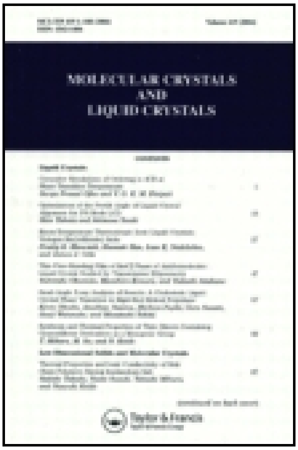

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An NMR and Specific Heat Study of the Spin-Peierls system TTF-Aubdt

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AN NMR AND SPECIFIC HEAT STUDY OF THE SPIN-PEIERLS SYSTEM TTF-AuBDT

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Abstract Field-induced transitions in the Spin-Peierls compound TTF-AuBDT are studied by means of NMR and specific heat measurements. The data agree qualitatively with the presence at low temperatures of an incommensurate soliton-lattice state above a critical field B_c = 2.25 T.

Experimental research on field-induced transitions in Spin-Peierls (SP) systems is of great current interest[1,2]. The SP-problem in a 1-dimensional (1-d), S=½, Heisenberg antiferromagnet can be mapped via the Jordan-Wigner transformation to that of a 1-d system of interacting spinless fermions coupled to a 3-d phonon lattice (the Peierls problem). Most importantly, the application of a magnetic field in the SP-problem corresponds with a decrease of the number of fermions, enabling a study of the Peierls system as a function of band filling. The dimerization wave-vector is expected to be pinned up to a critical field B_c of the order of $k_B T_{SP}/g\mu_B$, where T_{SP} is the B=0 transition temperature. Various models predict different intermediate phases for $B > B_c$, separating at low temperatures the dimerized phase from the paramagnetic phase[1]. In many of these models the phase immediately above B_c is predicted to be a soliton-lattice[3]. In an alternative approach the possible absence of a separate intermediate phase in the experimental systems has been discussed in terms of a domain-wall model[1,4].

In order to investigate these questions a systematic study of the field-dependent behavior of the SP system TFF-Au BDT has been started at our laboratory[5,6]. Here NMR and preliminary new specific heat data are reported. The compound has a low $T_{SP}(0)$ = 2.0 K, implying a conveniently low critical field B_c = 2.2 T.

The shape of the spin-echo in a pulsed NMR experiment is the Fourier transform of the distribution function of static local magnetic fields at the nuclei. In fig. 1a the inhomogeneous half-

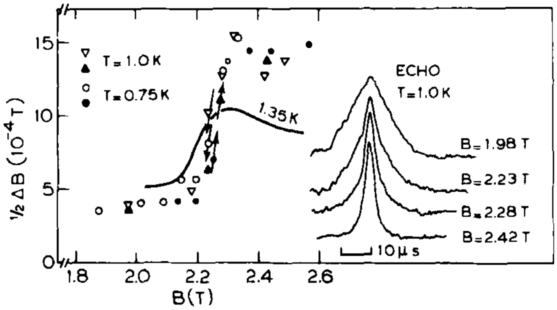

Fig. 1a

FIGURE 1: Inhomogenious NMR half-linewidth for the protons as obtained from the decay time of the echo measured at constant temperatures (a), and constant field (b).

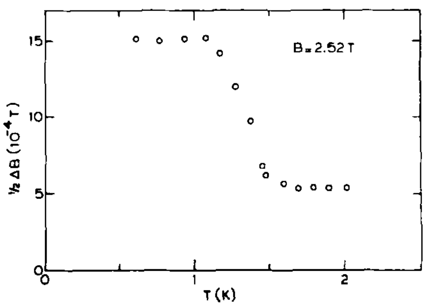

Fig. 1b

linewidth $\frac{1}{2}$ ΔB, obtained from the echo decay-time for the protons, is plotted versus field. For T = 0.75 K and 1.0 K, sharp increases were observed at B = 2.25 T. In the paramagnetic phase and also in the dimerized phase $\frac{1}{2}$ ΔB is found to be mainly determined by the inhomogeneity in the applied field. In a narrow field range ($\simeq$ 0.05 T) around 2.25 T the echo is composed of two components (insert fig. 1a), suggesting the coexistence of two phases. Together with the apparent hysteresis of about 30 mT, this strongly suggests a first-order transition. On the other hand, the behavior found at T = 1.35 K appears to be second-order like. Also, in the temperature scan of $\frac{1}{2}$ ΔB at a constant field B = 2.52 T (fig. 1b) a very gradual increase is seen as the temperature is lowered, implying that the transitional behavior is quite different along this path in the phase diagram. Previously[5] it was found that the magnetization is nearly temperature independent for B $\gtrsim$ 2.5 T. Thus we attribute the observed increases in $\frac{1}{2}$ ΔB to the development of an inhomogeneous distribution of the magnetic moments along the chains in the region B > B_c and T $\lesssim$ 1.2 K. Although the present NMR data cannot fully exclude the presence of an (ordered)

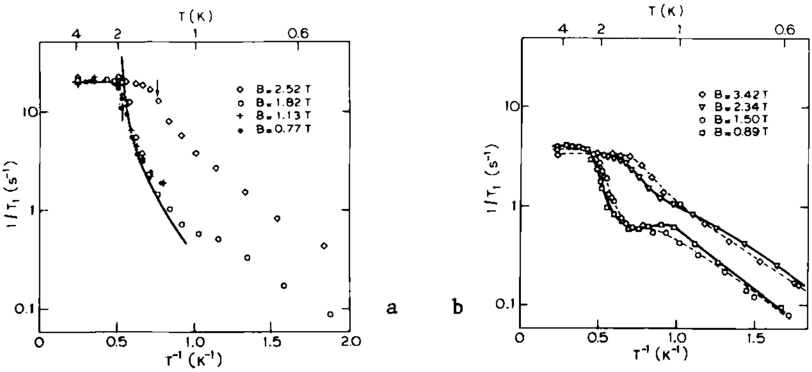

FIGURE 2: Nuclear spin-lattice relaxation rate for the protons (a) and the fluorine nuclei (b).

antiferromagnetic phase above B_c, they agree qualitatively with the soliton-lattice and/or domain wall models[3,4], in which a net spin ½ is localized near the centre of each soliton (wall). The soliton (wall) patterns should be static with respect to the NMR time scale. This contrasts with the case of an incommensurate phase with a sinussoïdal modulation of the dimerization pattern for which the magnetization is expected to remain uniformly distributed along the chain[3].

The spin-lattice relaxation rates $1/T_1$, (figs. 2a,b) of the protons on the TTF- and the fluorine nuclei on the BDT molecules decrease rapidly in the dimerized phase. For $B < B_c$ the behavior appears to be independent of B, and therefore the data for B = 0.77 T were fitted to the theory (for $\omega = 0$) of Ehrenfreund and Smith[7]. The coupling strength was adjusted so as to obtain the correct value for $1/T_1$ in the paramagnetic phase, and the correct height of the curve below T_{SP} was obtained by choosing the zero temperature gap Δ/k equal to 1.8 K. The slope of the curve at relatively high temperature reproduces the data reasonably well, but the anomalies at low temperature are not quite understood at present. For $B > B_c$ we also observe $1/T_1$ to decrease at low temperatures, indicating that this region of the BT-plane differs from the high-temperature paramagnetic phase.

Lastly, preliminary specific heat data are shown in fig. 3 for different fields. A broad short-range order maximum is seen to develop with increasing B, at the cost of the sharper anomaly marking the SP transition. Although also for $B > B_c$ some reminiscence of this peak seems to be present, the accuracy of the present data does not allow definite conclusions. Thus the existence or non-existence of the high-field phase boundary (dashed line in fig. 4) is still in doubt. The NMR, specific heat and

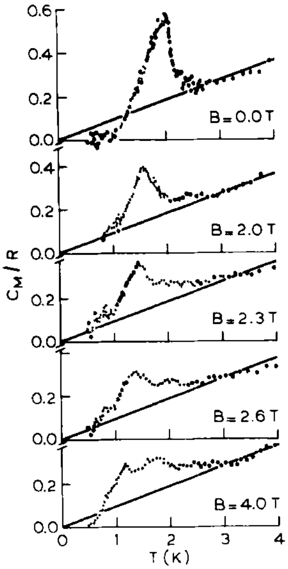

FIGURE 3: Specific heat versus temperature.

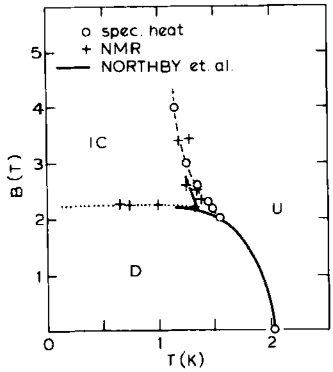

FIGURE 4: The phase diagram of TTF-AuBDT. The letters U,D and IC denote the uniform-, dimerized- and possibly incommensurate phase, respectively.

differential susceptibility experiments are currently being extended to solve this intriguing problem.

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