livant, P.; Lawer, R. G. *J. Am. Chem. Soc.* **1976**, *98*, 6044. (c) Bennett, J. E.; Gale, L. H.; Hayward, E. J.; Mile, B. *J. Chem. Soc., Faraday Trans. 1* **1973**, *69*, 1655.

(10) (a) Hermans, P.; Van Eyk, J. *J. Polym. Sci.* **1946**, *1*, 407. (b) Walling, C.; Gibian, M. *J. Am. Chem. Soc.* **1965**, *87*, 3361. (c) Shelton, J. R.; Uzelmeier, C. W. *Ibid.* **1966**, *88*, 5222.