

One-Dimensional Antiferromagnetic Properties of KCuF_3

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KCuF_3 is one of the typical example of the Heisenberg type one-dimensional antiferromagnet with $S=1/2$. It has a broad maximum in susceptibility curve at about 243°K but the antiferromagnetic ordering temperature is only 20°K (or 38°K). Recent experimental results on crystallographic, susceptibility, specific heat, neutron diffraction, ESR and NMR measurements etc. are summarised and more recent NMR studies of the dynamics of this system will be reported in greater detail. All these results are strongly characteristic of one-dimensionality of this spin system.

§1. Introduction

The importance of studying one-dimensional magnetic properties is in the fact that not only various physical quantities for instance susceptibility, specific heat, spin static correlation and some dynamic correlation can be calculated without serious mathematical difficulties as experienced in the 3-dimensional case, but also we can often find unexpected magnetic behaviour, which is difficult to see in 3-dimensional systems. Some of the quantities can be calculated without approximation analytically as well as numerically by starting from a finite number of atoms in a chain and extrapolating it to infinity. We have now quite a lot of theoretically calculated results on static and dynamic properties of the pure one-dimensional magnetic system.¹⁾ But non of real materials is truly one-dimensional. There are always a small amount of interchain interactions which cause three-dimensional phase transition at non-zero temperature though $T_N(T_c)$ might be unusually low compared to the three-dimensional case where the transition occurs at nearly the same temperature for interatomic (intra-chain) interaction J/k . In one-dimensional case, there is a broad maximum in specific heat as well as in susceptibility curves at T_{max}^c and T_{max}^x respectively.²⁾ These temperatures may be a measure of intra-chain exchange interaction and below which the SRO develops remarkably until the $T_N(T_c)$ is reached. The wide region $T_N(T_c) \sim T_{\text{max}}^x$ is the most interesting region because all sorts of critical phenomena may readily be seen. It should be noted that in this critical region even a weak interchain interaction can cause a great change in $T_N(T_c)$ ³⁾ and also even a much weaker anisotropy energy may change drastically the divergent nature of physical quantities in nearly one-dimensional system. The anomalously soft

dynamic behaviour persists even at temperatures much lower than the critical point. All these properties characteristic for the quasi-one-dimensional system can readily be seen in KCuF_3 as described later.

KCuF_3 is a very interesting substance not only because it is a one-dimensional antiferromagnet but also the one-dimensionality is one of the most ideal one ever found with $S=1/2$ though the crystal is of the perovskite type. This one-dimensionality is predominantly attributed to the cooperative Jahn-Teller effect which produces a special alignment of $3d$ wave functions of Cu^{2+} ions. As the critical region is very wide, it will help deeper understanding of the order-disorder transition in actual three dimensional case.

Ten years ago, Hirakawa et al.⁴⁾ reported on the magnetic properties of a series of perovskite type compounds KMF_3 measured on the powdered specimen. They found, in KCuF_3 , a very broad peak around 243°K suggesting the antiferromagnetic order below this temperature. Almost at the same time, Scatturin et al.⁵⁾ performed neutron diffraction at 4.2°K with the powdered specimen, but they could not get any information about magnetic LRO. They interpreted this curious fact as a result of the lack of ordering between one-dimensional antiferromagnetic chains. But no further attention had been called so far until bigger single crystals were grown and detailed measurements were made in our laboratory.^{6),7)} A lot of experimental studies performed since⁸⁾⁻¹¹⁾ has exposed new properties of this substance and some of the old data are to be corrected from the present stand point of view. So it seems better firstly to make clear that what are the correct properties and then to present our new data hitherto obtained.

§2. Experimental results

a) *Crystal Growth*

After a lot of test for growing conditions, we now came to grow fairly big crystals up to about $15 \times 15 \times 11$ mm in size. The direction is as follows: taking 75 gr of $\text{CuCO}_3 \cdot \text{Cu}(\text{OH})_2 \cdot \text{H}_2\text{O}$ with 900 cm^3 of 60°C water in a polyethylene beaker, 45% HF aqueous solution is slowly added until the ingredient becomes just transparent blue. Then 800 cm^3 of 70°C aqueous solution which contains 41 gr of KF is added. The solution is then kept in a furnace controlled at 37°C and the solvent is slowly evaporated in a room of low humidity. The method how to grow the (a) type only or the (d) type only (see the next paragraph) is not known as yet. Both types of crystals are pale violet and hard to distinguish in appearance.

b) *Crystallographic Properties*

Careful determination of the crystal structure and atomic positions has been made early by Okazaki and Suemune.¹²⁾ They found a typical coopera-

tive Jahn-Teller distortion due to Cu^{2+} ions as proposed by Kanamori.¹³⁾ Recently, however, Okazaki⁸⁾ found two different crystal types and named the (a) type and the (d) type.*⁾ The old X-ray datum¹²⁾ was for the (a) type structure. The difference of these two types are in the stacking manner of distorted $Cu-F_6$ octahedra as show in Fig. 1. The crystals are of tetragonally distorted and the parameters obtained are summarized in Table I.

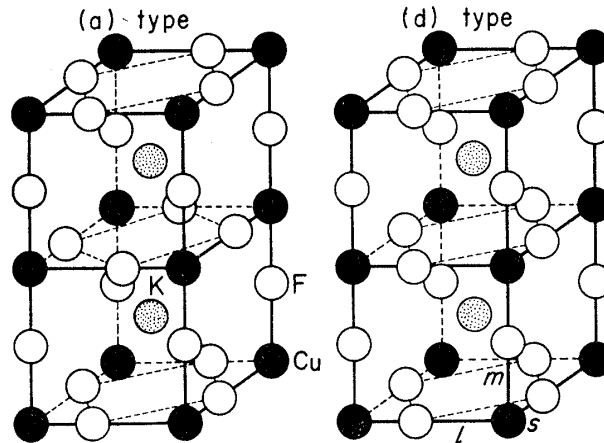


Fig. 1. Two crystal types of $KCuF_3$.

Table I. Various parameters of $KCuF_3$.

	(a) type	(d) type
Lattice parameters	$a_0 = 4.1410 \text{ \AA}$ $c_0 = 3.9237 \text{ \AA}$ $A = B = 2a_0$ $C = 2c_0$	$a_0 = 4.1396 \text{ \AA}$ $c_0 = 3.9303 \text{ \AA}$ $A = B = 2a_0$ $C = c_0$
T_{max}^z	243°K	243°K
T_N	38°K	22°K
J/k	-190°K	-190°K
J'/k	>0, very small (3°K after Oguchi's theory)	>0, very small (1°K after Oguchi's theory)
g_a } From ESR	2.28	2.25
g_c }	2.17	2.17
g_a } From χ_a, χ_c		2.27
g_c }		2.14
Magnetic moment per Cu^{2+} ion	0.54 μ_B	0.46 μ_B
From Neutron Diff.		

c) Wave Functions

According to the theory of Kanamori,¹³⁾ the ground state and the excited

*⁾ See the errata referred also.

state wave functions of a given Cu^{2+} orbital are written by the linear combination of two $d\gamma$ orbitals of the form

$$\frac{\sqrt{3}}{2}(x^2-y^2) \text{ and } \frac{1}{2}(3z^2-r^2), \text{ i.e.}$$

$$\psi_{\eta} = c\phi_{x^2-y^2} + c'\phi_{3z^2-r^2},$$

$$\psi_{\epsilon} = c'\phi_{x^2-y^2} - c\phi_{3z^2-r^2}.$$

The coefficients c and c' are determined if the normal modes Q_2 and Q_3 due to the displacements of ligand F^- ions are known. After Okazaki and Suemune,¹²⁾ the orthorhombically deformed Cu-F_6 octahedron has Cu-F distances $l=2.25 \text{ \AA}$, $m=1.96 \text{ \AA}$ and $s=1.89 \text{ \AA}$ as shown in Fig. 1, by which Q_2 and Q_3 are determined as

$$Q_2 = \frac{2}{\sqrt{2}}(l-s),$$

$$Q_3 = \frac{2}{\sqrt{6}}(2m-l-s).$$

From these Q_2 and Q_3 , c and c' are determined by

$$c/c' = \tan(\theta/2)$$

with

$$c^2 + c'^2 = 1,$$

where

$$\tan\theta = Q_2/Q_3.$$

Thus the wave functions are

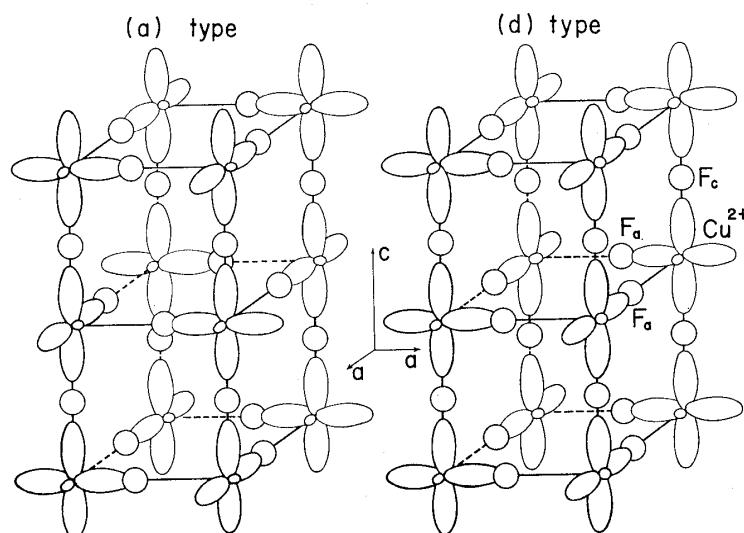


Fig. 2. Arrangements of the ground state wave function ψ_{η} in the (a) type and the (d) type crystals.

$$\psi_y = 0.092x^2 - 0.908y^2 + 0.816z^2$$

and

$$\psi_z = 0.995x^2 - 0.418y^2 - 0.577z^2.$$

The arrangements of the flat wave function ψ_y are shown schematically in Fig. 2.

d) *Magnetic Susceptibilities*⁶⁾

The measured magnetic susceptibilities along the a - and c -axes are shown in Fig. 3. The form is essentially the same as those obtained before on the powdered specimen.⁴⁾ The most peculiar is the extremely broad maximum around 243°K. When the temperature is lowered, no discontinuous change

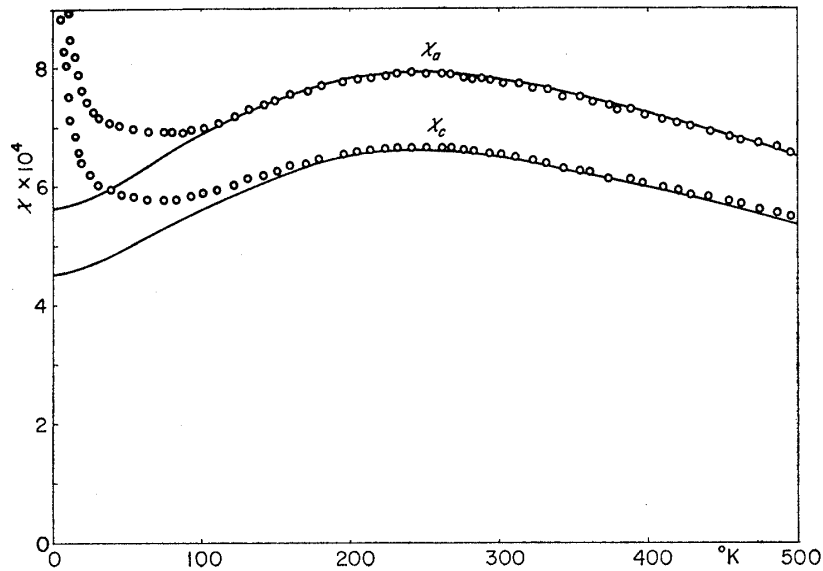


Fig. 3. Magnetic susceptibilities vs temperature curves measured along the c -axis (χ_c) and along the a -axis (χ_a) in the (d) type crystal. The open circles are the measured points and the curves are the calculated one using Bonner and Fishers results.

suggesting the onset of long range order appears, but the susceptibility increases gradually at low temperatures. At higher temperatures the curves are very close to the theoretical curve calculated by Bonner and Fisher²⁾ for the one-dimensional Heisenberg antiferromagnet. From the relation they found, we can estimate the intra-chain interaction J by

$$|J| = \frac{1}{1.282} k T_{\max}^{\chi}.$$

Using $T_{\max}^{\chi} = 243^{\circ}K$, we get $J/k = -190^{\circ}K$.

The curves for the a - and the c -axes are very nearly the same except for the differences of g -values and van Vleck paramagnetism $N\alpha$. These are

calculated to be

$$g_a = (g_x + g_y)/2 = 2.27,$$

$$g_c = g_z = 2.14,$$

where x, y and z are referred to the local coordinate and

$$g_x = 2.05,$$

$$g_y = 2.49,$$

$$g_z = 2.14$$

and

$$N\alpha_a = (N\alpha_x + N\alpha_y)/2 = 1.066 \times 10^{-4} \text{ emu/mole},$$

$$N\alpha_c = N\alpha_z = 0.550 \times 10^{-4} \text{ emu/mole}.$$

In this calculation, the energy separation ΔE_1 between the states ψ_0 and ψ_{xy} , ΔE_2 between the states ψ_0 and ψ_{xz} and ΔE_3 between the states ψ_0 and ψ_{yz} are chosen to be

$$\Delta E_1 = 9100 \text{ cm}^{-1},$$

$$\Delta E_2 = 10400 \text{ cm}^{-1},$$

$$\Delta E_3 = 12000 \text{ cm}^{-1},$$

so as to get the best fit to the experimental susceptibilities.

A little later, Ikebe and Date¹⁰⁾ determined the g -values from their ESR experiments and got

$$g_a = 2.25$$

and

$$g_c = 2.17.$$

The agreement to our values is quite satisfactory.

Although these magnetic data were taken on the (d) type crystal,^{*)} the (a) type has almost the same properties. The susceptibility can also be measured by the NMR shift. The results will be given later.

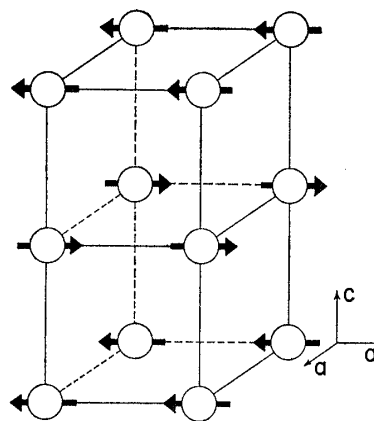
At low temperature, typically at 4.2°K, the susceptibility is field dependent. When the magnetic field is applied parallel to the c -plane, the susceptibility χ_a increases with increasing field strength. This nonlinear magnetization starts as low as 500 gauss. Such a nonlinear magnetization does not occur when the magnetic field is applied parallel to the c -axis. This point is important in connection with the anomalous dynamic behaviour at low temperatures, which will be described later.

*) The crystal on which our previous measurements^{6),7)} were made was thought to be the (a) type, but recently it has proved to be the (d) type.

e) *Neutron Diffraction*

Early neutron diffraction study⁵⁾ did not show any evidence for LRO even at liq. He temperature. But more careful study made by Hutchings et al.⁹⁾ at BNL on the single crystal we prepared showed existence of antiferromagnetic LRO on the two types (a) and (d) with the Néel point at 38°K and 22°K respectively. The spin structure for the (a) and (d) types are the same as shown in Fig. 4, although the spin direction in the *c*-plane is left unknown.

They looked for carefully, in the temperature range between T_N and T_{max}^x , the reciprocal lattice plane which is evidence for one-dimensionality, but they could not observe it. Instead, the observed spin moment at 4.2°K was nearly a half of the expected moment simply by assuming $g=2.2$, i.e. $1.1 \mu_B$ as listed in Table I. They argued that this abnormally small moment is attributed partly to the covalency effect and partly to the spin contraction effect originally proposed by Anderson.¹⁴⁾ More recently, Yamada¹⁵⁾ observed the moment of a similar but ferromagnetic compound K_2CuF_4 and found no serious deviation from $\sim 1 \mu_B$, so that the small moment in $KCuF_3$ must mostly be due to the Anderson contraction.



The spin structure of $KCuF_3$

Fig. 4. Antiferromagnetic structure below T_N which is determined by neutron diffraction. The spin direction in the *c*-plane is left unknown.

f) *Specific Heat*

As the ideal one-dimensional Heisenberg antiferromagnet does not have any LRO at finite temperatures, it has no sharp anomaly in the specific heat curve but has only a broad maximum at

$$T_{max}^c \approx 0.962 J/k,$$

as worked out by Bonner and Fisher.²⁾

In Fig. 5 is shown the measured specific heat curve which has no sharp peak at any temperature near 20°K and 38°K. The broad maximum is, if exist, difficult to distinguish from

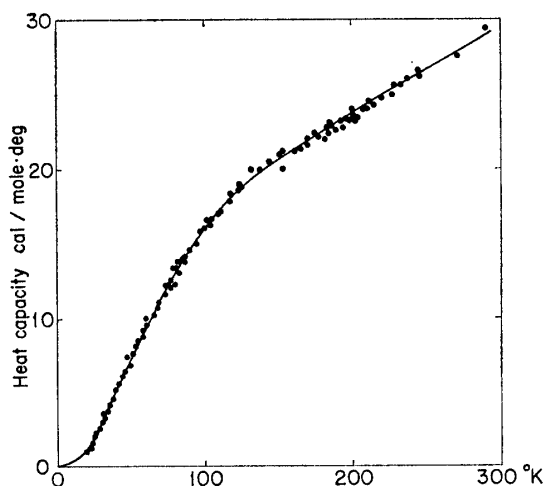


Fig. 5. Specific heat curve of $KCuF_3$ showing no sharp anomaly.

the lattice heat capacity.

g) *Electron Spin Resonance (ESR)*

Extensive ESR studies have been performed by Ikebe and Date.¹⁰⁾ The detail of their work should be referred to their paper, but we shall note their some important results. (i) As the spin system is very soft (interchain coupling being small) as indicated by the large spin contraction effect, its antiferromagnetic resonance line width is unusually broad and cannot be observed at frequencies lower than ~ 30 GHz (or $\sim 10^4$ Oe). (ii) They observed the paramagnetic resonance line width as a function of temperature and tried to interpret their results by Mori and Kawasaki's theory¹⁶⁾ by which the ESR line width in the critical region of MnF_2 had been explained satisfactorily. But among the substances ever examined, in KCuF_3 the deviation from the theory is most serious. This may be due to well developed SRO, while the effect is not properly taken into account in their theory. (iii) As stated in d), J can be estimated from T_{max}^x , but the interchain exchange interaction J' has been estimated only from Oguchi's theory³⁾ at present. From his theory, J' for the (d) type crystal is estimated to be $J'/k=1.0^\circ\text{K}$ by using $T_N=22^\circ\text{K}$. To know the exact value of J' is difficult at present, but in the (d) type crystal, as each chain has different principal axes of g -tensor, we may observe a single ESR line only when

$$J'/k \gtrsim |(g_x - g_y)| \mu_B H,$$

where H is the external field applied along the a -axis and $g_x=2.05$ and $g_y=2.49$ are as given before. They observed only one resonance line at $H \sim 10^4 \text{Oe}$. So that, they could give a lower limit of J' as

$$J'/k \gtrsim 0.3^\circ\text{K},$$

which does not contradict to the estimation given above. (iv) The anisotropy of this system is such that the c -plane is an easy plane. The origin is mainly caused by the anisotropic exchange interaction between the adjacent spins in a chain and this interaction is of the order of $(g-2)^2 J \sim 10^\circ\text{K}$ which is much larger than the dipole-dipole interaction. As the transition temperature is very low, the effect of anisotropy becomes very large near the transition point as described later.

h) *Nuclear Magnetic Resonance (NMR) on F^{19} and Cu^{63}*

(i) *Covalency Effect*

Our early work⁷⁾ was to study the covalency effect and to see whether our model on the special alignment of wave function in Fig. 2 is correct or not. As can be seen in Fig. 2, we have two kinds of fluorine nuclei F_c^{19} and F_a^{19} , where F_c^{19} is located at the centre of a couple of nearest Cu^{2+} ions in a

chain, and F_a^{19} is located at the asymmetric position of the displaced F^- ions. From the NMR shift and the susceptibility data, we determined at room temperature the fractions of the spin density on the 2s and 2p orbitals (σ -bonding) of F^- ions. They are

$$\begin{aligned} f_s &= 0.44\%, \\ f_\sigma &= 4.93\% \end{aligned}$$

for F_c^- ion and

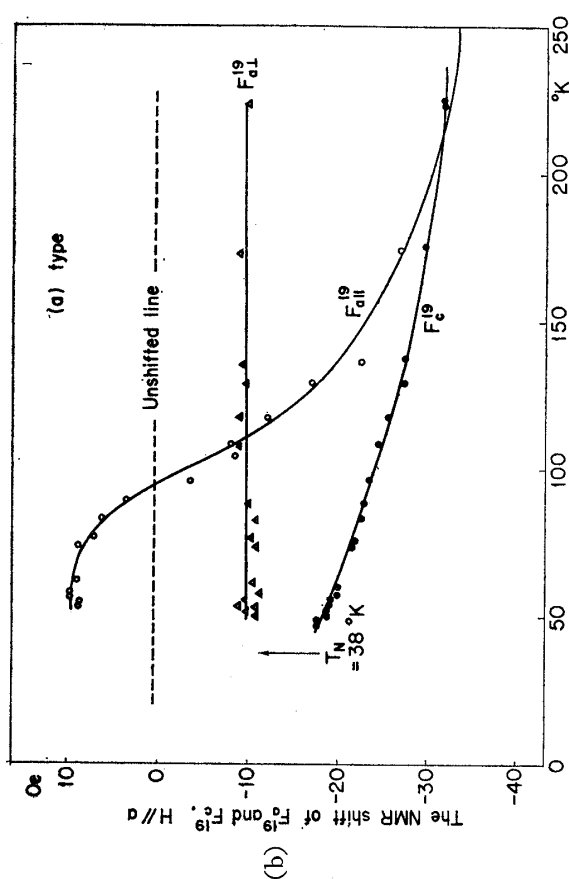
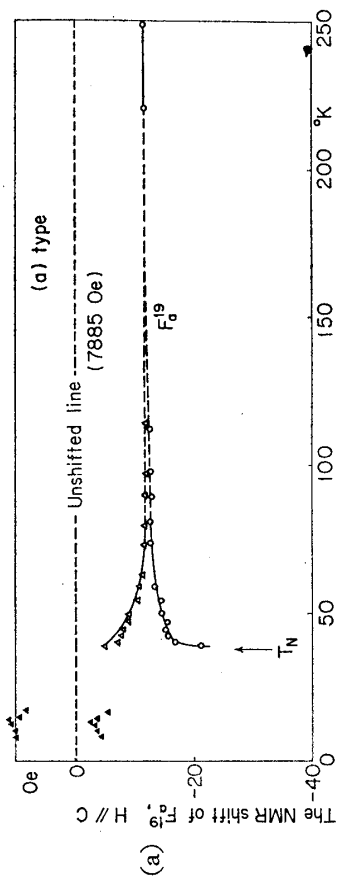
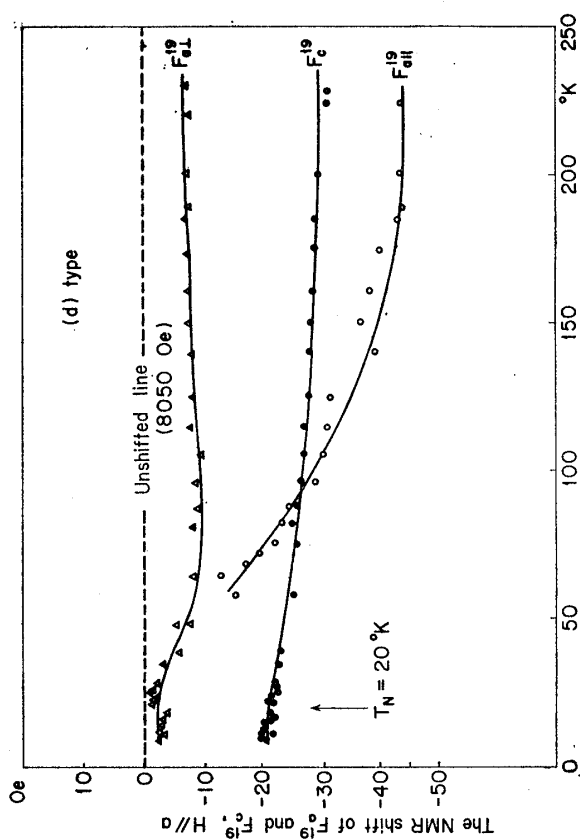
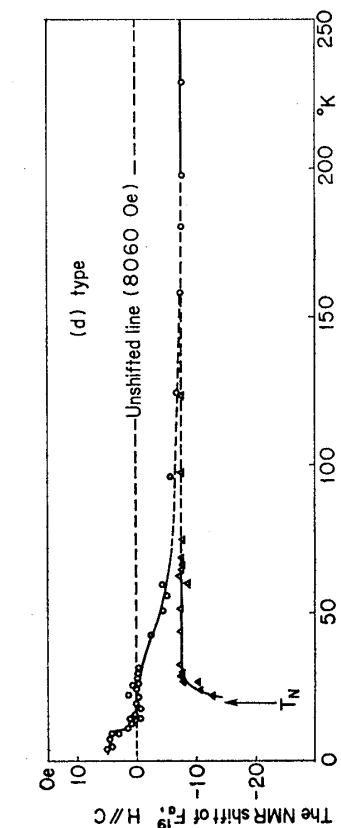
$$\begin{aligned} f_s &= 0.16\%, \\ f_\sigma &= 2.34\% \end{aligned}$$

for F_a^- ion. These data are on the (d) type crystal. The fact that f_σ for F_a^- is almost exactly a half of f_σ for F_c^- means that the overlap of wave functions on the F_a^- - Cu^{2+} is almost perfectly cut down on one-side. This supports the model we had in Fig. 2.

(ii) *NMR Shift of F_a^{19} and F_c^{19}*

In the previous work,⁷⁾ we saw that the hyperfine fields at F^- ions from neighbouring Cu^{2+} spins are nearly 5~10 times stronger than the dipole field, so that the observation of the temperature dependence of NMR shift in a paramagnetic state is a direct measure of the induced moment at the Cu^{2+} ion to which the nearest F^{19} is connected via hyperfine coupling. As stated before, we have two kinds of fluorine nuclei, but when the magnetic field is applied parallel to one of the a -axes, we come to have non-equivalent F_a^{19} nuclei again. We shall designate the fluorine nuclei which are on Cu-F-Cu line which is parallel to the external field as $F_{a\parallel}^{19}$ and those on the line normal to the field as $F_{a\perp}^{19}$. Of course we cannot distinguish $F_{a\parallel}^{19}$ and $F_{a\perp}^{19}$ when the field is parallel to the c -axis. All the data which were taken on the (a) type and on the (d) type are shown in Fig. 6.

Figure 6(a) shows the NMR shift of F_a^{19} when the magnetic field is applied parallel to the c -axis. It is a puzzling result that the line splits, because from the symmetry, it should not split. The splitting is symmetric in the (a) type, but asymmetric in the (d) type. As the intensities of both branches are the same, there must arise two non-equivalent F_a^{19} nuclei according presumably to the development of short range order, because the splitting starts almost just below T_{max}^X . In the (a) type crystal, when the short range order develops within a chain and if the preferred direction of spin is parallel to the a -axis, there arise just two non-equivalent spins according to the alternatively repeated non-equivalent site as shown in Fig. 1 or 2. If we dare to interpret this splitting, we have to assume spin direction dependent hyperfine coupling, which is difficult to suppose. This point should be clarified after further investigations.



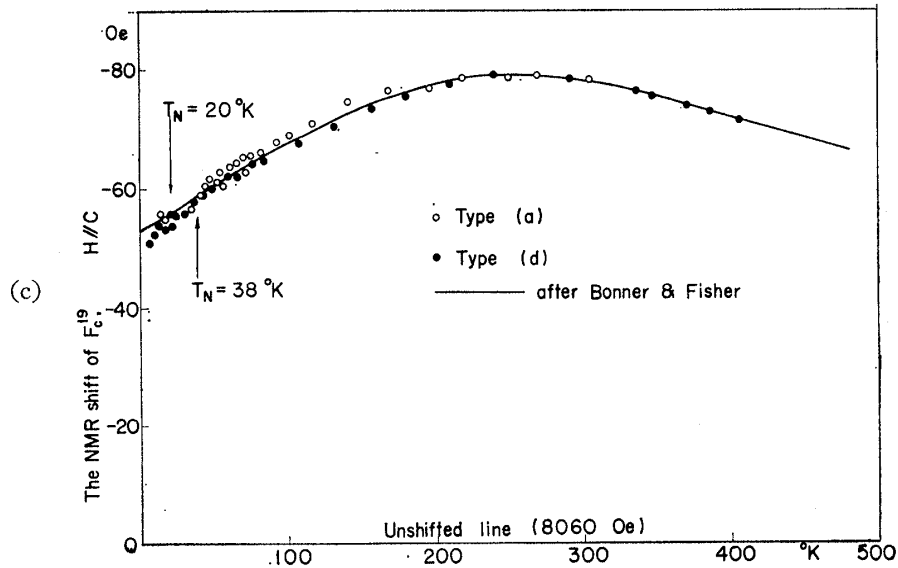


Fig. 6. The NMR shift of F_c^{19} and F_a^{19} with different directions of applied field.
 (a) The NMR shift of F_a^{19} when the field is applied parallel to the c -axis.
 (b) The shift of $F_{a\parallel}^{19}$, $F_{a\perp}^{19}$ and F_b^{19} when the field is parallel to the a -axis.
 (c) The NMR shift of F_c^{19} when the field is parallel to the c -axis. The curve is the calculated one by Bonner and Fisher.

Figure 6(b) shows the shift of the $F_{a\parallel}^{19}$ and $F_{a\perp}^{19}$ when the field is applied parallel to the a -axis. One of the interesting temperature variation is that for $F_{a\parallel}^{19}$ in the (a) type crystal, in which the sign of the shift is reversed (becomes positive) when the temperature approaches to T_N . This means that $g\mu_B\langle S \rangle_{\parallel}$ of a given Cu^{2+} spin that $F_{a\parallel}^{19}$ can see is at high temperature in the positive (parallel to the field) direction, but at low temperature $g\mu_B\langle S \rangle_{\parallel}$ becomes negative direction (antiparallel to the field). This change is explained in Fig. 7. As g_y is larger than g_x , when the SRO is well developed, a half of all atomic moments are induced parallel to the field direction but the rest will be induced antiparallel to the field direction. As the $F_{a\parallel}^{19}$ ion is hyperfine coupled only on the x -axis, $F_{a\parallel}^{19}$ sees only the Cu^{2+} magnetic moment which is in the antiparallel sense. At high enough temperature, where no SRO exists, all moments are induced to the same direction resulting in the negative shift. We think, thus, this is a typical evidence for the existence of well developed SRO in a chain in the paramagnetic phase.

Another interesting point is the tempera-

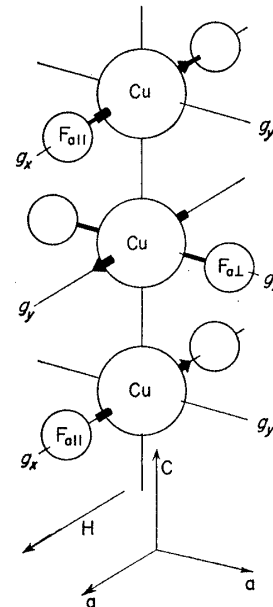


Fig. 7. A model for explanation of the positive shift of $F_{a\parallel}^{19}$.

ture dependence of the shift of F_c^{19} when the magnetic field is applied parallel to the c -axis. As F_c^{19} is at the centre of symmetry, it cannot directly see the SRO of the spin system as F_a^{19} can do. Instead, the shift is the direct measure of the static susceptibility of the chain. The shift of F_c^{19} depends little on the field direction, so we shall present the data when the field is applied parallel to the c -axis. The temperature dependences of the shift for the (a) and (d) types are almost exactly the same as shown in Fig. 6(c).*) A surprisingly excellent agreement to the curve calculated by Bonner and Fisher was obtained throughout the temperatures measured. No anomalous increasing in shift ($\propto \chi$) was observed at low temperature. The discrepancy between the NMR shift and previously mentioned susceptibility is difficult to understand. One may suggest that if some paramagnetic impurities are contained, as the NMR shift does not see the impurity, the difference may arise. If it is so, however, there must be considerable fluctuations of the anomalous increase due to the fluctuation of impurity contents. But the anomalous increase is always the same, so that this possibility must be ruled out.

(iii) NMR Line Width

NMR line width is a measure of dynamics of spin system. Figure 8(a) shows the measured results for the (d) type. Those for the (a) type were omitted because of the similar behaviour to the (d) type. As can be seen, the width ΔH for F_c^{19} decreases slightly when the temperature is reduced beyond T_{\max}^x until T_N is reached, where the width goes up suddenly. There is no remarkable change in width when the field direction is changed and the width is very narrow, being ~ 2.5 Gauss, and is the order of nuclear dipole-dipole interaction. So the effect of electron spins on the resonance width is almost completely eliminated. Generally, the resonance width depends partly on the amplitude of the fluctuating field (hyperfine field) and partly on the characteristic frequency of fluctuation. As F_c^{19} is just at the centre of the symmetry of two adjacent Cu^{2+} spins, the amplitude will be reduced to zero according to the development of short range order. The frequency of the mode which produces the fluctuation amplitude is so high that no influence on the width can be expected. This is the reason why ΔH of F_c^{19} 's becomes narrow at low temperatures.

The width for F_a^{19} is quite different. At $T > T_{\max}^x$, the width for F_a^{19} depends weakly on the field direction. As the temperature is reduced, the width for F_a^{19} , when the field is applied parallel to the a -axis, increases remarkably toward T_N , but, when the field is applied parallel to the c -axis, the width is independent of temperature until T_N is reached. So, near T_N , the width is extremely anisotropic as shown in Fig. 8(b). As the hyperfine field

*) When the field is applied normal to the c -axis, the curve becomes just a little bit more concaved.

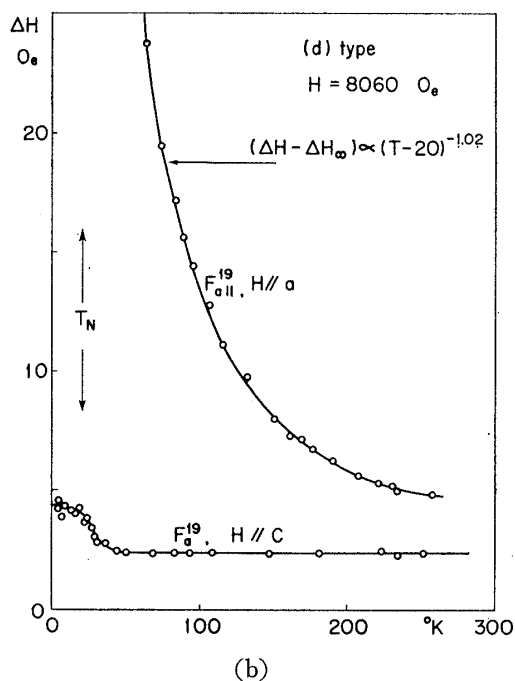
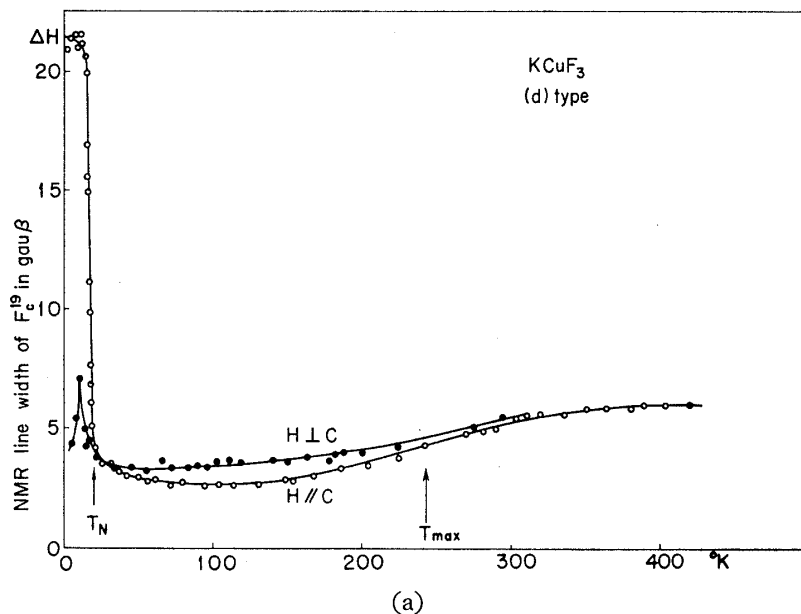


Fig. 8. The NMR line widths of (a) for F_c^{19} and (b) for $F_{a||}^{19}$ with the applied fields parallel to the c - or a - axes.

for F_a^{19} is always proportional to the spin of Cu^{2+} ion to which it contacts, the fluctuating amplitude is always the same throughout the temperatures. The temperature dependence of the width is consequently attributed to the slowing down of the system. The frequency of the fluctuating field component parallel to the c -axis will be high if the out of plane anisotropy is large. If it is so, the width will not be broadened until very nearly T_N is reached. As the anisotropy energy due to anisotropic exchange is such that to put the spins in the c -plane, the slowing down occurs only in the c -plane causing large and slowly fluctuating hyperfine field in the field direction when

the field is parallel to the a -axis. It is not so clear whether this slowing down is characteristic for an individual motion of a chain or a collective motion of all the spin system. We think, however, as the line width for $F_a^{19}(\Delta H - \Delta H_\infty)$ is well expressed by the simple form

$$(\Delta H - \Delta H_\infty) \propto (T - T_N)^{-1.02},$$

where $\Delta H_\infty = 2.0$ Gauss is the width for high temperature limit and $T_N = 20^\circ\text{K}$, we think that the slowing down must be of a collective motion of the chains. The motive force for this instability may possibly be the inter-chain dipole-dipole interaction which is only of the order of 2°K , but is sufficient magnitude to produce T_N at 20°K . This instability is connected smoothly to the low temperature spin pattern. All above studies were made by the steady method with the applied field of the order of 13 KOe.

(iv) *NMR below T_N*

NMR studies below T_N were made by the use of spin echo technique. The nature of loosely coupled chains remains even at temperatures as low as 1.5°K . For instance, we could observe zero field F_a^{19} NMR signal at 49 MHz. But the echo signals were observed in a wide range of ± 5 MHz and T_2 was of the order of only $1 \sim 10 \mu\text{sec}$. In Fig. 9, the peak height of the echo signal observed at a constant frequency and as a function of field strength is shown. The signal (band) at lower field is for F_a^{19} and the sharp peak at higher field is for F_c^{19} and this measurement was done on the (d) type single crystal. We can see qualitatively how the softness of the spin system is detected by F_a^{19} whereas not by F_c^{19} . The frequency of 49 MHz is, if we assume that the spin is in the direction of the a -axis, just consistent to the $\sim 50\%$ spin contraction. Another echo signal was also observed at 89.6 MHz at 1.5°K . This is likely to one of the quadrupole splitted Cu^{2+} signal, but the assignment is not so certain as yet.

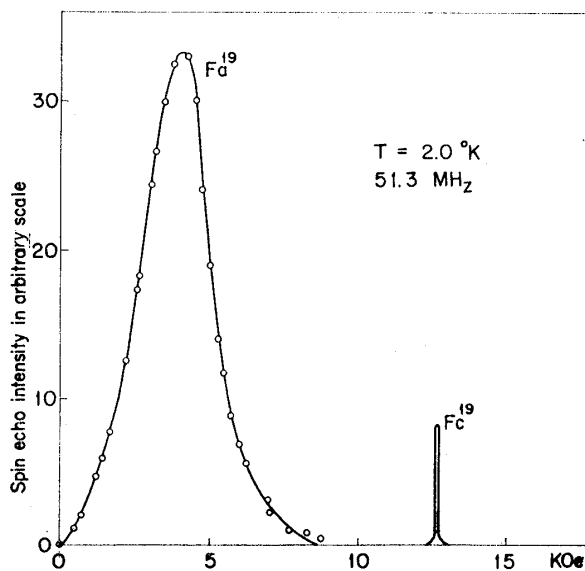


Fig. 9. The spin echo signal observed on F_a^{19} and F_c^{19} at 2°K (in the antiferromagnetic state) with fixing the frequency at 51.3 MHz and varying the field ($H//a$) strength.

§3. Conclusion

We observed great evidence for one-dimensional nature of the spin system in $KCuF_3$, i.e. short range order develops remarkably below T_{max} . Although the anisotropy energy ($\sim 10^\circ K$) is very low compared to the exchange interaction $J = -190^\circ K$, very low T_N brings the paramagnetic dynamical susceptibility which determines the NMR line width extremely anisotropic at the vicinity of the transition point. It should be noted that the asymptotic point of the softening is not $0^\circ K$ but T_N . This is consistent to the theoretical prediction that in quasi-one-dimensional system, LRO is caused by the inter-chain interaction, which brings the system to three dimensional problem. In $KCuF_3$, this three-dimensional softening seems to start as soon as the short range order within a chain develops. This is in strong contrast to the two-dimensional case (K_2NiF_4), where the susceptibility is again highly anisotropic near T_N ,¹⁷⁾ but divergence nature of the susceptibility seems to be the two-dimensional own.

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