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Synthesis and Mass Spectra of Some Hexa-, Hepta- and Octadeuterated Derivatives of Propane

BY FRANCIS E. CONDON

As part of a study of spectra and molecular structure, aimed at augmenting the usefulness of spectrometric methods of analysis, propane-1,1,1,3,3,3-d_6 and propane-d_8 were synthesized. The mass spectra of these compounds, and of acetone-d_6, propane-1,1,1,3,3,3-d_6-2-ol, propane-d_7-2-ol, 2-chloropropane-1,1,1,3,3,3-d_6 and 2-chloropropane-d_7, intermediates in the syntheses, are presented in comparison with the mass spectra of propane, acetone, 2-propanol and 2-chloropropane.

In a continuation of a study of spectra and molecular structure,[1] infrared and mass spectra have been obtained for propane-1,1,1,3,3,3-d_6 and propane-d_8, and for acetone-d_6, propane-1,1,1,3,3,3-d_6-2-ol, propane-d_7-2-ol, 2-chloropropane-1,1,1,3,3,3-d_6 and 2-chloropropane-d_7, intermediates in the syntheses of propane-1,1,1,3,3,3-d_6 and propane-d_8 from acetone by the reactions

$$CH_3COCH_3 \xrightarrow{\substack{\text{Excess } D_2O \\ K_2CO_3 \text{ catalyst}}} \underset{\text{I}}{CD_3COCD_3} \tag{1}$$

$$(\text{I}) \xrightarrow{\substack{\text{1. } LiAlH_4\text{, ether} \\ \text{2. } H_2O\text{, HCl}}} \underset{\text{IIa}}{CD_3CHOHCD_3} \tag{2a}$$

$$(\text{I}) \xrightarrow{\substack{\text{1. } LiAlD_4\text{, ether} \\ \text{2. } H_2O\text{, HCl}}} \underset{\text{IIb}}{CD_3CDOHCD_3} \tag{2b}$$

$$(\text{IIa}) \xrightarrow{\substack{ZnCl_2\text{, concd. HCl} \\ \text{mixture distilled}}} \underset{\text{IIIa}}{CD_3CHClCD_3} \tag{3a}$$

$$(\text{IIb}) \xrightarrow{\substack{ZnCl_2\text{, concd. HCl} \\ \text{mixture distilled}}} \underset{\text{IIIb}}{CD_3CDClCD_3} \tag{3b}$$

$$(\text{IIIa}) \xrightarrow{\substack{\text{1. Mg, ether} \\ \text{2. } H_2O}} \underset{\text{IVa}}{CD_3CH_2CD_3} \tag{4a}$$

$$(\text{IIIb}) \xrightarrow{\substack{\text{1. Mg, ether} \\ \text{2. } D_2O}} \underset{\text{IVb}}{CD_3CD_2CD_3} \tag{4b}$$

The syntheses and mass spectra are described here. The infrared spectra are presented elsewhere.[2]

Experimental Part

Acetone-d_6 was prepared from 0.66 mole of acetone by 21 successive D–H exchanges with about 0.6 mole of 99.8% deuterium oxide in the presence of about 0.1 g. of anhydrous potassium carbonate.[3] Each exchange was carried out in a 100-cc. kettle attached to a column, 30 × 0.7 cm., with Heligrid packing. The acetone-d_x and a small amount of the equilibrated water were distilled to a cut point about 95° and were collected in another 100-cc. flask, which became the kettle–reaction flask for the next exchange. The final product, obtained in 78% yield, gave a mass spectrum with the following C^{13}-corrected ion-intensity ratios: $63/(63 + 64) = 0.041$, $(2 \times 45)/(45 + 46) = 0.041$, and $(2 \times 17)/(17 + 18) = 0.041$, indicating 4.1% of acetone-d_5 and a $D/(D + H)$ content of 99.3%. This mass spectrum is not presented in full because it was not completely comparable with a mass spectrum of acetone obtained at the same time, apparently because of erratic behavior of the instrument. Found for acetone-d_6: $n^{20}D$ 1.3559; d^{20} 0.8749 g./cc.; molecular refractivity 16.00. Found for acetone: $n^{20}D$ 1.3589; d^{20} 0.7915 g./cc.; molecular refractivity 16.15. Calculated[4] for acetone: molecular refractivity 16.02.

Propane-1,1,1,3,3,3-d_6-2-ol was prepared by reduction of 0.2 mole of acetone-d_6 with about 0.1 mole of lithium aluminum hydride in about 50 ml. of ether[5]; and **propane-d_7-2-ol** was prepared by reduction of 0.3 mole of acetone-d_6 with about 0.1 mole of lithium aluminum deuteride ("94%")[6] in about 75 ml. of ether. Each reaction product was treated with about 75 ml. of water containing about 0.4 mole of hydrochloric acid. Each ether layer (dried with Drierite) and each water layer was distilled separately through a 30 × 0.7-cm. column with Heligrid packing. A heart cut from the ether layer, boiling at 80°, was used as a spectrometric sample. Material from the water layer distilled for the most part at 79°, presumably as an azeotrope corresponding to the known azeotrope (b.p. 80.38°) of 2-propanol (b.p. 82.44°) with water.[7] The yields were quantitative. Found for propane-1,1,1,3,3,3-d_6-2-ol (sample containing 10.45 weight per cent. water by Karl Fischer reagent): $n^{20}D$ 1.3753; d^{20} 0.8890 g./cc. Found for propane-d_7-2-ol (sample containing 7.11 weight per cent. water by Karl Fischer reagent): $n^{20}D$ 1.3750; d^{20} 0.9006 g./cc. Found for 2-propanol (sample containing 0.64 weight per cent. water by Karl Fischer reagent): $n^{20}D$ 1.3770; d^{20} 0.7874 g./cc. *Anal.* Calcd. for $C_3H_2D_6O$: C, 54.47. Found for propane-1,1,1,3,3,3-d_6-2-ol: C, 53.83, 53.67. Calcd. for C_3HD_7O: C, 53.66. Found for propane-d_7-2-ol: C, 53.92, 54.13.

2-Chloropropane-1,1,1,3,3,3-d_6 was prepared from 0.2 mole of propane-1,1,1,3,3,3-d_6-2-ol, 0.7 mole of concentrated hydrochloric acid and 1 mole of zinc chloride; and **2-chloropropane-d_7** was prepared from 0.3 mole of propane-d_7-2-ol, 1 mole of concentrated hydrochloric acid and 1.5 mole of zinc chloride. Each chloride was slowly distilled from the reaction mixture, caught in an ice-cooled receiver, washed with ice-water, dried with anhydrous potassium carbonate, and distilled through a 30 × 0.7-cm. column with Heligrid packing; material boiling 33–35° was obtained in about 70% yield; and a heart cut boiling at 34° was taken as a spectrometric sample. 2-Chloropropane, similarly distilled, boiled at 35.6°. Found for 2-chloropropane-1,1,1,3,3,3-d_6: $n^{20}D$ 1.3744; d^{20} 0.9247; molecular refractivity 20.90. Found for 2-chloropropane-d_7: $n^{20}D$ 1.3748; d^{20} 0.9386; molecular refractivity 20.86. Found for 2-chloropropane: $n^{20}D$ 1.3774; d^{20} 0.8621; molecular refractivity 20.97. Calculated[4] for 2-chloropropane: molecular refractivity 20.86.

Propane-1,1,1,3,3,3-d_6 was prepared from the Grignard reagent prepared from 0.13 mole of 2-chloropropane-1,1,1,3,3,3-d_6, 0.13 g. atom of magnesium and about 150 ml. of ether. After preparation of the Grignard reagent, the solution was refluxed for about one hour in order to eliminate by-product hydrocarbons (see below). Excess water was

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[1] (a) H. L. McMurry, V. Thornton and F. E. Condon, *J. Chem. Phys.*, **17**, 918 (1949); (b) H. L. McMurry and V. Thornton, *ibid.*, **18**, 1515 (1950); (c) F. E. Condon, H. L. McMurry and V. Thornton, unpublished work.

[2] H. L. McMurry and V. Thornton, in preparation.

[3] J. O. Halford, L. C. Anderson, J. R. Bates and R. D. Swisher, THIS JOURNAL, **57**, 1663 (1935).

[4] Bond refractivities given by K. G. Denbigh, *Trans. Faraday Soc.*, **36**, 936 (1940), were used.

[5] R. F. Nystrom and W. G. Brown, THIS JOURNAL, **69**, 1197 (1947).

[6] According to the supplier, Metal Hydrides, Inc., Beverly, Mass.: "Contains AlD_3 etherate as principal impurity, plus 1 per cent. Cl. Active H varies from zero to a few thousandths of a per cent."

[7] L. H. Horsley, *Ind. Eng. Chem., Anal. Ed.*, **19**, 508 (1947).

TABLE I

MASS SPECTRA (ION INTENSITIES)[a]

M/e	Propane	Propane-1,1,1,-3,3,3-d_6	Propane-d_8	Acetone	Acetone-d_6	2-Propanol[b]	Propane-1,1,1,-3,3,3-d_6-2-ol	Propane-d_7-2-ol	2-Chloropropane	2-Chloropropane-1,1,1-3,3,3-d_6	2-Chloropropane-d_7
14	7	2	2	36	11	17	3	3	4	..	2
15	27	5	1	242	2	57	7	7	12	..	2
16	..	6	6	4	42	3	20	17	..	4	6
17	..	6	1	..	9	3	27	17	..	4	2
18	..	13	20	1	290	7	57	60	4	14	18
19–24[c]	..	25	19	9	13	87	93	87	6	..	4
25	5	2	..	11	..	3	..	..	4	..	..
26	60	9	5	45	8	20	7	7	28	8	4
27	410	41	3	68	1	160	20	27	346	32	4
28	785	114	71	52	72	47	80	83	30	52	42
29	1255	273	24	44	3	90	170	70	12	212	26
30	28	610	409	2	75	7	147	283	4	166	386
31	1	326	71	8	2	80	70	97	..	20	16
32	..	1083	913	1	14	3	20	10	4	10	12
33	..	82	128	..	1	..	76	63	..	2	8
34	..	54	1310	..	22	..	17	17	..	..	4
35	..	1	29	..	..	..	..	..	6	6	6
36	6	3	3	7	4	..	3	3	36	20	24
37	38	4	1	23	..	17	3	..	32	8	8
38	61	21	17	27	19	23	10	10	48	22	22
39	199	16	3	49	2	77	7	3	120	12	4
40	33	37	32	13	26	17	20	20	28	28	28
41	191	58	9	27	2	117	30	13	246	38	6
42	79	83	113	97	60	67	63	80	80	74	92
43	463	22	7	1540	9	277	23	20	1280	34	8
44	548	43	17	38	81	80	57	40	46	46	24
45	18	97	12	4	45	1777	157	103	6	166	28
46		80	106	..	1575	60	263	253	2	128	259
47		70	13	..	38	3	93	66	2	82	32
48		108	53	..	9	..	2050	107	2	158	82
49		514	33	..	..	..	323	2183	16	1180	154
50		438	228	..	..	..	30	210	2	50	1340
51		51	63	..	..	..	3	17	4	10	52
52		8	492	..	..	..	..	..	..	2	4
53			15	..	..	3	..	..	..	2	4
54–57[c]				23	..	27	23	27	..	..	..
58				610	6	23	3	3	..	..	..
59				22	..	83	17	47	..	4	2
60					..	13	3	3	6	2	..
61					..		3	3	12	4	4
62					19		..	..	38	4	..
63					33		..	..	352	18	10
64					580		13	10	20	50	6
65					19		123	67	116	38	32
66					3		20	3	4	240	24
67							3	13	2	16	264
68									2	76	14
69									..	..	86
70									..	..	2
73–77[c]									14	2	..
78									350	6	..
79									14	6	2
80									114	6	2
81									4	10	4
82										14	6
83										46	12
84										274	34
85										26	288
86										90	20
87										4	94
88											4
43[d]	2025	2025	2025	1800	1800	1560	1560	1560	1850	1850	1850

[a] Obtained on a Westinghouse Type LV mass spectrometer with 75 volt electrons in the ionization chamber, an ion accelerating potential of about 550 volts, a catcher current of 6.5 microamps, and with the ionization chamber at about 210°. The observed values, corrected for background, are presented to the nearest whole number, and are adjusted to a sample pressure of 100 mm. for all compounds. [b] Corrected for 3.7% acetone found by infrared spectrophotometry.[1c] [c] Sum of intensities of all ions with these masses. [d] From the mass spectrum of *n*-butane obtained under the same conditions.

then added slowly and the generated propane-1,1,1,3,3,3-d_6, together with much entrained ether, was caught in a Dry Ice-cooled trap. The trapped material was distilled in a Podbielniak Heligrid low-temperature fractionating column to separate a C_3-fraction from the entrained ether, and the C_3-fraction was redistilled through the column with rejection of a forerun and a residue. The thus purified propane-1,1,1,3,3,3-d_6, obtained in 70% yield, distilled from −45° at 750 mm. to −58.5° at 370 mm.; and a heart cut, separately collected, distilled at −45.3° at 750 mm.

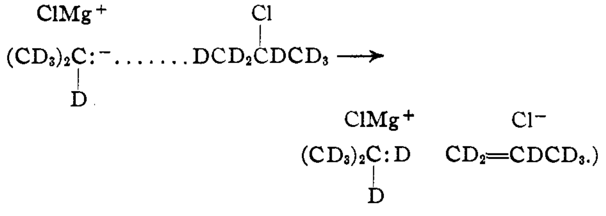
Propane-d_8 was similarly prepared from the Grignard reagent prepared from 0.19 mole of 2-chloropropane-d_7, 0.19 g. of atom of magnesium and about 150 ml. of ether, and excess deuterium oxide (0.3 mole 99.8% pure, was added at first and was followed by 0.2 mole, presumably 99.3% pure, from the final D–H exchange in the preparation of acetone-d_6). The purified propane-d_8, obtained in 77% yield, distilled from −46.0° at 750 mm. to −51° at 560 mm.; and a heart cut distilling at −51.2° at 560 mm. was collected separately.

Additional vapor–liquid equilibrium data are presented below in comparison with those for propane:

Compound	B.p., °K.	Pressure, mm.	Vapor pressure at 243.3° K.,[a] mm.	Approx. heat of vaporization, *L*, at 231°K., kcal./mole[b]
Propane	231.1[c]	760	878	4.8(4.5)[c]
Propane-2,2-d_2[1c]	229.2	748	903	3.9
Propane-1,1,1,3,3,3-d_6	227.9	750	932	3.6
Propane-d_8	227.2	750	943	3.4

[a] Melting mercury-bath. [b] Calculated from tabulated data by means of the formula, $L = 2.303RTT' \log (P/P')/(T - T')$, which assumes ideal gas law behavior for the vapor. [c] Literature value.

The by-product hydrocarbons produced during preparation of the Grignard reagents from 2-chloropropane-1,1,1,3,3,3-d_6 and 2-chloropropane-d_7, and eliminated by refluxing the ether solutions of the Grignard reagents, were caught in a Dry Ice-cooled trap and were distilled in a Podbielniak low-temperature fractionating column in order to separate a C_3-fraction from entrained ether. The C_3-material from 2-chloropropane-1,1,1,3,3,3-d_6 (0.004 mole) was 47% absorbed by silver nitrate–mercuric nitrate absorbent; and the unabsorbed portion gave a mass spectrum that indicated 13% propane-d_7, 60% propane-d_6, and, apparently, lower propane-d_x homologs. The C_3-material from 2-chloropropane-d_7 (0.005 mole) was 44% absorbed by silver nitrate–mercuric nitrate absorbent; and the unabsorbed portion gave a mass spectrum that indicated 28% propane-d_8, 40% propane-d_7 and, apparently, lower propane-d_x homologs. The mass spectral data indicate that the C_3-hydrocarbons were produced from the alkyl chlorides in part by an intermolecular process, for example[8]



$$\overset{\text{ClMg}^+}{\underset{\text{D}}{(CD_3)_2C}}:^- \ldots\ldots \text{DCD}_2\overset{\text{Cl}}{\text{C}}\text{DCD}_3 \longrightarrow \overset{\text{ClMg}^+}{\underset{\text{D}}{(CD_3)_2C}}:\text{D} \quad \overset{\text{Cl}^-}{\text{CD}_2{=}\text{CDCD}_3.)}$$

Discussion of Mass Spectra

The mass spectra of the synthesized propane-1,1,1,3,3,3-d_6, propane-d_8, acetone-d_6, propane-1,1,1,3,3,3-d_6-2-ol, propane-d_7-2-ol, 2-chloropropane-1,1,1,3,3,3-d_6 and 2-chloropropane-d_7 are presented in Table I together with similarly obtained mass spectra of propane, acetone, 2-propanol and 2-chloropropane. In the discussion to follow, it is supposed that the intensities of the parent peaks in the mass spectra of molecules differing only by isotopic substitution will be identical.

After correction for C^{13}-containing ions, the intensity of the parent peak in the mass spectrum of propane-d_8 in Table I is 91.6% of the intensity of the parent peak in the mass spectrum of propane; and the relative intensities of the mass 52 and 51 peaks indicate 10.3% propane-d_7 in propane-d_8. The intensities of all the odd-numbered peaks in the mass spectrum of the propane-d_8 are satisfactorily accounted for by about 10% of propane-d_7 (and C^{13}-containing ions).

The mass spectrum of propane-d_8, approximately corrected for C^{13}-containing ions and for 10% of propane-d_7 (simply by dividing the C^{13}-corrected intensities by 0.9),[9] is presented in Table II together with the mass spectrum of propane corrected for C^{13}-containing ions.

TABLE II

CALCULATED PARTIAL MASS SPECTRA OF ISOTOPICALLY PURE PROPANE AND PROPANE-d_8[a]

Ion	Intensity X = H	Intensity X = D	Ion	Intensity X = H	Intensity X = D
C_2X^+	5	6	$C_3X_2^+$	62	37
$C_2X_2^+$	61	80	$C_3X_3^+$	204	129
$C_2X_3^+$	418	465	$C_3X_4^+$	27	20
$C_2X_4^+$	794	1034	$C_3X_5^+$	196	121
$C_2X_5^+$	1265	1485	$C_3X_6^+$	76	61
C_3^+	6	3	$C_3X_7^+$	475	260
C_3X^+	39	19	$C_3X_8^+$	551	560

[a] Ion intensities of Table I corrected for contributions by propane-d_7 and C^{13}-containing ions.

The intensities of ions produced by loss of one or more deuterium atoms and an electron from propane-d_8 are 0.5–0.8 times the intensities of corresponding ions from propane, in qualitative accordance with previous knowledge that removal of deuterium is about one-half as likely as removal of hydrogen.[10] In spite of the lesser ease of removal of deuterium, the intensities of the ions produced from propane-d_8 by loss of a CD_3 group and an electron, with or without loss of one or more deuterium atoms, are 1.1–1.3 times the intensities of the corresponding ions from propane, analogous to previous knowledge that removal of hydrogen attached to carbon bearing deuterium is about 1.2 times as likely as removal of hydrogen attached to carbon bearing only hydrogen.[10b,c]

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[8] *Cf.* H. Gilman and R. E. Fothergill, THIS JOURNAL, **50**, 3334 (1928); H. Gilman and E. A. Zoellner, *ibid.*, **53**, 1583 (1931); F. C. Whitmore and D. E. Badertscher, *ibid.*, **55**, 1559 (1933); A. A. Morton, J. B. Davidson and B. L. Hakan, *ibid.*, **64**, 2242 (1942); W. V. Evans and R. Pearson, *ibid.*, **64**, 2865 (1942).

[9] This approximation gives ion intensities too high by amounts equal to 11% of the intensities of ions of corresponding masses in the spectrum of pure propane-d_7. However, even-numbered masses in the spectrum of propane-d_7 would be expected to be relatively small, because of the low probability of loss of the lone hydrogen.

[10] (a) J. M. Delfosse and J. A. Hipple, Jr., *Phys. Rev.*, **54**, 1060 (1938); (b) M. W. Evans, N. Bauer and J. Y. Beach, *J. Chem. Phys.*, **14**, 701 (1746); (c) J. Turkevich, L. Friedman, E. Solomon and F. M. Wrightson, THIS JOURNAL, **70**, 2638 (1948).

After correction for C^{13}-containing ions, the intensity of the parent peak in the mass spectrum of propane-1,1,1,3,3,3-d_6 in Table I is 79% of the intensity of the parent peak in the mass spectrum of propane. Thus, since the propane-1,1,1,3,3,3-d_6 apparently was only about 79% pure, its spectrum is not much help for interpreting the mass spectrum of propane. Noteworthy, however, are the mass 33 and 34 peaks, conceivably from CD_3CDH^+, $CD_2HCD_2^+$ and $CD_3CD_2^+$ ions produced by rearrangement within the mass spectrometer.[11] (Rearrangement during the synthesis appears to be ruled out by the lack of rearrangement during the synthesis of propane-2,2-d_2 by a similar route.[1])

An augmented ease of removal of a secondary hydrogen atom and an electron from propane-1,1,1,3,3,3-d_6 is inferred from the intensity of the mass 49 peak in its spectrum in Table I. From data in the table, on the basis that electron impact removes a secondary hydrogen atom and an electron from propane 13 times as readily as a primary hydrogen atom and an electron,[1c] assuming 21% of propane-d_5 in the propane-1,1,1,3,3,3-d_6, and neglecting the small correction for C^{13}, it was calculated that electron impact removes a secondary hydrogen atom and an electron from propane-1,1,1,3,3,3-d_6 $\frac{514 - (0.21 \times 548)}{0.79} \div 2 \div \frac{463}{(2 \times 13) + 6} = 17$ times as readily as a primary hydrogen atom and an electron from propane.

The fact that the propane-d_8 and propane-1,1,1,3,3,3-d_6 were not as pure as was anticipated from the purity of the acetone-d_6 from which they were made (see Experimental part) is believed to be due chiefly to some replacement of primary deuterium by hydrogen during conversion of the alcohols to chlorides by means of concentrated hydrochloric acid and zinc chloride. Such replacement of primary, but not of secondary,[1c] deuterium is analogous to the formation of 2-methylpropane from 2-methylpropane-1-*d*, but not from 2-methylpropane-2-*d*, and sulfuric acid.[12]

The mass spectrum of acetone-d_6 in Table I yields the following ion intensity ratios: $63/(63 + 64) = 0.054$; $(2 \times 45)/(45 + 46) = 0.056$; and $(2 \times 17)/(17 + 18) = 0.060$, indicating about 5.7% of acetone-d_5. After corrections for the effects of C^{13} and of 5.7% of acetone-d_5, the intensity of the $C_2D_3O^+$ ion is 1.09 times the intensity of the $C_2H_3O^+$ ion in the spectrum of acetone, and the intensity of the CD_3^+ ion is 1.26 times the intensity of the CH_3^+ ion, further exemplifying the likelier breakage of deuterium-loaded carbon–carbon bonds which was manifest in comparing the spectra of propane-d_8 and propane.

The parent-ion intensities in the mass spectra of 2-propanol, propane-1,1,1,3,3,3-d_6-2-ol, and propane-d_7-2-ol are too small for use in estimating the purities of the deuterium-containing substances. The intensities of the ions produced by loss of CD_3 and an electron are 1.3–1.4 times the intensity of the ions produced by loss of CH_3 and an electron, indicating that, for 2-propanol also, deuterium loading disposes the carbon–carbon bonds to dissociation by electron impact. The spectra indicate that, in forming the $C_3X_7O^+$ ion (X = H or D), the hydrogen (deuterium) attached to the hydroxyl-bearing carbon is removed almost solely.

After corrections for C^{13}-containing ions, a comparison of the intensities of the mass 83 and 85 peaks in the spectrum of 2-chloropropane-1,1,1,3,3,3-d_6 with the intensities of the mass 84 and 86 peaks, respectively, indicates 17–18% of 2-chloropropane-d_5; and comparison of the intensities of the mass 84 and 86 peaks in the spectrum of 2-chloropropane-d_7 with the intensities of the mass 85 and 87 peaks, respectively, indicates 10–11% of 2-chloropropane-d_6. These amounts of impurities are in concordance with the amounts estimated in the propane-1,1,1,3,3,3-d_6 and propane-d_8 prepared from these chlorides and support the view that some H–D exchange occurred during formation of the chlorides from the corresponding alcohols.

The intensities of the ions produced by loss of CD_3 and an electron from 2-chloropropane-1,1,1,-3,3,3-d_6 and 2-chloropropane-d_7 are (after correction for the effects of impurities) about 0.8–0.9 times the intensity of the ion produced by loss of CH_3 and an electron from 2-chloropropane. Apparently the chlorine atom causes deuterium loading to decrease, rather than to increase, the dissociation of carbon–carbon bonds by electron impact. When the chlorine is removed, however, as in formation of the $C_2X_3^+$ ion, deuterium-loaded carbon–carbon bonds are broken about 1.3 times as readily as hydrogen-loaded ones. Removal of the chlorine atom and an electron is about 1.15 times as likely for the deuterium-containing molecules as for 2-chloropropane. A similar ratio was observed for 2-chloropropane-2-d.[1c]

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[11] R. E. Honig, Paper presented before the Meeting of the Am. Phys. Soc., New York, N. Y., January, 1949.

[12] O. Beeck, J. W. Otvos, D. P. Stevenson and C. D. Wagner, *J. Chem. Phys.*, **17**, 418 (1949).