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MÖSSBAUER EFFECT OF $^{125}$Te IN MnTe$_2$ --- SPIN AXIS IN NON-COLLINEAR ANTIFERROMAGNETIC ORDERING OF THE FIRST KIND IN FCC LATTICE

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Abstract.- Mössbauer effect of $^{125}$Te in MnTe$_2$, which is antiferromagnetic below 85 K, was measured from 4.2 to 90 K. At 60 K the hyperfine field and the quadrupole splitting change discontinuously. The angle between the direction of hyperfine field and the principal axis of electric field gradient increases from 23° at 4 K to 30° at 60 K and decreases to 0° at 70 K.

1. Introduction.- Antiferromagnetic MnTe$_2$ and NiS$_2$ have the spin structure of the first kind ordering of fcc lattice /1,2/. Neutron diffraction patterns obtained from powders or multidomain single crystals cannot distinguish between the collinear and the non-collinear structures. In this case, Mössbauer experiment gives information for determination of the magnetic structure /3,4/.

Pasternak and Spijkervet measured the transferred hyperfine field at $^{125}$Te in MnTe$_2$ /3/. The analysis of the transferred hyperfine field from the nearest-neighbor Mn atoms revealed that MnTe$_2$ has the non-collinear first kind ordering and the direction of spin makes an angle $\theta$ which is about 30° at 4.2 K and nearly 0° at 77 K with the symmetry axis at Mn site /3/. A similar spin structure is observed in NiS$_2$ by the Mössbauer effect of $^{57}$Fe doped in NiS$_2$ /5/. Recently, the effect of the fourth order interactions between localized spins in antiferromagnetic fcc lattices has been studied theoretically by Yoshimori and Inagaki /6/. It appears that the effect of the higher order interactions is appreciable in 3d-metal dichalcogenides with the pyrite structure.

We have made Mössbauer measurement of $^{125}$Te to investigate the temperature dependence of spin direction in MnTe$_2$, and have found anomalous changes in the spin direction and the hyperfine field below $T_N$.

2. Mössbauer spectrum.- The sample was a polycrystalline MnTe$_2$ prepared by a sintering method. The Néel temperature of this sample was determined to be 85 K from resistivity and magnetic susceptibility measurements.

Fig. 1: Mössbauer spectra of $^{125}$Te in MnTe$_2$.

An average line width $2\Gamma$ of the Lorentzian was...
This value is 2% smaller than that of Pasternak and Spijkervet /3/. 

3. Results and Discussion.- Temperature dependences of the quadrupole splitting and hyperfine field are shown in figures 2 and 3, respectively.

At about 60 K both the quadrupole splitting and the hyperfine field change discontinuously. The hyperfine field decreases from about 125 to 75 kOe and the quadrupole splitting from -5.8 to -8.0 mm/s at 60 K with increasing temperature. These changes were not observed by Pasternak and Spijkervet /3/. This is because the value of quadrupole splitting was fixed at the value of 90 K in their analysis of the spectrum at 4.2 K.

The thermal expansion of MnTe₂ was measured by Kasai and Waki /7/. An anomalous increase in lattice constant of the order of 10⁻⁵ Å was observed at 60 K with increasing temperature. The hyperfine field at ¹²⁵Te is the transferred hyperfine field from the nearest neighbor Mn and the quadrupole splitting is caused by the molecular nature of the Te₁⁻⁻Te₂⁻⁻ anion pair in MnTe₂ /3/. Therefore, both the hyperfine field and the quadrupole splitting are sensitive to the position of tellurium atoms in the crystal. Since the crystal structure of MnTe₂ is the pyrite type down to 4.2 K /1/, it is most likely that the internal parameter of crystal lattice changes at 60 K in MnTe₂.

Figure 4 shows the temperature dependence of θ in MnTe₂. The θ increases from about 23° to 30° with increasing temperature up to 60 K. Above this temperature θ rapidly decreases to 0°. The experimental results show that the direction of Mn spin begins to rotate toward a body diagonal direction with increasing temperature above the temperature at which the internal parameter changes. Further work is required to make clear the mechanism of the temperature dependence of spin axis.

References
/7/ Kasai, N. and Waki, S., private communication.