PHYSICAL REVIEW B VOLUME 15, NUMBER 5 1 MARCH 1977

Addendum: New evidence for the formation of dynamic clusters at temperatures in the vicinity of a structural phase transition*

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(Received 3 September 1976; revised manuscript received 29 November 1976)

As a result of studying the chlorine nuclear spin-lattice relaxation time for crushed single crystals of K_2OsCl_6 it has been definitively shown that the difference in behavior of the nuclear relaxation data for powder and single-crystal samples of this compound is *not* a size effect.

Chlorine nuclear spin-lattice relaxation-time measurements in K_2OsCl_6 near the structural phase transition at $T_c = 45$ K have been reported.[1] Data taken for powder samples obtained from two different suppliers were consistent and revealed that below 120 K the recovery of the nuclear magnetization following a saturating pulse was not describable by a single exponential. However, it was possible to analyze the recovery curves on the basis of a two exponential model and to show that the number of nuclei which contribute to the short component increases dramatically as the sample temperature approaches T_c. These results were presented as evidence of the development of tetragonal-phase dynamic clusters in the cubic phase of K_2OsCl_6 near T_c. Some of the same K_2OsCl_6 powder was used to grow single crystals from an aqueous solution. Measurements of the recovery of the chlorine nuclear magnetization following a saturating pulse revealed that for the crystals the relaxation was describable by a single spin-lattice relaxation time at all temperatures.

An obvious and important question is the following: what is it that is different in the powder than in the crystal that gives rise to the difference in the behavior of the nuclear relaxation data? The most obvious difference is that of size. The single crystals have a linear dimension of ~0.025 mm. Is the difference in sample size responsible for the difference in the behavior of the nuclear relaxation data? To answer this question four single crystals were crushed and examined in stages until finally the average particle size was comparable to that of the crystallites in the powder sample. The original single crystal data,[2] the present single-crystal data and the crushed single-crystal data all agree and yield a single spin-lattice relaxation time T_1 at each temperature. The temperature dependence of T_1 is shown in Fig. 1 along with that for the long component T_1^l obtained from the powder.[1] It may be concluded that the difference in size between the crystals and the particles of the powder is *not* responsible for the difference in behavior of the nuclear relaxation data. Although this result was anticipated in the earlier publication it was *not* shown by direct experimental evidence. In view of the present controversy[3,4] regarding the mechanism responsible for central peaks this addition to our previous work is of particular significance.

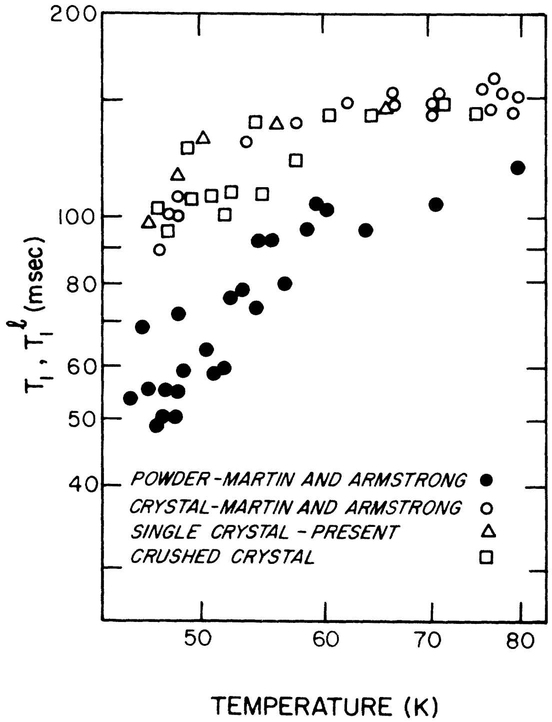

FIG. 1. Temperature dependence of T_1 for single-crystal samples and T_1^l, for a powder sample of K_2OsCl_6 near $T_c = 45$ K.

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*Research supported by the National Research Council of Canada.

[1] R. L. Armstrong and C. A. Martin, Phys. Rev. Lett. 35, 294 (1975).

[2] C. A. Martin and R. L. Armstrong, J. Magn. Reson. 20, 411 (1975).

[3] J. A. Krumhansl and J. R. Schrieffer, Phys. Rev. B 11, 3535 (1975); T. Schneider and E. Stoll, Phys. Rev. Lett. 31, 1254 (1973); 35, 296 (1975); R. A. Cowley, J. D. Axe, and M. Iizumi, *ibid.* 36, 806 (1976).

[4] B. I. Halperin and C. M. Varma, Phys. Rev. B 14, 4038 (1976); K. A. Müller and W. Berlinger, Phys. Rev. Lett. 37, 916 (1976).