

§1. Introduction

The magnetic properties of Eu chalcogenides with the NaCl structure are usually described by the first and second nearest neighbor exchange interactions J_1 and J_2 . The ferromagnetic J_1 is a strong function of the Eu-Eu distance, while J_2 is less sensitive to it [1,2]. EuSe shows various magnetic phases such as NNSS (4.6~2.8 K), NNS (2.8~1.8 K) and NSNS types (≤ 1.8 K) [3]. (Here, NSNS means that the spin directions within alternative (111) planes are north and south.) The complex magnetic phases of EuSe have been primarily attributed to $J_1 \approx -J_2$ [1], but have not yet been fully explained.

In a previous work [4] the effects of S, Te and Sr substitutions on the magnetic phases of EuSe were investigated by measuring NMR and AC susceptibilities χ_{ac} . In $\text{EuSe}_{1-x}\text{Te}_x$ and $\text{Eu}_{1-x}\text{Sr}_x\text{Se}$, where the lattice constant a increases with x , the NSNS phase is stabilized as shown in Fig.1. In $\text{EuSe}_{1-x}\text{S}_x$ the NNSS and ferromagnetic phases exist for $x \leq 0.1$ and $x \geq 0.1$, respectively. In this work we have reexamined the magnetic phases of $\text{EuSe}_{1-x}\text{S}_x$ near $x \approx 0.10$ and have investigated the effect of alloying with nonmagnetic compounds such as SrS, CaS and SmSe by NMR and χ_{ac} .

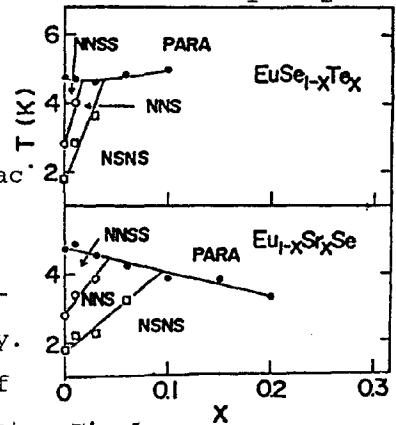


Fig.1

§2. Experimental Results

Polycrystalline specimens were prepared by heating a pressed mixture of powders of EuSe and appropriate compound in an enclosed tantalum crucible at 1700°C for ten hours. The lattice constants of the specimens, as shown in Figs. 2 and 3, decrease with increasing x , following Vegard's law, except for $\text{Eu}_{1-x}\text{Sm}_x\text{Se}$, where a exhibits an anomalous dependence on x owing to a valence change of Sm ions. With increasing x , θ_p increases in $\text{EuSe}_{1-x}\text{S}_x$, while it decreases in other alloys. The x dependences of θ_p are explained on the basis of the molecular field theory.

The line shape of the ^{153}Eu NMR in EuSe at 1.7 K consists of two peaks as shown in Fig.4. The high-frequency NMR comes from nuclei on the N sites of the NNS phase (NNS) and the low-frequency NMR comes from the nuclei in the NSNS phase and on the S sites of the NNS phase (NNS) [5]. The NMR of the NNSS phase, the frequency of which agrees with that of the NNS line, is observed above 2.5 K [5]. The difference in the frequencies of the two NMR is that in the transferred

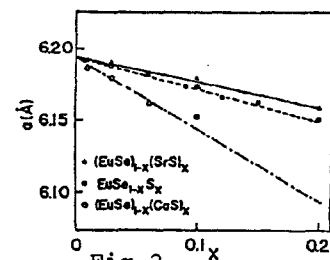


Fig.2

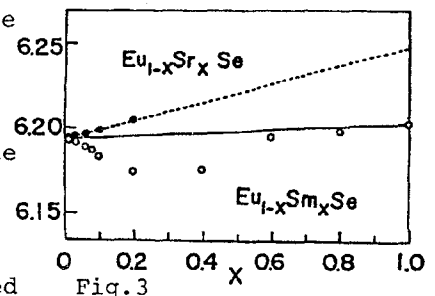


Fig.3

hyperfine fields at the two types of Eu sites. χ_{ac} of EuSe, as is shown in Fig.5, exhibits three abrupt changes at 2.0, 2.8 and 4.6 K, which correspond to the NSNS-NNS, NNS-NNSS and NNSS-paramagnetic transitions, respectively.

(1) $\text{EuSe}_{1-x}\text{S}_x$

The specimens of $x = 0.08, 0.09, 0.12$ and 0.15 were prepared and investigated. The results, together with previous ones, are shown in Figs.4 and 5. For $x \leq 0.10$ only the high-frequency NMR was observed and is attributed to the NNSS phase, since no NNS line was observed. For $x \geq 0.12$ no NMR was detected in the range of 100-200 MHz, probably because of short values of T_2 ($\leq 1 \mu\text{sec}$). χ_{ac} for $x \leq 0.06$ show only the change due to the first order transition at 4.6 K, indicating, together with NMR results, that only the NNSS phase exists in these specimens. χ_{ac} for $x \geq 0.15$ show typical ferromagnetic behaviors, indicating the existence of the ferromagnetic order, although the NMR confirmation was unsuccessful. The specimens of $x = 0.08$ and 0.09 are in the NNSS phase as suggested by NMR and χ_{ac} results, but show no first order transition at $T_N \approx 4.0$ K. The specimens with $0.10 \leq x \leq 0.12$ are in the transition from the NNSS to ferromagnetic phase. The magnetic phase diagram of $\text{EuSe}_{1-x}\text{S}_x$ is shown in Fig.6.

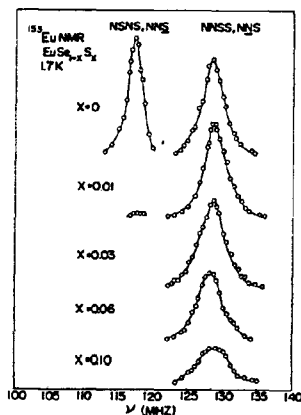


Fig.4

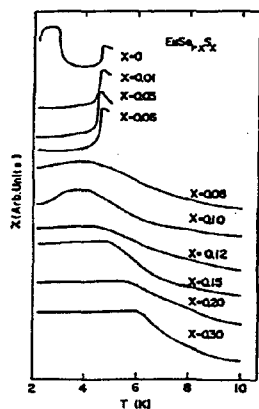


Fig.5

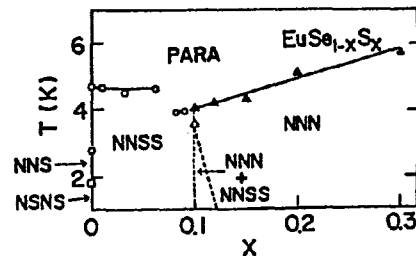


Fig.6

(2) Alloys with SrS, CaS and SmSe

The specimens of $(\text{EuSe})_{1-x}(\text{SrS})_x$ with $x \leq 0.2$, $(\text{EuSe})_{1-x}(\text{CaS})_x$ with $x \leq 0.1$ and $\text{Eu}_{1-x}\text{Sm}_x\text{Se}$ with $x \leq 0.2$ were investigated. Typical results of NMR and χ_{ac} are shown in Figs.7 and 8. Only the high-frequency NMR were observed in $(\text{EuSe})_{1-x}(\text{SrS})_x$ with $x \geq 0.06$ and in $(\text{EuSe})_{1-x}(\text{CaS})_x$ with $x \geq 0.01$. χ_{ac} for $(\text{EuSe})_{1-x}(\text{CaS})_x$ show only the changes at T_N , at which no first order transition occurs for $x \geq 0.06$. These indicate that the NNSS phase is stabilized with increasing x in these alloys as in $\text{EuSe}_{1-x}\text{S}_x$. But no ferromagnetic order appeared. The results for $\text{Eu}_{1-x}\text{Sm}_x\text{Se}$ are similar, except for the critical concentration of $x = 0.10$, above which only the NNSS phase exists.

In Fig.9 the Néel temperatures in various alloy systems are plotted against $\mathcal{C}$. The temperatures of the NSNS-NNS and NNS-NNSS transitions are also drawn by

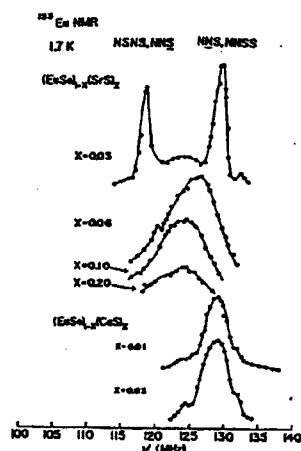


Fig. 7

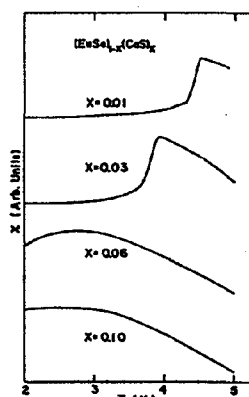


Fig. 8

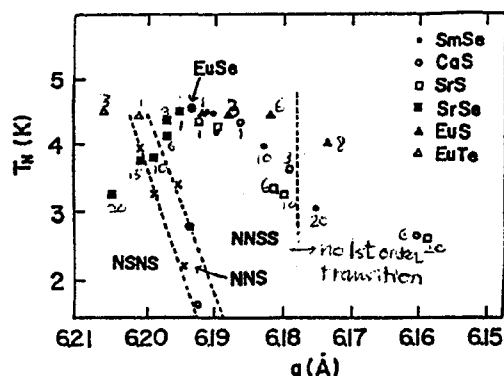


Fig. 9

the points (X) and broken curves. The transition temperatures exhibit different dependences on x among different alloy systems, but their dependences on a lie on smooth curves, suggesting that the changes of the magnetic phases in alloying are primarily induced by the changes in a . As seen in Fig. 9, the NSNS and NNS phases are stabilized with increasing and decreasing a , respectively, and the NNS phase is stable in a narrow range of a . The first order transition at T_N disappears below $a = 6.175 \sim 6.180$ Å.

§3. Discussions

In the molecular field approximation using J_1 and J_2 the magnetic phase is determined by J_2/J_1 [1]. Various phase changes of EuSe in alloying may be primarily due to $J_2/J_1 \approx -1$ and a strong dependence of J_1 on a . The NSNS phase in $\text{EuSe}_{1-x}\text{Te}_x$ and $\text{Eu}_{1-x}\text{Sr}_x\text{Se}$ and the ferromagnetic phase in $\text{EuSe}_{1-x}\text{S}_x$ are qualitatively understood by a decrease and an increase of J_1 , respectively, which are caused by the changes of a in alloying. But the NNS and NNS phases, including the first order transition at T_N , cannot be explained only in terms of J_1 and J_2 . Other mechanisms such as lattice distortion [6] and higher order exchanges [7] might play important role in determining the magnetic phases.

References

- [1] W. Zinn: J. Magn. Magn. Mater. 3(1976) 23.
- [2] T. Kasuya: IBM J. Res. Develop. 14(1970) 214.
- [3] R. Griessen, M. Landolt and H.R. Ott: Solid State Commun. 9(1971) 2219.
- [4] K. Kojima, T. Hihara and T. Kamigaichi: Proc. Intern. Conf. Ferrites(1981)
- [5] T. Komaru, T. Hihara and Y. Koi: J. Phys. Soc. Jpn 31(1971) 1391.
- [6] H. Callen and M.A. deMoura: Phys. Rev. B 16(1977) 4121.
- [7] G. Petrich and T. Kasuya: Solid State Commun. 8(1970) 1625.