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Influence of linear and nonlinear excitations in the ferromagnetic $S=1/2$ chain system $[C_6H_{11}NH_3]CuBr_3$ (CHAB)

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The magnetic properties of the title compound have been studied by measurements of the nuclear spin-lattice relaxation time T_1, the uniform magnetization M, and by quasielastic and inelastic neutron scattering. The field and temperature dependence of T_1 and the wave-number dependence of the magnon excitation energy can be satisfactorily described by linear spin-wave theory. A good description of M, however, requires a contribution from kink solitons. The available evidence suggests that, in contrast to the linear excitations, the nonlinear excitations in CHAB for $\mathbf{B}\|\mathbf{c}$ can be described very well by a classical model, which is consistent with the interpretation of earlier heat-capacity measurements.

INTRODUCTION

$[C_6H_{11}NH_3]CuBr_3$ (CHAB) is a system built up from ferromagnetic $S = 1/2$ chains with a nearest-neighbor interaction of about 55 K, which contains 5% easy-plane anisotropy. The interchain interactions are smaller by three orders of magnitude,[1] and induce a three-dimensional magnetic ordering below $T_c = 1.50$ K. If at temperatures above T_c a symmetry-breaking field is applied within the easy (XY) plane, the equation of motion of the spins in this compound can be mapped to a sine-Gordon (sG) equation, at least under certain approximations.[2] Apart from linear excitations (magnons), this equation has nonlinear solutions called kink solitons. We have analyzed the influence of the linear and nonlinear excitations on various experimentally observable properties of CHAB. As an extension of earlier investigations on the magnetic heat capacity of this compound,[3] we present in this paper an analysis of the nuclear spin-lattice relaxation time T_1 of the hydrogen nuclei, the magnetization for applied fields along the c axis (within the XY plane), the intrachain correlation length, and the magnon dispersion relation.

The crystallographic structure of CHAB is orthorhombic, space group $P2_12_12_1$. The magnetic properties of the individual chains can be described by the Hamiltonian[1,4]

$$\mathscr{H} = -2\sum_i \left(J^{xx}S_i^xS_{i+1}^x + J^{yy}S_i^yS_{i+1}^y + J^{zz}S_i^zS_{i+1}^z\right). \tag{1}$$

with $J^{xx}/k_B = 55 \pm 5$ K, $J^{zz}/J^{xx} = 0.95$, and $(J^{xx} - J^{yy})/J^{xx} = 5\times 10^{-4}$. The y axis coincides with the crystallographic c axis, whereas the x axis lies within the ab plane at an angle φ from the b axis. Two symmetry-related types of chains are present, with $\varphi = -25°$ and $\varphi = 25°$, respectively. We will confine ourselves to measurements collected with the external field $\mathbf{B}\|\mathbf{c}$, which is located in the XY plane for both types of chains.

NUCLEAR SPIN-LATTICE RELAXATION

The spin-lattice relaxation rate T_1^{-1} of the hydrogen nuclei was measured for $1.2\ \text{K} < T < 6.5$ K and external fields $0 < B < 70$ kG by means of a spin-echo technique. Following the usual approach,[5] we plotted the data as $\ln(TT_1^{-1})$ against $\sqrt{B}/T$ in Fig. 1. Since the relaxation of the nuclear spin system towards equilibrium is induced by fluctuations in the electron spin system, it can be expressed in terms of the various elementary magnetic excitations. In the figure we have included the calculated contribution of the two-magnon (Raman) process $(T_1^{-1})^R$, the three-spin-wave process $(T_1^{-1})^T$, and that arising from solitonlike excitations $(T_1^{-1})^{\text{sol}}$. It appears that for $\sqrt{B}/T > 1$ the field and temperature dependence of the relaxation rate can fully be explained by the two-magnon process. At lower values of $\sqrt{B}/T$, the data can be described fairly well by the sum of two- and three-magnon processes (dotted curve). All these processes are calculated from standard linear spin-wave theory, based on Eq. (1) and the parameters appropriate to CHAB. The inclusion of soliton processes seems to improve the description of the data below $\sqrt{B}/T = 0.6$, (see inset), but their effect on the relaxation rate is very small.

MAGNETIZATION

The magnetization (M) was measured with a commercial (PAR) vibrating sample magnetometer for $0 < B < 50$ kG and $1.4 < T < 10$ K. Since for several magnetic model systems, including the sG model, the reduced magnetization M/M_s is an universal function of $T/\sqrt{B}$, we plotted our ex-

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[a] Also at the Risø Research Centre, Roskilde, Denmark.

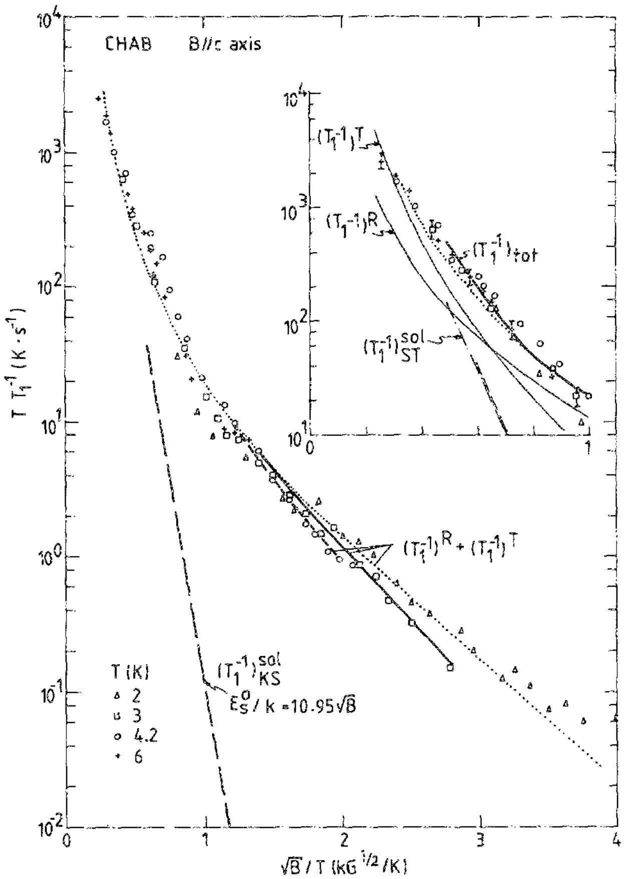

FIG. 1. ^{1}H nuclear spin-lattice relaxation rate T_1^{-1} in CHAB for B||c plotted as $\log(TT_1^{-1})$ against $\sqrt{B}/T$. The contributions from the Raman and three-spin-wave processes are denoted by $(T_1^{-1})^R$ and $(T_1^{-1})^T$, respectively. The contribution from solitons is represented by curves labelled $(T_1^{-1})^{\text{sol}}$. The subscripts of these curves refer to the soliton density used in the corresponding calculation (Ref. 6).

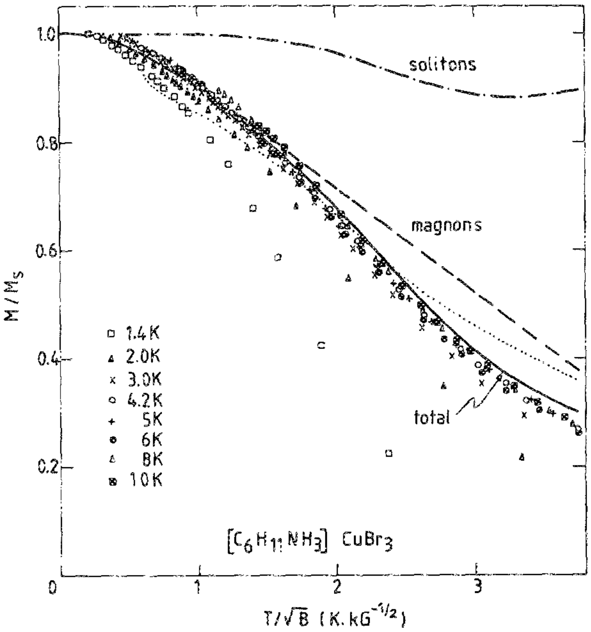

FIG. 2. Reduced magnetization of CHAB for B||c plotted against $T/\sqrt{B}$. The dotted curve reflects numerical calculations on a classical sG system (Ref. 7). The dashed curve represents the reduction of M according to linear spin-wave theory, whereas the dashed-dotted curve denotes the additional reduction from kink solitons. The solid curve reflects the sum of these two reductions.

perimental results in such a way in Fig. 2. Inspection of this figure shows that the data collected above 3 K almost perfectly collapse onto a single curve, suggesting universal behavior. At lower T systematic deviations occur, which are due to the small interchain interactions. In the figure we have included several theoretical predictions. The dashed curve represents the decrease of M from its saturation value M_s, calculated from linear spin-wave theory using Eq. (1) without any adjustable parameters. It is obvious that for $T/\sqrt{B} > 1$ systematic deviations between this prediction and the data occur, suggesting the presence of other excitations, which are not included in the theory. Within the framework of the classical sG model, the free energy can written[6] as $F = F_{\text{sol}} + F_m$, where F_{sol} is proportional to the soliton density n_{sol} and F_m reflects the contribution of linear excitations (magnons). The decrease of M resulting from solitons, calculated by differentiation of F_{sol} with respect to B, is denoted by the dashed-dotted curve. If we add this decrease to that calculated from linear spin-wave theory, thus replacing F_m in first order by its quantummechanical counterpart, we obtain the result reflected by the solid curve, which describes the experimental data very well. The dotted curve represents (exact) numerical calculations on the sG model.[7] These calculations, in which all elementary excitations are implicitly included, systematically deviate from the data, also at low values of $T/\sqrt{B}$, where the contribution of solitons is insignificant. Possibly, this is caused by the presence of spin components out of the XY plane, which are not taken into account in this model. However, attempts to describe the observed magnetization by other classical models, i.e., discrete systems of classical spins having, alternatively, two or three nonzero components, were also unsuccessful. One might therefore conclude that for a correct description of the present compound for B||c a quantum treatment of the linear excitations is necessary. Such a conclusion is supported by measurements of the heat capacity C,[3] which revealed that, in contrast to C itself, the excess heat capacity $\Delta C = C(B) - C(0)$, which is dominated by nonlinear excitations,[6,7] can be described fairly well by the classical sG model.

NEUTRON SCATTERING

The development of magnetic correlations within the individual chains in CHAB in zero field was investigated by quasielastic neutron scattering experiments on the deuterated compound. These correlations give rise to a diffuse scattering in planes in reciprocal space perpendicular to c^*. From measurements of the magnetic scattering cross section by scans perpendicular to such a plane the correlation length ξ can be deduced. The temperature dependence of the inverse correlation length κ evaluated by approximating the observed scattering profile by a Lorentzian is plotted in Fig. 3 as κ/T against T. In this figure we included several theoretical predictions, all calculated with the parameters appro-

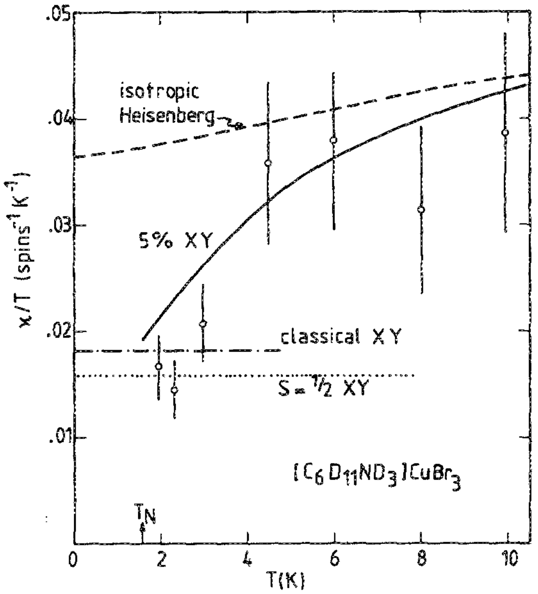

FIG. 3. Temperature dependence of the inverse correlation length κ along the chains of deuterated CHAB, plotted as κ/T against T. Experimental data are represented by open circles. The theoretical predictions are calculated without any adjustable parameters. Note the small difference between the classical (Ref. 8) and quantum-mechanical (Ref. 9) XY models.

priate to CHAB. From the figure it is obvious that the data show a crossover from Heisenberg behavior at high temperatures to XY behavior at temperatures below about 3 K. A good overall description of the data is given by the results of transfer-matrix calculations on a discrete classical system of spins having three nonzero components, i.e., the original spin Hamiltonian [Eq. (1)], which are reflected by the solid curve.

Although the correlation length has been determined at $B = 0$, in which case we deal with a different model system as that for $\mathbf{B} \| \mathbf{c}$, the good agreement of the classical prediction with the neutron scattering data might indicate that the spin-spin correlations in this case are governed by nonlinear excitations.

To obtain information on the linear excitations in the present system at $B = 0$ we studied the magnon-dispersion relation by means of inelastic neutron scattering experiments. Up till now, well-defined signals have been observed at 1.5 K and reduced wave vectors $0.07 \leqslant q_c \leqslant 0.25$ r.l.u. We tried to describe the wave-number dependence of the magnon energy by linear spin-wave theory, based on Eq. (1). Least-squares fits of the corresponding dispersion relation resulted in a good description of the data for an intrachain interaction $J/k_B = 66 \pm 1$ K, which is about 20% higher than the value $J/k_B = 55 \pm 5$ K deduced from heat-capacity measurements.[1] The reason for this discrepancy is not yet clear.

DISCUSSION

In concluding we would like to remark that a consistent description of various magnetic properties of CHAB for $\mathbf{B} \| \mathbf{c}$ seems possible if the linear excitations are described by conventional spin-wave theory and the nonlinear excitations by a classical model. In the presence of such an external field the sG model appears to be appropriate, as can be inferred from the magnetization data presented above as well as from measurements of the excess heat capacity reported before.[3] In view of the present results it may be worthwhile to investigate whether also in the case of $CsNiF_3$ the observed inadequacies of a classical model[10] can be explained by the poor description of the linear excitations by such a model.

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