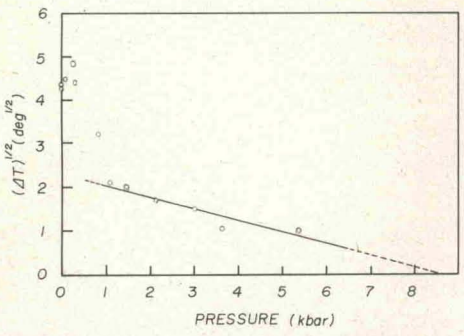

Fig. 14. Pressure dependence of the square root of the temperature hysteresis $(\Delta T)^{1/2}$ of the VI-VII transition in $\{(NH_4)_3H(SO_4)_2\}_{0.03}\{(ND_4)_3D(SO_4)_2\}_{0.97}$.

thermal hysteresis $(\Delta T)^{1/2}$ observed for $x=0.97$ compound as a measure of the first order nature of the transition. It is seen that the first order nature of the VI-VII transition disappears at around 8.5 kbar, and above this critical pressure the transition becomes to be continuous.

§4. Conclusive Remark

From the results described in the preceding section the followings are concluded:

(1) The two ferroelectric phases in the deuterated compound at atmospheric pressure correspond to the pressure-induced ferroelectric phases VI and VII in the normal compound, respectively.

(2) The two intermediate phases VIII and IX are newly found in high pressure region in the normal-deuterated compound system.

(3) In the deuterated compound with $x=0.97$ the critical point of the ferroelectric-to-ferroelectric VI-VII phase transition exists at a relatively low pressure around 8.5 kbar.

As to the stabilization of the ferroelectric phases of VI and VII, the effects of the deuterium-substitution and pressure application are analogous, namely, both the deuterium-substitution and the pressure application enhance the ferroelectricity. On the other hand, the II-III transition temperature varies very slightly by the deuterium-subsitution, but it is depressed by the pressure-application with a moderate rate. For the present, the nature of the II-III transition is not understood since the crystal structure of Phase III has not been known as yet. Although the dielectric anomaly associating the II-III transition is insignificant, there is a conspicuous specific heat anomaly with a wide critical region.[10] Probably a certain kind of super-lattice structure would exist in Phase III. The dielectric anomalies at the II-IX-VI transitions shown in Fig. 1 resemble those found for the paraelectric-incommensurate-ferroelectric phase transitions in $NaNO_2$.[7] So that an incommensurate long period structure may be expected in Phase XI by the analogy with the case of $NaNO_2$.

In the present study the nature of the critical point of the VI-VII transition is not clarified: Whether the critical point is tricritical or gas-liquid type? In order to solve the problem it is necessary to determine the transition parameter of the VI-VII transition. Since the ferroelectric VI and VII phases can be seen at atmospheric pressure in the deuterated compound, the transition parameter will be easily determined from the crystal structure analysis on the deuterated compound. The mechanism of the complicated phase transitions and the unique pressure and isotope effects observed in $(NH_4)_3H(SO_4)_2$ are the problem to be solved in future.

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