263. *Delocalization and Magnetic Properties of the Phosphonitrilic Halides.*

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The magnetic anisotropy due to π-electron delocalization is calculated for systems of alternating $p\pi$- and $d\pi$-orbitals, and compared with that for $p\pi$–$p\pi$ systems. Calculations on the London model show a strong dependence upon the electro-negativity difference between the $p\pi$- and $d\pi$-orbitals, and for realistic values the contribution is small compared with that in benzene. If the $d\pi$-orbitals are d_{xz}, tangentially directed, the π-electron contribution is small and paramagnetic; if d_{yz}-orbitals participate also, their influence is opposite in sign, leading to a steadily diminishing paramagnetic behaviour until, with equal participation by d_{xz}- and d_{yz}-orbitals, the π-electron ring current contribution is close to zero

Measurement of diamagnetic anistropy are reported for $(PNCl_2)_3$, and of average susceptibility for $(PNCl_2)_n$, $n = 3$, 4, 5, 6, and 7. These measurements are compatible with a small paramagnetic or zero contribution by π-electrons but cannot be interpreted in detail to settle the degree of participation by d_{yz}-orbitals.

Energy calculations including d_{yz}-orbitals suggest that in the phosphonitrilic compounds these orbitals are used to some extent as well as the dominant d_{xz}-type.

We first report two sets of calculations on cyclic systems of alternating $p\pi$- and $d\pi$-orbitals, such as occur in the phosphonitrilic and thiazyl halides,[1,2] and in some other ring systems. The effect on the π-electron energetics of the inclusion of the second, radially directed $d\pi$-orbital (d_{yz} in the convention of ref. 2) is first considered, and then the contribution to the diamagnetic susceptibility made by π-electrons which are delocalized in $p\pi$–$d\pi$ systems is calculated and compared with that in $p\pi$–$p\pi$ systems.

The method for dealing with the energetics of delocalization involving both d_{xz}- and d_{yz}-types of $d\pi$-orbitals in a $p\pi$–$d\pi$ system was given some time ago.[3] The systems treated are planar cyclic molecules of 4, 6, and 8 ring atoms, providing alternately $p\pi$-orbitals and a pair of $d\pi$-orbitals. The methods are those of simple molecular-orbital theory, framed in terms of a single resonance parameter δ, defined as the resonance integral for the interaction of neighbour $p\pi$- and $d\pi$-orbitals *oriented for maximum overlap.** It is easily shown that the resonance integrals for the atoms arranged in planar rings are $h_{xz}\delta$ and $h_{yz}\delta$, with the h's given by the cosines of the angles made by the internuclear axis with the tangential and radial directions respectively. The electronegativity of the tangential (d_{xz}) orbital is α; that of the $p\pi$-orbital, being more electronegative, is $\alpha + \rho\delta$, ρ being a parameter taking zero or positive values. The d_{yz}-orbital will have (i) electronegativity α, or (ii) $\alpha - \delta$, or (iii) for purposes of comparison it will be supposed not to take part at all, as in earlier calculations. Since the d_{xz}-orbital will be more strongly perturbed by neighbouring atoms than the d_{yz}, the only situations that are realistic are those in which d_{xz} is made the more electronegative atomic orbital. Thus cases (i) and (iii) are limiting cases, and (ii) is a representative intermediate case.

The general secular equation for orbital energies may be written as follows, with the substitution $x = (\alpha - E)/\delta$, and with an unspecified electronegativity parameter α_{yz} for the d_{yz}-orbital:

$$\begin{vmatrix} x + \rho & 2i \sin (l\pi/n)\, h_{xz} & 2 \cos (l\pi/n)\, h_{yz} \\ -2i \sin (l\pi/n)\, h_{xz} & x & 0 \\ 2 \cos (l\pi/n)\, h_{yz} & 0 & (\alpha_{yz} - E)/\delta \end{vmatrix} = 0 \quad . \quad . \quad . \quad (1)$$

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* This change from the definition of refs. 1, 2, and 3, helps the formulation of the two $d\pi$-orbital problem.

[1] Craig and Paddock, *Nature*, 1958, **181**, 1052.

[2] Craig, *J.*, 1959, 997.

[3] Craig, *Chem. Soc. Spec. Publ.* No. 12, 1958, p. 343.

in which l is the ring quantum number and $2n$ is the number of atoms in the ring. By solving this equation, summing the energies of the occupied orbitals, and subtracting the energy of a " structure " consisting of n two-electron bonds made up from the $p\pi$-orbital on the nitrogen atoms and the pair of appropriately directed $d\pi$-orbitals on the phosphorus atoms we get the delocalization energy. This quantity, divided by the number of π-electrons in the system, is listed in Table 1 for several cases.

TABLE 1. *Delocalization energy per π-electron in* pπ–dπ *systems.**
(*In units of the resonance parameter* δ).

	$\rho = 0$	$\rho = 0{\cdot}5$	$\rho = 1{\cdot}0$	$\rho = 2{\cdot}0$
$n = 2$	0·41	0·41	0·38	0·32
	0·35	0·36	0·34	0·29
	0	0·07	0·12	0·14
$n = 3$	0·39	0·38	0·36	0·30
	0·30	0·32	0·32	0·30
	0·13	0·19	0·22	0·22
$n = 4$	0·43	0·42	0·40	0·34
	0·29	0·30	0·31	0·28
	0·19	0·23	0·25	0·24

* The first value in each block is calculated for equal electronegativities of d_{xz} and d_{yz}, the second for d_{yz} less electronegative by δ, and the third for participation by d_{xz} alone.

The gain in delocalization energy due to inclusion of the d_{yz}-orbital is relatively greater in the systems of 4 π-electrons than in those of 6 and 8. Thus, if in the real molecules the electronegativities of the d-orbitals permit substantial participation by both types, the 4- and 8-electron systems could be more stabilized than the 6-electron system. This might be described as *anti*-Hückel behaviour, since it is contrary to the Hückel rule which, in the $p\pi$–$p\pi$ aromatic series, associates aromatic behaviour with 6, 10, 14 . . . electron systems, and not with 4, 8, 12 . . . systems. The delocalization energies per π-electron for the cases $\rho = 0$ and 1 are given in Fig. 1. It is necessary to emphasise that these results are in terms of a common unit δ; and it is an approximation to neglect variations of δ with ρ and to assume, in the second entry in each block of Table 1, that the same δ applies to the two different $p\pi$–$d\pi$ resonance integrals. Moreover, the total population of the d-orbitals is greater the more available the d_{yz}-orbital is assumed to be: this increase in population should perhaps be reflected in a promotion energy term. That is, the total energy of the system may be higher when both d-orbitals are included on account of increased intra-atomic electron repulsion. This would reduce the energetic advantage shown in Table 1 for the first two entries in each block. In spite of these points, however, it seems clear that use of the d_{yz}-orbitals results in some energy gain, and therefore that they will be used to some extent in the delocalized system.

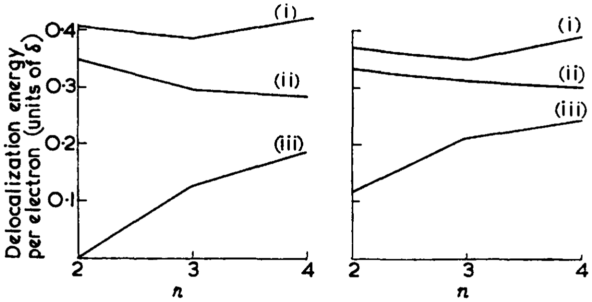

FIG. 1. *Delocalization energies for various* (AB)$_n$ *pπ–dπ systems.*

(i) *Including* d_{xz} *and* d_{yz} *orbitals with equal electronegativity* α. (ii) *Including* d_{xz} *and* d_{yz} *orbitals, with* d_{yz} *less electronegative by* δ. (iii) d_{xz} *Orbital alone.*

Left-hand diagram: $\rho = 0$
Right-hand diagram: $\rho = 1$

Although the qualitative features of the energy calculations are not much changed by the inclusion of the d_{yz}-orbital, there is a considerable influence on the contribution made by the delocalized electrons to the diamagnetic susceptibility. It was shown many years ago by London[4] that the exaltation of diamagnetism measured perpendicular to the ring plane in normal aromatic molecules may be accounted for by considering the induction of peripheral electron currents in the π-electron system. London's methods may be applied to deal with alternating $(AB)_n$ $p\pi$–$p\pi$ systems and $p\pi$–$d\pi$ systems. The first permit the calculation of the changed diamagnetism of molecules such as 1,3,5-triazine and borazole in which electronegativity differences occur in the ring atoms. The results in this case are in no way surprising (Fig. 2, upper half). They show that in $p\pi$–$p\pi$ delocalization the diamagnetism due to the induced ring current dies away as the electronegativity difference increases, *i.e.*, it decreases as the delocalization decreases. The dependence of the orbital energies on the magnetic field $\boldsymbol{H}$ is expressed in the secular equation (2):

$$\begin{vmatrix} x + \rho & 2\cos(l\pi/n + \theta) \\ 2\cos(l\pi/n + \theta) & x \end{vmatrix} = 0 \quad . \quad . \quad . \quad . \quad . \quad (2)$$

The two species of ring atom have electronegativities differing by ρ units of the appropriate resonance integral, and the field dependence is contained in the angle θ, $\theta = 2\pi \boldsymbol{H}\beta S/2n$, where S is the (signed) area of the ring, and $\beta = e/hc$.[5] The cosine form of the off-diagonal elements ensures that the binding energies of the occupied orbitals will be reduced or left unchanged in the presence of a field, leading to diamagnetic behaviour. Also, as ρ increases, corresponding to an increased electronegativity increase between the ring atoms, the influence of the off-diagonal terms is less, allowing the diamagnetism to fall away as shown in Fig. 2.

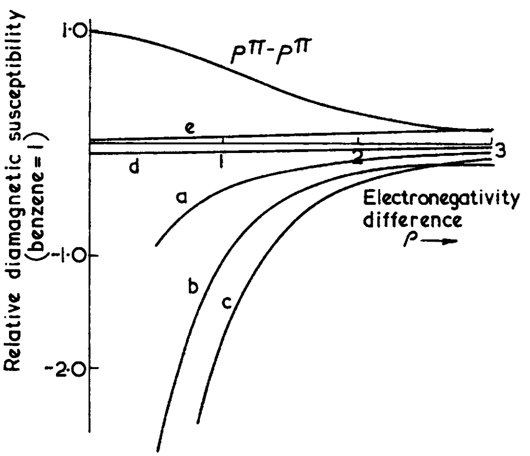

FIG. 2. *Contributions to the magnetic susceptibility normal to the ring plane for* $(AB)_n$ *ring systems.* ρ *is the electronegativity difference between the participating orbitals in units of the resonance integral. The susceptibility unit is the calculated value for benzene,* δ *being assumed constant.*

Top curve: normal six-membered $p\pi$–$p\pi$ *aromatic series.*

(a) d_{xz}- *and* $p\pi$-*orbitals only, in a* 4-*ring.* (b) d_{xz} *and* $p\pi$, 6-*ring.* (c) d_{xz} *and* $p\pi$, 8-*ring.* (d) d_{xz}, d_{yz}, *and* $p\pi$, 6-*ring.* (e) d_{xz}, d_{yz}, *and* $p\pi$, 8-*ring.*

For $p\pi$–$d\pi$ systems we first consider the magnetic properties of a set of $p\pi$- and d_{xz}-orbitals only, as in case (iii) heretofore. The secular equation for the energies in a magnetic field, in the units as before, is:

$$\begin{vmatrix} x + \rho & 2i\sin(l\pi/n + \theta) \\ -2i\sin(l\pi/n + \theta) & x \end{vmatrix} = 0 \quad . \quad . \quad . \quad . \quad (3)$$

For occupied orbitals with $l \neq 0$ the effect of the field is, again, to reduce the binding energy, giving a diamagnetic contribution, but a special situation occurs for the orbitals $l = 0$. In the absence of the field these two orbitals have different symmetries[2] and do not interact: the more stable (occupied) orbital consists solely of the more electronegative of the atomic orbitals, namely $p\pi$, and the less stable consists only of the less electronegative,

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[4] London, *J. Phys. Radium*, 1957, **8**, 397.

[5] McWeeny, *Proc. Phys. Soc.*, 1951, **64**, *A*, 921.

namely d_{xz}. However, the magnetic field causes these molecular orbitals to interact with one another with the result that the lower, occupied, one is increased in binding energy. Electrons in this orbital behave paramagnetically, and their contribution outweighs the diamagnetism of the other electrons in the occupied orbitals, giving an overall paramagnetic contribution perpendicularly to the ring plane. This behaviour is illustrated in the lower half of Fig. 2. This effect gets less as the parameter ρ increases, as expected. As ρ drops toward zero the paramagnetism apparently increases without limit, but this is a false result because near $\rho = 0$ the single configuration approximation breaks down, and it would be essential to consider the orbital degeneracy problem as in the analogous example of cyclobutadiene.[6] Thus the magnetic instability of the $p\pi$-d_{xz} system suggested by Fig. 2 is not real. The considerations outlined for $p\pi$–d_{xz} systems hold word for word in $p\pi$–$p\pi$ systems of $4m$-electrons, *e.g.*, the plane regular octagonal model of cyclo-octatetraenes and its $(AB)_4$ analogues.

The inclusion of both d_{xz}- and d_{yz}-orbitals changes the expected magnetic behaviour completely. The secular equation including the field is found from (1) by replacing the angle $(\pi l/n)$ by $(\pi l/n + \theta)$; as usual, the diamagnetism is equal to the coefficient of $-(1/2)\boldsymbol{H}^2$ in the expression for the total energy as a function of $\boldsymbol{H}$. The examples given in Fig. 2 are for equally electronegative $d\pi$-orbitals: it is found that the π-electrons in these cases make a negligibly small contribution through delocalization. In the 4-membered ring the contribution is zero for all values of ρ. In the 6-membered ring it is feebly paramagnetic and in the 8-membered feebly diamagnetic. The contributions are all small compared with those from $p\pi$–$p\pi$ delocalization or for $p\pi$–d_{xz}.

It will be observed that the effect of d_{yz}-orbital participation in the delocalization is to add to the stabilization produced by $p\pi$–d_{xz} interaction, but to oppose the paramagnetic effect by a diamagnetic one which cancels it almost exactly. The magnetic results, applying as they do to case (i) of equally electronegative $d\pi$-orbitals, are for the extreme situation. The magnetism of examples with less electronegative d_{yz}-orbitals is more difficult to calculate, but from the form of the relationship it is clear that the results would be intermediate between the $p\pi$–d_{xz} case and the magnetically isotropic case of equally electronegative $d\pi$ orbitals.

Dewar *et al.*[7] have recently argued that the situation in $p\pi$–$d\pi$ systems is in fact that of equally electronegative d_{xz}- and d_{yz}-orbitals, as illustrated in the curves labelled (i) (Fig. 1) and (d) and (e) (Fig. 2). Dewar *et al.* show that the π-electron system then consists of " islands " or conjugated zones stretching over a nitrogen atom and two adjacent phosphorus atoms, with little conjugation between different islands. This leads them to conclude that the delocalization is not over the entire ring and therefore that the systems should not be described as aromatic. We think that this conclusion is not well founded, because it depends upon an exact, or nearly exact, equality of the electronegativities of the two $d\pi$-orbitals. If they are unequal, irrespective of which is the greater, the delocalization does involve the whole cyclic system to an extent which increases the more the two orbitals differ. Now there is nothing in the problem to support the idea of *exactly* equal $d\pi$-orbitals. The weakly bound d-orbitals of free phosphorus are easily polarizable by ligand atoms, and the degree of polarization of an orbital depends upon its directional properties in relation to the ligand environment. This is not symmetrical in relation to the two $d\pi$-orbitals, which must be differently affected by it. Dewar's procedure of simply taking for granted, in spite of this, that the orbitals are of equal electronegativity, and then drawing conclusions which depend critically on their equality, is open to question. We think that the relative electronegativities of d_{xz} and d_{yz} is an open question that will have to be settled by testing theory against experiment. Magnetic properties are interesting in this connection but, as will be shown, are not yet conclusive.

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[6] Craig, *Proc. Roy. Soc.*, 1950, *A*, **202**, 498.

[7] Dewar. Lucken. and Whitehead, *J.*, 1960, 2423.

EXPERIMENTAL

Results.—The results to be described are (i) the diamagnetic anistropy of single crystals of $(PNCl_2)_3$ and (ii) the average susceptibility of $(PNCl_2)_n$ for $n = 3, 4, 5, 6$, and 7.

The diamagnetic anistropy of single crystals of $(PNCl_2)_3$, determined by the critical torsion method,[8] is: $\chi_c - \chi_b = -8\cdot5 \times 10^{-6}$, $\chi_c - \chi_a = -6\cdot8 \times 10^{-6}$, and $\chi_a - \chi_b = -1\cdot7 \times 10^{-6}$ c.g.s. unit. Coupled with the mean susceptibility $\frac{1}{3}(\chi_a + \chi_b + \chi_c)$ of $-149\cdot3 \times 10^{-6}$ given by the Gouy method, these values lead to the following susceptibilities in the directions of crystal axes: $\chi_a = -147\cdot6 \times 10^{-6}$, $\chi_b = -145\cdot9 \times 10^{-6}$, and $\chi_c = -154\cdot4 \times 10^{-6}$ c.g.s. unit.

The molecular susceptibilities may be found from the crystal values by using the results of a recent *X*-ray analysis.[9] The molecules of $(PNCl_2)_3$ have a nearly planar skeleton of phosphorus and nitrogen atoms and a three-fold symmetry axis normal to the plane; the two in-plane susceptibilities K_1 and K_2 should therefore be equal. The value measured along a crystal axis is the sum of contributions by the principal molecular susceptibilities each multiplied by the square of the appropriate direction cosine.[10] By inverting this relationship we find the molecular susceptibilities: $K_1 = -145\cdot8 \times 10^{-6}$, $K_2 = -145\cdot9 \times 10^{-6}$, and $K_3 = -156\cdot3 \times 10^{-6}$ c.g.s. unit. The molecular diamagnetic anistropy of $(PNCl_2)_3$ is then:

$$\Delta K = K_3 - \tfrac{1}{2}(K_1 + K_2) = -10\cdot5 \times 10^{-6} \text{ c.g.s. unit}$$

Measurements by the Gouy method of the average susceptibilities of a series of cyclic polymers $(PNCl_2)_n$ are summarized in Table 2. The preparation and characterization of these compounds are described elsewhere.[11]

TABLE 2. *Average susceptibilities* $\bar{\chi}_M$ *for* $(PNCl_2)_n$.

Compound	$(PNCl_2)_3$	$(PNCl_2)_4$	$(PNCl_2)_5$	$(PNCl_2)_6$	$(PNCl_2)_7$
$\bar{\chi}_M$ ($\times 10^{-6}$ c.g.s.)	$-149\cdot3$	$-196\cdot8$	$-248\cdot1$	$-300\cdot5$	$-342\cdot5$
Susceptibility per $PNCl_2$	$-49\cdot8$	$-49\cdot2$	$-49\cdot6$	$-50\cdot1$	$-48\cdot9$

DISCUSSION

The diamagnetic anistropy of benzene is -60×10^{-6} c.g.s. unit; that of $(PNCl_2)_3$ is $-10\cdot5 \times 10^{-6}$, and the analysis of this much smaller value requires an examination of the terms that can contribute to it. Hoarau[12] showed that in benzene only a part of the observed anistropy is due to ring currents. He discussed the anistropy of benzene and other aromatic hydrocarbons according to the scheme:

$$\Delta K = \Delta K_{\text{London}} + \Sigma\Delta k_\sigma + m\Delta k_\pi$$

The first term is the anisotropy according to the London theory, *i.e.*, the component due to ring currents in the delocalized π-electrons, and is the term of present interest. The second term represents the anisotropy due to σ-electrons, and the third arises from the π atomic orbitals,[13] and acts in the sense of reducing the in-plane components K_1 and K_2. It follows from Hoarau's analysis of the molecular susceptibilities in aromatic hydrocarbons that the ring current contribution is given to a good approximation by the expression:

$$\Delta K_{\text{London}} = K_3 - \Sigma\chi_A$$

We make the heuristic assumption, which cannot yet be adequately tested, that this formula applies generally, and use it in Table 4 to obtain ΔK_{London} for several molecules relevant to the discussion of $(PNCl_2)_3$. Table 3 lists the essential atomic susceptibility constants. The two benzene derivatives in Table 4 show large delocalization anisotropies,

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[8] Krishnan and Banerjee, *Phil. Trans.*, 1934, **234**, 265.
[9] Wilson and Carroll, *J.*, 1960, 2548.
[10] Lonsdale and Krishnan, *Proc. Roy. Soc.*, 1936, *A*, **156**, 597.
[11] Lund, Paddock, Proctor, and Searle, *J.*, 1960, 2542.
[12] Hoarau, *Ann. Chim.* (*France*), 1956, **13**, 544.
[13] Berthier, Mayot, Pullman, and Pullman, *J. Phys. Radium*, 1952, **13**, 15.

both somewhat reduced from benzene itself (-38×10^{-6}). Heterocyclic molecules, however, show very small values, reflecting the sensitivity to electronegativity differences apparent in the theoretical calculations illustrated in Fig. 2. These residual anisotropies are too small to be significant, as is shown by the negative result for cyanuric chloride.

TABLE 3. *Atomic susceptibility constants* ($-\chi_A \times 10^{-6}$ *c.g.s.*).

H	2·0	Cl	18·5
N	9·0	B	7·2 *
C	7·4	P	10·0 *

* Taken from "Selected Constants and Numerical Data, Diamagnetism and Paramagnetism," Masson, Paris, 1957, Vol. VII, p. 222.

TABLE 4. *Molecular susceptibilities* (*units*, -10^{-6} *c.g.s.*).

Compound	K_1	K_2	K_3	ΔK	$\frac{1}{2}(K_1 + K_2)$	$\Sigma\chi_A$	$K_3 - \Sigma\chi_A$
p-Dichlorobenzene [14]	75·8	63·8	117	47·2	69·8	89·4	27·6
Hexachlorobenzene [8]	128	128	182	54	128	155·4	26·6
2,4,6-Trichloro-1,3,5-triazine [15]	70·9	71·2	101·3	30·3	71	104·7	−3·4
1,3,5-Trichloroborazole [15]	97·6	97·3	115·3	17·9	97·4	110·1	5·2
$(PNCl_2)_3$	145·8	145·9	156·3	10·5	145·8	168	−11·7
Anthracene [17]	77·2	74·7	250·7	174·7	76	123·6	127·1
Phenazine [17]	55·9	54·7	222·4	167·1	55·3	122·8	99·6

The value $+11·7 \times 10^{-6}$ for $(PNCl_2)_3$ is based upon $-10·0 \times 10^{-6}$ for the atomic susceptibility of phosphorus. This value for the ring-current anisotropy, though uncertain in size, suggests that the π-electron contribution may in fact be of opposite sign to that in the benzene-like aromatic series, in agreement with theoretical expectation.

We must, however, consider further the atomic susceptibility of phosphorus upon which the result for $(PNCl_2)_3$ depends critically. It is possible to make use of the in-plane susceptibilities to get another estimate. For the molecules listed in Table 4 it is clear that $\frac{1}{2}(K_1 + K_2)$ is always less than $\Sigma\chi_A$ and especially so when sp^2-hybridized nitrogen is involved. Chlorine, however, makes its normal contribution in *p*-dichlorobenzene and in hexachlorobenzene, and will be assumed to do so also in $(PNCl_2)_3$. For the determination of the in-plane contribution by nitrogen we have the values for 2,4,6-trichloro-1,3,5-triazine and for phenazine. From the former we find that nitrogen contributes $-1·2 \times 10^{-6}$ to the in-plane susceptibility, and from the latter $+1·95 \times 10^{-6}$, instead of the average value of $-9·0 \times 10^{-6}$ given in Table 3 and based on studies of saturated nitrogen compounds. The difference between the two determinations of the in-plane value is disturbing but not surprising, in view of the crudity of the procedure. By using these values in $(PNCl_2)_3$ we can get estimates of the in-plane contribution by phosphorus. The six chlorine atoms contribute -111×10^{-6} c.g.s. unit, and the three nitrogens either $-3·6$ or $+5·9 \times 10^{-6}$. In the first case the difference between the measured in-plane susceptibility of $-145·8 \times 10^{-6}$ and the $-115·8$ from the chlorine and nitrogen atoms is $-30·0$, and in the second it is $-40·7$, leading to values for phosphorus of $-10·0$ and $-13·6 \times 10^{-6}$ respectively. The anistropy of the atomic susceptibility being supposed small, as is likely in a nearly tetrahedrally hybridised atom, the calculated in-plane value should be close to the average χ_A. The first value of $-10·0 \times 10^{-6}$ is, in fact, equal to the value given in Table 3 derived from non-cyclic saturated phosphorus compounds, and gives $+11·7 \times 10^{-6}$ for the (paramagnetic) ring-current anisotropy term. The second leads to an even higher paramagnetic anistropy of $+22·4 \times 10^{-6}$.

We consider finally the mean susceptibilities of the higher polymers, listed in Table 2. They show that the increment of susceptibility per $PNCl_2$ unit is closely constant. We

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[14] Lemanceau, *J. Chim. phys.*, 1959, **56**, 933.

[15] Lonsdale, *Z. Krist.*, 1936, **95**, 471.

[16] Lonsdale and Toor, *Acta Cryst.*, 1959, **12**, 1048.

[17] Leela, Thesis, University of London, 1958.

may deduce either that the ring-current contributions to $\bar{\chi}_M$ are small in all cases, or that the ring-current and the in-plane terms change in opposite, counteracting, senses as the size of the polymers increases. Although the latter seems unreasonable theoretically it cannot be ruled out by analysing experimental results now available. Our conclusion, which must therefore be tentative, is that both the diamagnetic anistropy of $(PNCl_2)_3$ and the mean susceptibilities of the several polymers are compatible with a ring-current contribution that is small, and probably paramagnetic, in all cases.

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