Mossbauer Effect Studies of Valence Fluctuating Compound Sm3Se4 in an Applied Magnetic Field at Low Temperature

I. 10 Mössbauer Effect Studies of Valence Fluctuating Compound Sm_3Se_4 in an Applied Magnetic Field at Low Temperature

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Physical Properties of Sm_3Se_4 have been studied by us in the various measurements.[1,2] It indicates the thermal activated valence fluctuation at high temperature similar as Eu_3S_4 in which the electrical resistivity shows an activation type. The activation type, however, originate from the 4f-electron, that is mainly evidenced by the lack of the observation of Hall effect in Sm_3Se_4. The problem is now concentrated to the anomalous valence fluctuating state at low temperature. There are possibilities to explain the anomaly by a quantum-mechanical tunneling valence fluctuation or the some ordering at low temperature.[3] To shed light on the above problems, we performed the Mössbauer Effect experiment of Sm_3Se_4 in an applied magnetic field at low temperature.

The Mössbauer absorption spectra of Sm_3Se_4 were obtained in a standard transmission geometry with the source and the absorber at 4.2 K, using a conventional constant acceleration spectrometer. The source, consisting of 22.5 keV γ-transition in ^{149}Eu, was produced in a (p,2n) reaction using the cyclotron accelerator at Tohoku University. The γ-rays were detected by a pure Ge LEPS of 5 cm^3. The direction of the external magnetic field of 5 tesla was parallel to the γ-direction. The other experimental details were described elsewhere.[1,2]

Mössbauer spectra at 4.2 K without external magnetic field are plotted in Fig. 1. It shows antisymmetrical in respect to the peak position. We analyze the data with the following assumption. i) The spectra consist of Sm^{2+} and Sm^{3+} ions, and they give the different absorption. This fact is proved by the various measurements,[1-3] and the intensity ratio of Sm^{2+}/Sm^{3+} is to be 1/2. ii) Each line shape is the Lorentzian as usual. iii) The isomer shift of Sm^{3+} is zero as is shown in the various compounds with the source of Sm_2O_3. iv) Sm^{3+} sites have the internal field and have the nuclear level splitting. Because we could not fit the data adequately by two Lorentzian curves of Sm^{2+} and Sm^{3+}. In this way we get the isomer shift of Sm^{2+}, linewidth and the internal magnetic field as the fitting parameter listed in Table 1. These are consistent with our reported results.[2] Here, we use the internal field analysis similar as Ref. 4.

Mössbauer spectra under the external magnetic field of 5 tesla at 4.2 K are shown in Fig. 2. We analyze the data with the same assumption above adding the existence of the internal field of Sm^{2+} site. The fitting data are also tabulated in Table 1. The isomer shift of Sm^{2+} is consistent with the other Sm^{2+}-compounds within the error. The internal field of Sm^{2+} is 1.8 tesla at 4.2 K with the external field of 5 tesla. It means that the internal field for Sm^{2+} acts as opposite sense to the external field as is expected by the simple theory.[5] The internal field of Sm^{3+} is 24 tesla at 4.2 K with the external field of 5 tesla. We have measured the magnetization of Sm_3Se_4 under high magnetic field.[1] It shows anomalously small value of 0.03 μ_B per Sm^{3+} ion under the magnetic field of 5 tesla at 4.2 K. The full moment of Sm^{3+} is 0.714 μ_B and it induces the internal field of 470 tesla at the nucleus.[6] Then the 0.03 μ_B corresponds to the internal field of 20 tesla. This value is consistent with the observed internal field of 24 tesla considering the external field of 5 tesla. This fact proved the anomalously small value of the magnetization by the microscopic standpoint of the Mössbauer effect measurement. The small value of magnetization is attributed to the anomalous valence fluctuation state at low temperature. The origin of the internal field observed at zero external magnetic field is still remained unknown, and more detailed experiments at very low temperature (less than 1 K) are needed to check this point.

References

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Table 1. Isomer shift, line width and the internal magnetic field at 4.2 K without and with the external magnetic field of 5 tesla, respectively, for $^{249}Sm^{3+}$ and $^{149}Sm^{2+}$.

Mössbauer effect		4.2 K	
		0 tesla	5 tesla
Sm^{3+}	isomer shift (mm/s)	0	0
	linewidth (mm/s)	4.3±1.2	3.9±0.7
	magnetic field (tesla)	13.8±4.8	24.0±6.0
Sm^{2+}	isomer shift (mm/s)	-1.0±0.3	-0.8±0.3
	linewidth (mm/s)	5.2±0.9	4.5±2.2
	magnetic field (tesla)	0	1.8

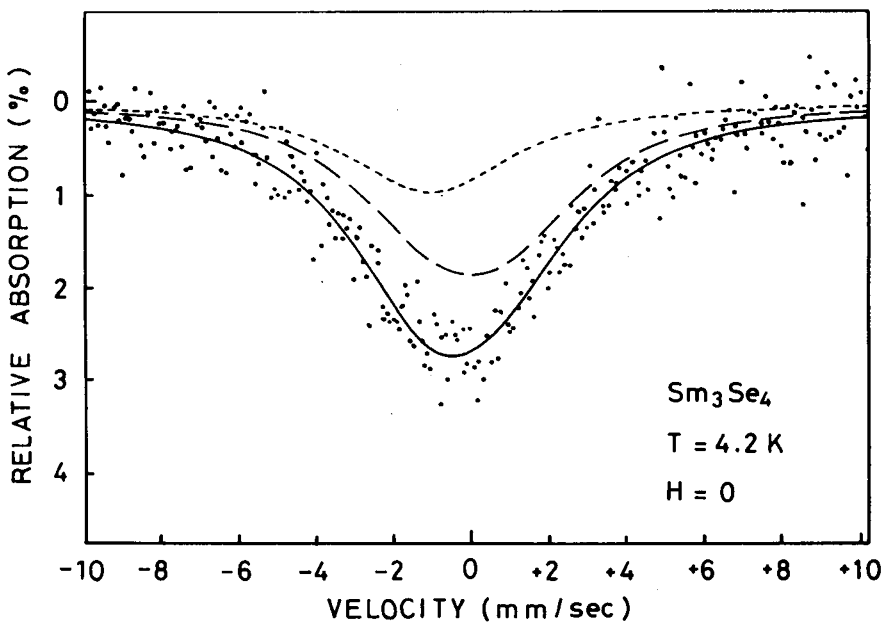

Fig. 1. Mössbauer spectra of Sm_3Se_4 at 4.2 K. The lines are least squares fits to the data. Upper line is for Sm^{2+}-ion, middle line is for Sm^{3+}-ion and the lowest line is a total of them.

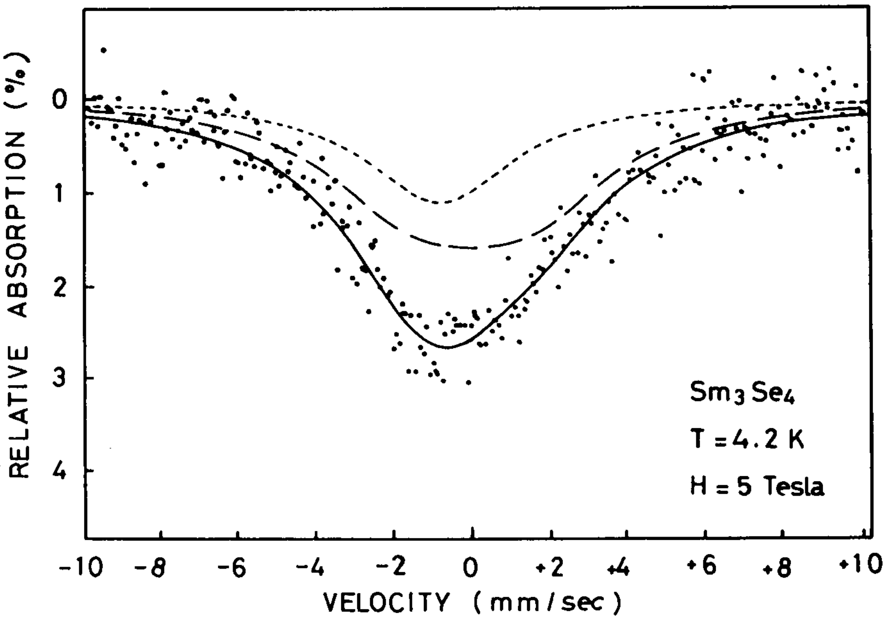

Fig. 2. As Fig. 1, but the external magnetic field of 5 tesla is applied.