## $\mu$ SR study of the spin correlation in iron-chalcogenide superconductors Fe<sub>1-y</sub>M<sub>y</sub>Se<sub>0.3</sub>Te<sub>0.7</sub> (M = Co, Ni, Zn)

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In recent years, much attention has been paid to the so-called iron-based superconductors due to their high superconducting (SC) transition temperatures,  $T_{\rm c}$ 's, in the research field of superconductivity. Hsu et al. have discovered superconductivity with  $T_{\rm c} = 8$  K in the iron-chalcogenide FeSe.<sup>1)</sup> It has been found that  $T_{\rm c}$ of FeSe increases through the partial substitution of Te for Se, shows a maximum of 14 K at  $x \sim 0.7$  in  $\operatorname{FeSe}_{1-x}\operatorname{Te}_{x}$  and the superconductivity disappears at x = 1, namely, in FeTe.<sup>2</sup>) The compound FeTe is not SC but develops an antiferromagnetic (AF) order at low temperatures below  $\sim 67 \text{ K}^{(2)}$  Therefore, one may guess the mechanism of superconductivity relating to the AF spin fluctuation in the iron-chalcogenide superconductors, which is similar to the case of high- $T_{\rm c}$ cuprate superconductors.

In order to investigate impurity effects on the SC properties, we have grown impurity-substituted single crystals of  $\text{Fe}_{1-y}M_y\text{Se}_{0.3}\text{Te}_{0.7}$  (M = Co, Ni, Zn) with y = 0 - 0.05 and have measured the in-plane electrical resistivity.<sup>3)</sup> Here, Co and Ni ions are expected to dope one and two electrons, respectively, while Zn is expected to dope no electrons. As a result, it has been found that  $T_{\rm c}$  is reduced through the Co substitution and that the reduction of  $T_{\rm c}$  is more significant in the Ni substitution than in the Co substitution. On the other hand, the decrease in  $T_{\rm c}$  with the Zn substitution has been found to be negligibly small. Moreover, it has been found that both  $T_{\rm c}$  and the residual resistivity depending on the impurity concentration cannot be explained by the Abrikosov-Gor'kov theory, suggesting that a glue to form electron pairs is not the spin fluctuations $^{4,5)}$  but the orbital fluctuations.<sup>6)</sup>

Therefore, in order to investigate the spin fluctuations directly, we have performed  $\mu$ SR measurements of Fe<sub>1-y</sub>M<sub>y</sub>Se<sub>0.3</sub>Te<sub>0.7</sub> (M = Co, Ni, Zn) with  $y = 0 - 0.05^{(7)}$  Zero-field (ZF) and longitudinal-field  $\mu$ SR measurements were carried out using a Variox cryostat at temperatures down to 1.6 K at RIKEN-RAL.

For the impurity-free crystal of y = 0, it has been found that ZF  $\mu$ SR spectra are independent of temperature at low temperatures down to 1.6 K, indicating that the crystal is in a paramagnetic state. Figure 1 shows ZF spectra of the Zn-substituted crystal of Fe<sub>1-y</sub>Zn<sub>y</sub>Se<sub>0.3</sub>Te<sub>0.7</sub> with y = 0.05. At 29 K, the spectrum shows slow depolarization of muon spins similar to that observed in the impurity-free crystal. On the other hand, the depolarization becomes fast with decreasing temperature and the initial asymmetry is missing at 1.6 K. These results suggest significant development of the spin correlation through the Zn substitution. For the Co and Ni substitution, it has been found that the spin correlation is enhanced at y = 0.02 and that further substitution of impurities leads to weakening of the spin correlation at y = 0.05. These  $\mu$ SR results depending on the amount and kind of impurities strongly suggest that not the spin fluctuations but the orbital fluctuations may be a glue to form electron pairs in the iron-chalcogenide superconductors.

In summary, we have found impurity-induced development of the spin correlation in  $\text{Fe}_{1-y}M_y\text{Se}_{0.3}\text{Te}_{0.7}$  (M = Co, Ni, Zn) from  $\mu$ SR measurements. The present  $\mu$ SR results strongly suggest that the formation of electron pairs is mediated by the orbital fluctuations in iron-chalcogenide superconductors.



Fig. 1. Zero-field  $\mu$ SR time spectra of Fe<sub>1-y</sub>Zn<sub>y</sub>Se<sub>0.3</sub>Te<sub>0.7</sub> with y = 0.05.

References

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