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High Field Mössbauer Study of Dilute Ir-(Fe) Alloys

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The magnetic behavior of very dilute Fe impurities in Ir has been studied by means of Mössbauer measurement in external fields up to 80 kOe at 4.2 K. The saturation hyperfine field increases in proportion to the external field up to the maximum magnetic field available. This means that for a localized spin fluctuation system *Ir*Fe, the effective magnetic moment associated with Fe impurities is induced in proportion to the external field. No anomalous spectrum was observed with a very dilute sample (~10 ppm ^{57}Co), indicating that the interaction between impurities is responsible for the anomalous spectrum previously observed with a less homogeneous sample.

KEY WORDS: Mössbauer effect/ Dilute alloys/ Iridium-iron/

I. INTRODUCTION

High field Mössbauer study is known as a very powerful method for investigation of magnetic properties of dilute alloys. Particularly, this method has provided valuable informations about Kondo alloy systems.[1,2]

Recently, we have reported the measurement of loclaized spin fluctuation systems *Tc*Fe, *Ru*Fe, and *Ir*Fe.[3] For the last alloy, the hyperfine field is much smaller than those of usual Kondo systems and the saturation field is almost proportional to the external magnetic field up to 50 kOe. In addition, an anomalous spectrum with an additional amplitude in the inner part was observed at low temperatures below 4.2 K and in external field of 30 and 50 kOe.

In order to study the origin of the anomaly and the field dependence of the saturation field, we have made two improvements on the measurement: (1) More homogeneous Mössbauer source of ^{57}Co in Ir was carefully prepared. (2) Hyperfine field measurements were extended to 80 kOe using newly prepared superconducting magnet system. In this paper, we present our new data on hyperfine field of *Ir*Fe and give some discussions.

II. EXPERIMENTS

1. Sample Preparation

The Mössbauer sources were prepared by diffusing ^{57}Co into Ir foils. In the previous experiment,[3] a 50 μm thick Ir foil (99.9%) was used. Approximately 100 μCi of ^{57}Co activity was deposited on the foil and was dried. Then it was heat

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treated at 1100°C for 25 hours in a flowing pure H_2 atmosphere. This source is here referred to as Ir-I.

To achieve more homogeneous distribution of ^{57}Co atoms in the host metal, we made some modifications in the procedure of source preparation. As the host, a 99.99% pure Ir foil of 50 μm thick was used. First, the foil surface was electropolished in HCl solution. Then, about 400 μCi of ^{57}Co activity was electroplated from an ammonical solution onto the foil area of 40 mm^2. Diffusion was carried out at 1400°C for 5 hours in the mixed atmosphere of H_2 and Ar. Finally, the surface was chemically etched. Assuming the average depth of diffusion was 20 μm, the average concentration of ^{57}Co corresponds to 9 atomic ppm. We refer to the present sample as Ir-II.

2. High Field Mössbauer Measurement

Mössbauer measurement was performed in a longitudinal geometry with a fixed cold source and a moving room temperature absorber. A schematic of the magnet and dewar system with the spectrometer is shown in Fig. 1. The sample was mounted in a superconducting magnet capable of attaining fields up to 80 kOe. The magnet was operated in the persistent current mode. The axis of the field was perpendicular to the sample surface and was parallel to the γ-ray direction. The sample temperature can be varied between 1.5 and 300 K. Temperatures below 4.2 K are obtained by pumping down the inner cell of the sample holder, into which liquid He distills from the He bath through a needle valve. Temperatures above 4.2 K can be controlled by regulating the heater current and the pressure of exchange gas.

The presence of the high field magnet causes difficult problems on both measurements of temperature and Mössbauer effect. The former problem arises from the fact that usual temperature sensors are field sensitive. We used a thermocouple of Au–0.07 at.% Fe *vs.* Ag–0.37 at.% Au and carbon resistors (Allen-Bradley and Matsushita). The thermocouple is insensitive to a magnetic field, while the resistors are slightly field sensitive.[4,5]

A fringing field affects an absorber located out of the dewar, resulting in a slight broadening of the line width of the spectrum. In the arrangement of Fig. 1, the fringing field acting on the absorber is about 2.5% of the field at the magnet center. As a single line absorber, potassium ferrocyanide containing 1.0 mg/cm^2 of ^{57}Fe was used.

Magnetic stray fields also affect a velocity transducer which contains permanent magnets, a driving coil, and a sensing coil. Correction of this effect is very important to estimate hyperfine fields as small as 10 kOe. In order to avoid the errors in the velocity, calibration of the velocity was made under the same condition with the hyperfine field measurement. For the calibration, another ^{57}Co source (15 mCi ^{57}Co in Rh) attached to the opposite side of the transducer and an Fe absorber were used. Since the shift in the velocity is proportional to the strength of the stray field, the transducer was kept apart from the dewar. In addition, as a magnetic shield, soft steel sheets (10 mm in total thickness) were placed between the dewar and the trans-

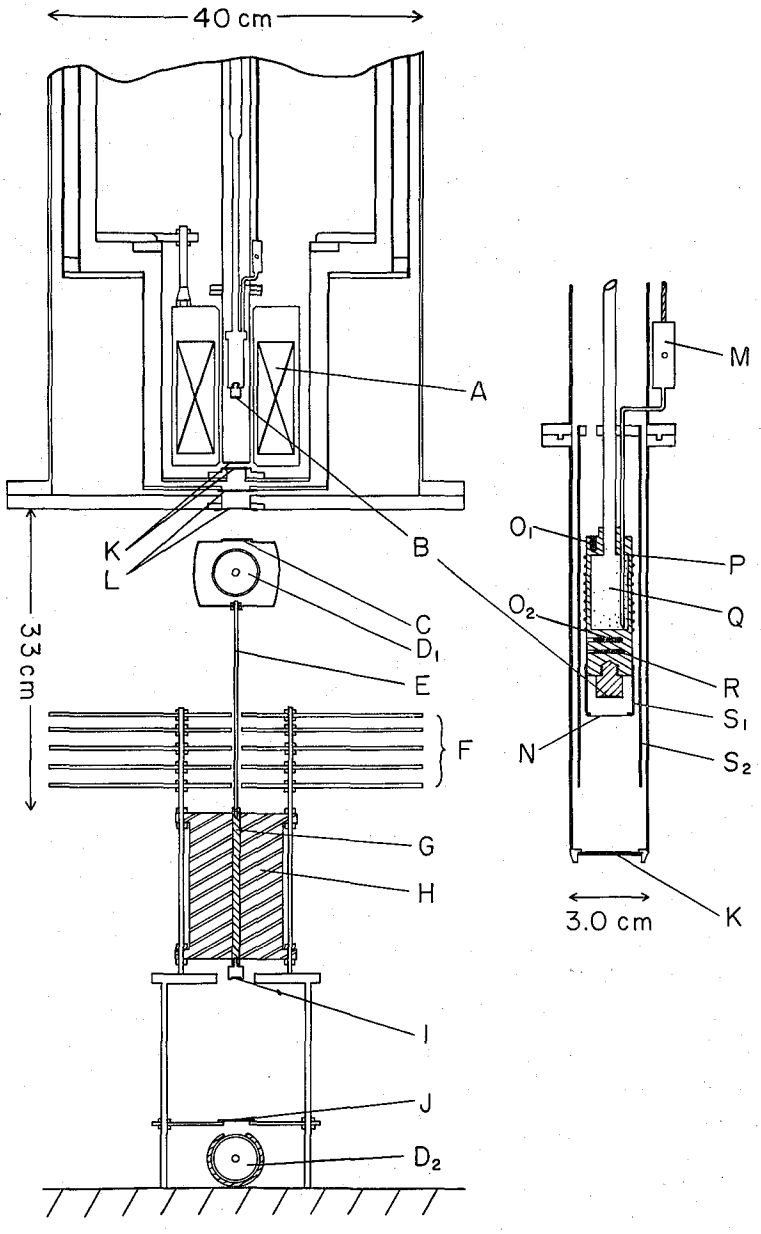

Fig. 1. Dewar system and spectrometer for high field Mössbauer measurement. A) Superconducting magnet, B) Sample (^{57}Co in Ir), C) Absorber (potassium ferrocyanide), D) Proportional counter, E) Al rod, F) Soft steel sheets, G) Drive rod, H) Velocity transducer, I) Calibration source, J) Fe absorber, K) Be windows, L) Mylar windows, M) Needle valve, N) Al foil, O) Carbon resistor thermometer, P) Heater, Q) Cu chips, R) Thermocouple (Au-0.07 at. % Fe *vs.* Ag–0.37 at. % Au), S) Cu radiation shield.

ducer. Even in this arrangement, the stray field at the top of the transducer is still 100 Oe, when the maximum field of 80 kOe is supplied. More effective shielding case of Permalloy is in preparation.

III. RESULTS AND DISCUSSION

Before dealing with the present data with Ir-II, we arrange the result of Ir-I. In the absence of external field, no appreciable braodening of linewidth was observed at low temperatures down to 1.4 K. The spectra obtained in fields of 30 and 50 kOe at 86 and 290 K were well fitted assuming the Lorentzian line shape, the ideal intensity ratio of 3:0:1:1:0:3, and equal line widths. On the other hand, spectra obtained at temperatures below 4.2 K under magnetic fields could not be fitted with the ideal ratio. The additional magnitude in the inner part of the spec-

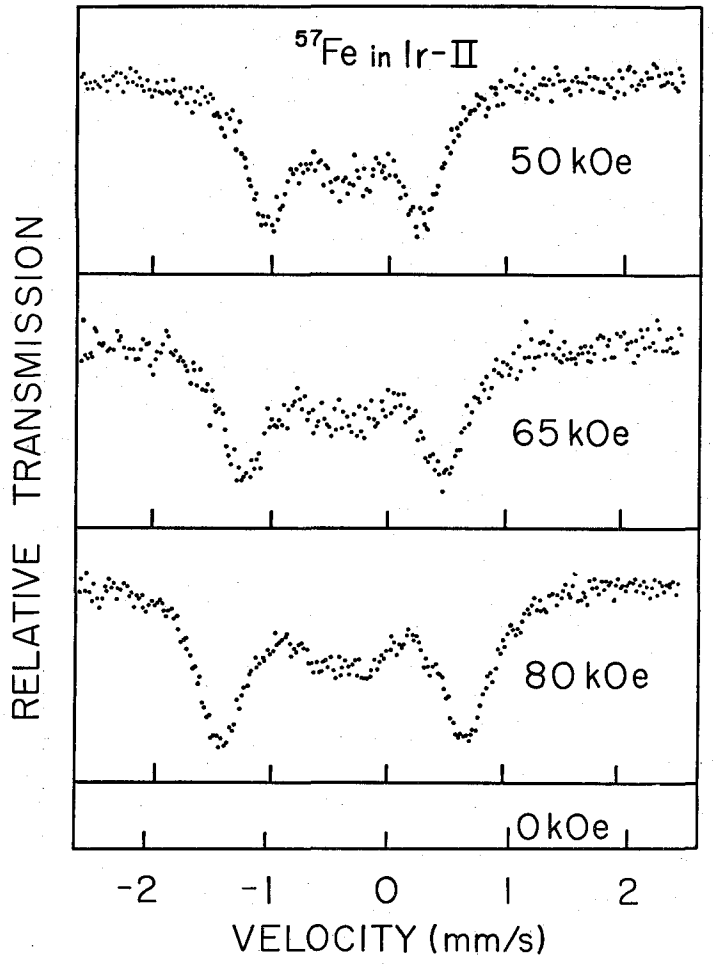

Fig. 2. Mössbauer spectra of Fe^{57} impurities in Ir-II at 4.2 K in various external magnetic fields. The direction of γ rays is parallel to the external field. The velocity calibration was made in the presence of external field (See Fig. 1).

trum was 40% at 4.2 K and 20% at 1.4 K in 50 kOe field. In the previous paper, we pointed out two possibilities which may cause this anomaly, *i.e.*, (1) relaxation effects of a single impurity, and (2) interaction effects between impurities.

Relaxation effects occur at low temperatures when the electronic relaxation time becomes comparable with the Larmor precession time of ^{57}Fe nucleus in the effective hyperfine field. In this case, the line width of the outer lines broadens than that of the inner ones, resulting in the additional magnitude in the inner part.[6]

Interaction effects between impurities also appear at low temperatures even at very low impurity concentration.[7,8] Distribution of the hyperfine field caused by the interaction can produce such an anomaly of the spectrum.

Typical Mössbauer spectra of Ir-II sample taken at 4.2 K in various external fields are shown in Fig. 2. Shift in the velocity scale due to stray fields is also shown. Contrary to the case of Ir-I, all the spectra of Ir-II can be well fitted with the ratio of 3:0:1:1:0:3 using a single line width.

The discrepancy between Ir-I and Ir-II is considered to be arised from the different processes of sample preparations. The distribution of ^{57}Co impurities in Ir-I is certainly less homogeneous than in Ir-II due to initial segregation of ^{57}Co atoms, insufficient hot annealing, and less pure Ir-I host. Inhomogeneous distribution of ^{57}Co atoms may cause the interactions between impurities, resulting in a distribution of hyperfine field. The hyperfine field of ^{57}Co atoms which have Fe or Co atoms in the vicinity is expected to be larger than that of isolated ^{57}Co atoms. The contribution from these ^{57}Co atoms may appear in the inner part of the spectrum. Above discussion leads us to the conclusion that interactions between impurities are responsible for the anomalous spectra found for Ir-I.

However, the anomaly is not attributable to the clustering of Co atoms, but is mainly due to long-distance interactions between impurities, Fe-Co and Fe-Fe. The reasons are: (1) At room temperature and in zero field, there is no difference be-

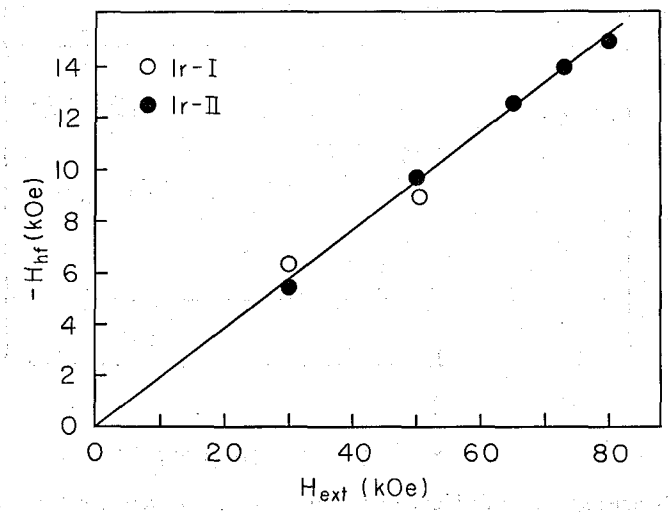

Fig. 3. Hyperfine fields (H_{hf}) of ^{57}Fe in Ir-I and Ir-II at 4.2 K as a function of external field (H_{ext}). Straight line is drawn as a visual aid.

tween the spectra of Ir-I and Ir-II. The spectrum of Ir-I has no shoulder corresponding to clusters. (2) At low temperatures down to 1.4 K in zero field, there is no appreciable broadening of linewidth, indicating that no spontaneous magnetic ordering due to the clustering occurs.

In Fig. 3 is shown the hyperfine field of Ir-I and Ir-II at 4.2 K as a function of external field. In our preliminary analysis of the spectra of Ir-II, the effect of stray fields on the velocity scale was not correctly taken into account.[9] The present analysis shown in Fig. 3 indicates that there is no significant difference in the hyperfine field of both samples. This suggests that impurity-impurity interactions do not affect the main hyperfine field of single impurities. This result is consistent with the work by Taylor and Steyert,[10] who used sources of ^{57}Co in Ir and $Ir_{999}Fe_1$.

As shown in Fig. 3, the hyperfine field of Ir-II increases in proportion to the external field up to 80 kOe. In our previous paper, we showed that the hyperfine field of Ir-I almost saturates when the value of H_{ext}/T exceeds 10, while the saturation field depends on H_{ext}. Therefore, the data of Ir-II at 4.2 K in external fields larger than 50 kOe can be regarded as the saturated value. Accordingly, the saturation field of Ir-II is almost proportional to the external field up to 80 kOe. This behavior closely resembles that of the Kondo system *Cu*Fe. In this case, almost linear behavior in saturation field was observed up to 136 kOe,[11] which was explained as follows: Since *Cu*Fe has a high Kondo temperature $T_K=28$ K, it is not possible to complete the breaking up of the spin compensation state with the available field strength. If we regard the localized spin fluctuation temperature of *Ir*Fe ($T_{sf}=225$ K)[12] as the Kondo temperature, our result is consistent with that of *Cu*Fe.

This saturation field is generally considered to be proportional to the magnitude of the effective magnetic moment of the impurity. In conclusion, for a localized spin fluctuation system *Ir*Fe, the effective magnetic moment associated with Fe impurities is induced in proportion to the external field and does not saturate even at magnetic fields as high as 80 kOe.

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