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NOTE

Study of Nuclear Spin Conservation in Ammonium Alkali Halide Crystals

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The mixed salt $(NH_4)_cK_{1-c}X$ (X = I, Br), prepared from aqueous solutions, can maintain a spin-temperature inversion. It is shown that both the mixed salts (1, Br) relax at similar rates and these rates increase with the temperature variation (T^3). The findings and results have been discussed in the context of other works on these salts.

Key Words: Aqueous solutions, Spin-temperature inversion, Temperature variation.

The ammonium-alkali halide crystals are considered to be popular samples for the study of interactions in solid state. Recently it has been shown that the onset of the orientational glass phases in these materials is moderated through the strain field[1]. At low temperature of 4 K and at very low NH_4^+ ion concentrations C (C *ca.* 0.02), a sharp rotational tunneling spectrum can be observed in the INS spectrum. As the value of C increases the sharp line is accompanied by a broad shoulder of intensity to lower energies[2–4]. Alternatively, as the temperature is raised the tunneling line remains at a fixed position but quickly loses intensity and is completely absent at 15 K. Previously, it was reported that a sample of $(NH_4)_{0.005}K_{0.995}$,Br, grown from the melt, could maintain a non-equilibrium spin-temperature[5]. Recently there has been increased interest in spin conservation in rotational tunneling systems. A number of experimental investigators[6] were reported and theoretical models have also been suggested for the spin conservation processes[7–9].

In this article we shall be concerned with the study of the temperature dependence of the spin relaxation dynamics of low C samples.

Theoretical Analysis

Analysing the findings, three distinct transitions are seen in the INS spectrum of $(NH_4)_{0.01}K_{0.99}I$ at 1.4 K. The principal line at 600 μeV is the neutron energy loss transition labelled as 0–1.[2]

The corresponding neutron energy gain line, 1–0, is at –600 μeV. At thermal equilibrium the ratio of the intensities of these two lines {R = I(0–1)/I(1–0)} should be *ca.* 140; the observed intensity ratio is *ca.* 2. Therefore, the sample is not in thermal equlibrium; specifically the proton-spin temperature of the ammonium ion is too high. This confirms the 1–2 transition seen at 750 μeV. As time evolves, the intensity of the 0–1 transition increases but that of 1–0 and 1–2 decreases and the samples slowly recover thermal equilibrium. The same 0–1, 1–0 and 1–2 features are observed in the bromide but at different energy transfers. The time evolution of the line intensities was followed for several hours at three temperatures for the bromide but only at the lowest temperature for the bromide. The derived characteristic decay times for the 1–0, at different temperatures, are shown in Fig. 1. The value reported previously for the bromide, *ca.* 37 ± 4 h at 1.8 K,[5] compares well with our value of 3.33 h at 1.4 K. We have compared the temperature variations with those predicted by different theories.

If I suppose that relaxation is a thermally activated process, then I find an Arrhenius activation energy of 200 μeV. In the bromide system, at temperatures above 8 K, calorimetric measurements are well described by an Arrhenius law with a large activation energy. In the present analysis the temperatures are lower and 200 μeV is less than the 0–1 transition energy. It would be difficult to rationalize this low activation energy in terms of the energies of isolated ammonium ions and we reject the simple Arrhenius model. Alternately, relaxation proceeding through a Raman process should demonstrate a variation with temperature as T^{-7}.[9] This theory again fails to provide a reasonable fit to the data. The failure of these models is in keeping with the published work on bromide where the relaxation rate fell smoothly towards a constant at the lowest temperature[5]. In Fig. 2 we show our results where $1/\tau$ is plotted against T^3. The published data for bromide[3] are also plotted on the same axes.

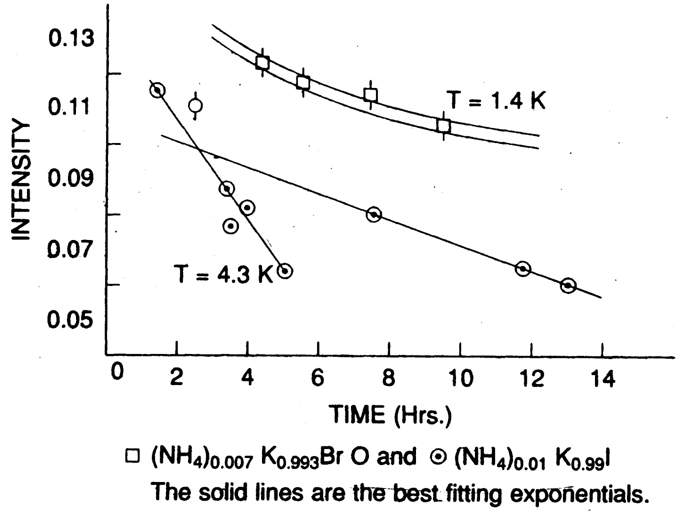

Fig. 1. Decrease in intensity, with time, of the 1–0 transition in the INS spectrum at different temperatures (□)-(NH$_4$)$_{0.007}$K$_{0.993}$Br; (*), (0) and (•) – (NH$_{0.01}$) K$_{0.001}$. The solid lines are the best fitting exponentials

Conclusion

It is known that the phonon population is well represented by the Debye T^3 at low temperatures. We believe its appearance here reflects the important role played by the number of phonons in the salt. From Fig. 2 we see that the relaxation process, at a given temperature, occurs at the same rate in the bromide and the iodide.

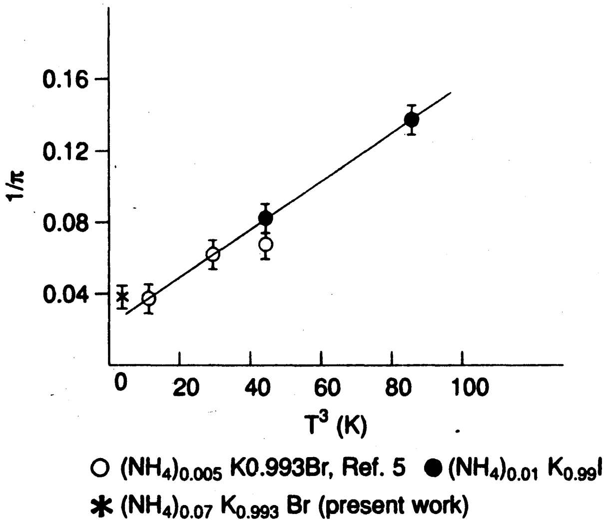

Fig. 2. Variation of characteristic decay times, (0) $(NH_4)_{0.005}K_{0.995}Br$; Ref. [5], (•) $(NH_4)_{0.01}K_{0.99}I$ and (*) $(NH_4)_{0.07}K_{0.993}Br$ (present work).

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