DIAMAGNETIC SUSCEPTIBILITY OF LEAD ION

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Previous systematic investigations on the magnetic properties of a large number of inorganic and organic compounds of Ca, Sr, Ba, Mg and Zn deal with the determination of the diamagnetic susceptibilities of these cations by the method of averages and other methods. An important result accruing from these investigations is the establishment of the fact that the mean ionic susceptibility values of cations deduced from the salts of organic acids are higher than those obtained from the salts of inorganic acids.

The ionic susceptibility of Pb given in the 'International Critical Tables' is $- 46 \cdot 0 \times 10^{-6}$ (Pascal) while a mean value deduced from lead salts studied by Kido[1] comes out to be $- 30 \cdot 07 \times 10^{-6}$. The values calculated according to Slater and Angus are $- 50 \cdot 2 \times 10^{-6}$ and $- 49 \cdot 9 \times 10^{-6}$, respectively. Hence in order to obtain an accurate experimental value of Pb^{++}, which could be compared with the theoretically calculated values without any ambiguity the molecular susceptibilities of several lead salts of inorganic and organic acids were measured by the same method and technique as followed in previous investigations. The results obtained have been utilised to examine the findings of Prasad and co-workers[2] regarding the differences in susceptibilities of cations deduced from their salts of inorganic and organic acids.

Experimental and Results

All the salts used in this investigation were chemically pure; they were either of Merck's Extra-Pure quality or were prepared in a pure state in the laboratory. The results obtained are given in column 3 of Table I. Each susceptibility value given in the Table is the mean of six closely agreeing measurements, each being the mean of three readings. Each measurement was made with a fresh packing of the substance.

In order to compare the observed values with those computed by adding the empirical values for susceptibilities of the anion and the cation given by the same investigator (column 5) the observed molar susceptibilities for the anhydrous salts were obtained by deducting the susceptibility value of water molecules ($X_{H_2O} = 12 \cdot 96$) from the experimental molecular susceptibilities of the hydrated salts. The experimental values obtained by previous

workers are given in column 4 of Table I (*a*). Since no determinations of susceptibilities of organic compounds have been made by previous workers, column 4 has been dropped in Table I (*b*); only lead acetate has been

TABLE I (*a*)

Lead Salts of Inorganic Acids

Compounds	N	Authors' values	χ_{ma} Other workers	χ_{ma} Computed experimentally	χ_{ma} Computed theoretically
PbCO₃ ..	112	0·2259 60·36 60·36	.. .. ..	68·2(4) 58·2(1)	$Pb^{+2}+(CO_3)^{-2}$ 89·8 (S) 83·8 (A)
PbCl₂ .. Pb Found = 74·10% Pb Theory = 74·46%	116	0·2923 .. 81·30	89·00(4) 73·48(1)	86·2(4) 74·3(1)	$Pb^{+2}+2(Cl)^{-1}$ 101·78 (S) 95·62 (A)
PbSO₄ ..	130	0·2420 .. 73·36	69·01(1) 77·96(5)	79·6(4) 69·1(1)	$Pb^{+2}+(SO_4)^{-2}$ 104·21 (S) 96·21 (A)
PbCrO₄ .. Pb found = 64·10% Pb theory = 64·07%	138	0·0563 .. 18·18	..	..	$Pb^{+}+(CrO_4)^{-2}$ 108·30 (S) 100·05 (A)
PbS₂O₃ .. Pb found = 64·78% Pb theory = 64·89%	138	0·2619 .. 83·60	..	89·4(4) 73·1(1)	$Pb^{+2}+(S_2O_3)^{-2}$ 113·91 (S) 105·63 (A)
Pb(CNS)₂ .. Pb found = 64·02% Pb theory = 64·07%	140	0·3111 .. 100·60	..	..	$Pb^{+2}+2(CNS)^{-1}$ 214·20 (S) 183·74 (A)
Pb(NO₃)₂ ..	144	0·2320 .. 76·85	82·15(4) 71·20(1) 77·03(5)	74·4(4) 70·3(1)	$Pb^{+2}+2(NO_2)^{-1}$ 129·32 (S) 117·62 (A)
PbBr₂ .. Pb found = 56·51% Pb theory = 56·39%	152	0·2776 .. 101·90	102·80(4)	107·4(4) 99·5(1)	$Pb^{+2}+2(Br)^{-1}$ 130·22 (S) 123·20 (A)
PbI₂ .. Pb found = 44·7% Pb theory = 44·89%	188	0·3019 .. 139·20	152·20(4)	135·2(4) 136·5(1)	$Pb^{+2}+2(I)^{-1}$ 169·86 (S) 160·54 (A)
Pb(BrO₃)₂·H₂O Pb found = 43·10% Pb theory = 43·03%	200	0·2462 118·40 105·44	..	..	$Pb^{+2}+2(BrO_3)^{-1}$ 152·92 (S) 141·08 (A)
Pb(IO₃)₂ .. Pb found = 37·22% Pb theory = 37·15%	236	0·2356 .. 131·20	..	..	$Pb^{+2}+2(IO_3)^{-1}$ 173·66 (S) 161·74 (A)
Pb₃(PO₄)₂ ..	340	0·2239 .. 181·70	..	208·8(4) 184·7(1)	$3Pb^{+}+2(PO_4)^{-3}$ 259·12 (S) 242·78 (A)

TABLE I (b)

Lead Salts of Inorganic Acids

Compounds	N	Authors' values	χ_{ma} Computed experimentally	χ_{ma} Computed theoretically
PbC_2O_4 Pb found $=70 \cdot 18\%$ Pb theory $=70 \cdot 20\%$	126	$0 \cdot 2416$ .. $71 \cdot 32$	$73 \cdot 94(4)$	$Pb^{+2} + (C_2O_4)^{-2}$ $77 \cdot 90$ (S) $77 \cdot 60$ (A)
$Pb(HCOO)_2$ Pb found $=69 \cdot 1\%$ Pb theory $=69 \cdot 7\%$	128	$0 \cdot 2439$ .. $72 \cdot 49$	$79 \cdot 8$ (4) $70 \cdot 5$ (1)	$2Pb^{+2} + (HCOO)^{-1}$ $83 \cdot 70$ (S) $83 \cdot 40$ (A)
$Pb(CH_2COO)_2$ Pb found $=63 \cdot 74\%$ Pb theory $=64 \cdot 10\%$	142	$0 \cdot 2839$ .. $91 \cdot 77$	$97 \cdot 66(4)$	$Pb^{+2} + (CH_2COO)_2^{-2}$ $101 \cdot 40$ (S) $101 \cdot 10$ (A)
$Pb(CH_3COO)_2 \cdot 3H_2O$	144	$0 \cdot 3573$ $135 \cdot 50$ $96 \cdot 62$ $93 \cdot 82(1)$	$103 \cdot 8$ (4) $94 \cdot 1$ (1)	$Pb^{+2} + 2(CH_3COO)^{-1}$ $107 \cdot 20$ (S) $106 \cdot 90$ (A)
$Pb(CHOHCOO)_2$ Pb found $=58 \cdot 3\%$ Pb theory $=58 \cdot 34\%$	158	$0 \cdot 3102$ .. $110 \cdot 30$	$109 \cdot 8$ (4)	$Pb^{+2} + (CHOHCOO)_2^{-2}$ $110 \cdot 6$ (S) $110 \cdot 3$ (A)
$PbC_6H_4(COO)_2$ Pb found $=55 \cdot 70\%$ Pb theory $=55 \cdot 82\%$	166	$0 \cdot 3336$ .. $123 \cdot 90$	$127 \cdot 14(4)$	$Pb^{+2} + C_6H_4(COO)_2^{-2}$ $126 \cdot 70$ (S) $126 \cdot 40$ (A)
$Pb(C_6H_5COO)_2 \cdot H_2O$ Pb found $=44 \cdot 1\%$ Pb theory $=44 \cdot 4\%$	208	$0 \cdot 4250$ $198 \cdot 60$ $185 \cdot 64$	$181 \cdot 20(4)$	$2Pb^{+2} + (C_6H_5COO)^{-1}$ $181 \cdot 30$ (S) $181 \cdot 00$ (A)
$Pb(C_6H_4OHCOO)_2 \cdot H_2O$ Pb found $=40 \cdot 9\%$ Pb theory $=41 \cdot 5\%$	224	$0 \cdot 0485$ $208 \cdot 90$ $195 \cdot 94$	$191 \cdot 38(4)$	$2Pb^{+2} + (C_6H_4OHCOO)^{-1}$ $190 \cdot 5$ (S) $190 \cdot 2$ (A)
$Pb_3[CH_2COO \cdot C(OH)COO CH_2COO]_2 3H_2O$ Pb found $=58 \cdot 80\%$ Pb theory $=58 \cdot 98\%$	440	$0 \cdot 3437$ .. $330 \cdot 80$ $291 \cdot 92$	$296 \cdot 34(4)$	$3Pb^{+2} + 2(CH_2COOCOH COO CH_2COO)^{-3}$ $307 \cdot 60$ (S) $306 \cdot 70$ (A)

studied by Kido and its value has been entered in column 3 along with the authors' values. The specific and the molecular susceptibilities of the hydrated and the anhydrous salts are given one below the other in column 3 of Table I. The molar susceptibilities of salts were also obtained by adding up the susceptibility values of the cations and the anions according to the method followed by Angus and Farquharson.[3] They are given in column 6 of Table I, and are marked as (S) and (A) according as Slater's and Angus' values are used. All the susceptibility values are expressed in $- 1 \times 10^{-6}$ c.g.s. units.

DISCUSSION OF RESULTS

It will be seen that in many cases values calculated according to Angus and Farquharson's[3] method do not agree with the experimental values. The observed differences are probably due to bond effects which have not been taken into consideration while calculating the susceptibility values of ions. This is borne out by the fact that there is definitely a better agreement in the case of organic compounds where the anion values have been computed by using Pascal's corrective constants than in the case of inorganic ones where purely theoretical values are used. There is a very good agreement between the experimental values of the authors and those calculated by using the values for the anions and the cations obtained by the same investigator (column 5).

(a) Comparison of lead salts with those of barium and thallium

Lead ordinarily behaves as a bivalent element giving salts isomorphous with those of bivalent alkaline earth metals, particularly those of barium. There is a marked resemblance in the physical properties such as crystalline form and stability of salts like chromates, oxalates, sulphates and phosphates of the two elements. Prasad, Dharmatti and Gokhale[2] have shown that on plotting the values of χ_{ma} against ΣZ for salts of calcium a pattern is obtained which is repeated in the case of the same salts of strontium and barium which are chemically similar. Hence values of χ_{ma} were plotted against ΣZ for the same salts of bivalent lead and barium. These curves are shown in Fig. 1.
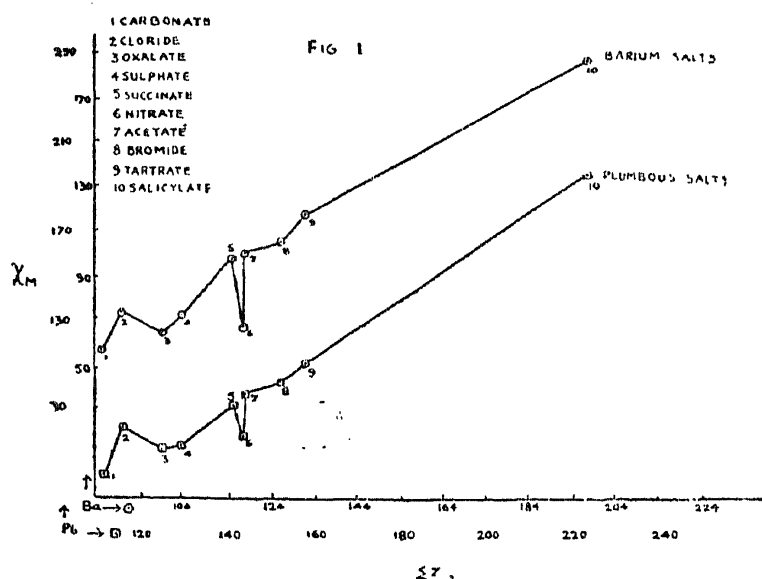

FIG. 1

The striking similarity between the two curves indicates the analogy between the two elements as suggested by the properties mentioned above.

Mellor[27] has pointed out similarity in physical properties of thallous and plumbous salts, such as appearance, specific gravity, hardness, melting point and electrical conductivity, and in their chemical behaviour, such as sparingly low solubility, the colour of the halides and chromates and the behaviour of aqueous solutions towards zinc. Electronically, the thallous and the plumbous ions possess identical electronic shells but differ only in atomic numbers. This analogy between the two elements has been further brought out by the identical patterns of the graphs between χ_{ma} and ΣZ shown in Figs. 2 and 3. In drawing these graphs a distinction has been made

Thallous salts with monovalent anions   Plumbous salts with monovalent anions

FIG. 2
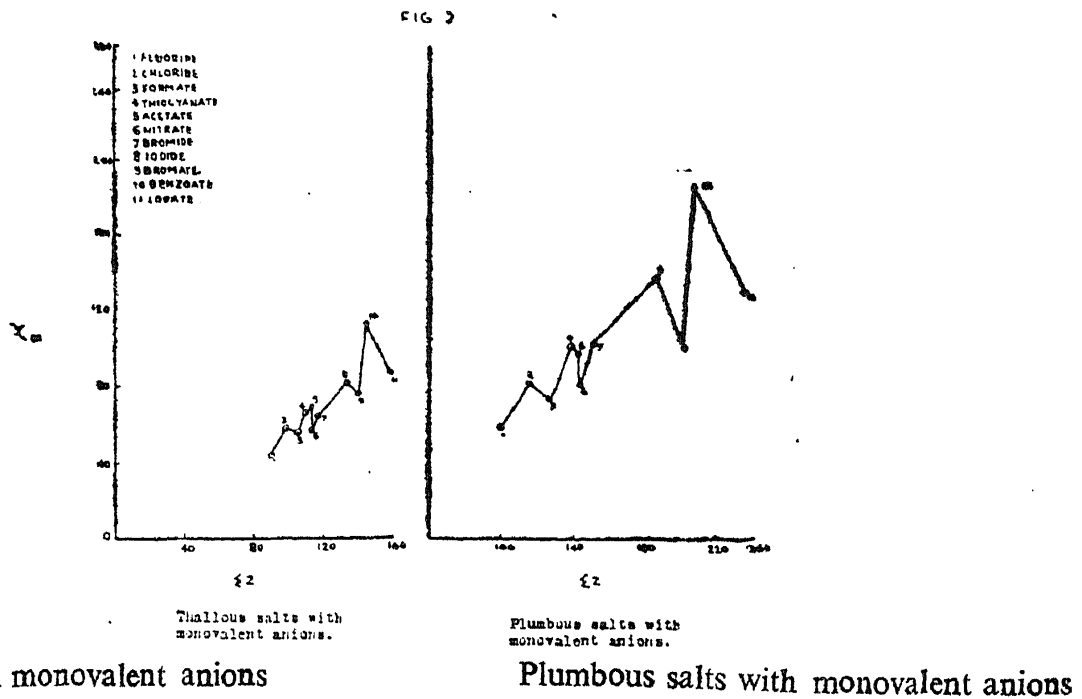

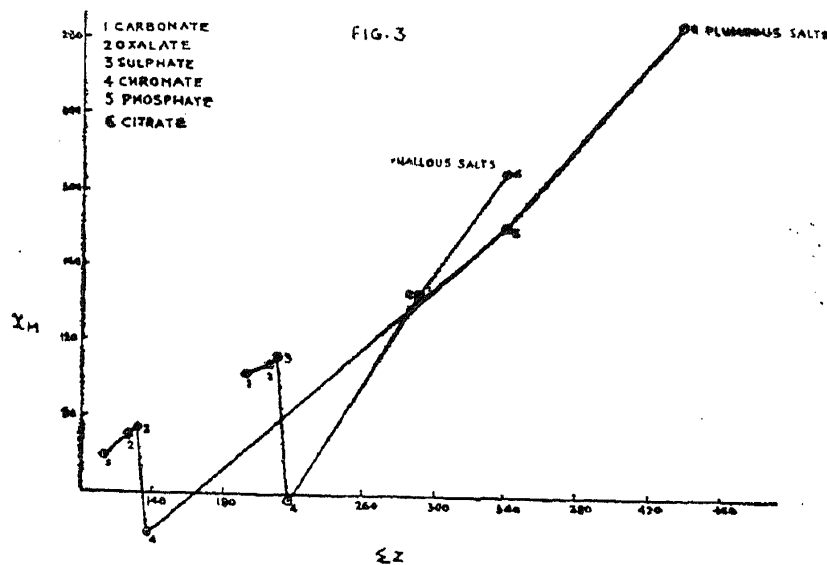Thallous and plumbous salts with Di- and trivalent anions

FIG. 3

between the salts of monovalent anions and those of divalent anions since the valencies of the plumbous and thallous ions are two and one respectively.

(b) Ikenmeyer's relation[6]

It will be seen from Fig. 1 that the plot of χ_{ma} against ΣZ is not a straight line and hence the Ikenmeyer's relation $(-\chi_M = C_1 N + C_2)$ does not hold in the case of plumbous salts. Same general conclusion has been drawn from the study of the several salts of some cations from this laboratory. However it was observed in the case of plumbous salts that if the values of χ_{ma} for (i) organic salts, (ii) $PbCl_2$, $PbBr_2$, PbI_2 and $Pb(CNS)_2$ and (iii) $PbSO_4$, $PbCO_3$, $Pb(NO_3)_2$, $Pb(IO_3)_2$ and $Pb(BrO_3)_2$ are separately plotted against ΣZ the curves obtained are straight lines. The inclinations (C_1) and the intercepts (C_2) of these straight lines on χ_{ma} axis are given below:

	C_1	C_2
(i)	$1 \cdot 32 \times 10^{-6}$	$-95 \cdot 37$
(ii)	$0 \cdot 75 \times 10^{-6}$	$-6 \cdot 00$
(iii)	$0 \cdot 57 \times 10^{-6}$	$-3 \cdot 54$

It may therefore appear that Ikenmeyer's relation holds for definite groups of anions. Further work concerning the elucidation of this viewpoint and assigning meanings to the values of C_1 and C_2 is in progress in this laboratory.

(c) Electronic Isomers

Values of the molecular susceptibilities of some of the lead salts were calculated, wherever possible, according to Bhatnagar and Mathur's[7] equation for electronic isomers, and are given in the following table.

TABLE II

No. of atoms	Total electrons	Electronic isomers	$\chi_{ma\,(obs.)}$	Radius in Å. U.	K	$\chi_{ma\,(cal.)}$
5	112	$TlNO_3$ $PbCO_3$	$56 \cdot 50$ $60 \cdot 36$	$2 \cdot 63$ $2 \cdot 45$	$2 \cdot 87$	$49 \cdot 1$
6	130	$TlClO_3$ $PbSO_4$	$72 \cdot 50$ $73 \cdot 36$	$3 \cdot 08$ $2 \cdot 94$	$2 \cdot 68$	$66 \cdot 0$

It will be seen from the Table that the calculated and the observed values differ from each other by amounts larger than the experimental error and hence lend support to similar conclusions arrived at by Prasad and co-workers.

(d) Ionic susceptibility of plumbous ion

The ionic susceptibility of the plumbous ion was determined by the same method as followed by Prasad and co-workers.[2] Various values of

Mata Prasad and others

TABLE III (a)

$\chi_{Pb^{++}}$ *from Salts of Inorganic Acids*

Compounds	χ_{ma}	χ_{anion}	$\chi_{Pb^{++}}$
$Pb(BrO_3)_2 \cdot H_2O$ ..	105·44	39·6 (1) 35·6 (8)	26·24 34·24
$PbBr_2$ ..	101·90	34·7 (9) 33·9 (10) 34·8 (6) 30·5 (4) 32·5 (11) 31·0 (12) 33·9 (13) 37·1 (1) 31·4 (14)	32·50 34·10 32·30 40·90 36·90 39·90 34·10 27·70 39·10
$PbCO_3$ ..	60·36	22·20(4) 28·10(1) 24·59(5) 29·50(8)	38·16 32·26 35·77 30·86
$PbCl_2$ ..	81·30	23·10(10) 22·10(1) 20·40(6) 20·10(4) 20·00(12) 23·10(13) 23·10(15) 22·20(16) 21·90(11) 25·30(17) 24·00(17) 24·20(18) 19·50(19)	35·10 37·10 40·50 41·10 41·30 35·10 35·10 36·90 38·70 30·70 33·30 32·90 42·30
$PbCrO_4$ ..	18·18	+21·86(20) +33·00(8)	40·04 51·18
$Pb(IO_3)_2$ ..	131·20	45·00(21) 47·40(8) 47·30(22) 46·75(5) 52·20(1)	41·20 36·40 36·60 37·70 26·80
PbI_2 ..	139·20	49·5 (10) 53·20(1) 44·60(4) 49·25(6) 50·20(11)	40·20 32·80 50·00 40·70 38·80
$Pb(NO_3)_2$ ..	76·85	20·10(1) 18·00(12) 18·40(23) 18·10(22) 19·50(22) 17·79(5) 14·20(4) 18·90(8)	36·65 40·85 40·05 40·65 37·85 41·27 48·45 39·05

TABLE III (a)—(Contd.)

Compounds	χ_{ma}	χ_{anion}	$\chi_{Pb^{++}}$
$Pb_3(PO_4)_2$ ..	181·70	35·40(4) 40·43(5) 47·20(1) 43·49(4)	36·97 33·61 29·10 31·57
$PbSO_4$ ..	73·36	33·60(4) 39·00(1) 39·00(8) 37·50(5) 40·10(8) 37·00(8)	39·76 34·36 34·36 35·86 33·26 36·36
$Pb(CNS)_2$ ..	100·60	30·00(8) 38·60(1)	40·60 24·60
PbS_2O_3 ..	83·60	43·00(1) 43·40(4)	40·60 40·20

TABLE III (b)

$\chi_{Pb^{++}}$ *from Salts of Organic Acids*

Compounds	χ_{ma}	χ_{anions}	$\chi_{Pb^{++}}$	Mean $\chi_{Pb^{++}}$
$Pb(CH_3COO)_2 \cdot 3H_2O$ ..	96·62	28·90(4) 32·00(1) 30·00(8) 30·00(21) 29·80(22) 28·76(4) 31·40(1) 30·20(22)	38·82 32·62 36·62 36·62 37·02 39·10 33·82 36·22	36·38
$Pb(C_6H_5COO)_2 \cdot H_2O$ ..	185·64	67·60(4) 66·10(8)	50·44 53·44	51·94
$Pb_3 \left(\begin{array}{c} CH_2{-}COO \\ \| \\ COH{-}COO \\ \| \\ CH_2{-}COO \end{array} \right)_2 \cdot 3H_2O$ ..	291·92	79·17(4)	44·53	44·53
$Pb(HCOO)_2$ ..	72·49	16·90(4) 20·20(1) 17·30(22) 17·60(22)	38·69 32·09 37·89 37·29	36·49
PbC_2O_4 ..	71·32	27·94(4) 28·50(21)	43·38 42·82	43·10
$PbC_6H_4(COO)_2$ ..	123·90	81·14(4)	42·76	42·76
$Pb(C_6H_4[OH]COO)_2 \cdot H_2O$ ..	195·94	72·69(4)	50·56	50·56
$Pb(CH_2COO)_2$ ..	91·77	51·66(4)	40·11	40·11
$Pb(CHOHCOO)_2$ ..	110·30	63·80(8) 60·88(4)	46·50 **49·42**	47·96

anions used are taken from literature and are given in column 3 of the Tables III (a) and III (b). Values of $\chi_{Pb^{++}}$ thus obtained are given in the last column of these tables.

Two separate mean values of $\chi_{Pb^{++}}$ were taken, one for the values deduced from plumbous salts of inorganic acids and the other for those deduced from salts of organic acids. These mean values are 36·83 and 40·94 respectively. It will be seen from these values that the value deduced from plumbous salts of organic acids is significantly higher than the one obtained from salts of inorganic acids. This difference was statistically established by the standard method. These results are in agreement with those obtained by Prasad, Dharmatti and co-workers, in the case of Ca, Sr and Ba. If, however, the mean values of $\chi_{Pb^{++}}$ deduced from individual organic salts are examined, it will be observed that the values found from formate and acetate agree fairly well with the mean value deduced from inorganic salts. In other cases the values of $\chi_{Pb^{++}}$ obtained are fairly high.

It can be seen from Table III (b) that the value of $\chi_{Pb^{++}}$ roughly increases with the number of carbon atoms in the anion. Trew,[8] who also observed such an increase, attributes it to the increase in the effective ionic radius of the cation which results from an increase in the size of the anions. These observations can also be explained on the basis of equilibrium inter ionic distances in a compound which again depend on (i) co-ordination number, (ii) radius ratio, (iii) amount of covalent bond character and (iv) crystal structure.

(e)   Susceptibility and Co-ordination[24] Number

Trew[8] has argued that according to Goldschmidt's theory regarding the influence of the co-ordination number on the interatomic distances and hence on the diamagnetic susceptibility, the diamagnetic susceptibility of a cation in combination with a univalent anion should be greater than that deduced from salts of anions of higher valencies. She found that the mean diamagnetic susceptibility of thallous ion deduced from salts containing univalent anion is $-38\cdot4 \times 10^{-6}$ while that deduced from salts containing divalent and trivalent anions is $-37\cdot4 \times 10^{-6}$. According to her these results support Goldschmidt's theory. Similar data were collected by the authors in the case of plumbous salts containing (a) univalent and (b) di- and trivalent anions, and are presented in Table IV.

It will be seen that the mean values of $\chi_{Pb^{++}}$ in the two cases are slightly different but the difference is not statistically significant. Hence it cannot be said with any definiteness that the results given in this paper for plumbous salts support Goldschmidt's hypothesis.

TABLE IV

(a) *Salts with Univalent Anions*

Compounds		$\chi_{Pb^{++}}$	Mean $\chi_{Pb^{++}}$
$Pb(NO_3)_2$	..	40·60	
$PbCl_2$	..	36·91	
$PbBr_2$	..	35·28	
PbI_2	··	40·50	37·12
$Pb(IO_3)_2$	..	35·74	
$Pb(BrO_3)_2$	..	30·24	
$Pb(CNS)_2$	..	40·60	

(b) *Salts with di- and trivalent anions*

$PbSO_4$	..	35·66	
$PbCO_3$	..	34·26	
$PbCrO_4$	..	45·61	37·75
PbS_2O_3	··	40·40	
$Pb_3(PO_4)_2$	..	32·81	

(f) *Theoretical and Experimental Ionic Susceptibilities*

The susceptibility values of Pb^{++} according to Slater's and Angus' methods are 50·2 and 49·9, respectively. Employing the mass susceptibility data for plumbous compounds quoted in the I.C.T. (Vol. VI, page 357), Trew finds the susceptibility for the plumbous ion to be 44·3. She considers that this value is probably high " due to the tendency to convalency " in plumbous compounds. According to her, the value of the plumbous ion deduced from strictly polar compounds should be 38·0. This value agrees fairly well with the experimental value, 36·83, deduced from inorganic salts which is slightly less than 4/5 of the theoretical value. In the case of the thallous ion, which is an electronic isomer of Pb ion, Trew[8] observes that the experimental value was less than $\frac{2}{3}$ of the theoretical value. However, Prasad, Dharmatti and co-workers[2] have found that in the case of calcium, strontium and barium ions, the ionic susceptibilities deduced from the inorganic salts are in fair agreement with the theoretical ones determined by the methods of Slater and Angus. Brindley[25] has observed that there is an increasing divergence between the theoretical and experimental susceptibilities with the increasing atomic weight of the element concerned. This is, however, not borne out from the results obtained in the case of barium. The discrepancies noticed in the case of plumbous and thallous ions and those observed by Brindley suggest that some modification in Slater's method of calculation, probably in the evaluation of the screening constants, is necessary.

(g) *Ionic Radius*

The ionic radius of Pb^{++} has been calculated from the mean ionic suscepti-bilities, from inorganic as well as organic salts. The results are given below:

TABLE V
Radius of Pb^{++} ion

From χPb^{++} deduced from Inorganic Salts	From χPb^{++} deduced from Organic Salts	From X-ray data (Goldschmidt's value)
1·260 Å.U.	1·328 Å.U.	1·320 Å.U.

It will be seen that the experimental values agree very well with Goldschmidt's[24] value of Pb^{++} from X-ray data.

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