

Fig. 8. Pressure-temperature phase diagram of  $\{(NH_4)_3H(SO_4)_2\}_{0.03}\{(ND_4)_3D(SO_4)_2\}_{0.97}$ . Vertical bars show the temperature hysteresis of first order transitions.

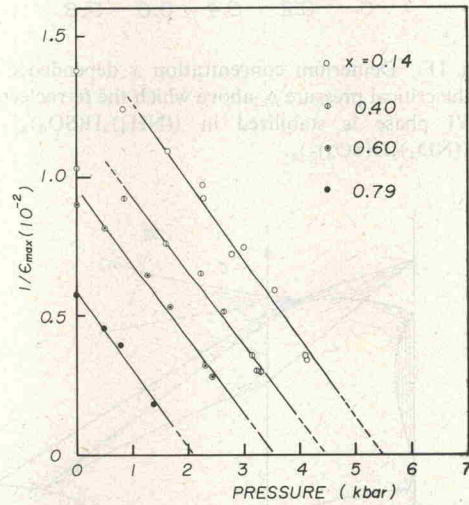
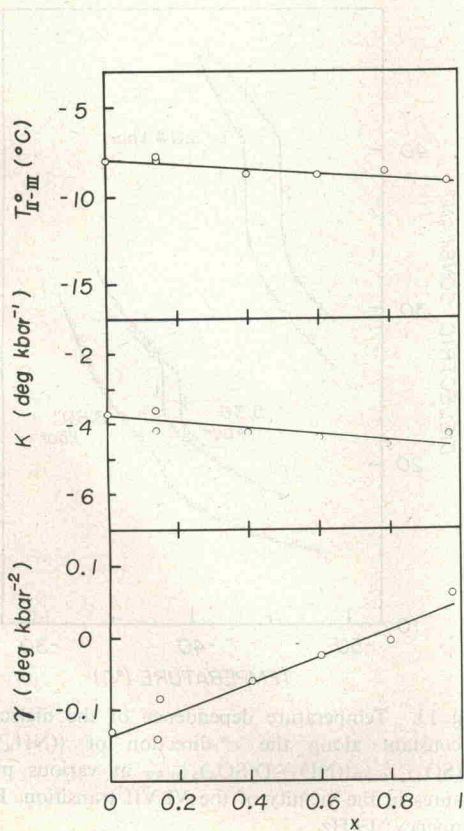


Fig. 10. Pressure dependence of the reciprocal of the maximum value  $1/\epsilon_{max}$  at the broad dielectric constant peak in Phase III for the compounds  $\{(NH_4)_3H(SO_4)_2\}_{1-x}\{(ND_4)_3D(SO_4)_2\}_x$  with various  $x$ .

← Fig. 9. Deuterium concentration  $x$  dependence of the parameters which describe the II-III phase boundary in the  $p$ - $T$  plane for the system  $\{(NH_4)_3H(SO_4)_2\}_{1-x}\{(ND_4)_3D(SO_4)_2\}_x$ : The II-III transition temperature  $\theta_{II-III}$  is represented by  $\theta_{II-III} = T_{II-III}^0 + Kp + \gamma p^2$ .

ferroelectric phase VI appears at atmospheric pressure for  $x > 0.9$ .

From the above results of the  $p$ - $T$  phase diagrams for various  $x$ , we can draw the  $x$ - $p$ - $T$  three dimensional phase diagram of the  $\{(NH_4)_3H(SO_4)_2\}_{1-x}\{(ND_4)_3D(SO_4)_2\}_x$  system. Figure 12 shows schematically the three dimensional phase diagram. For simplicity, the region of Phase IX is not demonstrated in

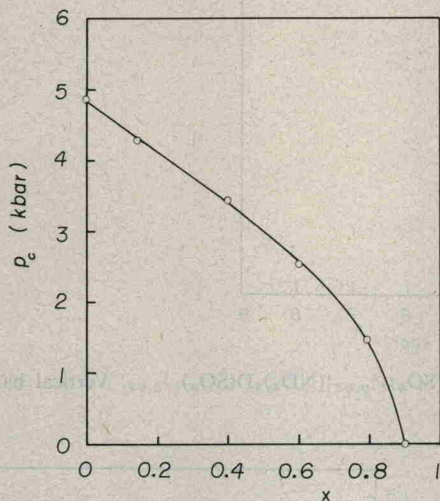


Fig. 11. Deuterium concentration  $x$  dependence of the critical pressure  $p_c$  above which the ferroelectric VI phase is stabilized in  $\{(NH_4)_3H(SO_4)_2\}_{1-x}\{(ND_4)_3D(SO_4)_2\}_x$ .

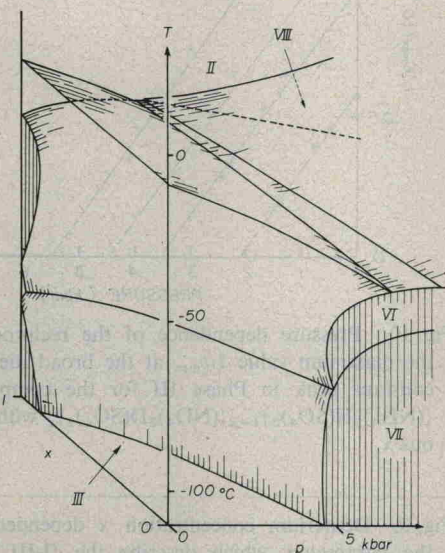


Fig. 12. Schematic picture of the three dimensional  $x$ - $p$ - $T$  phase diagram of the system  $\{(NH_4)_3H(SO_4)_2\}_{1-x}\{(ND_4)_3D(SO_4)_2\}_x$ . For simplicity, the region of Phase IX is not shown.

the figure. This figure shows the relation between the phases at atmospheric pressure in deuterated compound and those at high pressures in the normal compound. Especially the correspondence is well established between the two ferroelectric phases of VI and VII in the deuterated crystal and the pressure-induced ferroelectric phases in the normal compound. Also Phases III and III' in the deuterated compound at atmospheric pressure should be isostructural.

The critical point of the ferroelectric VI to ferroelectric VII transition was expected to be found in a lower pressure region in deuterated compound.<sup>6)</sup> Figure 13 shows the temperature dependence of the dielectric constant at different pressures in the vicinity of the VI-VII transition for  $x = 0.97$  compound. At 3.04 kbar a marked discontinuous change in the dielectric constant accompanied with a thermal hysteresis is seen. The discontinuous change in the dielectric constant becomes smaller as pressure increases, and at 8.6 kbar it is hardly observable. Figure 14 indicates the pressure dependence of the square root of the

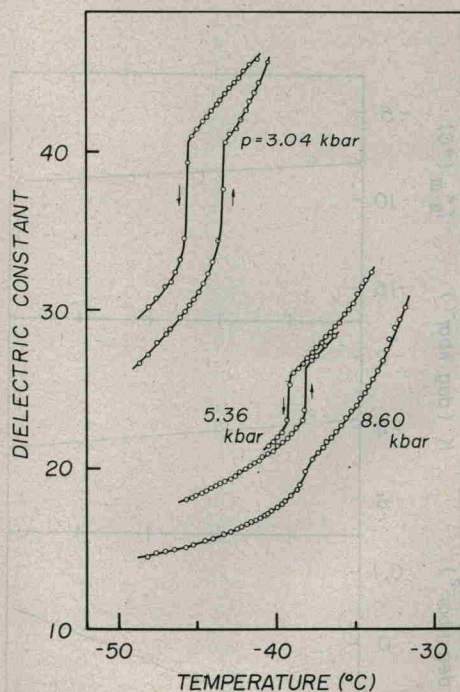


Fig. 13. Temperature dependence of the dielectric constant along the  $c^*$ -direction of  $\{(NH_4)_3H(SO_4)_2\}_{0.30}\{(ND_4)_3D(SO_4)_2\}_{0.97}$  at various pressures in the vicinity of the VI-VII transition. Frequency: 1 kHz.