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Thermoelectric power in Fe-based granular alloys

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Abstract. We have investigated the field dependence of thermoelectric power S(H) in Fe–Ag and Fe–Cu granular alloys to compare with the theoretical models to explain the S(H) correlation with the giant magnetoresistance. The sign for the magnetic field dependent part of the S(H) is found to be negative for both systems in agreement with the virtual-bound-state model prediction.

1. Introduction

Since the discovery of the giant magnetoresistance (GMR) [1], various transport properties have been investigated to characterize the conduction electron scattering responsible for the GMR on multilayers and granular alloys [2–7]. However there still remain several controversial issues concerning the thermoelectric power in the GMR systems.

One of the interesting issues in the GMR effect is whether the dominant scattering occurs in the bulk or at the interfaces. The spin-split density of state (DOS) model [5, 6] is based on the former; i.e., the bulk scattering within the ferromagnetic layers (granules), while the virtual bound state model [8] is based on the latter, i.e., the scattering at the virtual bound states near the interface. Both models have predicted the sign of the field dependent part of the thermoelectric power ($\Delta S(H)$) correlated with the GMR in T–Cu (T, 3d transition metals) systems. The sign of $\Delta S(H)$ for multilayers and granular alloys containing Co has been reported to be always negative [2–7], which has been considered to be a supporting proof for the spin-split DOS model, i.e., the bulk scattering as an origin of the GMR. On the other hand, for the Fe-based system, an experiment on only a single Fe–Cu multilayer sample has been reported, in which the sign agrees rather with the virtual bound state model [7]. In order to confirm the discrepancy between the theoretical models and the experiment on the thermoelectric power, further experiments on Fe-based granular alloys are necessary.

In this paper, we report the experimental results on the thermoelectric power in Fe–Ag and Fe–Cu granular alloys to clarify the issues described above.

2. Experimental procedure

Fe–Ag granular samples were prepared by the following three methods: molecular-beamepitaxy (MBE) [9], DC sputtering (SP) [10] and ion-cluster-beam (ICB) [11] methods. Fe– Cu granular samples were prepared by DC sputtering [12]. The averaged cluster radius was

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Figure 1. The temperature dependence of thermoelectric power (*S*) at zero field for Fe–Cu granular alloy, along with the data for Co–Cu granular alloy from [4].

estimated to be of the order of 100 Å by a transmission-electron-microscopy observation for SP and ICB samples [10, 11]. We found no essential difference in the transport properties due to the sample preparation methods, except a slight difference in the magnitude of the resistivity and the thermoelectric power. Therefore we only show the results for the SP samples in this paper.

Magnetoresistances were measured by a standard DC four-probe method using a computer-controlled current source and Keithley 181 nanovoltmeters. Magnetic field was applied up to 5 T using a superconducting magnet and up to 1 T using a conventional iron core electromagnet. Thermoelectric power was measured by a differential method using AuFe–chromel differential thermocouples. The field dependence of the thermoelectric power of the thermocouples is less than 1% for temperature from 77 to 300 K and field up to 1 T.

3. Results and discussion

Shi and coworkers proposed that the thermoelectric power (S) measurements can be used to test the theoretical models for the GMR effect [5, 6]. According to their spin-split-DOS model, *S* can be scaled as

$$S(H) = A + B/\rho(H) \tag{1}$$

where A and B are independent of H. In their model, the sign of the constant B is determined solely by the DOS for the bulk ferromagnetic component, and is negative for Co systems and positive for Fe systems. In contrast, in the virtual-bound-state model proposed by Inoue and coworkers for T–Cu multilayers, the sign of B is determined by the virtual bound states of the magnetic elements in Cu, and is predicted to be positive for Co and negative for Fe [8].

Many experiments have been performed on the giant magnetothermoelectric power in the Co-based GMR systems, where B was found to be negative [2–7] in agreement with



Figure 2. The field dependence of S and ρ for Fe₂₇Cu₇₃ granular samples at T = 296 and 77 K.

the spin-split-DOS model prediction. This fact has been argued to be proof for the bulk scattering rather than the interfacial scattering as an origin of the GMR, since the spin-split-DOS model is based on the bulk scattering within the ferromagnetic components [5, 6] while the virtual-bound-state model is based on the scattering at the virtual bound state near the interface [8]. On the other hand, the reported experiment on the Fe-based system is quite limited; only one on a single Fe–Cu multilayer sample has been reported, where B was found to be negative in agreement with the virtual-bound-state model [7].

To confirm the sign of *B* in Fe-based systems, we have performed the magnetothermoelectric power experiment on Fe–Cu and Fe–Ag granular alloys. Figure 1 shows the temperature dependence of *S* at zero field for an Fe₂₇Cu₇₃ sample along with that reported for Co–Cu granular alloy [4]. In the Fe–Cu alloy, the influence of the surface oxidization, which is slightly larger compared with that in the Co–Cu alloy, might increase the experimental noise leading to the scatter of the data points. The temperature dependences are similar: a non-linear component which almost saturates near 70 K plus a linear component. Examples of the field dependence of *S* are shown in figures 2 and 3 along with the corresponding the $\rho(H)$ curves. The resemblance between S(H) and $\rho(H)$ is evident also in the Fe-based systems. The magnitude of the field dependent part of *S* for



Figure 3. The field dependence of S and ρ for Fe₂₈Ag₇₂ granular samples at T = 296 and 77 K.

the Fe–Cu granular alloy is similar to that reported for the Fe–Cu multilayer [7]. The sign of the field dependent part of *S* is negative for both Fe–Cu and Fe–Ag systems. The scaling relation is nicely fulfilled for both alloys as shown in figure 4. For both the Fe–Cu and the Fe–Ag systems, the sign of the constant *B* is negative in agreement with the virtual-boundstate model prediction and disagrees with the spin-split-DOS model one. The present result suggests that the negative sign of the constant *B* is common to the Fe-based GMR systems. This fact indicates that neither the spin-split-DOS model nor the virtual-bound-state model can explain the sign of *B* for Fe and Co systems consistently: in other words, the bulk scattering and the interface scattering cannot solely explain the scattering responsible for the GMR. Comparing the cluster size and/or layer thickness with the mean free path of conduction electrons, it may not be reasonable to assume that only either the bulk scattering or the interface scattering dominates in the GMR effect. To explain the sign of *B* in the scaling relation for both Fe and Co systems consistently, a new theoretical model is needed in which both types of scattering are taken into account.



Figure 4. The *S*/*T* versus ρ_{max}/ρ plots for Fe–Cu and Fe–Ag granular samples at T = 296 and 77 K.

4. Conclusion

We have measured the thermoelectric power in Fe–Ag and Fe–Cu granular alloys. We confirmed that the sign of the constant *B* in the scaling relation between *S* and ρ is negative for the Fe-based systems in agreement with the virtual-bound-state model. Taking into account the results for the systems containing Co, it is clear that neither the spin-split-DOS model nor the virtual-bound-state model can explain the thermoelectric power in Fe and Co systems consistently.

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