tailed discussion of the reasons for this which was earlier given for three-coordinate phosphorus compounds[4] is completely transferable to the present four-coordinate silicon compounds, with the substitution of one of the groups attached to silicon for the phosphorus lone-pair electrons. Since the purpose of this paper is to discuss rotational barriers, it follows from the above that the observed $\Delta G_c^{\ddagger}$(max) must be an upper limit for the rotational barrier about the silicon-nitrogen bond.

This work also suggests that rotational barriers about silicon-nitrogen bonds are substantially lower than those about analogous phosphorus-nitrogen and sulfur-nitrogen bonds. In a set of (very roughly) analogous compounds the following barriers are observed: $CCl_3SN(CH_2Ph)_2$, 15.0 kcal/mol;[1] $CClH_2P(Cl)N(CH_3)_2$, 11.3 kcal/mol;[8] $CClH_2Si(CH_3)(Cl)(N(CH_3)_2)$, <7.2 kcal/mol (present work).

Recent theoretical analyses of the origins of rotational barriers in compounds amenable to *ab initio* molecular orbital calculations have shown that net rotational barriers are the result of a fine balance between attractive and repulsive interactions involving adjacent electron pairs and polar bonds.[13] In analyses of the "gauche effect" in second-row compounds ("the tendency of species containing adjacent electron pairs and/or polar bonds to exist preferentially in that conformation which has the maximum number of gauche interactions between these electron pairs or polar bonds"[14]) it seems that polar bond-polar bond interactions are greater than polar bond-lone pair interactions which are, in turn, greater than lone pair-lone pair interactions.

The compounds being compared in this study contain a third-row atom, bonded to other groups by polar bonds. The third-row atom (Si, P, S) is linked to nitrogen, bearing a lone pair of electrons, and also linked to other groups by polar bonds. There are many different types, and strengths, of interactions in these molecules, and it is premature to attempt to make a detailed analysis of them in view of the limited data available. Two observations may be germane; the observed barrier increases as the electronegativity of the atom attached to nitrogen (and the number of lone pairs of electrons it formally carries) increases. However in the phosphorus system, at least, the barrier decreases sharply[5] in going from a phosphonamidous chloride, $RP(Cl)NR'_2$, to the corresponding fluoride, $RP(F)NR'_2$. We have previously argued that this effect is best interpreted in terms of a p_π-d_π multiple-bond contribution to the rotational barrier,[5] but it must be conceded that the present negative observations in the silicon-nitrogen system do not indicate any very substantial p-d interaction with a geometric preference in this system.

In view of the relatively primitive level of our understanding of the bonding of third-row elements, it may be best to summarize these observations on rotational barriers as a set of provocative data in search of a reasonable explanation.

Registry No. Table I $R = CH_3$, $R' = CH_3$, $R'' = N(CH_3)_2$, 3768-58-9; $R = CH_3$, $R' = CH_2{=}CH$, $R'' = N(CH_3)_2$, 13368-45-1; $R = CH_3$, $R' = C_6H_5$, $R'' = N(CH_3)_2$, 33567-83-8; $R = CH_3$, $R' = CH_2{=}CH$, $R'' = Cl$, 25374-07-6; $R = CH_3$, $R' = CH_2Cl$, $R'' = Cl$, 36245-72-4; $R = CH_3$, $R' = C_6H_5$, $R'' = Cl$, 36245-73-5. $(CH_3)_3SiN(CH_2CH_3)_2$, 996-50-9; $(CH_3)_3Si\text{-}N\text{—}CH_2\text{—}CH_2$, 2116-90-7.

Acknowledgments. This work was supported by Public Health Service Grant No. CA-07182 from the National Cancer Institute and by a grant from the President's Fund, California Institute of Technology. F. Fong was a participant in a National Science Foundation Undergraduate Research Program.

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Raman Spectra of Molten Bismuth Trichloride and Antimony Trichloride and of Their Mixtures with Potassium Chloride or Aluminum Trichloride[1a]

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Raman spectra have been obtained for solid and molten $SbCl_3$, molten equimolar $SbCl_3$-$AlCl_3$, $SbCl_3$-KCl mixtures, pure $BiCl_3$, equimolar $BiCl_3$-$AlCl_3$, and $BiCl_3$-KCl mixtures. The major species present in these samples have been postulated and their structures discussed.

Introduction

The crystal structure of solid $SbCl_3$ has been studied by X-ray diffraction.[2] The results show that there are discrete $SbCl_3$ molecules in the crystal located at sites with C_s symmetry. Nuclear quadrupole resonance experiments[3] indicate that the molecule in the crystal has a lower symmetry than C_{3v} because of intermolecular forces. However, only the four Raman lines of a typical C_{3v} molecule were reported for crystalline $SbCl_3$ by Krishnamurti.[4] Raman data on molten $SbCl_3$ are reported in several papers,[4-6] but the results show considerable disagreement. We, therefore, reinvestigated the Raman spectrum of $SbCl_3$ in order to clarify the situation. As a result of conductance measurements[7]

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(1) (a) Research sponsored by the United States Atomic Energy Commission under Contract At-(40-1)-3518 and under contract with Union Carbide Corp. (b) Oak Ridge University of Tennessee. (c) Oak Ridge National Laboratory.

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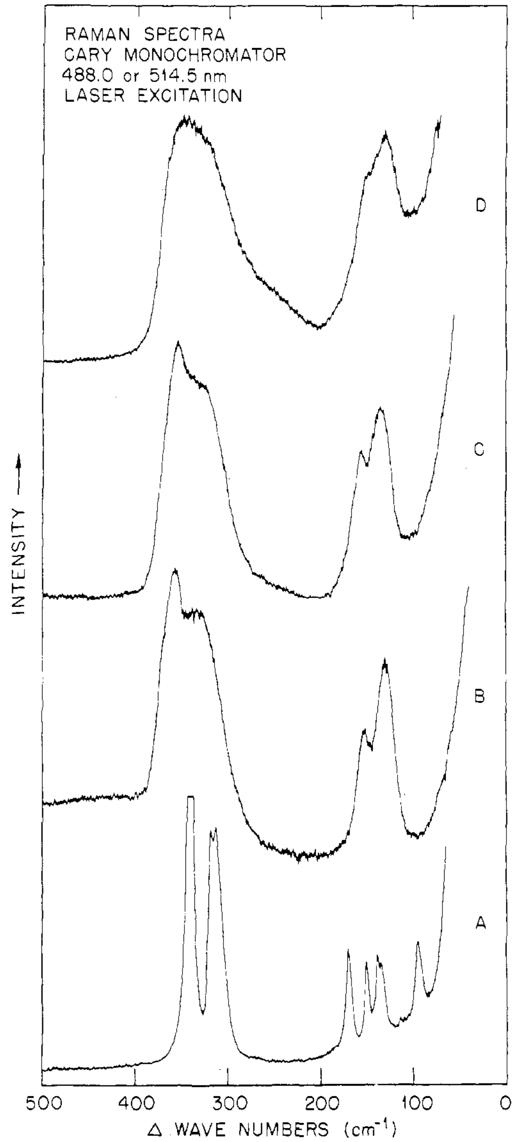

Figure 1. Raman spectra of $SbCl_3$ containing samples: A, solid $SbCl_3$; B, molten $SbCl_3$; C, molten $SbCl_3$-KCl (50:50 mol %); D, molten $SbCl_3$-KCl (33:67 mol %).

of $AlCl_3$ in molten $SbCl_3$ and of KCl in molten $SbCl_3$,[8,9] the presence of $SbCl_2^+$, $AlCl_4^-$, and $SbCl_4^-$ in these mixtures has been suggested. We studied the Raman spectra of these mixtures in the hope of characterizing the major species present. The Raman spectra of molten $BiCl_3$ and $BiCl_3$ in mixtures with alkali chlorides were investigated by Kenney and Powell;[10] however, the spectra were weak, and the conclusions were tentative. Recently, Oertel and Plane[11] made an extensive study of chlorobismuthates in aqueous solution and in solid compounds. We, therefore, reexamined the molten systems with the hope of obtaining better spectra which could be compared with the aqueous data.

Experimental Section

The anhydrous chlorides used were prepared in the laboratory of G. P. Smith at Oak Ridge National Laboratory. All preparative operations and transfers were conducted in vacuum systems or in a nitrogen-filled drybox with the H_2O content below 10 ppm. Bismuth and antimony trichlorides were prepared by treating reagent grade metals with dry chlorine gas and purifying the products by repeated distillations. Reagent grade KCl was treated with anhydrous HCl and processed in a manner described elsewhere[12] to remove any residual impurities. Aluminum chloride was synthesized from high-purity aluminum by passing specially prepared HCl vapor over the molten metal.[13]

Table I. Raman Frequencies of Solid and Gaseous $SbCl_3$ (cm^{-1})[a]

Gaseous Ref 16	Solid Ref 4	Solid Ref 16	Solid This work	Assignment C_{3v}	Assignment C_s
384 s, P	338	340 s	340 vs	$\nu_1(A_1)$	A′
361 w, sh	312	314 ms	318 s	$\nu_3(E)$	A′
			312 s		A″
		168 m	169 m		*b*
154 m, P	166	150 mw	150 m	$\nu_2(A_1)$	A′
127 m	146	137 mw	138 m	$\nu_4(E)$	A′
			133 m		A″
		93 w	94 m		Lattice
		60 m			

[a] Key: s, strong; m, medium; sh, shoulder; v, very; w, weak; P, polarized. [b] Long Sb-Cl intermolecular stretching.

The salts were sealed in evacuated Pyrex tubes, 6-mm o.d. by ~25 mm. The samples were melted by wrapping a platinum wire spirally around the sample tube and passing an electric current through the wire. Temperatures were not closely controlled or measured. For $BiCl_3$ mixtures, temperatures ranged from around 250° for the sample of high $BiCl_3$ content to nearly 600° for the 25:75 mol % $BiCl_3$-KCl sample.[14] Equimolar $BiCl_3$-$AlCl_3$ is liquid[15] above 184°. Temperatures for the $SbCl_3$-containing melts ranged from near 80° for pure $SbCl_3$ to 200° for the $SbCl_3$-KCl (33:67 mol %) sample. Visual inspection of the melts in the laser beam ensured that no solid phases were present.

Samples were clamped into position in the Raman spectrometer before melting. A laser beam was focused at right angles to the long axis of the sample tubes and the spectra were observed at right angles to both the laser beam and the sample tube. Qualitative polarization measurements were made by rotating the plane of polarization of the laser beam with a half-wave plate. Appropriate spike filters were employed to attenuate the background laser lines in the Raman spectra.

A Spectra-Physics Model 141 argon ion laser was used to illuminate the sample. In most cases observations were made with both the 488.0- and 514.5-nm laser lines. Despite very careful preparation, small amounts of impurities caused discoloration of the $BiCl_3$-KCl samples, and a Spectra-Physics Model 125 helium-neon laser emitting light at 632.8 nm was therefore used for observation of these spectra. Power levels at the sample were approximately 80 mW for the argon ion laser lines and 20 mW for the helium-neon laser line. A Cary Model 81 monochromator, coupled with a cooled 9558 EMI photomultiplier tube with a photon counting system, was used to record the Raman spectra.

Results and Discussion

Solid and Molten $SbCl_3$. The Raman spectrum of solid $SbCl_3$ at room temperature is shown in Figure 1A. The band positions and their assignments are given in Table I with the results of previous studies for comparison. The data are different from the old results[4] and in good agreement with recent work,[16] except for better resolution. From comparison with the gas- and liquid-phase data (see below) and the relative intensity, we assign the highest frequency band at 340 cm^{-1} to the symmetric stretching ν_1

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Table II. Raman Frequencies of Molten $SbCl_3$ and Molten $SbCl_3$-KCl Mixtures (cm^{-1})[a]

$SbCl_3$ This work	$SbCl_3$ Ref 5	50:50 mol % $SbCl_3$-KCl	33:67 mol % $SbCl_3$-KCl	Assignment C_{3v}
358 vs, P	350	358 s, P	350 s, P	ν_1
332 s	315	334 m	327 sh	ν_3
153 m	150	155 m	150 sh	ν_2
130 m	123	136 m	132 m	ν_4

[a] Key: v, very; sh, shoulder; s, strong; m, medium; P, polarized.

(A′) mode. The two doublets were assigned to asymmetric stretching (ν_3) and bending (ν_4) modes. The splitting of these E modes is caused by the lowering of the symmetry from C_{3v} to C_s in the solid crystalline phase. By comparison with the gas-phase and melt spectra, the band at 150 cm^{-1} was assigned to ν_2. The band at 169 cm^{-1}, which is absent in the gas and melt spectra, is, we believe, the result of the stretching of the intermolecular, long Sb-Cl bond indicated by X-ray analysis.[2] The band at 94 cm^{-1} is in the frequency region for a crystal lattice mode. We conclude that the Raman data for solid $SbCl_3$ can be well interpreted in terms of the crystal structure.

The Raman spectrum of molten $SbCl_3$ is shown in Figure 1B. The band positions, the assignment, and the previous results are given in Table II. The specific conductance (0.8×10^{-6} ohm^{-1} cm^{-1}) and viscosity (0.0184 P) measured[17] for molten $SbCl_3$ indicate that the melt contains discrete molecular units. The spectrum is, indeed, well interpreted by assuming $SbCl_3$ molecules with C_{3v} symmetry. The polarization of ν_2 was not observed. However, ν_2 in other C_{3v} molecules, such as $AsCl_3$, is only weakly polarized.[11]

Molten $SbCl_3$-KCl. The spectra of molten $SbCl_3$-KCl (50:50 and 33:67 mol %) are shown in Figure 1C and 1D, respectively, and the data are given in Table II. The solid spectra (glasses) are similar to those of the molten state. The mixture spectra are very close to the spectrum of pure molten $SbCl_3$, with slight shifts in band positions. At high KCl concentrations the resolution of the $SbCl_3$ bands is poorer. Antimony trichloride is a Lewis acid and a good Cl^- acceptor. The complex anion $SbCl_4^-$ in aqueous solution[18] and in the molten salt[8] has been postulated. The Raman spectrum of $[Et_4N]SbCl_4$ has been reported[19] for the solid and in $MeNO_2$ solution. In this case a C_{2v} or distorted T_d symmetry is postulated for the $SbCl_4^-$ ion. With C_{2v} symmetry one should observe four Sb-Cl stretching frequencies in the Raman spectrum: two symmetric, A_1, polarized frequencies and two antisymmetric bands. In solution, Ahlijah and Goldstein[19] assigned bands at 342 and 297 cm^{-1} as symmetric stretches and a band at 249 cm^{-1} as an antisymmetric stretch with the other antisymmetric stretch near 313 cm^{-1} (observed in the ir solid spectrum only). If $SbCl_4^-$ were produced in our melt, we would expect to see a strong polarized line near 297 cm^{-1} and another medium strength depolarized line near 249 cm^{-1}. The $SbCl_3$-KCl (50:50 mol %) sample, Figure 1D, shows some general broad emission in this region but polarization studies did not show any definite polarized bands in this region. We believe the close resemblance between the Raman spectrum of pure molten $SbCl_3$ and the spectra of the $SbCl_3$-KCl molten mixtures is evidence against the formation of appreciable quantities of $SbCl_4^-$ in the molten mixtures. The broad bands appearing in the region 250-300 cm^{-1} in the mixtures with highest KCl concentrations do indicate, however, the possible formation of some quantity of higher chloride complexes of antimony.

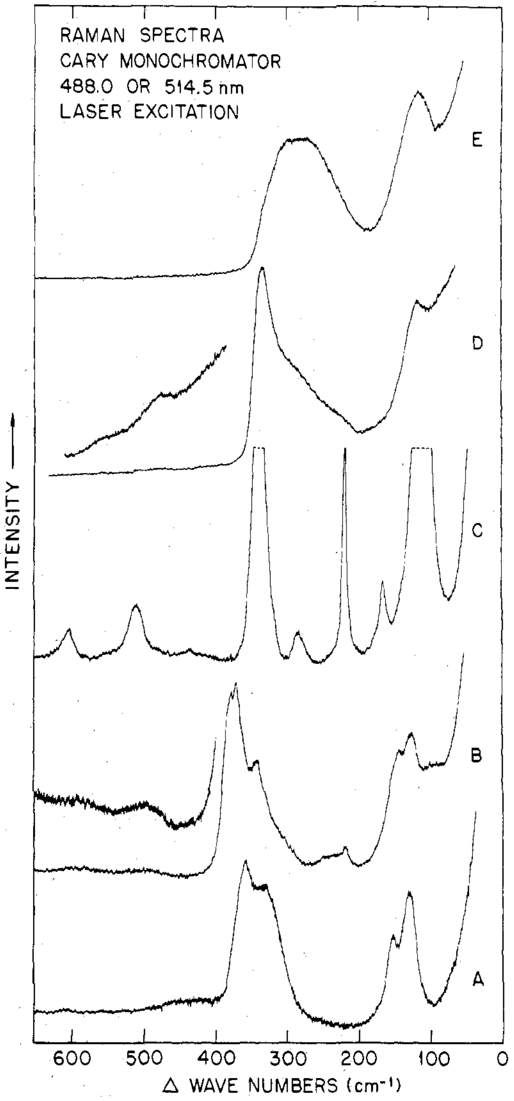

Figure 2. Raman spectra of molten $SbCl_3$ and $BiCl_3$ and their mixtures with Al_2Cl_6: A, $SbCl_3$; B, $SbCl_3$-$AlCl_3$ (50:50 mol %); C, Al_2Cl_6; D, $AlCl_3$-$BiCl_3$ (50:50 mol %); E, $BiCl_3$.

Molten $SbCl_3$-$AlCl_3$. Texier[20] measured the equivalent conductance of $AlCl_3$ in molten $SbCl_3$ and proposed the reaction

$$SbCl_3 + AlCl_3 \rightleftharpoons SbCl_2^+ + AlCl_4^-$$

The $AlCl_4^-$ species is well characterized in chloroaluminates.[21] In nonaqueous systems[22] $AlCl_3$ is a stronger Lewis acid than $SbCl_3$, and the formation of $AlCl_4^-$ appears quite reasonable. The spectra of molten Al_2Cl_6 and equimolar $SbCl_3$-$AlCl_3$ are shown in Figure 2C and 2B, and the band positions are given in Table III. The results do not support the formation of $AlCl_4^-$ (complete absence of the

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Table III. Raman Frequencies of Molten Equimolar $SbCl_3$-$AlCl_3$ (cm^{-1})[a]

$SbCl_3$	$SbCl_3$-$AlCl_3$	Al_2Cl_6[b]	$AlCl_3$[c]
	595 vw	608 m	
	497 w, P	512 m	
		440 w	
	376 s, P		~371 s
358 vs, P	344 m, P	341 vs, P	
332 s		290 m, P	
	~255 b, w		
	220 w, P	218.5 s, P	
153 m		166 w	
	145 w, P		~146 m
130 m	128 w		
		119 s	
	99 vw	104 s, P	

[a] Key: v, very; w, weak; m, medium; s, strong; P, polarized; b, broad. [b] Liquid, ref 21. [c] Gas, ref 23.

strong $AlCl_4^-$ band at 186 cm^{-1}).

If we examine the spectrum of the equimolar mixture we easily see the main features, perhaps altered slightly, of the liquid $SbCl_3$ spectrum. However, it is much harder to find the Al_2Cl_6 components of the spectrum. The weak lines are entirely missing and the strong lines are either obscured or very weak. The very strong doublet at 104 and 119 cm^{-1} is probably present but not resolved. The very sharp line at 218.5 cm^{-1} is represented by a weak line at 220 cm^{-1}, and the strong band at 341 cm^{-1} is either represented by a small peak at 344 cm^{-1} or shifted to 376 cm^{-1}. One might say that the $SbCl_3$ spectrum is relatively much more intense and that Al_2Cl_6 is a much poorer scatterer. However, this is contradicted by the ease with which a strong spectrum is obtained from molten Al_2Cl_6 (Figure 2C). It appears most likely that Al_2Cl_6 is present at a much lower concentration in the mixture than indicated by its mole per cent. The one new feature of the mixture spectrum is the appearance of a new polarized line at 376 cm^{-1}. Beattie and Horder[23] have examined the Raman spectra of Al_2Cl_6 vapor at high temperatures and assigned two lines to monomeric $AlCl_3$. These lines are ν_1 371 cm^{-1} (s) and ν_4 146 cm^{-1} (m). The region near 146 cm^{-1} is obscured in our spectrum but the line at 376 cm^{-1} could be assigned to monomeric $AlCl_3$. If a polymeric species $(SbCl_3 \cdot AlCl_3)_n$ were formed, one might expect a relatively featureless spectrum with rather broad bands. Certainly our spectrum indicates interaction between $SbCl_3$ and $AlCl_3$. This interaction shifts either the Al-Cl or the Sb-Cl stretching frequency to a higher value. In the first case this means the $AlCl_3$ is more nearly monomeric, and this view is supported by the disappearance of many of the Al_2Cl_6 lines. In the second case the $SbCl_3$ would be assuming a more nearly planar structure rather than its usual pyramidal form. We were not able to choose between these two interpretations. Spectra at varying mole ratios might clarify the situation.

Molten $BiCl_3$. Recently, Denchik, *et al.*,[16] examined the structure of solid $BiCl_3$ and concluded that Bi is strongly bonded to three chlorine atoms and less strongly bonded to five more. From the viscosity data[24] (~0.3 P), one should expect the long-range Bi-Cl interaction to be present to a certain degree in the molten state, but it should be quite weak. The Raman spectrum of molten $BiCl_3$, shown in Figure 3A and Table IV, is quite different from that of solid $BiCl_3$. It is more similar to the spectrum of

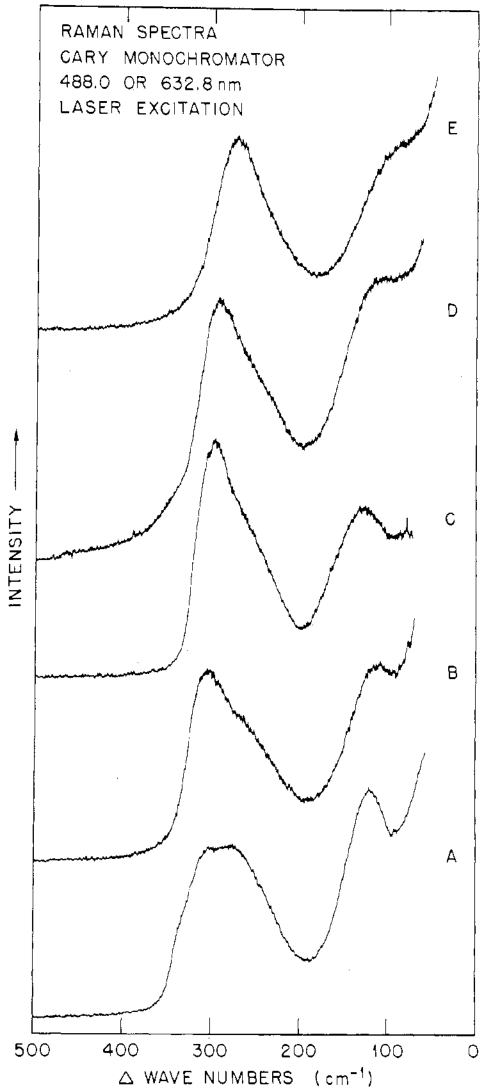

Figure 3. Raman spectra of molten $BiCl_3$-containing samples: A, $BiCl_3$; B, $BiCl_3$-CsCl (75:25 mol %); C, $BiCl_3$-KCl (67:33 mol %); D, $BiCl_3$-KCl (50:50 mol %); E, $BiCl_3$-KCl (25:75 mol %).

monomeric gaseous $BiCl_3$, which has C_{3v} symmetry. The spectrum is insensitive to temperature,[10,15] and it is therefore reasonable to ascribe it to discrete molecular vibration modes rather than to quasilattice structure. Because of the similarity of our Raman spectrum to that reported in aqueous solution,[11] we conclude that the species present in molten $BiCl_3$ are similar to those in aqueous solution. The simplicity of the spectra of molten $BiCl_3$ indicates that C_{3v} symmetry is still maintained although the considerable decrease in the stretching frequencies shows general intermolecular interactions which would account for the high viscosity but not for the reported specific conductivity (~0.44 ohm^{-1} cm^{-1}).[24] In addition, our data show the conventional C_{3v} intensity pattern rather than that found by Kenney and Powell.[10] Since the spectra of $BiCl_3$ in H_2O and alcohols[11] are similar to those of the pure molten state, the species is unlikely to be solvated in these solvents.

Molten $BiCl_3$-KCl Mixtures. The appearance of the spectra for molten $BiCl_3$-KCl and $BiCl_3$-CsCl mixtures, as shown in Figure 3B-E, is similar to that of molten $BiCl_3$: a strong polarized band at 300 cm^{-1}, a depolarized shoulder on the low-energy side, and another depolarized band at ~100 cm^{-1} which probably contains two bands. The band positions for the molten mixtures are summarized in Table IV.

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Table IV. Raman Frequencies for $BiCl_3$ and $BiCl_3$-Containing Melts (cm^{-1})[a]

$BiCl_3$			67:33 mol % molten $BiCl_3$-KCl	Aqueous $BiCl_4^-$ [c]	50:50 mol % molten $BiCl_3$-KCl	Aqueous $BiCl_6^{3-}$ [c]	25:75 mol % molten $BiCl_3$-KCl
Gas[b]	Aqueous[c]	Molten					
342 s, P	307 s, P	304 s, P	300 s, P	293 s, P	294 s, P	263 s, P	276 s, P
322 w, sh	272 m	277 m	272 m, sh, b	256 m	252 m, sh, b	220 w	236 m, sh, b
123 m, P	120 m	122 m	116 m	115 w	108 m	110 w	80 m, sh, b
107 mw							

[a] Key: s, strong; m, medium; w, weak; b, broad; sh, shoulder; P, polarized. [b] Reference 16. [c] Reference 11.

From the comparison with the results in aqueous media,[11] as shown in Table IV, and the spectra of solid $(N_2H_5)_3BiCl_6$ and $C_6H_{13}N_2BiCl_4$,[11] we believe that the anionic species $BiCl_4^-$ and $BiCl_6^{3-}$ are formed by mixing appropriate amounts of $BiCl_3$ and KCl. Their structures are similar to the species in aqueous solution, *i.e.*, C_{2v} or distorted T_d for $BiCl_4^-$ and O_h for $BiCl_6^{3-}$. The presence of $BiCl_4^-$ and $BiCl_6^{3-}$ in those systems is also supported by the phase diagram studies.[14] Oertel and Plane[11] have made a considerable case for a $BiCl_5^{2-}$ ion in aqueous solution and such an ion might be postulated here. Certainly the evidence is not conclusive, but we tend to favor the simpler picture without $BiCl_5^{2-}$, since such a structure would produce more complex Raman spectra. The melting temperatures of the various $BiCl_3$-KCl mixtures varied considerably, and it was not possible entirely to eliminate the possibility that differences in the spectra were caused by the shifting of chemical equilibria with temperature changes. Observations on the lower melting mixtures made at higher temperatures, however, showed no appreciable changes in the spectra. In addition, in several cases, glassy solids were formed upon cooling rather than the usual polycrystalline solids, and the spectra of these glasses were indistinguishable from the melt spectra. Our main conclusions are that there is a remarkable similarity between the aqueous and molten Raman data and that $BiCl_3$ in both cases forms higher species of the type $BiCl_n^{(n-3)-}$

Molten $BiCl_3$-$AlCl_3$. The Raman spectrum of the molten equimolar $BiCl_3$-$AlCl_3$ mixture is shown in Figure 2D. As with $SbCl_3$, the absence of a band at 186 cm^{-1} shows that there is very little $AlCl_4^-$ present. The spectrum is poorer than the $SbCl_3$-$AlCl_3$ spectrum and may be interpreted as being produced by a mixture of $BiCl_3$ and Al_2Cl_6, although we see only the strongest Al_2Cl_6 bands. In contrast to the $SbCl_3$ case, we do not find evidence of $AlCl_3$ monomer or complex production. The congruently melting solid compound obtained in the phase studies[15] does not seem to be present in the melt.

Registry No. Bismuth trichloride, 7787-60-2; antimony trichloride, 10025-91-9; potassium chloride, 7447-40-7; aluminum trichloride, 7446-70-0; $BiCl_6^{3-}$, 15977-99-8; $BiCl_4^-$, 20057-70-9.

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The Synthesis and Spectroscopic Properties of Vanadium(III) Squarate Trihydrate

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Vanadium(III) squarate trihydrate has been prepared; and the infrared, X-ray diffraction, and electronic spectra of the complex indicate that the central metal atom is in a slightly distorted octahedral field. A thermogravimetric study has shown that the complex loses three water molecules upon heating. A hydroxy-bridged, dimeric structure has been proposed from these data.

The preparation and spectroscopic properties of several metal complexes of diketocyclobutenediol[1] (1,2-dihydroxy-3,4-diketocyclobutene) have been previously reported.[2-4] These reports have usually presented preparations and little spectral data. In this report the preparation is similar to the one reported by West and Niu.[2] Also, the powder X-ray diffraction, thermogravimetric, magnetic, and infrared and electronic spectral data are presented and discussed.

Experimental Section

Materials. Squaric acid and hexachloropropene were purchased from Aldrich Chemical Co., Milwaukee, Wis. Vanadium pentoxide was purchased from Alfa Inorganics, Beverly, Mass. Vanadium trichloride hexahydrate was prepared by the hydrolysis of vanadium trichloride.[5] Vanadium trichloride was prepared by the chlorination of vanadium pentoxide with hexachloropropene according to published procedure.[5]

Preparation of Vanadium(III) Squarate Trihydrate. A solution containing 0.76 g (2.9 mmol) of vanadium trichloride hexahydrate was mixed with a solution containing 0.50 g (4.5 mmol) of squaric acid. The dark brown color of the vanadium trichloride solution was almost immediately replaced by an olive green precipitate. The solution was stirred for about 24 hr before the precipitate was filtered, washed with acetone and ether, and dried in a vacuum desiccator over calcium chloride. *Anal.* Calcd for $V^{III}(OH)C_4O_4 \cdot 3H_2O$: V, 21.8; C, 20.5; H, 2.99. Found: V, 20.2; C, 20.7; H, 3.17.

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(1) Hereafter, diketocyclobutenediol will be referred to by its accepted common name, squaric acid, and the dianion, diketocyclobutenediolate will be referred to as the squarate anion.

(2) R. West and H. Y. Niu, *J. Amer. Chem. Soc.*, **85**, 2589 (1963).

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