Vibrational Spectroscopy at Very High Pressures. Part 15.[1] Raman and Infrared Study of Mercury(II) Chloride †

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Raman spectra at 295 and 150 K, and i.r. spectra at 295 K, are given for $HgCl_2$ (phases I, II, and IV) at hydrostatic pressures up to 30 kbar. The ultramicrosampling technique used with the diamond-anvil high-pressure cell is described. The small spectral differences between phases I and IV are interpreted in terms of a second-order transition with retention of space group. The profound spectral differences accompanying the IV–II transition are accommodated by postulating that $HgCl_2$(II) has the cubic $CO_2(T_h^6)$ structure. Mechanisms for the transitions are proposed.

FROM compression measurements Bridgman found that $HgCl_2$ showed only one phase transition up to 45 kbar † at ambient temperature.[2] A recent n.q.r. study has revealed a second-order transition below this[3] as indicated in Figure 1. We have investigated all the three phases by both i.r. and Raman spectroscopy under conditions of, or near, hydrostatic compression up to *ca.* 40 kbar. This is the first study of combined i.r. and Raman spectroscopy under such conditions. The principal results have been outlined elsewhere.[4] The phase diagram of Figure 1 is based on that given by Pistorius,[5] with addition of the data of ref. 3 and of our own further investigations of the I–IV transition line.

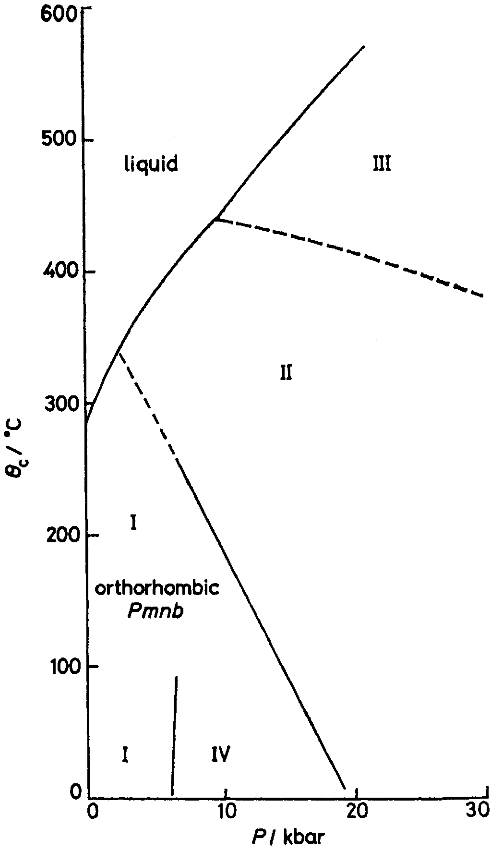

FIGURE 1 Phase diagram of $HgCl_2$

EXPERIMENTAL

The high-pressure device used was a diamond anvil cell (d.a.c.) developed from one described earlier.[6] A principal difference from the older design was that the fixed-anvil holder was extended so that either a heater or a cooling coil could be mounted on it. An insulating ring of either Paxolin (for low-temperature use) or Sindanio asbestos (for high temperatures) was inserted between the cell body and the barrel in which the anvil holders are mounted.[7]

Samples were contained within molybdenum gaskets initially 0.05-mm thick and 0.4 mm in diameter. Inconel gaskets could not be used as there was a reaction with $HgCl_2$. Within this hole the solid $HgCl_2$ was immersed in a pressure-transmitting fluid. This was either Nujol (Raman, i.r.), silicone oil (Raman; i.r. in 20—200 cm^{-1} region), or oct-1-ene (i.r. 40—400 cm^{-1}). These liquids all freeze at lower pressures than methanol–ethanol (4 : 1),[8,9] but this mixture cannot be used for our experiments as it dissolves $HgCl_2$. Moreover, methanol–ethanol is effectively useless for i.r. work as it has strong i.r. absorption.

Pressure calibration was carried out using the R_1 and R_2 fluorescence lines of a ruby[8,9] chip which was loaded together with the $HgCl_2$. This is simple and direct for Raman experiments but there is, as yet, no equivalent internal calibrant for i.r. use. Accordingly, after obtaining an i.r. spectrum at elevated pressure the cell, still under pressure, was transferred to the Raman spectrometer and the R_1 and R_2 line positions were determined. The ruby chip was too small to show i.r. absorption.

I.r. spectra were obtained using a Beckman-RIIC FS-620 Fourier spectrometer extensively modified for use in the off-axis mode. We emphasise the severe technical difficulties involved in obtaining spectra through a gasket hole of 0.4-mm in diameter which, moreover, decreases in diameter as pressure is applied. Less than 0.1% incident light is transmitted to the detector.[7] Raman spectra were obtained using a Coderg T800 triple-monochromator instrument with an E.M.I. 9558B tube and d.c. detection. 514.5-nm Excitation was used. Details of the optical system used to couple the d.a.c. to the monochromator are given elsewhere.[1] The low-temperature spectra were obtained with the d.a.c. mounted inside a large vacuum enclosure (in fact, the sample module of a defunct Beckman-RIIC FS520 Fourier spectrometer) provided with hydraulics,

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† No reprints available.

‡ Throughout this paper: 1 bar = 10^5 Pa.

[1] Part 14, D. M. Adams, R. Appleby, and S. K. Sharma, *Appl. Optics*, 1977, **16**(7), in the press.

[2] P. W. Bridgman, *Proc. Amer. Acad. Arts Sci.*, 1937, **72**, 45.

[3] D. B. Balashov and D. A. Ikhenov, *Fiz. Tverd. Tela*, 1975, **17**, 2693.

[4] D. M. Adams and R. Appleby, *J.C.S. Chem. Comm.*, 1976, 975.

[5] C. W. F. T. Pistorius, *Progr. Solid-State Chem.*, 1976, **11**, 1.

[6] D. M. Adams, K. Martin, and S. J. Payne, *Appl. Spectroscopy*, 1973, **27**, 377.

[7] D. M. Adams and S. K. Sharma, *J. Phys.* (*E*), in the press.

[8] G. J. Piermarini and S. Block, *Rec. Sci. Instr.*, 1975, **46**, 973 and refs. therein.

[9] D. M. Adams, R. Appleby, and S. K. Sharma, *J. Phys.* (*E*), 1976, **9**, 1140.

TABLE 1

Raman bands (cm^{-1}) for $HgCl_2$ at various pressures

Pressure/kbar	ν_L (translatory)					ν_L (rotatory)			ν_1	ν_3	Phase
(*a*) At 295 K											
0.001	18	26	43		74	124		168	315	384	I
2.0	18.5	26	41.5	49	75	128		167	315	384	I
4.6	20	26	42.5	51.5	76.5	129	139	168.5	315	385	IV
8.6	21.5		41	51	77	133	144	170	316	386	IV
13.8	22		41	53	80	139	152		316		IV
18.4	22		42		79		161		314		II
23.0							171		312		III
29.8							178		312		II
(*b*) At 150 K											
0.001	18.5	26, 29	48		77	126	134		315	388	I
0.8	18.5	29	49		78	126	135		317, 325 (sh)	388	I
9.3	22		54, 58		80	136	150		318	390	IV
21.0	24				83	146.5	163.5		318	393	IV
37.3	27				87	160	184		318	395	IV + II
38.1	27				88		184		318	396	IV + II

electrical leads, and a flow of liquid nitrogen which was passed through a coil wrapped around the extended nosepiece of the fixed-anvil holder of the d.a.c. Temperature was measured with a thermocouple attached to the fixed diamond anvil with silver paste. The diamonds used were type IIB stones chosen according to our criteria.[10,11]

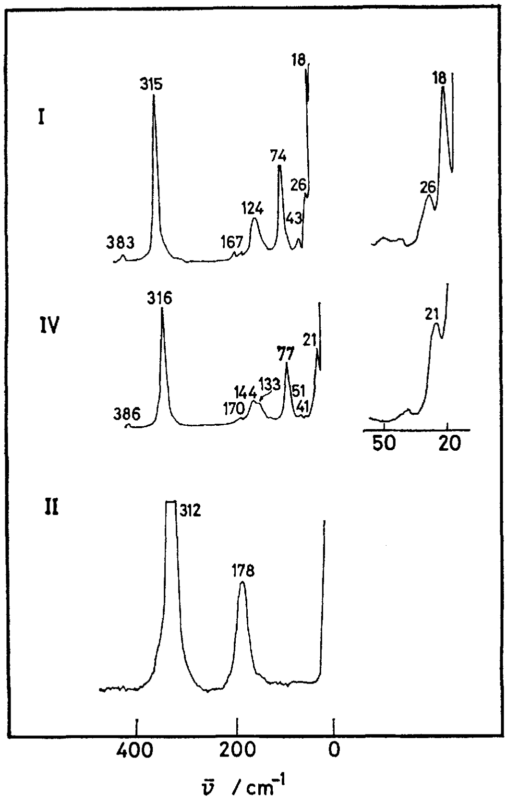

FIGURE 2 Raman spectra of $HgCl_2$ at 295 K and 0 (phase I), 8.5 (IV), and 30 kbar (II). Spectral slit width, 1.5 cm^{-1}; 60-mW 514.5-nm radiation at the sample

RESULTS AND DISCUSSION

Assignment for $HgCl_2$ (*Phase* I).—The results are illustrated in Figures 2—4, and Tables 1 and 2. Surprisingly, there is no satisfactory literature assignment of the entire vibrational spectrum of the ambient-pressure phase, $HgCl_2$(I), despite many partial studies. This phase has the symmetry[12] *Pnma* (D_{2h}^{16} with $Z = 4$). All the atoms are on sites 4*c* and are arranged in planar sheets stacked along *b*. A factor-group analysis (f.g.a.) for $HgCl_2$(I) is given in Table 3. The two Hg–Cl bonds are not required, crystallographically, to be equal (they have lengths 2.23 and 2.27 Å), neither is the molecule required to be linear. The actual distortions from strict linear centrosymmetric geometry are sufficiently slight that $D_{\infty h}$ forms a good basis for understanding the spectra.

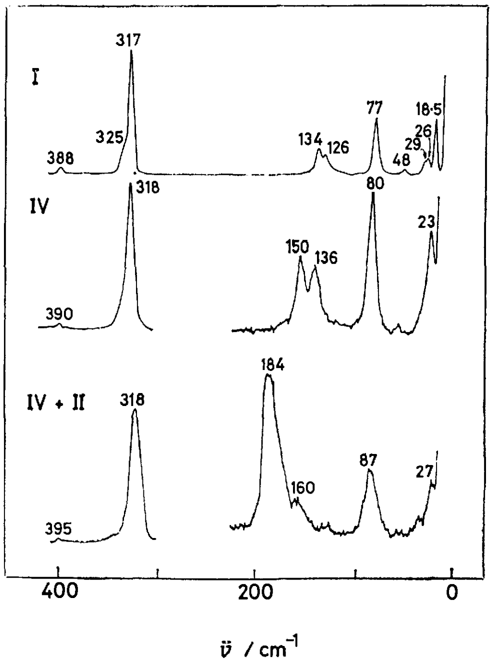

FIGURE 3 Raman spectra of $HgCl_2$ at 150 K and 0 (phase I), 9.3 (IV), and 37.0 kbar (II + IV). Conditions as for Figure 2

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[10] D. M. Adams and S. J. Payne, *J.C.S. Faraday II*, 1974, 1959.

[11] D. M. Adams and S. K. Sharma, *J. Phys.* (*E*), 1977, **10**, in the press.

[12] R. W. G. Wyckoff, 'Crystal Structures,' 2nd edn., Wiley–Interscience, New York, 1964, vol. 1.

ν_1, ν_{sym}(Hg–Cl), is present in the Raman spectra as an intense band at 315 cm^{-1} with asymmetry on the high-frequency side: these are the expected $A_g + B_{2g}$ components [Table 3 (*b*)]. The i.r. counterpart of this correlation quadruplet, $B_{1u} + B_{3u}$, appears as a pair of sharp medium-intensity bands at 310 and 330 cm^{-1}. In contrast, ν_3, ν_{asym}(Hg–Cl), is intense in the i.r. spectra but very weak ($A_g + B_g$ components coincident) in Raman spectra.

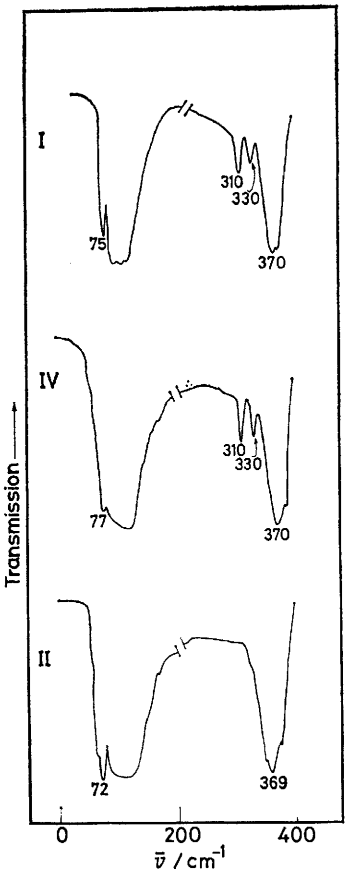


FIGURE 4 Far-i.r. spectra at 295 K of $HgCl_2$ at 0 (phase I), 9.0 (IV), and 30 kbar (II)

Under $D_{\infty h}$ symmetry ν_2, $\delta(HgCl_2)$, is i.r.-active only. Although it may attain Raman activity in the crystal by virtue of both site and correlation fields, it is expected to be intense in the crystal i.r. spectra but weak in Raman spectra. Accordingly we associate it with the region of intense i.r. absorption at *ca.* 100 cm^{-1} which has no Raman counterpart: this i.r. band shows three unresolved components in exact agreement with theory which requires $B_{1u} + B_{2u} + B_{3u}$. By an analogous argument, the molecular rotatory modes, which originate as π_g (Raman-active only) in $D_{\infty h}$, are expected to be weak in the i.r. spectra of the solid but intense in Raman spectra. These considerations indicate that Raman bands in the interval 120—180 cm^{-1} are of rotatory type. Finally, bands below 85 cm^{-1} in both the Raman and i.r. spectra are attributed to translatory modes of which five of the theoretical six were found at room temperature in the Raman spectra, and both of the predicted ($B_{1u} + B_{3u}$) i.r. bands (40 * and 75 cm^{-1}). This assignment differs from others [13-16] chiefly in the ν_2-rotatory mode description. If bands from 75 to 135 cm^{-1} are all ascribed to ν_2 components an unreasonably large factor-group splitting is implied.

TABLE 2

I.r. bands (cm^{-1}) for $HgCl_2$ at various hydrostatic pressures

Pressure/kbar	0.001	10	30
ν_3, ν_{asym}(Hg–Cl)	370	370	359
ν_1, ν_{sym}(Hg–Cl)	330 310	330 310	
ν_2, δ(ClHgCl)	106 *	108 *	109 *
ν_L	75	77	72
Phase	I	IV	II

* Band centre recorded.

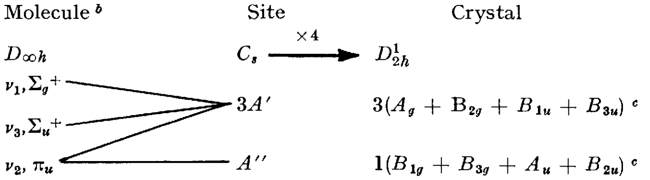
TABLE 3

Factor-group analysis (*a*) and mode-correlation scheme (*b*) for $HgCl_2$(I)

(*a*)

D_{2h}^1	N_T [a]	T_A	T	$R(x,y)$	ν_i	Activities
A_g	6		2	1	3	Raman
B_{1g}	3		1	1	1	
B_{2g}	6		2	1	3	
B_{3g}	3		1	1	1	
A_u	3		1	1	1	Inactive
B_{1u}	6	1	1	1	3	I.r.
B_{2u}	3	1	0	1	1	
B_{3u}	6	1	1	1	3	

(*b*)



[a] N_T = Total number of modes in unit cell, T_A = acoustic branch modes, T = optic branch translatory lattice modes, $R(x,y)$ = rotatory lattice modes, ν_i = molecular internal modes. [b] Approximate geometry (see text). [c] These sum to give column ν_i in (*a*).

In the absence of a single-crystal assignment we cannot be sure of the symmetry labels of the Raman translatory modes. Nevertheless it is helpful for interpretation of the high-pressure data to have at least an outline understanding of them. The crystal translatory modes originate from molecular motions classified in $D_{\infty h}$ as $\Sigma_u{}^+$ (along the molecular axis) and π_u (degenerate, normal to the axis). The π_u is both site- and correlation-field split, yielding a crystal vector $A_g + B_{1g} + B_{2g} + B_{3g}$ for the Raman-active components. It is not unreasonable that these should be in mutual proximity as the correlation field, in particular, is unlikely to be large in a molecular crystal such as this. Accordingly

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* Seen in mull spectra but not under the conditions used with the d.a.c.

[13] H. Poulet and J. P. Mathieu, *J. Chim. phys.*, 1963, 442.

[14] G. Randi and F. Gesmundo, *Atti. Accad. naz. Lincei., Rend. Classe Sci. fis. mat. nat.*, 1964, **41**, 200.

[15] Y. Mikawa, R. J. Jacobsen, and J. W. Brasch, *J. Chem. Phys.*, 1966, **45**, 4528.

[16] S. D. Ross, 'Inorganic Infrared and Raman Spectra,' McGraw-Hill, London, 1972 and refs. therein.

we assign the bands at 26 (split at 150 K) and 18 cm^{-1} (rather broad both at 295 and 150 K) to translatory modes of π_u origin, implying a site splitting of π_u of *ca.* 10 cm^{-1}. This leaves an $A_g + B_{2g}$ pair which originates from Σ_u^+: due to its intensity the band at 74 cm^{-1} is assigned as A_g; that at 43 cm^{-1} is, by elimination, B_{2g}.

The High-pressure Results.—$HgCl_2$(II). Bridgman[2] located a single-phase transition in $HgCl_2$, at *ca.* 18 kbar at room temperature, which became increasingly sluggish at lower temperatures. Our Raman spectra, obtained on successively increasing the pressure at ambient temperature, show clearly that at 19.3 kbar a new phase has appeared and exists in equilibrium with $HgCl_2$(IV). The spectrum at 30 kbar shows only features due to $HgCl_2$(II): this spectrum is much simpler than those of either phases I or IV (discussed below); the ν_3 component is no longer present at *ca.* 386 cm^{-1}, ν_1 is greatly increased in intensity, and the translatory lattice-mode region is clear.

These experiments were repeated, having first cooled the sample to 150 K (Figure 3). In the 21-kbar spectrum there is still no sign of phase II and even at 37 kbar, although $HgCl_2$(II) is now clearly present as judged by a strong band at 184 cm^{-1}, it is in equilibrium with phase I. This behaviour is typical of a sluggish phase transition: nevertheless $HgCl_2$(II) is unambiguously characterised by these experiments as having a much simpler spectrum than $HgCl_2$(I).

In the i.r. spectra (room temperature only) the IV–II transition is most clearly characterised by the prompt disappearance of the two sharp ν_1 factor-group components at 310 and 330 cm^{-1}, and by a *decrease* in the frequency of ν_3, a most unusual result of increasing pressure. Concurrently the lattice mode at 75 cm^{-1} gains intensity substantially and the region of intense absorption at *ca.* 100 cm^{-1} associated with ν_2 shifts to higher energy. We note that there is no significant i.r. absorption by $HgCl_2$(II) in the interval 120—340 cm^{-1}, and certainly none corresponding to the strong Raman band at 180 cm^{-1} (at 30 kbar).

$HgCl_2$(IV). Careful examination of the Raman spectra at 295 K obtained at pressures $<$10 kbar shows a number of small but significant and repeatable changes in the region below 180 cm^{-1}. No such changes were found in the i.r. spectra, although the extreme experimental difficulties encountered in the 20—200 cm^{-1} region with a gasketed d.a.c. limits the detail that can be reliably observed. Bridgman did not find a phase transition in $HgCl_2$ until *ca.* 18 kbar at ambient temperature, *viz.* the so-called I–II transition. However, a very recent report[3] of some n.q.r. measurements shows clearly that there is a second-order transition at 6.9 kbar at 312 K.

In our Raman spectra at 295 K for the range 0—10 kbar the changes effected by increase of pressure are: (*i*) a positive shift of the bands at 18 and 77 cm^{-1}, assigned to translatory modes (this is normal and does not necessarily indicate a phase change); (*ii*) a rapid loss of intensity of the band at 28 cm^{-1} which either disappears or breaks into two or three weak components which merge into the edge of the band at 18 cm^{-1}, now shifted to *ca.* 23 cm^{-1}; (*iii*) rapid upward shift by *ca.* 10 cm^{-1} of the higher-frequency component of the weak unresolved doublet at 43 cm^{-1} (having reached 56 cm^{-1} this band is unaffected by a subsequent pressure increase of 6.5 kbar); and (*iv*) a similar differential shift of the components of the unresolved band at 124 cm^{-1} resulting in its resolution. Apart from progressive frequency shifts with pressure, the spectra obtained at $>$10 kbar remain remarkably constant in appearance until development of phase II ensues. Our Raman data therefore provide independent evidence for the existence of a new phase which we label $HgCl_2$(IV). In contrast with the IV–II change, it is evident from the spectral changes that the structural differences between phases I and IV are subtle.

Balashov and Ikhenov[3] showed that the I–IV transition pressure decreases linearly with temperature by 3.6 kbar K^{-1} in the range 256.5—365.0 K and remarked that if the linear dependence were maintained to 0 K the transition pressure would be 5.45 kbar. Even allowing for pronounced deviation from linearity, this strongly suggests that $HgCl_2$(IV) cannot be prepared at ambient pressure at any temperature. Accordingly our ambient-pressure Raman spectrum at 150 K must be that of $HgCl_2$(I). Comparison with the spectrum at 295 K shows that the bands at 28 and 128 cm^{-1} have been resolved into doublets: this also happened with increase of pressure. In contrast, the band at 48 cm^{-1} remains single at 150 K until pressures of *ca.* 9 kbar are applied: this is the only real Raman evidence at 150 K for the existence of a new phase.

The Structure of $HgCl_2$(II).—The strict i.r.-Raman exclusions plainly indicate that this phase has a centrosymmetric unit cell. Further, the great simplification of the Raman spectrum in the lattice-mode (translatory and rotatory) region, and the disappearance of the ν_1 factor-group components from the i.r. spectrum, strongly imply a structure of higher symmetry than that of $HgCl_2$(I). It is, however, significant that HgCl(II) appears to be composed of the same kind of molecular unit as the other phases in that the ν_1 and ν_3 molecular internal modes remain in similar regions throughout. These three phases therefore represent three different solutions to the problem of packing the same linear molecules. In the higher-pressure phases it must be presumed that the intermolecular interactions are increased. An indication of this is given by the decrease in ν_3 which accompanies the IV–II transition.

Other known solutions to the problem of packing linear AB_2 molecules are represented by the structures of $HgBr_2$,[12] CO_2,[12] CS_2,[17] XeF_2,[18] and KrF_2,[19] and our high-pressure polymorphs might well adopt one of them.

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[17] N. C. Baenziger and W. L. Duax, *J. Chem. Phys.*, 1968, **48**, 2974.

[18] L. L. Lohr and W. N. Lipscomb, *J. Amer. Chem. Soc.*, 1963, **85**, 240, 241.

[19] R. D. Burbank, W. E. Falconer, and W. A. Sunder, *Science*, 1975, **178**, 1285.

$HgCl_2$(II) and $HgCl_2$(IV) plainly do not adopt the $HgBr_2$ structure as it has the space group C_{2v}^{12} and requires i.r.–Raman coincidences. Moreover it has so different a structure from $HgCl_2$(I) that a transition to it would require a reconstructive process of catastrophic proportions. Factor-group analyses for the other structures are shown in Table 4. The XeF_2 structure may be rejected immediately: it requires only one ν_1 component as opposed to the two observed, and only one i.r. absorption below 150 cm^{-1}.

TABLE 4

Factor-group analyses for the crystals of some linear triatomic molecules

	$KrF_2(D_{2h}^{14}, Z' = 2)$ [a]						$XeF_2(D_{2h}^{17}, Z' = 1)$					
D_{4h}	N_T [b]	T_A	T	R	ν_i	ν(Hg–Cl)	N_T	T_A	T	R	ν_i	ν(Hg–Cl)
A_{1g}	1				1	1	1				1	1
A_{2g}												
B_{1g}												
B_{2g}	1				1	1						
E_g	2			2			1			1		
A_{1u}												
A_{2u}	2	1			1	1	2	1			1	1
B_{1u}	2		1		1	1						
B_{2u}												
E_u	4	1	1		2		2	1			1	

	$CS_2(D_{2h}^{18}, Z' = 2)$							$CO_2(T_h^6, Z' = 4)$					
D_{2h}	N_T	T_A	T	R	ν_i	ν(Hg–Cl)		N_T	T_A	T	R	ν_i	ν(Hg–Cl)
A_g	2			1	1	1	A_g	1				1	1
B_{1g}	1			1			E_g	1			1		
B_{2g}	1			1			T_g	3			2	1	1
B_{3g}	2			1	1	1	A_u	2		1		1	1
A_u	2		1		1		E_u	2		1		1	
B_{1u}	4	1	1		2	1	T_u	6	1	2		3	1
B_{2u}	4	1	1		2	1							
B_{3u}	2	1			1								

[a] Z' = Number of molecules in primitive cell. [b] N_T = Total number of modes of primitive cell, T_A = acoustic modes, T = optic branch translatory modes, R = rotatory modes, ν_i = internal modes of molecules; ν(Hg–Cl) subset of ν_i due to ν_1 and ν_3 molecular modes.

The CO_2, CS_2, and KrF_2 structures predict almost identical far-i.r. spectra. Each requires four or five bands below 150 cm^{-1} and the observed spectrum of $HgCl_2$(II) is compatible with either. Similarly, all the three structures are compatible with the absence of translatory modes from the Raman spectrum of phase II. The only basis for a decision is therefore in the relative predictions for the Raman-active rotatory and ν_1 mode regions.

The CS_2 structure requires four rotatory modes spread among four non-degenerate symmetry species and the spectrum does not seem to be compatible with this, although the possibility cannot be altogether excluded. A decision between the KrF_2 and CO_2 structures is delicate but we consider the latter to be more probable chiefly because it more naturally accounts for the increased intensities associated with the ν_1 and rotatory mode regions. In the CO_2 structure the cell remains tetramolecular: the four ν_1 modes couple *via* S_6 site symmetry to yield the components $A_g + T_g$ in T_h^6, both of which are Raman-active. The very high observed intensity of ν_1 in $HgCl_2$(II) is correlated with the T_g species, and the unresolved shoulder with A_g. In contrast, the KrF_2 structure requires only the components $A_g + B_{2g}$ (in D_{4h}), and the cell is halved.

Since the CO_2 structure forms an acceptable basis in accounting for the spectra of $HgCl_2$(II), we do not at this stage search for new hypothetical structures. We note that, whatever the new structure is, the change to it appears to involve the modes which we have assigned as molecular rotatory.

The Structure of $HgCl_2$(IV) *and the Mechanisms of the Phase Transitions.*—We have already noted that the similarity of the spectra of phases I and IV implies a subtle structural difference. $HgCl_2$(I) has the symmetry of space group D_{2h}^{16} and all the atoms lie in mirror planes. Accordingly, there is no crystallographic requirement that the Hg–Cl bond lengths be equal or the molecule linear; the bond lengths are different and the molecule is not quite linear. We propose that the second-order transition to $HgCl_2$(IV) consists of a rearrangement of the molecules relative to each other within each mirror plane with retention of space group. Expressed crystallographically, this takes advantage of the fact that the sets of Wyckoff sites 4*c* have no geometrical restrictions on them other than that they remain in the mirror plane. Such a rearrangement would require a combination of translatory motions, and rotary motion about axes normal to the plane. Credence is lent to this view by knowledge that the small spectral changes accompanying the I–IV transition are all in the region of the translatory and rotatory lattice modes. The only precedent for this kind of transition, so far as we are aware, is in cerium which undergoes a phase transition believed to originate in a change in band structure.[20]

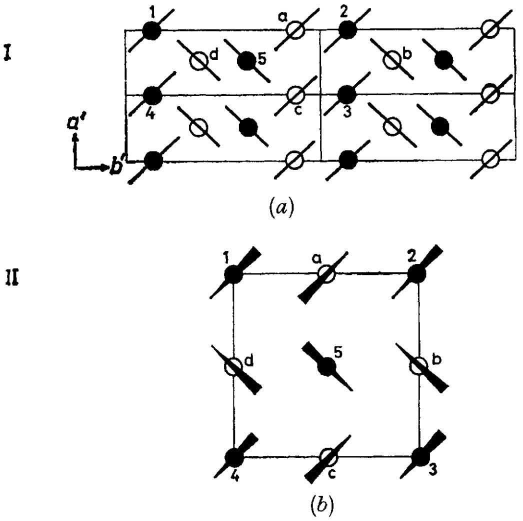

FIGURE 5 Projections of the structures of (*a*) $HgCl_2$(I), (*b*) CO_2, with an indication of the mechanism of the transition IV–II

Since the structures of phases I and IV are almost identical we consider the mechanism of the IV–II

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[20] M. K. Wilkinson, H. R. Child, C. J. McHargue, W. C. Koehler, and E. O. Wollan, *Phys. Rev.*, 1961, **122**, 1409.

transition in terms of a hypothetical I–II transition since the structure of phase I is fully characterised. The spectroscopic results are consistent with the view that $HgCl_2$(II) has the CO_2 structure (cubic, T_h^6). To move from the orthorhombic phase I to a phase with the CO_2 structure the following changes are necessary: (*i*) translate the layers of $HgCl_2$(I) relative to each other by *c*/4; (*ii*) alter the relative lengths of the three axes such that they become equal (*c* is halved due to the change in symmetry); and (*iii*) rotate each molecule out of the *ac* plane. In practice these changes must occur in one concerted movement. Examination of a model of $HgCl_2$(I) indicates that the first result of compression is likely to be the translatory and rotatory motions specified above as leading to phase IV. Further compression would then increase the repulsive energy rapidly but this could be relieved by allowing adjacent molecules to ' dive ' under one another by means of rotation about axes parallel to *c* passing through the mercury atoms, accompanied by translations, to give the cubic CO_2 structure. These movements are indicated in Figure 5. In principle this mechanism could be tested by explicit calculation of lattice energies but this is frustrated at present by the lack of suitable potential functions. It would be simple to calculate the energetics based on an electrostatic model with fractional atomic charges but, even should that support our proposed mechanisms, we would regard the agreement as fortuitous because dispersive forces must be dominant in a crystal of this type.

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