Fig. 3. Monomer packing requirements with limits $3.4 < S_1 < 4.0$ Å and $\gamma_1 \simeq 45°$.

than 4·0 Å between the reacting atoms [C(6) and C(6')] and there is a unique reacting axis the topochemical polymerization reaction can occur. In DP, $D = 4.237$ (3) Å, well outside the limit. However, in terms of Wegner's monomer packing requirements for d_1, γ_1 and S_1 ($= d_1\sin \gamma_1$) the values for DP are 3·973 (1) Å (along the b axis), 66·2 (2)° and 3·635 (3) Å, respectively. Though D and γ_1 (in Fig. 3) are well outside the limits ($D < 4.0$ Å, $3.4 < S_1 < 4.0$ Å and $\gamma_1 \approx 45°$), S_1 is within the prescribed limit suggesting low polymerization reactivity for DP. However, we note that DP undergoes thermal and photochemical polymerization to form a coloured amorphous polymer. Although formation of poly-

diacetylenes of poor crystallinity in topochemical reaction is known (Bloor, 1982), especially in the absence of a unique reaction direction (axis), the possibility that the reaction is non-topochemical cannot be eliminated. Details of the polymerization of DP are being studied.

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The Structures of (R,S)-2,2'-(1,2-Ethanediyldiimino)bis-1-butanol Dihydrochloride at 295 and 333 K. A Reversible Phase Transition

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Abstract.   (R,S)-N,N'-Bis(1-hydroxymethylpropyl)-ethylenediammonium chloride, $C_{10}H_{26}N_2O_2^{2+}.2Cl^-$, (RSET), $M_r = 277.23$, monoclinic, $P2_1/n$, $a = 6.509$ (1), $b = 23.292$ (4), $c = 5.1453$ (8) Å, $\beta = 92.41$ (1)°, $V = 779.4$ (4) Å^3, $Z = 2$, $D_x = 1.181$, $D_m = 1.160$ Mg m^{-3}, $\lambda(\text{Mo } K\alpha) = 0.71069$ Å, $\mu = 0.399$ mm^{-1}, $F(000) = 300$, $R = 0.046$ for 1397 unique observed reflections at $T = 295$ K (RSET–295). $a = 6.671$ (3), $b = 22.75$ (1), $c = 5.179$ (3) Å, $\beta = 92.76$ (3)°, $V = 785$ (1) Å^3, $D_x = 1.173$ Mg m^{-3}, $R = 0.087$ for 715 unique observed reflections at $T = 333$ K (RSET–333). The main effect of the phase transition is a fifteen degree rotation of the ions around the centres of symmetry. The differences in bond lengths and bond angles between (RSET–295) and (RSET–333) are less than 0.06 Å and 4.6° respectively.

Introduction.   (S,S)-2,2'-(1,2-Ethanediyldiimino)bis-1-butanol dihydrochloride, ethambutol dihydrochloride,

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(SSET), is a drug substance used against tuberculosis. This compound, melting at 473 K, shows an enantiotropic phase transition at 343 K that is reversible by cooling (Kuhnert-Brandstätter & Moser, 1979). (R,S)-2,2'-(1,2-Ethanediyldiimino)bis-1-butanol dihydrochloride (RSET) was synthesized according to Wilkinson, Shepherd, Thomas & Baughn (1961). (RSET) shows a phase transition at 313 K analogous to (SSET). The present work has been undertaken to clarify the structural changes which take place in the crystals of (RSET) at the phase transition.

Experimental. (RSET–295). Crystal: $0.10 \times 0.25 \times 0.30$ mm. Density measured by flotation. Picker FACS-1 diffractometer, graphite-monochromatized Mo $K\alpha$, lattice parameters from setting angles for 30 reflections with $16.0 < \vartheta < 22.5°$. 4288 reflections measured, 1852 unique reflections ($h\,0\rightarrow8$, $k\,0\rightarrow30$, $l\,-6\rightarrow6$) with $2.0 < \theta < 27.5°$. $R_{int} = 0.018$. 455 reflections with $|F| < 2\sigma(F)$ excluded from the refinement process. Four standard reflections (110, 060, 0120, 004) were measured for every 60 reflections and showed no significant variations. The intensities were corrected for Lorentz and polarization effects. No absorption correction was applied. The structure was solved by direct methods, full-matrix least-squares anisotropic refinement of non-H atoms. H-atom positions were calculated and included in the last cycles of refinement but were not allowed to vary. $\sum w(\Delta|F_o|)^2$ was minimized to a final $R = 0.046$ using unit weights, $wR = 0.043$, $S = 2.6$, $(\Delta/\sigma)_{max} = 0.10$. Minimum and maximum in final $\Delta\rho$ map -0.3 and $0.2\,e\,\text{Å}^{-3}$. Scattering factors from *International Tables for X-ray Crystallography* (1974), computer programs from *XRAY*76 (Stewart, Machin, Dickinson, Ammon, Heck & Flack, 1976) and *SYBYL* (Tripos Associates Inc., 1986). The positional and thermal parameters are given in Table 1.

(RSET–333). Crystal: $0.13 \times 0.33 \times 0.33$ mm. Density measured by flotation. Enraf–Nonius CAD-4F diffractometer, graphite-monochromatized Mo $K\alpha$, lattice parameters from setting angles for 21 reflections with $8.08 < \theta < 13.81°$. 1947 unique reflections ($h\,0\rightarrow8$, $k\,0\rightarrow29$, $l\,-6\rightarrow6$) with $2.0 < \theta < 27.5°$, 715 with $I > 2.5\sigma(I)$ used in refinement process together with 791 'less-than' reflections $[I < 2.5\sigma(I)]$ with calculated values greater than the observed. $\omega/2\theta$ scan technique, scan angle $\Delta\omega = 1.60° + 0.35\tan\theta$. 062 and $\bar{2}81$ used for orientation control every 100 reflections, intensity check every third hour of exposure time by using $1\bar{7}1$. The crystal was mounted on a special goniometer head (Enraf–Nonius RF 559 Crystal Heater). The temperature was maintained with a heated flow of air surrounding the crystal. The heating device, an inherent part of the goniometer head, produced heated air with a flux direction parallel to the goniometer head rotation axis. The temperature was

Table 1. *Fractional coordinates* $(\times 10^4)$ *and equivalent isotropic thermal parameters of non-*H *atoms with e.s.d.'s in parentheses*

	x	y	z	$B_{eq}(\text{Å}^2)$
(RSET–295)				
Cl(1)	7774 (1)	4189 (1)	479 (1)	4.38 (6)
O(1)	2002 (3)	3823 (1)	8560 (4)	5.5 (3)
N(1)	5073 (3)	4198 (1)	5236 (4)	3.0 (1)
C(1)	1636 (4)	3759 (1)	5850 (5)	4.0 (2)
C(2)	3689 (4)	3689 (2)	4605 (4)	3.4 (2)
C(3)	4886 (5)	3152 (1)	5460 (6)	4.3 (2)
C(4)	3860 (6)	2604 (2)	4505 (9)	7.0 (2)
C(11)	4151 (4)	4778 (1)	4818 (5)	3.5 (2)
(RSET–333)				
Cl(1)	6844 (1)	3998.3 (5)	350.9 (2)	5.85 (8)
O(1)	1069 (4)	4135 (2)	8331 (6)	10.5 (4)
N(1)	4320 (4)	4204 (1)	5107 (4)	4.4 (2)
C(1)	734 (7)	4049 (3)	5762 (10)	5.8 (5)
C(2)	2530 (6)	3818 (2)	4492 (6)	5.6 (3)
C(3)	3244 (9)	3202 (2)	5107 (11)	9.0 (6)
C(4)	1729 (13)	2736 (3)	3991 (17)	15. (1)
C(11)	4018 (5)	4848 (1)	4800 (6)	4.9 (3)

controlled (± 1 K) with a thermocouple near the crystal Correction for decay was applied as the standard intensity was reduced by 18% during the data collection. Absorption was ignored. The structure was solved with direct methods, full-matrix least-squares refinement of anisotropic non-H atoms. The positional H parameters were calculated and included in the last cycles of refinement but were not allowed to vary. $\sum w(\Delta|F_o|)^2$ minimized, final $R = 0.087$, $wR = 0.090$. Average $(w\Delta|F_o|)^2$ was almost independent of $|F_o|$ by using $w = 1$ when $|F_o| < 40$ else $w = (40/F_o)^2$. $S = 1.50$, $(\Delta/\sigma)_{max} = 0.06$. Minimum and maximum in final $\Delta\rho$ map -0.6 and $0.4\,e\,\text{Å}^{-3}$. Scattering factors from *International Tables for X-ray Crystallography* (1974). Computer programs from *SHELX*76 (Sheldrick, 1976) and *XRAY*76 (Stewart, Machin, Dickinson, Ammon, Heck & Flack, 1976). The positional and thermal parameters are given in Table 1.*

Discussion. The spontaneous and reversible phase transformation between the crystal forms (RSET–295) and (RSET–333) affects mainly the packing of the ions whereas the geometry of a single ion is unchanged. Bond distances and angles are given in Fig. 1. The differences in bond lengths between corresponding bonds in (RSET–295) and (RSET–333) are less than 0.035 Å except for O(1)–C(1) ($\Delta = 0.06$ Å). The differences are probably not significant. Fig. 2 shows the molecular geometries and thermal motion; corrections for thermal libration have not been applied to the

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distances. The differences between corresponding bond angles (Fig. 1) and corresponding torsion angles in (RSET–295) and (RSET–333) (Table 2) are less than 4·6 and 5·3°, respectively. The angle differences are considered as significant if they exceed 2·0°.

The most outstanding consequence of the phase transition is a rotation of the ions around the centres of symmetry, *i.e.* around the *z* axis. A least-squares line in (RSET–295), defined by the centre of symmetry ($\frac{1}{2},\frac{1}{2},\frac{1}{2}$) and the atoms C(11), N(1), C(2), C(3) and C(4), makes an angle of 14·7 (5)° with a corresponding line in (RSET–333). The rotation involves a movement of C(4), the extreme carbon atom of the chain, of about 1·5 Å.

The Cl⁻ ion is H-bonded to two N atoms and one O atom (Table 3), showing a pyramidal arrangement around Cl⁻. In the rotation movement the Cl⁻ ion follows the N atoms resulting in an inversion of the pyramidal arrangement through a 'transition state' as shown in Fig. 3. The Cl···O and N···O distances are very similar in the two crystalline forms. A plot displaying a section of the van der Waals surfaces is also given in Fig. 3. The black areas are outside all the van der Waals surfaces and their presence makes the reversible transformation from one crystalline form to the other possible. The H bonds compare well with those found by Hämäläinen, Lehtinen & Ahlgren
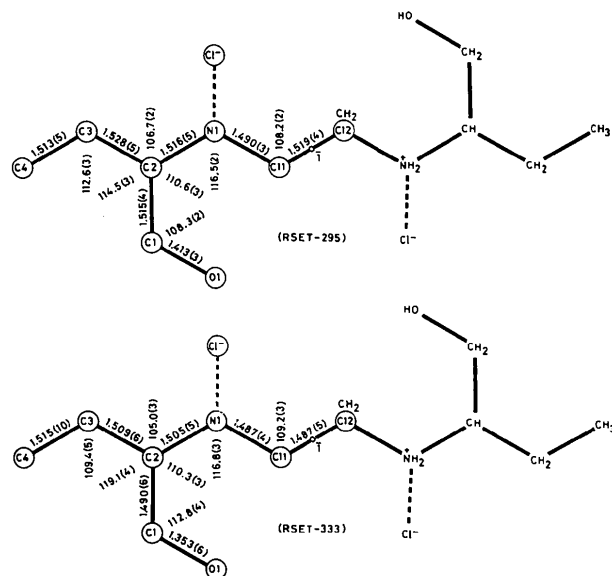

Fig. 1. Bond distances (Å), angles (°) and the atom-numbering scheme for non-H atoms in (RSET–295) and (RSET–333). Estimated standard deviations are given in parentheses.
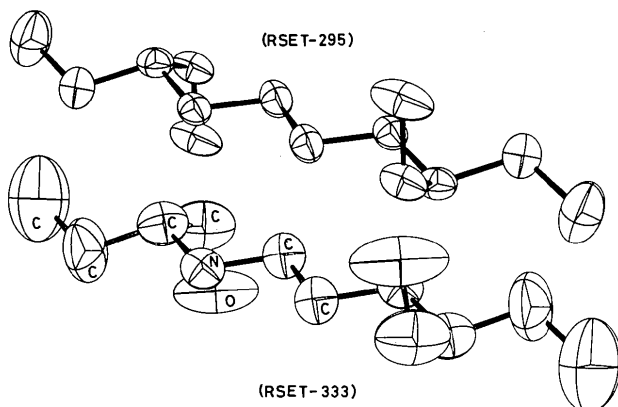

Fig. 2. Perspective drawing (*ORTEP*; Johnson, 1965) of the cations in (RSET–295) and (RSET–333). Atoms are represented by ellipsoids enclosing 50% of the thermal displacements.

Table 2. *Torsion angles* (°) *with e.s.d.'s in parentheses*

	(RSET–295)	(RSET–333)
C(12)–C(11)–N(1)–C(2)	−172·0 (2)	−173·7 (3)
C(11)–N(1)–C(2)–C(1)	−48·9 (3)	−47·5 (4)
C(11)–N(1)–C(2)–C(3)	−174·0 (2)	−177·1 (3)
N(1)–C(2)–C(1)–O(1)	−57·4 (3)	−53·1 (5)
N(1)–C(2)–C(3)–C(4)	−169·4 (3)	−167·0 (5)
O(1)–C(1)–C(2)–C(3)	63·1 (3)	68·4 (6)
C(1)–C(2)–C(3)–C(4)	67·9 (3)	68·9 (6)

Table 3. *A comparison between hydrogen bonds* (Å) *in* (*RSET*-333), (*RSET*-295) *and* (*SSET*). *The superscript* (i) *refers to the* O *position* (*x*+1, *y*, *z*−1) *and* (ii) *to the* N *position* (*x*, *y*, *z*−1)

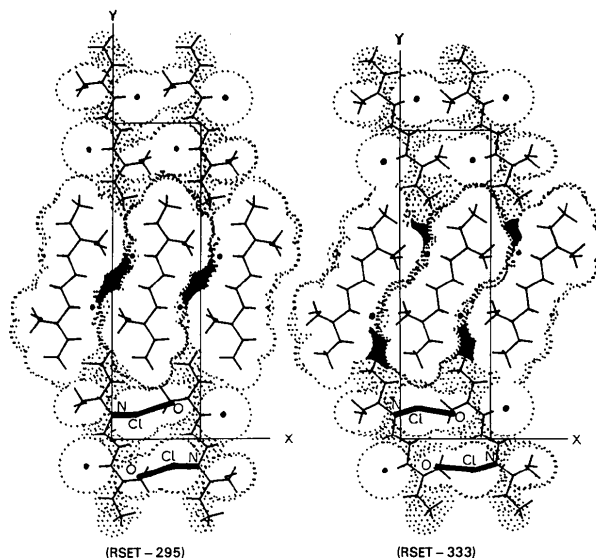|	(RSET–333)	(RSET–295)	(SSET)
O(1ⁱ)···Cl⁻	3·069 (3)	3·082 (2)	3·073 (4)
N(1ⁱⁱ)···Cl⁻	3·086 (3)	3·074 (2)	3·066 (4)
N(1)···Cl⁻	3·161 (3)	3·157 (2)	3·174 (4)

Fig. 3. The packing of the ions combined with a section of the van der Waals surfaces, showing in black the volume that is outside these surfaces. The thick lines in the lower part of the figures illustrate the N—H···Cl and O—H···Cl bonds.

(1985) in the crystal structure of S,S-ethambutol dihydrochloride (SSET) (Table 3).

The structures of (SSET) and (RSET–295) are closely related. Although (SSET) crystallizes in the orthorhombic space group $P2_12_12$ ($Z = 2$), the cell constants are, however, nearly the same [$a = 23\cdot183$ (22), $b = 6\cdot555$ (5), $c = 5\cdot176$ (5) Å for (SSET)]. The differences between corresponding bond lengths and bond angles range from 0·001 to 0·027 Å and 0·6 to 1·2°, respectively.

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Structure of Ethyl 1,2-Dihydro-2-oxo-3,6-diphenyl-4-pyridinecarboxylate

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Abstract. $C_{20}H_{17}NO_3$, $M_r = 319\cdot3$, monoclinic, $P2_1/n$, $a = 9\cdot0385$ (8), $b = 9\cdot613$ (2), $c = 18\cdot879$ (3) Å, $\beta = 92\cdot895$ (9)°, $V = 1640$ (2) Å^3, $Z = 4$, $D_m = 1\cdot301$ (3) g cm^{-3}, $D_x = 1\cdot292$ g cm^{-3}, Cu $K\alpha$, $\lambda = 1\cdot5418$ Å, $\mu = 6\cdot22$ cm^{-1}, $F(000) = 672$, $T = 295$ K, final $R(F) = 0\cdot0643$, $wR = 0\cdot0663$ for 1852 observed reflections, [$I \geq 2\cdot5\sigma(I)$]. In the unit cell the molecules are stabilized by N—H⋯O and C—H⋯O hydrogen bonds. Centrosymmetrically related molecules form dimers about the centre of inversion, and the hydrogen-bond pattern is reminiscent of pyrimidine-purine base pairing in nucleotides.

Introduction. Pyridine is generally used in the synthesis of vitamins and drugs and pyridine derivatives like isonicotinyl hydrazide are used in the treatment of tuberculosis (Metzler, 1977). The title compound is a metabolite of the anti-tubercular drug ethambutol [(R)-2,2'-(1,2-ethanediyldiimino)bis-1-butanol]. The structure solution was undertaken to verify the chemical structure.

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† DCB contribution No. 722.

Experimental. Colourless, transparent crystals, 0·4 × 0·3 × 0·25 mm, rectangular, prismatic, were crystallized from methanol/water. Density measured by flotation. Three-dimensional intensity data using Enraf–Nonius CAD-4 automated diffractometer; $\omega/2\theta$ scan mode with max. $2\theta = 130°$, graphite-monochromated Cu $K\alpha$ radiation. Intensity data were corrected for polarization and Lorentz effects, not for absorption ($\mu t < 1$). Unit-cell parameters were refined from 25 reflections in the range $30 \leq 2\theta \leq 58°$. The intensities of 2968 unique reflections were measured within the limits $-10 \leq h \leq 10$, $0 \leq k \leq 11$, $0 \leq l \leq 28$; 1852 observed with $I \geq 2\cdot5\sigma(I)$; three standard reflections monitored every 100 reflections showed no significant variation. Structure solution by direct methods using *SHELXS*86 (Sheldrick, 1986). The structure was refined using full-matrix least-squares technique. The function minimized is $\sum w(|F_o| - |F_c|)^2$ where $w = 1/[\sigma^2(|F_o|) + 0\cdot036066|F_o^2|]$ in *SHELX*76 (Sheldrick, 1976); H-atom positions from $\Delta\rho$ syntheses. Atoms C(23) and C(24) are disordered; two peaks of height 1·5 e Å^{-3} and 0·6 e Å^{-3} were located in the $\Delta\rho$ map at a distance of 0·7 Å to C(23) and 0·6 Å to C(24), respectively, and they have been included in the