## Spin-Disorder and Non-Degenerate Energy States in Geometrically Frustrated Materials

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We have studied chalcogenide Ni<sub>1-x</sub>Fe<sub>x</sub>Ga<sub>2</sub>S<sub>4</sub>, showing geometrical frustration effect. M-H curves at 4.2 K reveal that the disordered spins cannot be rotated completely along the direction of high external field of 5 T, since these spins are strongly constrained in the triangular lattice.  $H_{\rm C}$  increases with increasing Fe concentration, being consistent with the enhanced antiferromagnetic (AFM) spin-spin interactions and the suppressed spin-fluctuation due to the increase in freezing temperature  $T_{\rm f}$ . The specific heat  $(C_{\rm p}/T)$  measurement do not show any phase transformation between 2 and 160 K and there is no clear indication of gap in the temperature dependent  $C_{\rm p}$  curve between 0 and 7 T, because the atomic short-range ordering of the strained spin in the geometrically frustrated triangular lattice exists at low temperature. Though typical geometrically frustrated magnet shows degeneracy, Mössbauer analysis at 4.2 K shows that 5D of 3d orbit in the samples studied here is splitted into  $^5T_{2g}$  and  $^5T_{2g}$ , and  $^5T_{2g}$  is further splitted into singlet and doublet by large electric quadrupole splitting. This suggests that Fe<sup>2+</sup> ionic state has no absolute degenerate energy states in all samples due to Jahn-Teller effect.

Index Terms—Geometrically frustrated magnet, Mössbauer spectroscopy, non-degenerated energy, spin-disorder.