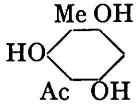
193. *Rottlerin. Part VI. A Spectrographic Study of Rottlerin and its Derivatives.*

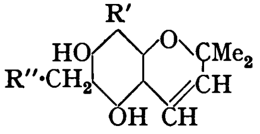
By R. A. Morton and Z. Sawires.

The absorption spectra of rottlerin and its derivatives are largely determined by those of methylphloracetophenone. If the $\mathrm{CH_3{\cdot}CO}$ group is replaced by $\mathrm{C_6H_5{\cdot}CH{:}CH{\cdot}CO}$, a new factor enters into the spectra, and a less important effect occurs when chromen structures occur. There is qualitatively a predominant additive effect in the spectra, but constitutive effects are more significant in determining intensities of absorption. A large number of relatively simple substances have been examined, and the data used as a basis for an extended correlation between structure and absorption. The methyl ethers give rise to interesting anomalies which are shown to have rather wide implications.

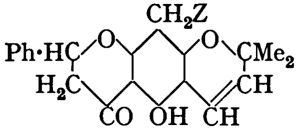
The investigation of rottlerin and its derivatives (Robertson and co-workers, J., 1937, 748; 1938, 309; 1939, 1257, 1579, 1587; Brockmann and Maier, *Annalen*, 1938, **535**, 149;

$\mathrm{X = CHPh{:}CH{\cdot}CO{\cdot}}$; $\mathrm{Y = CH_2Ph{\cdot}CH_2{\cdot}CO{\cdot}}$; Z =

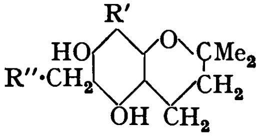




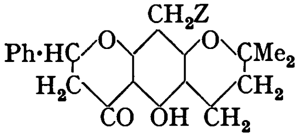
(I; Rottlerin, R′ = X; R″ = Z.)



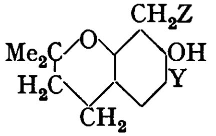
(II; *iso*Rottlerin.)



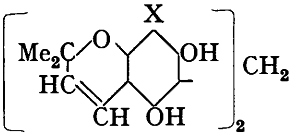
(III; Tetrahydrorottlerin, R′ = Y; R″ = Z.)



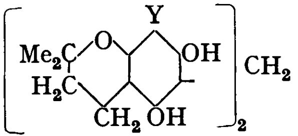
(IV; Dihydro*iso*rottlerin.)



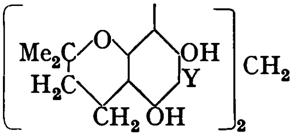
(V; Tetrahydro*allo*rottlerin.)



(VI; Rottlerone.)

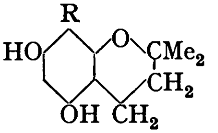


(VII; Octahydrorottlerone.)

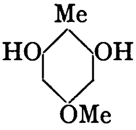


(VIII; Octahydro*allo*rottlerone.)

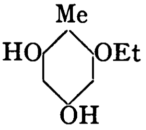
(IX.) $\mathrm{Z{\cdot}CH_2{\cdot}Z}$



(X; R = Y.)
(XI; R = H.)



(XII.)



(XIII.)

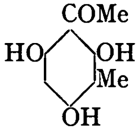
1939, **541**, 53) has now reached a stage when it is profitable to discuss absorption spectra (obtained on compounds prepared by the Liverpool workers) in relation to the structural formulæ which have been established. A large number of substances have been studied, mainly in 96% ethyl alcohol, and the spectra usually exhibit broad, unresolved absorption bands in the ultra-violet. The work is concerned with rather large molecules containing a number of chromophoric groups, and the problem as a whole affords an excellent opportunity for illustrating the mingling of additive and constitutive effects in the spectra of complicated compounds.

The principal compounds are shown in (I)—(XIII), and the spectra are given in Table I. The latter can best be understood if methylphloracetophenone (XIV) is taken as a starting point, and as a preliminary it is necessary to account for the spectrum of this substance.

TABLE I.

Summary of Spectrographic data.

Compound.	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$	$\lambda_{min.}$, mμ.	$\epsilon_{min.}$	Fig.
Rottlerin (I)	350	27,100	327	24,250	1
	294	34,800	257	16,100	
	231·5	33,000			
*iso*Rottlerin (II)	~366	5,600	244·5	14,200	2
	~302	22,450			
	273	41,300			
Tetrahydrorottlerin (III)	~345	6,600	253	3,250	3
	295	29,000	224·5	24,900	
	232	26,900			
Dihydro*iso*rottlerin (IV)	~345	5,800	253	2,590	4
	293	30,600	221·5	28,750	
	225	30,300			
Tetrahydro*allo*rottlerin (V)	345	5,500	252·5	4,420	5
	289·5	32,250	221·5	23,140	
	229	27,300			
Rottlerone (VI)	358	35,000	307	2,190	1
	290	23,200	265	12,500	
Octahydrorottlerone (VII)	~345	7,470	255	5,260	6
	297·5	34,300			
	232	28,500			
Octahydro*allo*rottlerone (VIII)	~345	5,130	252	3,790	7
	291·5	35,900	225	25,200	
	233·5	31,100			
(IX)	~345	5,860	252	5,860	8
	291	25,750			
	227	21,800			
(X)	~345	3,400	250	1,590	9
	293·5	22,500			
5 : 7-Dihydroxy-2 : 2′-dimethylchroman (XI)	272	624	252	220	10


(XIV.)

$\lambda_{max.}$, mμ.	$\epsilon_{max.}$
~333	2,510
291	17,800
223	12,600

*cyclo*Hexane is transparent in the ultra-violet and *cyclo*hexene has maxima at 220 and 190 mμ, whereas *cyclo*hexadiene shows two regions of intense selective absorption, *ca.* 260 mμ and *ca.* 190 mμ (Allsopp, *Proc. Roy. Soc.*, 1934, *A*, **143**, 618). Benzene, with three conjugated double bonds, shows resolved absorption 230—270 mμ of low intensity, ε = 200—300, and it is important that the absorption, although qualitatively similar to that of *cyclo*hexadiene, is very much weaker (one-twentieth). It is also quite distinct from that shown by compounds containing the triene group A·CH:CH·CH:CH·CH:CHB (cf. elæostearic acid). The spectra of phenol and anisole exhibit a small displacement of $\lambda_{max.}$, and a roughly 10-fold increase in $\varepsilon_{max.}$, as compared with benzene (the spectra show resolution into narrower bands):

	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$
Phenol (in hexane)	271	2200
(in alcohol)	272·8	2050
Anisole (in hexane)	272	2510
(in alcohol)	271	2240

Relatively high ε values are maintained in the dihydroxybenzenes, but phloroglucinol and its trimethyl ether exhibit a striking reversion to low ε values:

	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$	
Pyrocatechol in alcohol	277·8	2600	
Resorcinol in alcohol	276·1	2000	
Quinol in alcohol	294·2	3100	
Phloroglucinol in alcohol	266·5	380	Morton and Rogers, J., 1925, **127**, 2700.
O-Trimethoxyphloroglucinol in alcohol	264·6	465	
1 : 2 : 4-Trihydroxybenzene in alcohol	292	3160	Goslawski and Marchlewski, *Bull. Acad. Polonaise, A*, 1931, 383.
1 : 2 : 4-Triacetoxybenzene in alcohol	266·5	708	
1 : 2 : 3-Trihydroxybenzene in alcohol	266	800	
1 : 2 : 3-Triacetoxybenzene in alcohol	259	240	
1 : 3 : 5-Trihydroxybenzene in alcohol	268·5	356	
1 : 3 : 5-Triacetoxybenzene in alcohol	261	290	

1 : 2 : 4-Trihydroxybenzene shows a spectrum practically indistinguishable from that of quinol, whereas that of pyrogallol approaches the low ε values shown by phloroglucinol.

In all these compounds the electronic change concerned in the act of absorption is made possible by the presence of a double bond in the benzene ring, and it is clear that, just as the third double bond in benzene reduces the probability of the electronic transition occurring in both *cyclo*hexadiene and benzene, so also the distortion set up by mono- and di-substitution (OH or OMe) partly restores it. Trisubstitution may, but need not necessarily, reverse the effect. Although symmetry considerations obviously have a bearing on the matter, it is not satisfactory to attribute the decreased absorption merely to 1 : 3 : 5-substitution, since the neutralisation of the effects of substituents is equally clear in pyrogallol triacetate.

The methylphloracetophenone spectrum may now be approached from the standpoint of the acetyl substituent:

	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$		$\lambda_{max.}$, mμ.	$\epsilon_{max.}$
Acetophenone	320	45	*o*-Hydroxyacetophenone	327	3,550
	278	1,000		251·5	9,330
	243	12,600	*p*- ,, ,,	330	60
	199	20,000		277	13,500
				221	10,950

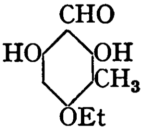
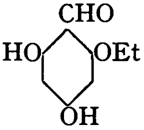
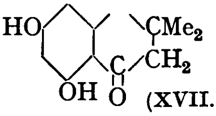
The origin of selective absorption in the hydroxyacetophenones will be discussed elsewhere by Morton and Stubbs. The 327 mμ and 251·5 mμ maxima of the *o*-isomeride and the 277 mμ maximum of the *p*-derivative are all benzenoid in origin, whereas the low-intensity bands at 320 and 330 mμ in the above table are due to the carbonyl group. In resacetophenone the superposition of effects due to *o*- and *p*-hydroxyls is manifest (Fig. 14):

$\lambda_{max.}$, mμ	315	278	236	231
$\epsilon_{max.}$	7,890	13,780	7,980	8,460

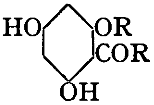
In methylphloracetophenone the inflexion at 333 mμ corresponds with the 327 mμ band of *o*-hydroxyacetophenone, and the 291 mμ maximum with the 277 mμ band of *p*-hydroxyacetophenone. It is less easy to locate the site in the molecule corresponding with the 223 mμ maximum, but it is provisionally regarded as the carbonyl group.

The aldehydes (XV) and (XVI) exhibit spectra very similar to that of methylphloracetophenone, and when comparison is made with *o*- and *p*-hydroxybenzaldehyde (Table II) it is clear that the greatly modified benzenoid electronic transitions of (XIV), (XV), and (XVI) are essentially predetermined, since their absorption curves are little different from the summation of those of the two simple hydroxyaldehydes.

TABLE II.

	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$		$\lambda_{max.}$, mμ.	$\epsilon_{max.}$
 (XV.)	345 295 230	3,550 21,950 7,850	 (XVI.)	332 297 229	4,100 * 20,000 11,650
o-Hydroxybenzaldehyde	325 255	3,020 10,000	*p*-Hydroxybenzaldehyde	332 284 221	180 17,400 13,800
(XII) (XIII) (XI)	270·5 274 272	625 800 624	 (XVII.)	~345 292·5	2,400 18,000

* Very feeble inflexion.


(XVIII.)

The absence of a substituent containing a carbonyl group (XII and XIII) effects a reversion to the simple low-intensity benzenoid absorption of phloroglucinol, and the chroman (XI) is chromophorically identical with (XII), whereas chromanone (XVII) is to be regarded as (XVIII) from the point of view of absorption, and shows a curve almost identical with that of methylphloracetophenone.

One of the best attested phenomena in absorption spectra is the insulating effect of CH_2 or $[CH_2]_n$, for a molecule $A\cdot CH_2\cdot B$, where A and B are chromophoric groups, shows normally a spectrum obtained by the simple summation of the absorption due to A and B. This insulating effect can be tested in many ways from the data for rottlerin derivatives.

(*a*) The diphenylmethane derivative (IX) shows qualitatively the same absorption as methylphloracetophenone, but the ϵ values do not reach twice those of this substance:

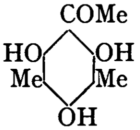
	$\lambda_{max.}$	$\epsilon_{max.}$ × 2.		$\lambda_{max.}$, mμ.	$\epsilon_{max.}$
(XIV)	~333 291 223	5,020 35,600 25,200	(IX)	~345 291 227	5,860 25,760 21,800

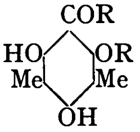
The test is not quite a strict one, since (IX) ought perhaps to be compared with 3 : 5-dimethylphoracetophenone.

(*b*) The phenyl group in (X) is insulated by two CH_2 groups, and it would therefore be expected to exert little effect on the spectrum of methylphloracetophenone, and octahydrorottlerone (VII) should have twice the intensity of absorption:

	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$
5 : 7-Dihydroxy-8-acetyl-2-*gem*dimethylchroman	~350 293·5 227	3,250 19,700 12,650
5 : 7-Dihydroxy-8-β-phenylpropionyl-2-*gem*dimethylchroman (X)	~345 293·5	3,400 22,490
Octahydrorottlerone (VII)	~345 297·5 232	7,470 34,300 28,500

(*c*) Tetrahydrorottlerin (III) is chromophorically equivalent to 3 : 5-dimethylphloracetophenone (XIX) plus (XX). The spectrum does in fact show the expected absorption

(XIX.)




(XX.)

maxima, but, as in (*b*), the maximum *ca.* 295 mμ fails to reach the expected two-fold rise in ε value (see Table I), *viz.*, 29,000 as compared with *ca.* 35,600 expected. From another point of view the spectrum of (III) may be expected to be given by the mean of those of octahydrorottlerone (VIII) and the diphenylmethane derivative (IX):

$\lambda_{max.}$, mμ, obs.	$\epsilon_{max.}$, obs.	$\epsilon_{max.}$, calc.
~345	6,600	6,665
295	29,000	30,035
232	26,900	25,150

and the spectrum of tetrahydro*allo*rottlerin (V) is, as would be expected, very similar.

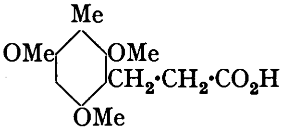
The element of additivity in the spectra is obvious, but extinction coefficients cannot be predicted accurately in every case. The significant variable is the degree of substitution in the benzene ring, and a definite effect shown by methyl groups on ε values is already known. The data in Table III (cf. Conrad-Billroth, *Z. physikal. Chem.*, 1935, *B*, **29**, 170; Wolf and Strasser, *ibid.*, 1933, *B*, **21**, 389) make this clear for the benzenoid chromophore:

TABLE III.

Methylbenzenes.

Methyl substituents	(None.)	1.	1 : 2.	1 : 3.	1 : 4.	1 : 2 : 3.	1 : 2 : 4.	1 : 2 : 5.
$\lambda_{max.}$, mμ	254	260	260	265	265	260	275	265
$\epsilon_{max.}$	230	300	330	300	440	360	830	300

Methyl substituents	1 : 2 : 3 : 5.	1 : 2 : 4 : 5.	1 : 2 : 3 : 5 : 6.	1 : 2 : 3 : 4 : 5 : 6.
$\lambda_{max.}$, mμ	265	275	270	270
$\epsilon_{max.}$	420	820	370	330



(XXI.)

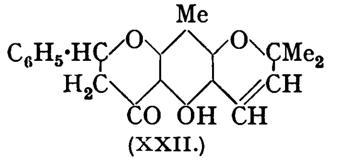
In 2 : 4 : 6-trimethoxy-*m*-tolylpropionic acid (XXI) the carboxyl group is effectively insulated, but the absorption spectrum (Fig. 18) ($\lambda_{max.}$ 280 mμ, $\epsilon_{max.}$ 2290) is about 5 times as intense as that shown by phloroglucinol trimethyl ether, *viz.*, $\lambda_{max.}$ 264 mμ, $\epsilon_{max.}$ 465.

The departures from quantitative additivity thus appear to indicate that the probability of a given electronic transition is influenced considerably by chromophorically inert substituents like methyl groups.

The spectra of rottlerin and rottlerone differ from those already discussed in that a very intense band $\epsilon_{max.}$ 27,000—35,000 at 350—360 mμ replaces the rather weak band $\epsilon_{max.}$ 3,000—7,500 at about 345 mμ. Both compounds contain additional unsaturation, as they possess the chromen structure and the cinnamoyl group in which there is no CH_2 group to insulate the phenyl and carbonyl groups. The two types of additional unsaturation may be taken separately:

	$\lambda_{max.}$	$\epsilon_{max.}$
(*a*) 5 : 7-Dihydroxy-8-cinnamoyl-2-*gem*dimethylchroman	347	23,970
(XI; R = CHPh:CH·CO·)	330	19,400

This shows greatly enhanced absorption at 350 mμ and there is obviously a second band of high intensity not far away, near 300 mμ, but the superposition of the effects of different chromophores makes it risky to be dogmatic about the interpretation of absorption in the region 270—325 mμ.



(XXII.)

(*b*) The effect of the chromen linkage is best approached indirectly by comparing the spectra of *iso*rottlerin and dihydro*iso*rottlerin (II and IV; Figs. 2 and 4). The additional unsaturation gives rise to a new and intense absorption band with $\lambda_{max.}$ *ca.* 273 mμ, but the absorption in the region 325—400 mμ is displaced somewhat in the direction of longer wave-lengths without significant change in intensity.

The spectrum of (XXII) (not examined or prepared) may be deduced by subtracting an absorption curve obtained by halving all the ε values for (IX) from the curve for *iso*rottlerin;

$\lambda_{max.}$, mμ	370	305	270
$\epsilon_{max.}$	4,000	10,200	34,000

Fig. 1.

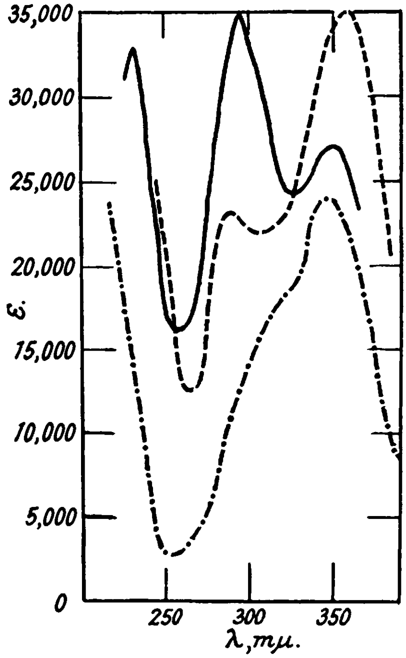
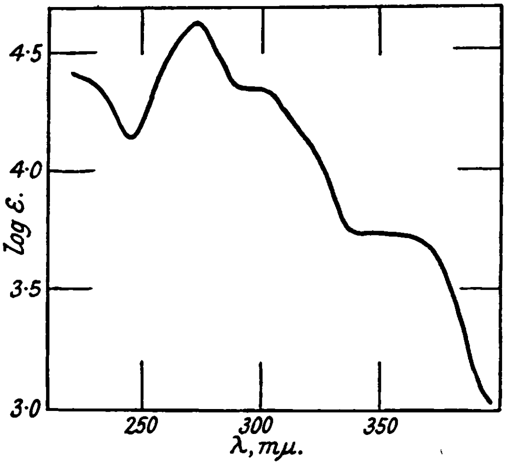


Fig. 2.



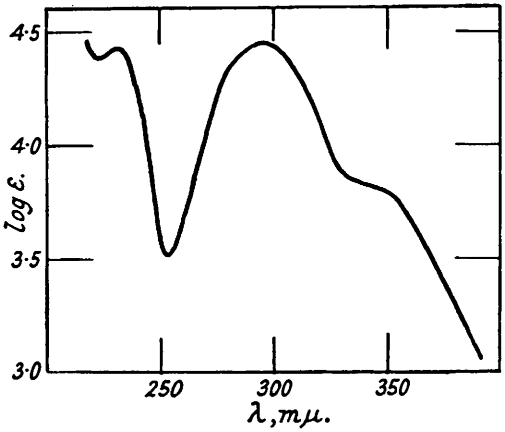


Fig. 3.

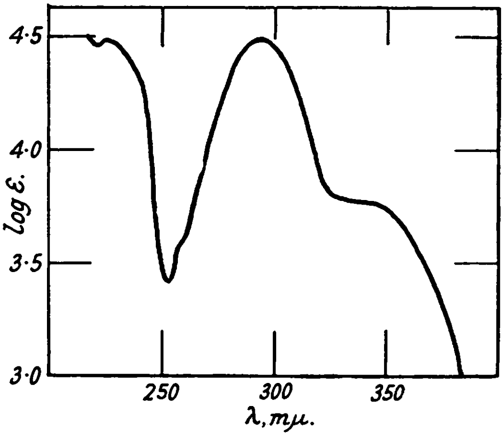


Fig. 4.

Fig. 1. ———— Rottlerin.
– – – – – Rottlerone.
—·—·— 5 : 7-Dihydroxy-8-cinnamoyl-2-*gem*dimethylchroman (XI; R = CHPh:CH·CO·).
Fig. 2. *iso*Rottlerin.
Fig. 3. Tetrahydrorottlerin.
Fig. 4. Dihydro*iso*rottlerin.

The curve characteristic of methylphloracetophenone has thus been changed from 345 to 370 mμ, with little change in ε values, and new bands occur at 305 and 270 mμ. It is thus clear that the cinnamoyl grouping of rottlerin is more important in determining its absorption spectrum than the chromen linkage. The styryl ketones are themselves very strongly absorbing:

	$\lambda_{max.}$	$\epsilon_{max.}$	$\lambda_{max.}$	$\epsilon_{max.}$	$\lambda_{max.}$	$\epsilon_{max.}$
Styryl methyl ketone	287	20,900	—	—	—	—
Styryl phenyl ketone	307	22,900	~270	11,200	225·5	10,700

FIG. 5.

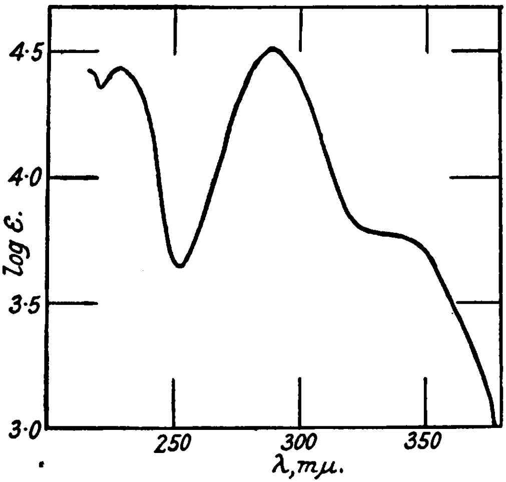
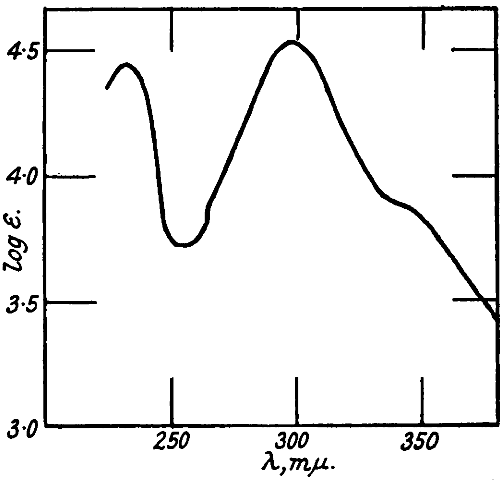


FIG. 6.



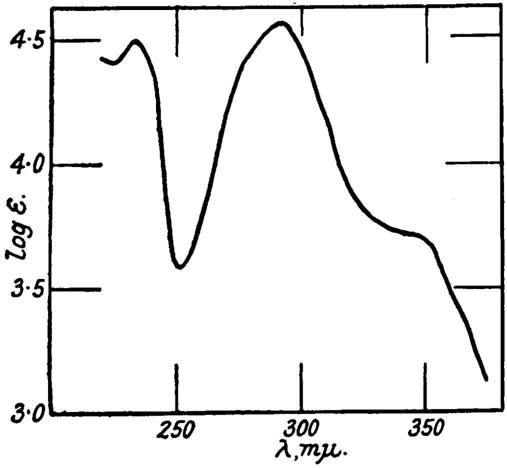


FIG. 7.

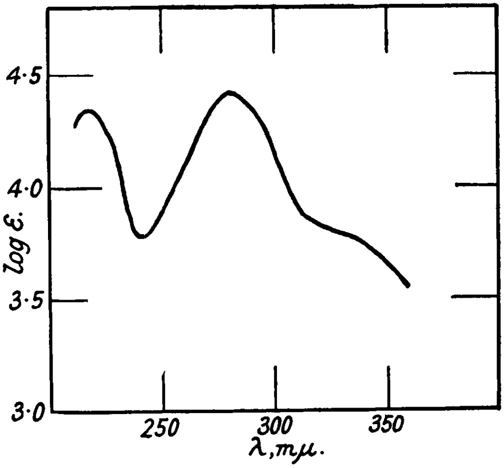


FIG. 8.

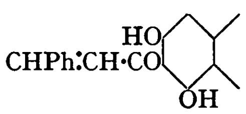
FIG. 5. Tetrahydro*allo*rottlerin.
FIG. 6. Octahydrorottlerone.

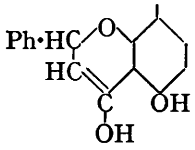
FIG. 7. Octahydro*allo*rottlerone.
FIG. 8. Compound (IX).

It is interesting that the spectrum of rottlerone (Fig. 1) is similar to that of (XI; R = CHPh:CH·CO) but is not quantitatively given by doubling intensities of absorption.

The absorption curve of rottlerin should, on the basis of insulation of two ring systems by the linking CH$_2$, be intermediate between those of rottlerone and methylphloracetophenone. The absorption at 350 mμ is, in fact, much less than that of rottlerone and qualitatively the summation effect holds good, but quantitatively it breaks down, again presumably because the CH$_2$ is a substituent in both ring systems as well as a link between them.

(XXIII.)





(XXIV.)

FIG. 9.

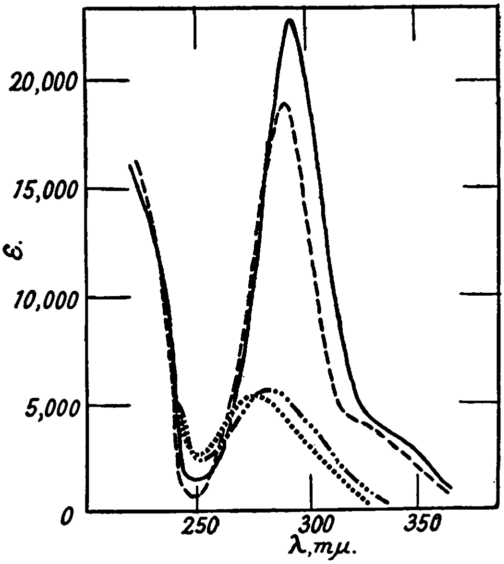
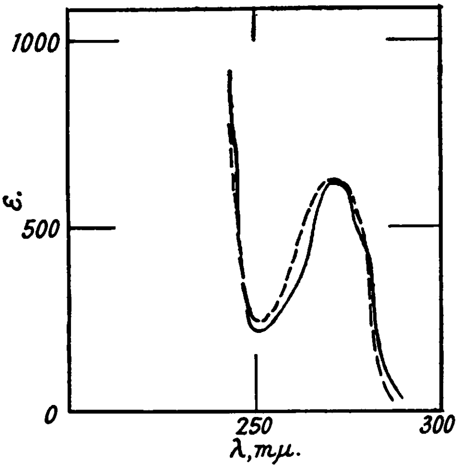


FIG. 10.



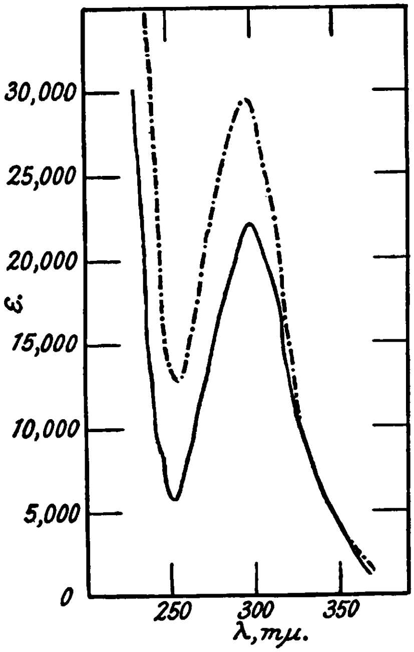


FIG. 11.

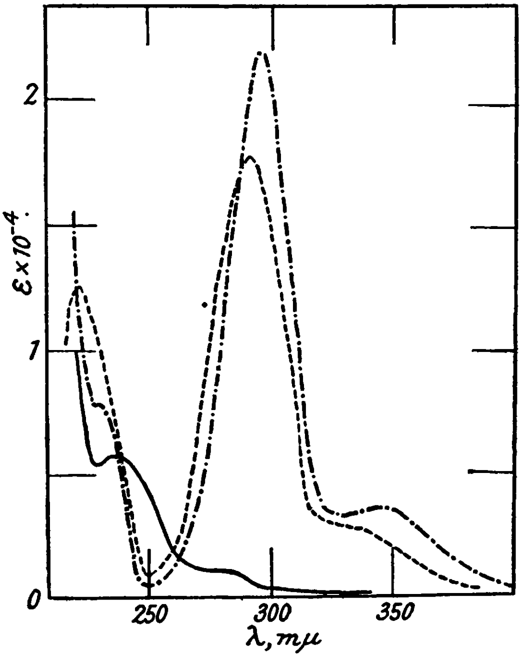


FIG. 12.

FIG. 9. Ethers of (X; R = Y):
————— Unsubstituted.
– – – – – – 5-Methyl ether.
—...—... 7- ,,
. . . . . . 5 : 7-Dimethyl ether.

FIG. 10. ————— (XI; R = H).
– – – – – (XII).

FIG. 11. —·—·— *allo*Rottlerin pentamethyl ether.
————— Dihydrorottlerin pentamethyl ether.

FIG. 12. —·—·—· Compound (XV).
– – – – – – ,, (XIV).
————— *O*-Triacetyl deriv. of (XIV).

FIG. 13.

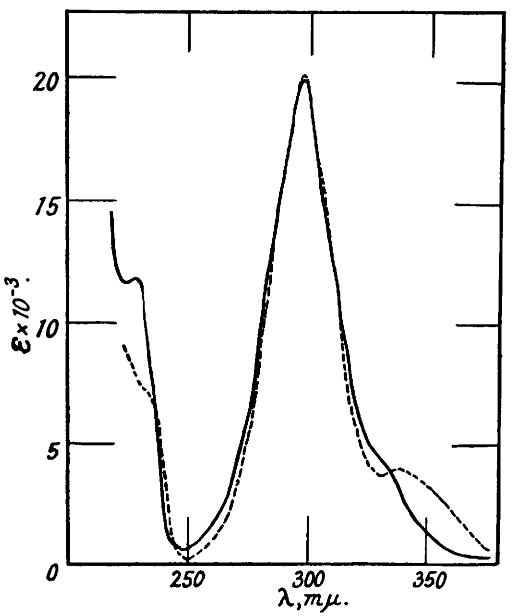
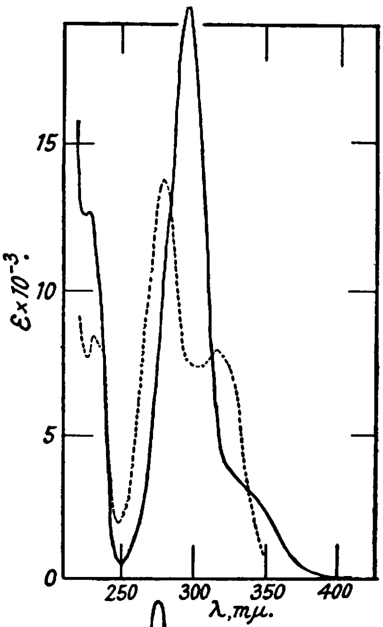


FIG. 14.



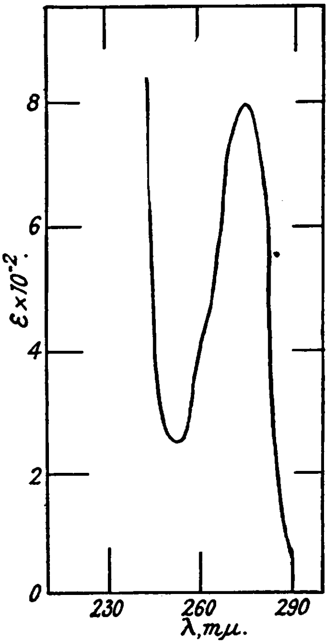


FIG. 15.

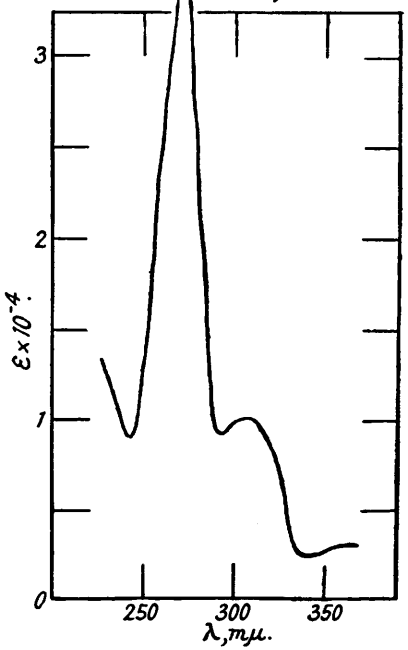
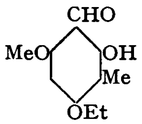


FIG. 16.

FIG. 13. - - - - -



——————

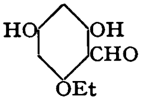


FIG. 14. —————— (XI; R = Ac).
- - - - - Resacetophenone.

FIG. 15. Compound (XIII).
FIG. 16. Compound (XXII) (predicted curve).

FIG. 17.

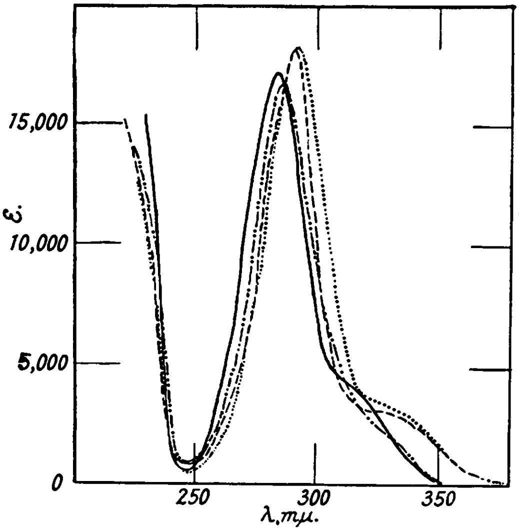
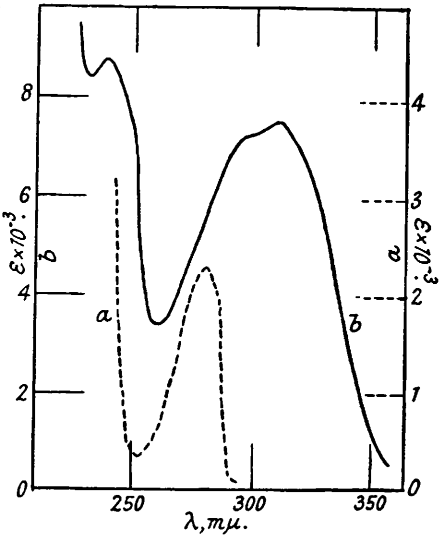


FIG. 18.



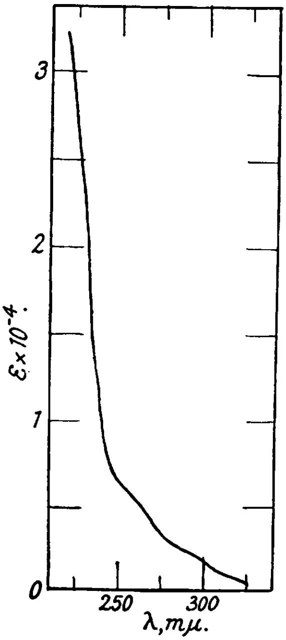


FIG. 19.

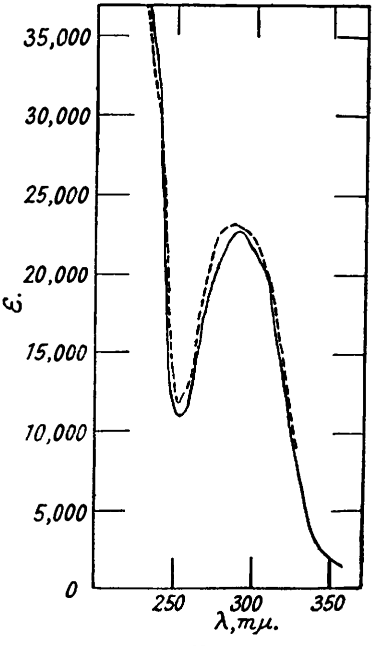
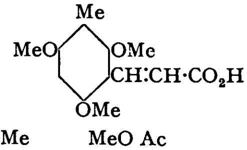


FIG. 20.

FIG. 17. . . . . . 5 : 7-Dihydroxy-2-*gem*dimethylchromanone.
– – – – – 5-Hydroxy-7-methoxy-2-*gem*dimethylchromanone.
—··—·· 5-Methoxy-7-hydroxy- ,, ,,
———— 5 : 7-Dimethoxy- ,, ,,

FIG. 18. ————



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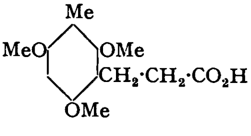


FIG. 19.

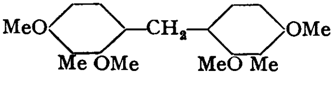


FIG. 20. Rottlerin pentamethyl ether :
———— Experimental.
– – – – – Predicted.

FIG. 21.

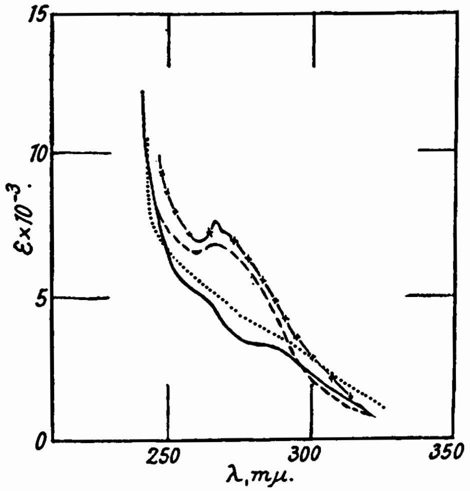
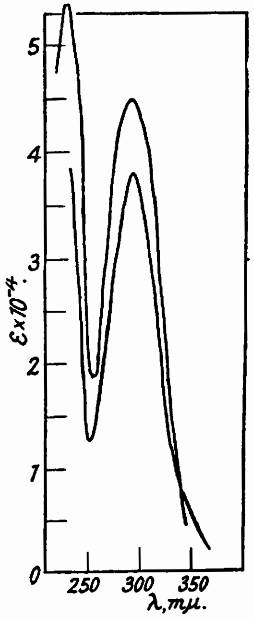


FIG. 22.



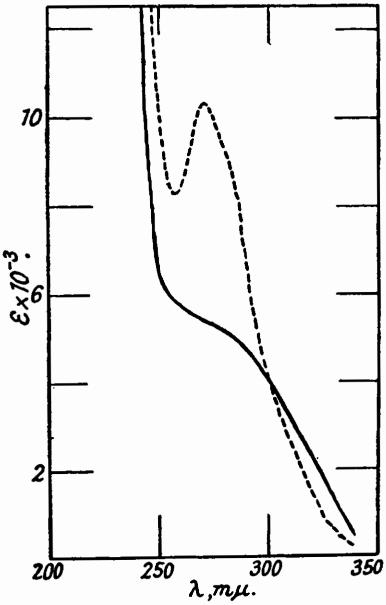


FIG. 23.

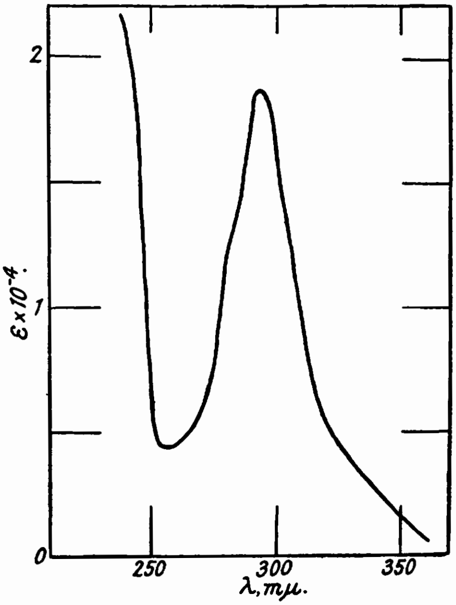


FIG. 24.

FIG. 21. Tetrahydrorottlerin pentamethyl ether:
———— Experimental. . . . . Predicted.
*allo*Tetrahydrorottlerin pentamethyl ether:
– – – – – Experimental. —×—×— Predicted.

FIG. 22. Upper curve: rottlerone tetramethyl ether.
Lower curve: rottlerin dimethyl ether.

FIG. 23. ———— Octahydrorottlerone tetramethyl ether.
– – – – – *allo*Octahydrorottlerone tetramethyl ether.

FIG. 24. Dihydro*iso*rottlerin tetramethyl ether.

Robertson and his co-workers (see Parts III, IV, and V) attribute the structure (XXIII) to the *allo*-compounds, whereas Brockmann and Maier (*loc. cit.*) make use of the formulation (XXIV). Absorption spectra data bearing upon this point of difference are inconclusive and the decision must rest on other evidence (J., 1939, 1587).

Spectra of Ethers derived from Rottlerin and its Derivatives.—It is generally accepted that the replacement of hydrogen in a hydroxyl group by methyl does not directly involve any material change in absorption spectrum; for instance,

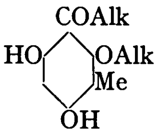
	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.
Phenol in alcohol	273	2,050
Anisole	271	2,240
Resorcinol	276	2,000
Resorcinol dimethyl ether	274	2,190
Phloroglucinol	266·5	380
Phloroglucinol trimethyl ether	264·5	465

Acetylation, on the other hand, may effect a great decrease in intensity of absorption; *e.g.*, phenyl acetate shows ε values of the order of one-tenth of those of phenol, and methylphloracetophenone triacetate is vastly more transparent than the parent substance (XIV):

$\lambda_{max.}$	281	238
$\epsilon_{max.}$	1,000	5,800 (persistence poor)

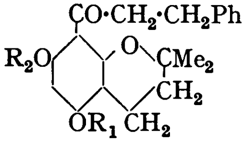
In the rottlerin series the effects are much more complicated, and the facts have rather wide implications in the general problem of correlating structure with absorption. They are therefore worth recording in some detail, even if the phenomena are capable of only partial explanation. The chroman (XI) is chromophorically equivalent to a monomethyl phloroglucinol:

	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.		$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.
Phloroglucinol	266·5	380	(XII)	270·5	625
(XI)	272	625	(XIII)	274	800


(XXV.)

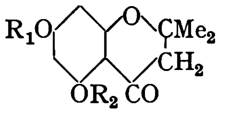
and compound (X) is chromophorically equivalent to (XXV), and, as compared with methylphloracetophenone, it shows an increased ε value (17,800 to 22,500). In these cases mono-etherification brings about a definite increase in intensity of absorption.

Introduction of a second methyl group, however, lowers the ε values, substitution of H in a hydroxyl ortho- to the carbonyl being very effective:



	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.
$R_1 = CH_3$, $R_2 = H$	291	18,700
$R_1 = H$, $R_2 = CH_3$	284	5,570
$R_1 = CH_3$, $R_2 = CH_3$	277	5,440

The hydroxychromanones are, however, less sensitive to methylation:



R_1.	R_2.	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.
H	H	292·5	18,000
H	CH_3	290·5	18,060
CH_3	H	285·5	16,650
CH_3	CH_3	283·5	17,100

The derivatives of cinnamic acid are also interesting in this connection:

Cinnamic acid.	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.	Cinnamic acid.	$\lambda_{max.}$, mμ.	$\epsilon_{max.}$.
Unsubstituted	269	18,200	*p*-Hydroxy-	299	15,500
o-Hydroxy-	327	10,230 *	*p*-Methoxy-	291	15,150
	272·5	14,800	2 : 4 : 6-Trimethoxy-3-methyl-	309	7,550
o-Methoxy-	319·5	3,700		~295	7,250
	274	6,300		238	8,820

* Ley and Dreinhofer, *Z. wiss. Phot.*, 1930, **29**, 134.

The striking decreases in $\epsilon_{max.}$ which occur as a result of methylation are made all the more difficult to explain by the absence of such effects in the simple chromanones. It is also significant that simple compounds like *o*- and *p*-hydroxyacetophenone maintain their characteristic absorption (ε values) on methylation.

An interesting point arises in considering the spectra of fully methylated compounds in the rottlerin series. In most cases (Figs. 19, 21, 23) selective absorption disappears and only moderately intense continuous end absorption remains. Ethers with chromen or cinnamoyl groups, however, show a "residual" selective absorption, but at 290 mμ rather than 350 mμ. The probable explanation is that the 350 mμ band has been displaced in the direction of shorter wave-lengths as a result of methylation and by a coincidence is liable to be confused with the 290—295 mμ band present in the hydroxy-compounds.

The conjugation of the styryl group through carbonyl with phloracetophenone residues normally enhances the 350 mμ absorption; methylation may damp down (reduced ε values) the absorption due intrinsically to phloracetophone, and at the same time cause the 350 mμ absorption to be displaced in the direction of lower wave-lengths :

	$\lambda_{\max.}$, mμ.	$\epsilon_{\max.}$
Rottlerin	294	34,800
Rottlerin pentamethyl ether	(general absorption of low intensity)	
Rottlerin dimethyl ether *	293	38,100
Tetrahydrorottlerin	295	29,000
Tetrahydrorottlerin pentamethyl ether	291	18,300
*allo*Rottlerin pentamethyl ether	294	29,300
Rottlerone	290	23,200
Rottlerone tetramethyl ether *	291	45,000

* Position of methyl groups uncertain.

These considerations are sufficient to disclose the existence of a very difficult problem, the core of which is a profound influence exerted by substituents (which are themselves transparent) primarily on the probability of a given electronic transition, and in more extreme cases on its very possibility.

The authors wish to express their gratitude to Professor A. Robertson, Dr. A. McGookin, and their colleagues, and also to the Egyptian Educational Mission for allowing one of them (Z. S.) to take part in this work.

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