お茶の水女子大学理学部物理教室

2. Magnetic property of randomly mixed antiferromagnet with competing anisotropies: $Fe_xNi_{-x}Cl_2$ —Field induced phase transitions—

（異方性の競合する反強磁性体混晶 $Fe_xNi_{1-x}Cl_2$ の磁性）— 磁場誘起の相転移 —

Mariko Kitazawa（北 沢 真理子）

Abstract

$Fe_xNi_{1-x}Cl_2$ is a randomly mixed antiferromagnet with orthogonal spin anisotropies. The magnetic behavior of this system under high magnetic field parallel to the hexagonal c-axis has been studied in the wide concentration range ($0.02<x<0.5$) using the ^{57}Fe Mössbauer spectroscopy and the magnetization measurement. $FeCl_2$ is a typical antiferromagnet with the strong Ising anisotropy along the c-axis and undergoes the spin flip (metamagnetic) transition at 10.6 kOe. On the other hand, $NiCl_2$ has the weak XY anisotropy perpendicular to the c-axis. In the intermediate concentration range, the magnetic anisotropy of the system is expected to reduce, in the average, with decreasing x owing to the competition of these orthogonal spin anisotropies.

The magnetization measurement at 4.2 K has revealed that the spin flop transition, which is characteristic of the system with weak uniaxial anisotropy, occurs in specimens with the concentration near x=0.1, while the spin flip transition similar to $FeCl_2$ occurs in specimens with higher Fe concentration ($x>0.3$). In the specimens of $0.03<x<0.1$, which are in the oblique antiferromagnetic (OAF) phase in zero magnetic field, a noticeable but gradual increase was observed in magnetization (M) below 35~38 kOe, where the flop of the spin direction occurred. This feature is very similar to the magnetization process of the substance with weak uniaxial anisotropy such as MnF_2 under a magnetic field pointing a direction other than the easy axis. In the specimens of $0.1<x<0.3$, which are in the $S_{/\!/}$ ordered phase in zero field, the marked jump of M was observed at H_{SF} =38 kOe following to the small rounded rise of M below H_{SF}. Then M increased linearly up to about 110 kOe, where M attained the saturation. This fact strongly suggests that the successive transition of $S_{/\!/}$-ordered AF→OAF→SF-like→paramagnetic phase predicted by Matsubara and Inawashiro using the mean field theory has been realized in the intermediate concentration range of this system. In the specimens of $x>0.3$, the jumps of M were also observed at H=36 kOe and they were larger than those in the specimens with less Fe concentrations. The magnetization process is qualitatively similar

to that of $FeCl_2$ except for the rounding observed both below and above the transition field. Therefore, we consider that in the specimens of $x>0.3$, the transition from the $S_{/\!/}$-ordered to the OAF phase and that from the OAF to the paramagnetic phase occur successively. From the variation of the magnetization process with concentration x mentioned above, it is concluded that the magnitude of the magnetic anisotropy field changes widely with concentration x across that of the exchange field.

In order to get the microscopic property of the phases induced by magnetic fields, we applied the Mössbauer spectroscopy to the specimens with various concentrations. Among them, the specimen of $x=0.15$ was examined in detail. From the precise analysis of the Mössbauer spectra, the following results were obtained. On the application of magnetic fields, the direction of the down spins is rotated apart from the c-axis toward the field direction gradually while that of the up spins is nearly along the c-axis, and the spin axis flops abruptly when the canting angle of the down spins from the c-axis exceeds 25°. These observations are the first and direct evidence for that the OAF phase is induced by magnetic fields as an intermediate phase between the $S_{/\!/}$-ordered AF phase and the spin flop phase.

Comparing the magnetization of the Fe^{2+} ions (M_{Fe}) derived from the Mössbauer measurements with the magnetization (M_{Fe+Ni}) obtained by magnetization measurements, we found that M_{Fe} is significantly larger than M_{Fe+Ni} under every field between H_{SF} and 50 kOe, the maximum field examined by our Mössbauer study. This fact implies that the direction of the Fe^{2+} spins makes a larger angle from the c-axis than that of Ni^{2+} spins, against the magnetic anisotropy of Fe^{2+} ions. The magnetic structure of this phase is now in open question.