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Electron spin resonance study of the two-dimensional randomly mixed system $K_2Cu_{1-x}Co_xF_4$

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Abstract. We report the experimental results of electron spin resonance (ESR) studies made on $K_2Cu_{1-x}Co_xF_4$, a randomly mixed system of the two-dimensional Heisenberg ferromagnet K_2CuF_4 and the two-dimensional Ising antiferromagnet K_2CoF_4 which have orthogonal anisotropies to each other. From both the temperature and the angular dependence of the resonance field at low temperatures, three different kinds of ferromagnetic phase are confirmed, i.e. XY, oblique and Ising types, which coincides well with the results obtained by neutron scattering and susceptibility measurements. Critical behaviours of both linewidth and resonance field are also reported and discussed. In addition, it is reported that the temperature dependence of the linewidth and the resonance field of the sample on the Co concentration of the spin-glass region show a common behaviour generally observed in many other spin glasses.

1. Introduction

The mixed system $K_2Cu_{1-x}Co_xF_4$ is one of the candidates for the investigation of the competing effect of ferromagnetic and antiferromagnetic exchange interactions. In this mixture, there are competing anisotropies as well. The end members are well known representative two-dimensional magnets; one of the end members, K_2CoF_4, orders antiferromagnetically below $T_N = 109$ K with the easy axis parallel to the crystallographic c axis owing to the strong Ising-type anisotropy, and the other one, K_2CuF_4, is a Heisenberg ferromagnet having $T_c = 6.25$ K with the preferred spin direction perpendicular to the c axis caused by a weak XY-like anisotropy.

The magnetic phase diagram of the system was determined by the measurements of neutron diffraction [1] and susceptibility [2]. That is, antiferromagnetic and spin-glass phases were found on the Co-rich side ($1 \geqslant x > 0.5$) and the intermediate range of x ($0.5 > x > 0.16$), respectively. The Cu-rich side, on the other hand, ($0.16 > x \geqslant 0$) was found to have three types of ferromagnetic order which were classified depending on the direction of the magnetization; i.e. an XY, an oblique and an Ising type. Both experiments [1, 2] confirmed the successive transitions from the paramagnetic phase to the oblique one through the Ising one with decreasing temperature, which proved the existence of the critical line between the Ising and the oblique phases. The boundary between the oblique and the XY phases was, however, left unconfirmed. In other words, two possible transitions with decreasing temperature, i.e. from the XY to the oblique or from the oblique to the XY, were not confirmed.

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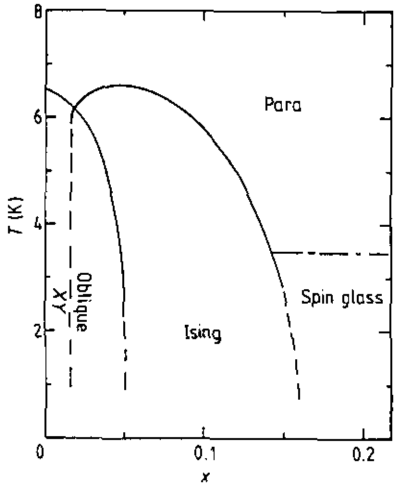

Figure 1. Schematic drawing of the transition temperature versus concentration phase diagram of $K_2Cu_{1-x}Co_xF_4$ for the Cu-rich side reproduced from [1] and [2].

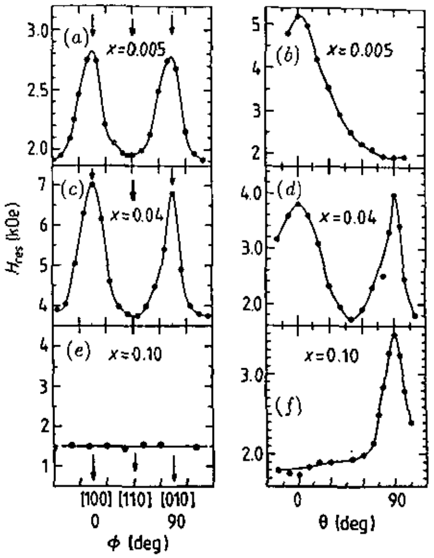

Figure 2. Angular (ϕ and θ) dependence of the resonance field H_{res}. (*a*)–(*d*) are obtained at 1.3 K, and (*e*) and (*f*) at 4.2 K. ϕ is an angle of the direction of the external field with respect to the [100] direction in the *c* plane, while θ is that with respect to the *c* axis ([001]) in the plane which includes the [110] and the [001] axes. Characteristic directions in the *c* plane, [100], [110] and [010] are indicated by arrows. The solid lines are guides to the eye.

These three ferromagnetic ordered states are brought about by the competition between the XY- and Ising-type anisotropies. The ferromagnetic phase diagram given in [1,2] is reproduced schematically in figure 1.

If a ferromagnetic sample is shaped into a sphere, the easy direction of the magnetization is easily determined by measurement of the ferromagnetic resonance field because the complicated effect from the demagnetization field can be avoided. Thus the ferromagnetic resonance (FMR) can be another useful method to investigate the ferromagnetic ordered states in such a mixed magnet having competing anisotropies.

We previously studied [3] the FMR of the present mixed system with very low Co concentration and clarified the spin structure representing the XY phase more precisely than neutron diffraction or susceptibility measurements. That is, the FMR revealed even the direction of the magnetization in the *c* plane, whereas the neutron diffraction or the susceptibility measurements only indicated the spin ordering perpendicular to the *c* axis.

The XY-like anisotropy field of K_2CuF_4 evaluated as 2.4 kOe [4] arises from both the dipolar interaction and the anisotropic exchange interaction [5], while the in-plane one is about 5 Oe [6], so small as to be negligible. The random displacement of Cu by Co brings about not only the Ising-type anisotropy but also an additional XY-type one. The intuitive picture we form is as follows. Each Cu site of K_2CuF_4 is surrounded by the distorted F octahedron, the distortion of which is due to the cooperative Jahn–Teller effect. The F octahedron is rhombic when projected onto the *c* plane and has the longest principal axis along the [100] and its equivalents where [ ] indicates the crystal axis in the unit cell of a K_2NiF_4-type structure. The Co impurities, as long as they are isolated from one another, should be in the rhombic F octahedra and their spins are pinned within the c plane by the antiferromagnetic exchange interaction J(Co–Cu) between the Co^{2+} and the Cu^{2+}. The direction of the Co spins should be along one of the principal axes of the F rhombus because the Co spins are strongly affected by the rhombic crystal field. Then there are two choices for their directions in the c plane, i.e. along the [100] or the [010] axis. Since the inherent in-plane anisotropy of the Cu spins is negligibly small, the nearest neighbouring Cu spins will be pulled nearly within the same direction of Co spins because of the relatively strong antiferromagnetic J(Co–Cu). Thus around each Co site, there will be formed two kinds of clusters of Cu with their spins polarized both along the [100] and the [010] directions. At larger distances from the Co sites where the majority of Cu sites are located, the Cu spins are obliged to align themselves along the [110] direction because this direction is a compromise between the two conflicting directions, [100] and [010]. This is the origin of the additional in-plane anisotropy observed for very low x as previously reported [3].

When x is increased further, the probability of adjacent Co sites increases. Then the strong antiferromagnetic J(Co–Co) will begin to govern the spin structure of the system. That is, the Co spins, when adjacent to one another, line up along the c axis and they will force the neighbouring Cu spins to align themselves with the c axis. In such a situation, the spins of the isolated Co also align themselves with the c axis but in the opposite direction to the neighbouring Cu spins because $J(\text{Co–Cu}) < 0$. Once the spins of Cu and the isolated Co are forced to align along the c axis by J(Co–Co), the fourfold XY-like anisotropy in the c plane due to the isolated Co sites will disappear. Although the system will be of highly disordered character on a microscopic level, the experimental results presented below show that it is possible to consider average quantities such as magnetization and anisotropy to describe the macroscopic properties.

Besides the three ferromagnetic ordered states, the paramagnetic critical region of the present mixed system is worth investigation. The EPR signal of K_2CuF_4 is recognized as a representative example which gives a concrete form to the effect of the long-wavelength fluctuation of spins described by the spin diffusion process [7]. In particular, its critical broadening of the linewidth reflects the increasing importance of the slowing down of the long-wavelength mode. That is, for instance the $(3\cos^2\theta - 1)^2$-like angular behaviour of the linewidth (θ is the angle of the external field with respect to the c axis) observed at high temperatures is enhanced near T_c. As far as we know, such phenomena as a two-dimensional Heisenberg ferromagnet were observed only in this compound as reviewed in [8, 9]. The problem to be clarified is, therefore, the influence of the Co spins on such a long-wavelength fluctuation.

In the present paper, we shall present the results of FMR measurements made on several samples with x covering all three ferromagnetic phases and discuss the results based on the change of average spin anisotropy. Furthermore, the spin dynamics near T_c apparent from the temperature or angular dependence of the EPR linewidth and the resonance field will be compared among three different ferromagnetic phases. In addition, the ESR lines observed for the sample of spin-glass phase will be mentioned briefly.

2. Experimental procedure and results

Single crystals of the present mixed system were grown by the Bridgman method as explained in [3]. We employed an atomic absorption method to analyse the concentration of Cu and Co. Because of the growing process, the concentration is uniform in the c plane, but a small concentration gradient along the c axis is unavoidable. We confirmed the inhomogeneity Δx along the c axis to be $\Delta x/\mathrm{mm} < 10^{-3}$, which was found to pose no problem to the present study. To get precise FMR signals, the samples should be formed into spheres. We stress that the single crystals of K_2CuF_4 and $K_2Cu_{1-x}Co_xF_4$ which were grown by ourselves can be made spherical with sufficiently small size. Since most single crystals of two-dimensional systems are easily cleavable along the layers and are almost impossible to shape spherically, the present case is rare. Each sample used was polished to form an almost complete sphere of diameter about 1 mm or less with approximate surface unevenness of 20 μm, this being sufficient to prevent the magnetostatic mode. By a combination of rotating the sample in an X-band cavity around the horizontal axis and rotating the external field $\boldsymbol{H}$ in the horizontal plane, we could obtain the condition of $\boldsymbol{H}$ being in the c plane and also in a plane including the [110] and the [001] crystal axes (we call it the a–c plane). From now on θ and ϕ denotes the angles of $\boldsymbol{H}$ with respect to the c axis in the a–c plane and to the [100] axis in the c plane, respectively.

As pointed out in [7], the apparent transition to the ferromagnetic ordered state in the measurement of ESR made on K_2CuF_4 seems to occur at a temperature (denoted as $\tilde{T}_c$ in [7]) which is quite a bit higher than the actual T_c. This is due to the strong external field necessary for resonance, i.e. the external field couples directly with the magnetization and wipes out the true ferromagnetic transition point which can be determined under zero external field. In the K_2CuF_4 case, $\tilde{T}_c$ was defined as a temperature at which the linewidth reaches its peak value. Below the temperature thus defined, the resonance field begins to deviate remarkably from the value obtained at high temperatures. In the present report we also use $\tilde{T}_c$ to indicate the transition from paramagnetic to ferromagnetic ordered state under the external field necessary for resonance. As will be shown below, $\tilde{T}_c$ for the present mixed system is also quite a bit higher than T_c, i.e. $\tilde{T}_c \simeq$ 11–12 K and is almost independent of x over the three ferromagnetic ranges.

2.1. XY phase

The detailed experimental results and analysis for the XY phase were reported in [3]. We thus merely show here the characteristic result representing the XY phase which is necessary to compare with those observed in the samples of oblique and Ising phases. In figures 2(*a*), (*b*), we show the angular dependence of the resonance field H_{res} of the sample with $x = 0.005$ measured at 1.3 K, the lowest temperature available in the present experiments. Since T_c for $x = 0.005$ is estimated to be approximately 6 K from figure 1, 1.3 K is low enough for the investigation of the ordered state. Figure 2(*a*) is the change of H_{res} when the direction of the external field $\boldsymbol{H}$ is varied in the c plane. As can be seen in this figure, the minimum of H_{res} appears when $\boldsymbol{H}\|[110]$ and its equivalents. After obtaining the result shown in figure 2(*a*), we measured H_{res} in the a–c plane, the result of which is given in figure 2(*b*). These two figures clearly indicate that the easy direction of the magnetization is along the [110] directions.

The dependence of H_{res} and the derivative peak-to-peak linewidth ΔH_{pp} on temperature T is shown in figures 3(*a*), (*b*), respectively. We find that H_{res} deviates from the value at high temperatures after turning into the ordered state, i.e. below $\tilde{T}_c$. On the other hand, ΔH_{pp} keeps the $(3\cos^2\theta - 1)^2$-like angular dependence at high temperatures and shows anomalous critical broadening when $\theta = 0°$, i.e. the $(3\cos^2\theta - 1)^2$-like behaviour is enhanced as shown in the inset of figure 3(*b*). Both

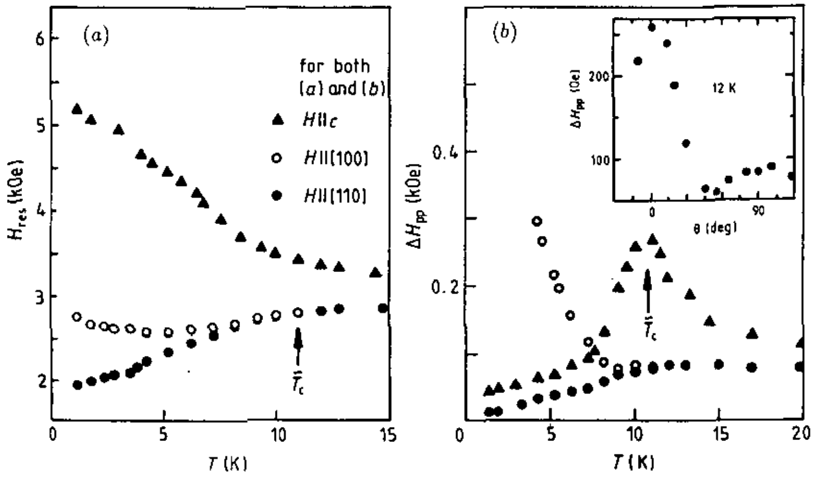

Figure 3. $x = 0.005$: (*a*) temperature dependence of the resonance field H_{res}; (*b*) temperature dependence of the linewidth ΔH_{pp} for the three directions of the external field. The inset in (*b*) is the θ-dependence of ΔH_{pp} obtained at a temperature near $\bar{T}_c$.

the angular and temperature dependences of ΔH_{pp} above $\bar{T}_c$ indicate that the spin relaxation is still governed by the diffusional process. The g-values necessary to analyse the FMR data were determined from H_{res} at high temperatures, i.e. $g_a = 2.29$ for $\boldsymbol{H} \perp c$ and $g_c = 2.08$ for $\boldsymbol{H} \| c$, which are not different from those of K_2CuF_4.

2.2. *Oblique phase*

The angular dependences (on ϕ and θ) of H_{res} measured at 1.3 K for the sample with $x = 0.04$ is shown in figures 2(*c*), (*d*), respectively. The H_{res} observed with $\boldsymbol{H}$ rotating in the c plane has a fourfold symmetric change with ϕ as shown in figure 2(*c*) and has its minimum value when $\phi = 45°$, i.e. $\boldsymbol{H} \| [110]$. On the other hand, H_{res} measured in the a–c plane shows a minimum value at an angle intermediate between $\theta = 0°$ and $90°$ as shown in figure 2(*d*), which is quite different from that observed for $x = 0.005$. These angular behaviours of H_{res} indicate that the magnetization lies in the a–c plane but its direction is oblique to the c axis.

The H_{res} versus T is shown in figure 4(*a*). As can be seen in this figure, H_{res} changes remarkably below $\bar{T}_c$. We find that H_{res} for $\theta = 0°$ is the lowest compared with that of other directions of $\boldsymbol{H}$ over a narrow temperature range indicated by $\longleftrightarrow$, i.e. approximately 6–8 K, which indicates that the magnetization is aligned along the c axis and thus the system is in the Ising phase over this range of temperature. With further decreasing temperature, H_{res} is smallest for $\theta \simeq 45°$, indicating that the system turns into the oblique phase. When T is decreased over the range of the oblique phase, the oblique angle changes with T. The most pronounced one was found for $x = 0.03$, which we show in figure 5. That is, the $H_{res}(\theta)$ observed at 1.3 K has its minimum around $\theta = 63°$, whereas its minimum at 4.2 K appears around $\theta = 45°$.

The $\Delta H_{pp}(T)$ does not show anomalous broadening around $\bar{T}_c$ as shown in figure 4(*b*). The $\Delta H_{pp}(T)$ for all directions of $\boldsymbol{H}$ increases with decreasing temperature below $\bar{T}_c$. This broadening, as will be discussed later, arises from the fluctuation of the anisotropy field. At high temperatures, ΔH_{pp} is broader than that observed for $x = 0.005$, but still has the $(3\cos^2\theta - 1)^2$-like angular dependence. When T approaches

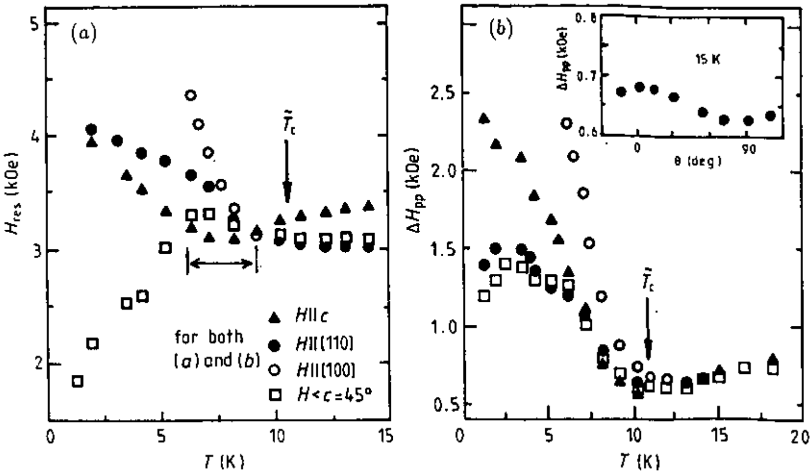

Figure 4. $x = 0.04$: (*a*) temperature dependence of H_{res}; (*b*) temperature dependence of ΔH_{pp} for the three directions of the external field. In the inset in (*b*), the θ-dependence of ΔH_{pp} obtained at a temperature near $\tilde{T}_c$ is shown.

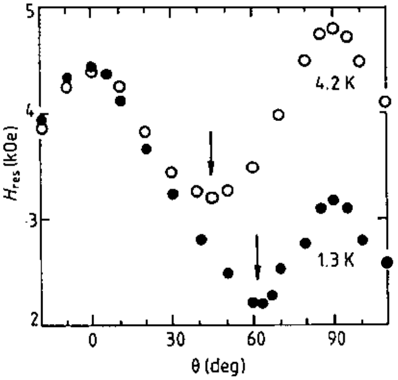

Figure 5. $x = 0.03$: angular dependence of H_{res} obtained at two temperatures. The minimum position indicated by an arrow changes with temperature.

$\tilde{T}_c$, however, a $(1 + \cos^2\theta)$-like angular behaviour appears as shown in the inset of figure 4(*b*). The g-values were found to be the same as those of K_2CuF_4.

2.3. *Ising phase*

The sample with $x = 0.10$ was chosen as one representing the Ising phase. According to figure 1, the sample should have $T_c \simeq 6$ K. At 4.2 K, a single absorption line appeared when $\theta = 0°$, whereas double resonance lines were observed when $\theta = 90°$. The value of H_{res} of the higher field peak changes continuously when the temperature goes down from the paramagnetic phase to the ordered phase. This peak, however, loses its intensity with decreasing temperature and then it becomes impossible to identify its H_{res} at 1.3 K. In contrast, the lower-field resonance line increases in intensity with decreasing T.

Such double FMR lines are common in ferromagnets or ferrimagnets having relatively strong anisotropy. According to several old and established experiments [10] and theories [11], a single FMR line can be expected only when the anisotropy field is fairly small compared with the effective field necessary for resonance. For a sample exhibiting an anisotropy field comparable to the effective field for resonance, an additional resonance line appears at the lower-external-field side of the ordinary line. In other words, an absorption occurs before the magnetization is lined up with $\boldsymbol{H}$ besides the one which corresponds to the aligned state as the usual FMR single signal. Furthermore, the theories show that such double resonances appear only when $\boldsymbol{H}$ is along or near the hard axis and successfully explain their origin in terms of magnetic domains. The present double resonances observed for $x = 0.1$ are thus surely due to the magnetic domains. The appearance of them itself also reflects a relatively strong Ising-type anisotropy. Since a single line appeared when $\theta = 0°$, this direction should be the easy axis, which agrees with the experiment at 4.2 K as developed below. We plotted the ϕ and θ dependence of H_{res} obtained at 4.2 K in figures 2(*e*), (*f*), respectively. As can be seen in these figures, no change of H_{res} with respect to ϕ can be seen, while H_{res} reaches its minimum value at $\theta = 0°$ when $\boldsymbol{H}$ is rotated in the *a*–*c* plane. These results clearly indicate that the magnetization is along the *c* axis. We show $H_{\text{res}}(T)$ in figure 6(*a*) and $\Delta H_{\text{pp}}(T)$ in figure 6(*b*). Below $\tilde{T}_c \simeq 11$ K, H_{res} for $\theta = 0°$ goes down, while that for $\theta = 90°$ goes up with decreasing temperature, which indicates the growing of the anisotropies below $\tilde{T}_c$.

At temperatures far above $\tilde{T}_c$, ΔH_{pp} shows a $(1 + \cos^2\theta)$-like change with θ instead of a $(3\cos^2\theta - 1)^2$-like one. Even near $\tilde{T}_c$, this θ dependence is maintained as shown in the inset to figure 6(*b*). No anomalous broadening of ΔH_{pp} is seen around $\tilde{T}_c$, which should be compared with the case of the XY phase.

2.4. *Successive transitions from Ising to XY through the oblique phase*

Since the competition of anisotropies produces two critical boundaries of ordering of parallel and perpendicular components of spins which intersect at the tetracritical point, successive transitions can be expected for certain values of x. Actually, the transition from the Ising to the oblique phase was confirmed for the samples with $x = 0.03$ and 0.025 by the measurements of both neutron diffraction [1] and susceptibility [2]. As shown in figure 4(*a*) and explained in 2.2, we also confirmed the transition from Ising to oblique phase, measuring $H_{\text{res}}(T)$ for $x = 0.04$. Here we show evidence of the transitions from the Ising to the XY through the oblique phase which appeared for $x = 0.02$. The dependence of H_{res} on the change of the direction of H obtained at three different temperatures is shown in figure 7. The $H_{\text{res}}(\theta)$ observed at 6.5 K has a minimum when $\theta = 0°$, while those obtained at 4.2 K and 1.3 K have minima at $\theta \simeq 45°$ and 90°, respectively. The dependence of H_{res} on T is shown in figure 8. These

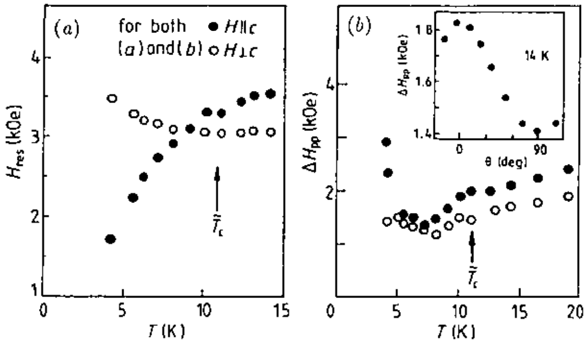

Figure 6. $x = 0.10$: (*a*) temperature dependence of H_{res}; (*b*) temperature dependence of ΔH_{pp}. The inset in (*b*) indicates the θ-dependence of ΔH_{pp} obtained at a temperature near $\tilde{T}_c$.

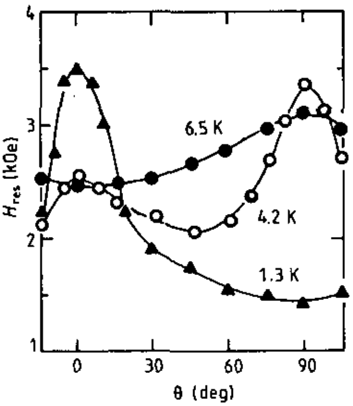

Figure 7. $x = 0.02$: Change of θ-dependence with temperature, indicating the successive transition from the Ising phase (6.5 K) to the XY phase (4.2 K) through the oblique one (1.3 K) with decreasing temperature. The solid lines are guides to the eye.

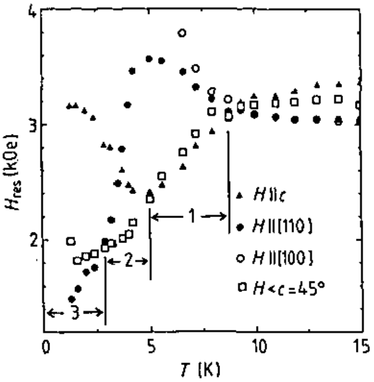

Figure 8. $x = 0.02$: temperature dependence of H_{res}. H_{res} for $\boldsymbol{H} \| [001]$ (c axis) is the minimum over the temperature range shown by ⟵ 1 ⟶, indicating that the system is in the Ising phase over this temperature range. Similarly, ⟵ 2 ⟶ is the oblique phase and ⟵ 3 ⟶ is the XY phase.

three $H_{res}(\theta)$ and $H_{res}(T)$ clearly indicate the successive transitions from Ising to XY through the oblique phase.

2.5. *Spin-glass phase*

When x becomes larger than approximately 0.16, a spin-glass phase is expected as the preceding studies [2] revealed. That is, the zero-field-cooled DC susceptibility for $0.16 < x < 0.5$ measured in a fixed field of 10 Oe applied parallel to the c direction shows a peak around 3.5 K. The peak position is almost independent of x [2].

For the present study, we chose a sample with $x = 0.2$. Since ΔH_{pp} observed at high temperatures was extremly broad, we employed a K-band spectrometer instead of the X-band one and operated it at 24.5 GHz. When $\theta = 0°$, we could not observe any absorption line above 1.3 K. With a slight change of θ from 0°, a broad line appeared. The $H_{res}(T)$ and $\Delta H_{pp}(T)$ are shown in figure 9 together with the individual θ dependence. It seems that $\Delta H_{pp}(T)$ (○) has a peak around 3–4 K, a temperature at which the zero-field-cooled susceptibility shows a peak, while $H_{res}(T)$ (●) goes down monotonically with decreasing temperature, showing no anomaly around 3–4 K.

3. Discussion

We first discuss the FMR results developed above based on the conventional phenomenological FMR theory. Since the analysis for the XY phase is given in [3], we here pay attention to the oblique and Ising phases. Solving a torque equation for a ferromagnet having tetragonal anisotropy energy, we obtain a frequency-field relation for $\boldsymbol{H}$ rotated in the a–c plane as

$$\omega/\gamma = \{[H_{res} - H_{A1}\cos 2\theta + \tfrac{1}{2}H_{A3}(3\sin^2\theta\cos^2\theta - \sin^4\theta)][H_{res} + H_{A3}\sin^2\theta]\}^{1/2} \qquad (1)$$

where H_{A1} and H_{A3} represent the out-of-plane and the in-plane anisotropy fields, respectively. On the other hand, the relation for the XY structure is given by

$$\omega/\gamma = \left\{(H_{res} - H_{A3}\cos 4\phi)[H_{res} + H_{A1} - \tfrac{1}{4}H_{A3}(\cos 4\phi + 3)]\right\}^{1/2} \qquad (2)$$

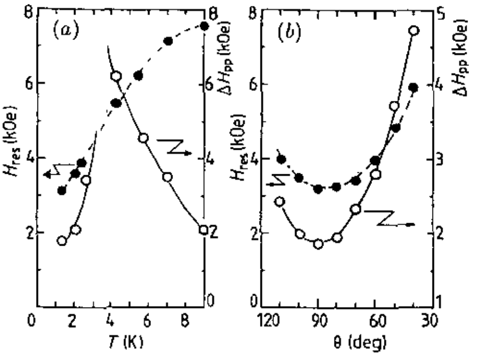

Figure 9. $x = 0.2$: (a) temperature dependence of H_{res} and ΔH_{pp} for the sample which turns into the spin-glass phase; (b) change of H_{res} and ΔH_{pp} with θ obtained at 1.3 K. The solid lines are guides to the eye.

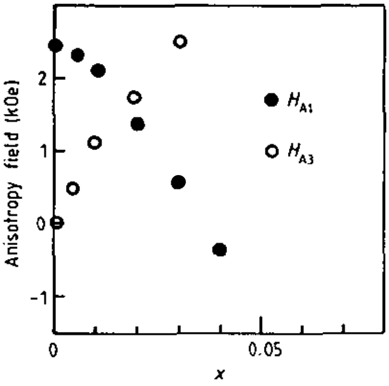

Figure 10. Concentration (x) dependence of the out-of-plane anisotropy field H_{A1} and the in-plane one H_{A3}.

under the condition of $\boldsymbol{H}$ being in the c plane. The operating microwave frequency throughout the FMR experiments was 9.25 GHz. The g-factors of the samples used were determined from the resonance field of the EPR lines observed at high temperatures. Introducing the experimental values of $H_{res}(T \rightarrow 0\,\mathrm{K})$ and the g-values calculated as $g = (g_a \sin^2\theta + g_c \cos^2\theta)^{1/2}$ into the equations (1) and (2), we obtained the anisotropy fields H_{A1} and H_{A3}, the variation of which with x is shown in figure 10, namely H_{A1} decreases from the inherent value of K_2CuF_4 with increasing x and become negative at a certain value of x, while H_{A3} increases with x. A simulation based on both equation (1) and the experimental data revealed that the oblique phase appears when $x > 0.013$, the value at which H_{A1} intersects H_{A3} and persists up to $x = 0.05$. In the oblique phase, the oblique angle of the magnetization with the c axis depends on the value of H_{A1} and H_{A3}. Thus, the angle changes with x and also with T because these two anisotropy fields change with T, which was confirmed by the experiment developed above.

We next discuss the temperature behaviour of the linewidth over the critical region. As shown in figure 3(b), the anomalous broadening of $\Delta H_{pp}(\theta = 0°)$ can be seen in the samples which turn into the XY structure, but ΔH_{pp} does not show such a critical broadening in the samples, the ordering state of which is the oblique or the Ising structure. According to the EPR linewidth theories [9] for a pure ferromagnet, an Ising ferromagnet regardless of whether it is two- or three-dimensional, should show a critical broadening of the EPR line. Our present data for the oblique or the Ising sample, however, do not show a critical broadening. We interpret the behaviour of ΔH_{pp} of these samples as follows. As explained above, the Co impurities become isolated from one another in the system of the XY phase. Then the correlation among the Cu spins is not interrupted severely by Co and the long-wavelength fluctuation can propagate. That is why the $(3\cos^2\theta - 1)^2$-like behaviour of ΔH_{pp} is maintained. When x is increased further, the Co–Co bondings begin to play a role in producing the oblique or Ising phase. Then the correlation of Cu spins is highly frustrated and the propagation of the long-wavelength fluctuation is impeded, which results in the collapse of the $(3\cos^2\theta - 1)^2$-like behaviour of ΔH_{pp}. Moreover, ΔH_{pp} does not show critical broadening for any direction of $\boldsymbol{H}$ as can be seen in figures 4(b) and 6(b). The ESR theories reviewed in

[9] indicate that the critical broadening should occur in a two-dimensional Ising system. Those theories, however, stand on the assumption that the spin relaxation is only due to the spin–spin interactions, which is not allowed in actual Ising compounds because the spins in them couple strongly with the lattice. In the present case, the Ising anisotropy owes its origin to the Co^{2+} ions which are strongly connected with the lattice. Although we cannot give a quantitative explanation for the present result, we think the spin–lattice relaxation should be taken into account when interpreting the present result.

It is instructive to compare the present results of ΔH_{pp} around $\bar{T}_c$ with those of $K_2Cu_{1-x}Mn_xF_4$ [12]. This mixed system also shows a ferromagnetic phase for the Cu-rich side, but the ferromagnetic state is wholly XY-like because the competition of the anisotropies is very weak. According to [12], the samples with x of the ferromagnetic phase show critical broadening of ΔH_{pp}, indicating the absence of the spin–lattice relaxation owing to the S state of the Mn impurities.

Once the system of Ising phase turns into the ordered state, however, ΔH_{pp} shows a broader value for the hard axis than for other axes. This is because the fluctuation of the anisotropy field disturbs the uniform precession of the magnetization, which results in the line broadening.

We finally refer to the result of the spin-glass phase. The downward shift of H_{res} with decreasing temperature without showing any anomaly at the freezing temperature is similar to other several spin glasses, for instance, $CdCr_{2x}In_{2-2x}S_4$ [13], $Rb_2Mn_{1-x}Cr_xCl_4$ [14] and so on. The origin of the shift of H_{res} is discussed in detail in [14], and so we do not repeat it here. However, it is surprising for us that the critical broadening appears in the sample of the spin-glass phase. Nevertheless the spin–lattice relaxation should not be so different in the samples of the spin-glass phase and those of the oblique and the Ising phase. It suggests that the critical broadening of $\Delta H_{pp}(T)$ at the freezing temperature in the spin glasses is very common, i.e. it appears regardless of the Ising- or Heisenberg-type interactions and the spin–lattice relaxation does not prevent its appearance.

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