II.— β -Aldehydopropionic Acid, $\text{CHO}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{COOH}$,
and β -Aldehydoisobutyric Acid, $\text{CHO}\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)\cdot\text{COOH}$.

By W. H. PERKIN, jun., and C. H. G. SPRANKLING.

SOME time since (Trans., 1896, 69, 162), a paper was published by one of us in conjunction with Messrs. W. H. Bentley and E. Haworth which had for one of its objects the discovery of a method for introducing the group $\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{OH}$ into organic substances, a synthetical process which, if it could be easily carried out, would be very valuable as a means of forming ring compounds. It was found that the action of glycol chlorhydrin, $\text{Cl}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{OH}$, on the sodium compounds of substances like ethylic acetoacetate, ethylic malonate, and their derivatives, did not, except in isolated cases, yield the desired result; ultimately, however, a method was devised which in the few cases tried gave fairly satisfactory results, and which may be briefly stated in the form of an example thus.

β -Bromethyl phenyl ether, $\text{C}_6\text{H}_5\cdot\text{O}\cdot\text{CH}_2\cdot\text{CH}_2\text{Br}$, was prepared by acting on sodium phenoxide with ethylene bromide, and this, when digested with the sodium derivative of ethylic methylmalonate, yielded ethylic γ -phenoxyethylmethylmalonate,

The acid corresponding to this ethereal salt, when heated at 100° , loses

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one molecule of carbon dioxide with formation of γ -phenoxyethyl-methylacetic acid, $\text{C}_6\text{H}_5\cdot\text{O}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)\cdot\text{COOH}$, from which hydrobromic acid eliminates the phenyl group and forms γ -bromethyl-methylacetic acid, $\text{CH}_2\text{Br}\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)\cdot\text{COOH}$. This brom-acid when boiled with sodium carbonate, gives the sodium salt of hydroxyethyl-methylacetic acid, $\text{OH}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)\cdot\text{COONa}$, from which, on acidifying, methylbutyrolactone $\begin{array}{c} \text{CH}_2\cdot\text{CH}_2\cdot\text{CH}\cdot\text{CH}_3 \\ | \quad \quad | \\ \text{O} \quad \quad \text{CO} \end{array}$, is at once

obtained. When this method was tried in more complicated synthetical experiments, it did not work well, partly owing to the number of operations involved, but principally on account of the smallness of the yield obtained in some of these operations. For these reasons, experiments were made with a view to discover a more direct method, and ultimately we found in *bromacetal*, $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\text{Br}$, a substance which seems likely to answer the purpose.

Bromacetal reacts readily with the sodium derivative of ethylic malonate, yielding *ethylic acetalmalonate* according to the equation

This ethylic salt, which distils without decomposition at $151\text{--}154^\circ$ (15 mm.), yields, on hydrolysis, the corresponding *acetulmalonic acid*, $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{COOH})_2$, and this, when heated with water at 180° is decomposed into alcohol, carbon dioxide, and β -aldehydopropionic acid, $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{COOH})_2 + \text{H}_2\text{O} = \text{CHO}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{COOH} + 2\text{C}_2\text{H}_5\cdot\text{OH} + \text{CO}_2$. β -Aldehydopropionic acid is a new and very interesting substance, since it is the "half-aldehyde" of succinic acid and belongs to the class of aldehyde acids of which, so far, very few have been prepared. Its properties show that it is a true aldehyde, and not a hydroxymethylene compound of the formula

it therefore, does not belong to the class of substances which Claisen has investigated with such brilliant results. Aldehydopropionic acid is an almost colourless liquid which reduces Fehling's solution and gives a violet coloration with a solution of rosaniline hydrochloride decolorised by sulphurous acid. It is slowly oxidised in contact with air, rapidly by nitric acid, with formation of succinic acid, and when reduced with sodium amalgam it yields *butyrolactone*,

When boiled with caustic soda solution in a flat basin, aldehydopropionic acid undergoes a most interesting change, yielding small quantities of *terephthalic acid*, the dihydroterephthalic acid, which may be assumed to be the first product of the condensation, being oxidised to tere-

phthalic acid by the action of the air,

and this is, so far, the only experiment which has been instituted with the object of testing the value of aldehydopropionic acid in condensation experiments.

The action of bromacetal on sodium compounds is probably a general one, since we have found that the reaction proceeds equally well when the sodium derivative of ethylic methylmalonate is substituted for that of ethylic malonate in the above experiments.

The *ethylic acetalmethylmalonate*,

thus obtained yields *acetalmethylmalonic acid*,

on hydrolysis, and this, when heated with water at 180° , is converted into β -aldehydoisobutyric acid, $\text{CHO} \cdot \text{CH}_2 \cdot \text{CH}(\text{CH}_3) \cdot \text{COOH}$, a liquid acid which, on oxidation, yields methyl succinic acid,

Further experiments on the action of bromacetal on the sodium compounds of ethereal salts are in progress. It should be mentioned, in conclusion, that C. Harries (*Ber.*, 1898, 31, 42) obtained a substance which is probably the methylal of the half-aldehyde of succinic acid, $(\text{CH}_3\text{O})_2\text{CH} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{COOH}$, by the action of sodium hypobromite on levulin methylal, $(\text{CH}_3\text{O})_2\text{CH} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{CO} \cdot \text{CH}_3$, but he does not appear to have further investigated this substance.

Action of Bromacetal on the Sodium Derivative of Ethylic malonate.

Formation of Ethylic Acetalmalonate, $(\text{C}_2\text{H}_5\text{O})_2\text{CH} \cdot \text{CH}_2 \cdot \text{CH}(\text{COOC}_2\text{H}_5)_2$.

Ethylic acetalmalonate is conveniently prepared as follows. Sodium (14.2 grams) is dissolved in absolute alcohol (170 grams) and the solution, while still warm, is mixed with ethylic malonate (100 grams) and bromacetal (80 grams) and the mixture, inclosed in sealed tubes, is heated at 130 — 140° for 4 hours; when as much alcohol as possible has been removed from the product by distillation on the water bath, water is added to the residue and the precipitated oil extracted several times with ether. The ethereal solution is washed, well dried over calcium chloride, the ether distilled off, and the residual oil fractionated under reduced pressure (15 mm.). More than half passes over below 145° and consists of a mixture of unchanged ethylic malonate and bromacetal, whilst the fraction distilling between 145° and 165°

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contains the ethylic acetalmonate. The oil boiling below 145° (15 mm.), and which was assumed to contain about 50 per cent. of bromacetal, was again heated in sealed tubes with the calculated quantity of the sodium derivative of ethylic malonate, the temperature, however, being now allowed to rise to 160° .

In this way, practically the whole of the bromacetal was converted into crude ethylic acetalmonate boiling at 145 — 165° (15 mm.), and from this fraction the almost pure ethereal salt could be obtained by repeated fractionation as a colourless oil of a peculiar and not unpleasant odour, and boiling at 151 — 154° (15 mm.), or at 166 — 168° (at 26 mm.). Analysis.*

0.1821 gave 0.3714 CO_2 and 0.1418 H_2O . C = 55.62; H = 8.65.

0.1640 „ 0.3360 CO_2 „ 0.1282 H_2O . C = 55.87; H = 8.67.

$(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{COOC}_2\text{H}_5)_2$ requires C = 56.50; H = 8.69 per cent.

Acetalmalonic Acid, $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{COOH})_2$.

Acetalmalonic acid is obtained by hydrolysing its ethereal salt with alcoholic potash, but the operation has to be carefully performed, since prolonged boiling with the alkali decomposes the acid with elimination of the group $\cdot\text{CH}_2\cdot\text{CH}(\text{OC}_2\text{H}_5)$, and formation of malonic acid. It is not clear how this decomposition takes place, but there can be no doubt as to the formation of malonic acid, since, in one instance, when a quantity of this acid was obtained melting at 131° , an analysis was carried out with the following results.

0.2687 gave 0.3480 CO_2 and 0.0972 H_2O . C = 35.32; H = 4.26.

$\text{CH}_2(\text{COOH})_2$ requires C = 34.63; H = 3.86 per cent.

It was very soluble in water, and when heated decomposed with evolution of carbon dioxide and formation of acetic acid.

If, however, the action of the potash is only allowed to proceed for a short time, hydrolysis takes place normally, and a good yield of acetalmalonic acid is obtained. After many experiments, we found that the following process gave the best results. Ten grams of the pure ethereal salt is mixed with a solution of 6 grams of pure potash in alcohol, and the mixture heated on the water-bath for 10 minutes, the alcohol is then rapidly driven off on the water-bath and the cold residue mixed with an excess of dilute sulphuric acid and extracted repeatedly with ether. The ethereal solution, after drying and evaporating, deposits a thick oil which shows no sign of crystallising, even after standing for some days over sulphuric acid in a vacuum. On

* The numbers obtained are slightly low, on account of the oil containing traces of bromine, which it was found impossible to remove by fractionation.

analysis,* it gave numbers agreeing approximately with those required for acetalmalonic acid.

- I. 0.1150 gave 0.2040 CO_2 and 0.0722 H_2O . C = 49.15 ; H = 6.97.
 II. 0.1779 „ 0.3177 CO_2 „ 0.1200 H_2O . C = 48.72 ; H = 7.47.
 III. 0.1974 „ 0.3529 CO_2 „ 0.1227 H_2O . C = 48.75 ; H = 6.91.
 IV. 0.1537 „ 0.2719 CO_2 „ 0.0915 H_2O . C = 48.26 ; H = 6.61.
 $(\text{COOH})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{OC}_2\text{H}_5)_2$ requires C = 49.05 ; H = 7.27 per cent.

Silver salt.—Acetalmalonic acid is very soluble in water, and if the aqueous solution is neutralised with ammonia, and silver nitrate added, a dense, white precipitate of the silver salt is precipitated, which, after washing first with water and then with alcohol and ether, gave the following results on analysis.

- 0.2177 gave, on ignition, 0.1092 Ag. Ag = 50.16.
 $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{COOAg})_2$ requires Ag = 49.79 per cent.

Distillation of Acetalmalonic Acid.—When this acid is heated in a fractionating flask, decomposition soon sets in with evolution of carbon dioxide, and the residue, after repeatedly fractionating under reduced pressure, yields a liquid which boils, apparently constantly, at $157\text{--}161^\circ$ (15 mm.), and which was at first thought to be acetalacetic acid $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{COOH}$. The analyses, however, show that this substance consists, for the most part, of aldehydopropionic acid (p. 16), elimination of alcohol having taken place during the distillation owing, probably, to the unavoidable presence of small quantities of water.

- 0.2658 gave 0.4740 CO_2 and 0.1607 H_2O . C = 48.63 ; H = 6.71.
 0.2514 „ 0.4372 CO_2 „ 0.1437 H_2O . C = 48.53 ; H = 6.39.
 $\text{COOH}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{CHO}$ requires C = 47.05 ; H = 5.88 per cent.
 $\text{COOH}\cdot\text{CH}_2\cdot\text{CH}_2\cdot\text{CH}(\text{OC}_2\text{H}_5)_2$ requires C = 54.54 ; H = 9.09 per cent.

In order to confirm this view, the liquid was heated with an equal quantity of phenylhydrazine for 10 minutes at 150° and poured into ether, when, on standing, a white, crystalline substance separated, which, after crystallisation from acetic acid, melted at 191° and gave the following results on analysis.

- 0.0601 gave 0.1502 CO_2 and 0.0350 H_2O . C = 68.17 ; H = 6.47.
 0.1258 gave 21.3 c.c. nitrogen at 19° and 768 mm. N = 19.42.
 $\text{C}_{16}\text{H}_{18}\text{N}_4\text{O}$ requires C = 68.09 ; H = 6.38 ; N = 19.85 per cent.

This substance, on examination, was found to be identical with the condensation product formed by heating aldehydopropionic acid with

* The four analyses given here were carried out with four different preparations of the acid.

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phenylhydrazine (see below), and thus it is probable that this aldehydo-acid was present in the oil obtained by the distillation of acetalmalonic acid.

β -Aldehydopropionic acid, $\text{CHO} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{COOH}$.

In order to prepare this substance, acetalmalonic acid is heated with about four times its weight of water at $180-190^\circ$ for 4 hours, the solution evaporated on the water-bath, and the residue allowed to stand over sulphuric acid in a well exhausted desiccator for 4 or 5 days.

The yellow, oily residue thus obtained, on analysis, gave numbers agreeing approximately with those required for β -aldehydopropionic acid.*

- I. 0.2084 gave 0.3611 CO_2 and 0.1127 H_2O . C = 47.40; H = 6.01.
 II. 0.2124 „ 0.3675 CO_2 „ 0.1132 H_2O . C = 47.20; H = 5.92.
 III. 0.1703 „ 0.2928 CO_2 „ 0.0890 H_2O . C = 46.89; H = 5.82.
 IV. 0.1379 „ 0.2300 CO_2 „ 0.0806 H_2O . C = 45.47; H = 6.47.
 V. 0.1576 „ 0.2649 CO_2 „ 0.0909 H_2O . C = 45.90; H = 6.40.
 $\text{CHO} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{COOH}$ requires C = 47.05; H = 5.88 per cent.

β -Aldehydopropionic acid is a slightly brownish liquid which dissolves readily in water, freshly prepared, its solution reduces Fehling's solution, and produces a pink colour when mixed with a solution of rosaniline hydrochloride which has been decolorised with sulphur dioxide. When heated at 150° with phenylhydrazine for 10 minutes, condensation readily takes place, and if the product is poured into ether a white, crystalline substance separates on standing, which, after recrystallisation from acetic acid, gave the following results on analysis.

0.0590 gave 10.1 c.c. nitrogen at 18° and 758 mm. N = 20.00.

This substance, of which a full analysis is given on p. 15, melts at 192° and is evidently the phenylhydrazone of the phenylhydrazide of aldehydopropionic acid, $\text{C}_6\text{H}_5 \cdot \text{NH} \cdot \text{N} : \text{CH} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{CO} \cdot \text{NH} \cdot \text{NH} \cdot \text{C}_6\text{H}_5$, which contains 19.85 per cent. of nitrogen.

Oxidation of Aldehydopropionic Acid. Formation of Succinic Acid.

When aldehydopropionic acid is left exposed to the air, it darkens in colour and gradually deposits crystals, ultimately being converted into a brown, pasty mass, which in contact with porous porcelain slowly

* As this substance would not crystallise, analyses of each preparation were made, and some of these varied as much as 3 per cent. from the theoretical; we therefore wish it to be distinctly understood that we do not consider that the aldehydo-acid made in this way is pure. All the preparations contained a small amount of ash derived from the tube in which they were prepared; this was allowed for in the analyses.

becomes a nearly colourless, crystalline mass, the sticky, oily impurity being only very gradually absorbed.

The crystals were purified by recrystallisation from hydrochloric acid with the aid of animal charcoal, and in this way colourless plates were obtained which melted at 181° and had all the properties of succinic acid. Analysis.

0.1201 gave 0.1786 CO_2 and 0.0561 H_2O . $\text{C} = 40.56$; $\text{H} = 5.18$.

$\text{COOH} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{COOH}$ requires $\text{C} = 40.68$; $\text{H} = 5.08$ per cent.

This experiment shows that aldehydopropionic acid is slowly converted into succinic acid by the oxygen of the air, and this change takes place very rapidly when oxidising agents are employed. A small quantity of the aldehydo-acid, after being heated to boiling with dilute nitric acid (20 per cent.) for some hours until no further oxidation took place, was evaporated repeatedly on the water-bath with the addition of small quantities of water, until a colourless, crystalline residue was left. This, after recrystallisation from hydrochloric acid, melted at 181 — 184° and consisted of succinic acid.

0.0598 gave 0.0898 CO_2 and 0.0279 H_2O . $\text{C} = 40.96$; $\text{H} = 5.18$.

$\text{COOH} \cdot \text{CH}_2 \cdot \text{CH}_2 \cdot \text{COOH}$ requires $\text{C} = 40.68$; $\text{H} = 5.08$ per cent.

Reduction of Aldehydopropionic Acid. Formation of Butyrolactone,

Five grams of the pure aldehydo-acid were dissolved in water and treated with three times the calculated quantity of 4 per cent. sodium amalgam, carbon dioxide being passed through the liquid and the temperature kept below 10° during the whole operation, in order to avoid, as far as possible, risk of polymerisation or condensation; after separating the mercury, the solution was made strongly acid with sulphuric acid, heated to boiling for half an hour in a reflux apparatus, and then repeatedly extracted with ether. The ethereal solution, when dried and evaporated, deposited a colourless oil, which, after twice fractionating, boiled at 203 — 208° . On analysis, it gave numbers agreeing with those of butyrolactone, which, according to Fittig and Roeder (*Annalen*, 227, 1885, 22), boils at 206° .

0.1356 gave 0.2760 CO_2 and 0.0888 H_2O . $\text{C} = 55.51$; $\text{H} = 7.27$.

$\text{C}_4\text{H}_6\text{O}_2$ requires $\text{C} = 55.81$; $\text{H} = 6.97$ per cent.

18 PERKIN AND SPRANKLING: β -ALDEHYDOPROPIONIC ACID*Action of Caustic Soda on β -Aldehydopropionic Acid. Synthesis of Terephthalic Acid.*

This synthesis, which is explained in the introduction to this paper, was carried out as follows. β -Aldehydopropionic acid (10 grams) was dissolved in an excess of dilute sodium hydroxide and the solution evaporated in a flat glass basin nearly to dryness, water was then added and the solution again evaporated, this operation being continued during three days; the concentrated liquid, after being acidified and allowed to stand over-night, deposited a small quantity of a brownish powder, which, on examination, was found to be crude terephthalic acid. This was purified by dissolving it in dilute sodium carbonate, boiling with animal charcoal until most of the colour had been removed, and then treating the solution at 0° with permanganate until the violet colour remained permanent for 2 minutes; the filtrate from the manganese precipitate was concentrated, and, while still hot, acidified with hydrochloric acid; the colourless, crystalline precipitate, which separated rapidly, had all the properties of terephthalic acid. It was almost insoluble in water and ether, did not melt at 270° , and on heating in a small test tube it sublimed apparently without melting.

The small quantity of acid remaining (about 0.2 gram) was converted into its methylic salt by Baeyer's (*Annalen*, 1888, 245, 140) method by heating with phosphorus pentachloride, mixing the product with methylic alcohol, and purifying the crystals which separated, by recrystallisation from methylic alcohol, with the aid of animal charcoal; the colourless plates thus obtained were very sparingly soluble in methylic alcohol and melted sharply at 140° , the melting point of the methylic salt of terephthalic acid. A specimen of the methylic salt prepared from pure terephthalic acid was found to be identical with the synthetical substance in every respect, moreover, an intimate mixture of the two preparations melted sharply at 140° . These experiments prove conclusively that the acid formed by the action of sodium hydroxide on β -aldehydopropionic acid is terephthalic acid, and it is unfortunate that, in spite of a number of experiments, we have been unable to devise a better method for the condensation of the aldehyde, as the yield of terephthalic acid obtained was certainly not more than 5 per cent.

β -Aldehydoisobutyric Acid, $\text{CHO} \cdot \text{CH}_2 \cdot \text{CH}(\text{CH}_3) \cdot \text{COOH}$.

The first step in synthesising this substance was to prepare ethylic acetaldehydomalonic acid, and this was readily accomplished by heating the sodium derivative of ethylic methylmalonate with bromoacetal

under the conditions already described in the case of ethylic acetal-malonate. The ethylic malonate employed in these experiments was made by the etherification of pure methylmalonic acid; the quantities of the substances used in the synthesis of ethylic acetal-methylmalonate were,

Ethylic methylmalonate	54 grams.
Sodium.....	7.2 „
Bromacetal	40 „

After isolating the product in the way described in the case of ethylic acetal-malonate, the crude oil was submitted to careful fractionation and the fraction 165° (26 mm.), which consisted of nearly pure ethylic acetal-methylmalonate, was analysed with the following result.

0.1491 gave 0.3056 CO_2 and 0.1204 H_2O . $\text{C} = 57.20$; $\text{H} = 8.97$.
 $(\text{C}_2\text{H}_5\text{O})_2\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)(\text{COOC}_2\text{H}_5)_2$ requires $\text{C} = 57.92$; $\text{H} = 8.96$ per cent.

Acetalmethylmalonic Acid, $(\text{C}_2\text{H}_5\text{O})_2\cdot\text{CH}\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)(\text{COOH})_2$.

This acid, which was prepared by the careful hydrolysis of its ethereal salt, is a colourless syrup readily soluble in water.

0.1965 gave 0.3624 CO_2 and 0.1404 H_2O . $\text{C} = 50.30$; $\text{H} = 7.94$.

$\text{C}_{10}\text{H}_{18}\text{O}_6$ requires $\text{C} = 51.28$; $\text{H} = 7.69$ per cent.

The *silver salt*, prepared by precipitating a neutral solution of the ammonium salt with silver nitrate, is a white, amorphous powder, which readily darkens when exposed to light.

0.2026 gave, on ignition, 0.0980 Ag. $\text{Ag} = 48.37$.

$\text{C}_{10}\text{H}_{16}\text{Ag}_2\text{O}_6$ requires $\text{Ag} = 48.19$ per cent.

Aldehydoisobutyric acid, obtained by heating acetalmethylmalonic acid with water at 180° for 4 hours, and evaporating the liquid on a water bath, is an oil which, after standing for some days over sulphuric acid in a vacuum, was analysed with the following results.*

0.1933 gave 0.3653 CO_2 and 0.1238 H_2O . $\text{C} = 51.53$; $\text{H} = 7.28$.

$\text{CHO}\cdot\text{CH}_2\cdot\text{CH}(\text{CH}_3)\cdot\text{COOH}$ requires $\text{C} = 51.72$; $\text{H} = 6.89$ per cent.

This substance is very similar to aldehydopropionic acid in its properties, and its constitution is proved by the fact that, when oxidised with nitric acid, it yields *methylsuccinic acid*; this, after recrystallisation, melted at 110 — 112° , and gave the correct numbers on analysis.

0.2142 gave 0.3558 CO_2 and 0.1238 H_2O . $\text{C} = 45.30$; $\text{H} = 6.40$.
 $\text{COOH}\cdot\text{CH}(\text{CH}_3)\cdot\text{CH}_2\cdot\text{COOH}$ requires $\text{C} = 45.45$; $\text{H} = 6.06$ per cent.

OWENS COLLEGE,
 MANCHESTER.

* See footnote, p. 16.