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I. 11. NMR/ON (Nuclear Magnetic Resonance in Oriented Nuclei) Study of Fe-Si Single Crystal

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1. Introduction

NMR/ON method is a powerful technique to investigate the magnetic or electrical properties of ferromagnetic materials. It has much better resolution to determine the hyperfine field than Mössbauer spectroscopy. Moreover it has an advantage over the conventional pulsed NMR technique that a single crystal can be used for the specimen. Because of the smallness of the specimen (4 mm diameter) needed, many kind of materials are available in a form of a single crystal form for the NMR/ON investigation. Different from the powder specimens employed in the pulsed NMR method, a bulk specimen can be studied free from the surface effects.

In a previous report by Nishimura et al.[1] on Fe-Si single crystal, where ^{111}In nuclei were implanted by a recoil implantation technique, no well defined resonance was detected although the temperature was low enough to give rise to oriented nuclei.

In this report the same NMR/ON technique is applied to Fe-Si single crystals with ^{57}Co and ^{60}Co[2] as the probe nuclei.

The purpose of the experiment is: (1) to determine the Hf at the Co nucleus site, (2) to resolve the satellites due to Si atoms in the 1st, 2nd and so on neighboring sites which are observed as only a shoulder or buried in the main absorption line in the Mössbauer spectrum and (3) to seek the dipolar field due to Si atoms in the 1st or 2nd neighbor site, which has been observed as the change of the Mössbauer line width by changing the magnetization axis.[3]

2. Experimental

Disks of Fe-Si (6 at %) alloy with 4 mm diameter and 0.1-0.2 mm thickness were

prepared by chemically etching a single crystal sheet supplied by Dr. A. Sakakura, Nisshin Steels Co. The specimen has (110) surface and hence it can be magnetized along to the three simple axes of <100>, <110> and <111>. A few drops of ^{57}Co or ^{60}Co chrolide solution were applied on a surface and the specimen was annealed at 1123 K for 1.8×10^3 sec in an atmosphere of hydrogen. With an available diffusion data of Co in Fe[4], the penetration depth, $\sqrt{2Dt}$, was estimated as 0.6 μm. After washing out the chrolide solution, the other surface of the specimen was soldered to a copper cold finger by a Wood metal. The cold finger was then set to the 400 μW dilution refrigerator with a top-loading facility, placed at Niigata University. The crystal orientation relative to the magnetization field supplied by two superconductive solenoid, was carefully adjusted at RT prior to the experiment at low temperatures.

The specimen was cooled down to 7-10 mK, where the temperature was determined by measuring ^{54}Mn γ ray anisotropy. With applying the magnetization field of 2 kG either to <100> or <111> direction, the resonance was sought by sweeping the radio frequency between 220-310 MHz for ^{57}Co and 120-190 MHz for ^{60}Co. The step of the sweep is either 1 MHz or 0.5 MHz with the modulation frequency of 100 Hz. The resonance was detected by the destruction of the γ ray (133 keV for ^{57}Co or 1.17 and 1.33 MeV for ^{60}Co) anisotropy which was monitored by four Ge detectors placed at 0, 90, 180 and 270 degree relative to the magnetization direction. The difference of the anisotropy between the frequency modulation on and off will be plotted as the NMR/ON spectrum in the followings.

3. Results and Discussion

Figure 1 shows an example of the results for ^{60}Co. Here the results of the runs with the 0.5 MHz step are shown.

In Fig. 1 for ^{60}Co, two large resonances are present, one at 166 MHz (R1) and the other at 151 MHz (R2). A similar measurement on a pure Fe single crystal shows a sharp resonance at 166 MHz. So the R1 for Fe-Si alloy should correspond to the ^{60}Co probe atoms without Si atoms in the 1st neighbor. The R2 should correspond to the ^{60}Co probe atoms with one Si atom in the 1st neighbor site. This is because the reduction of (151-166)/166 = 8.5 % of Hf is comparable with the value for a satellite corresponding to the ^{57}Fe with one Si atom in the 1st neighbor site in Mössbauer spectra of Fe-Si alloys.[2] Also the half width of both resonance lines are several times larger than that of pure Fe. This indicates several other ^{60}Co feeling different Hf also contribute to these two lines. Adding to the two lines, a shoulder is present at about 162 MHz (R3), -2.5 % reduction in Hf. This one is considered to be due to the ^{60}Co atom with either a Si atom at the 2nd or the 3rd

neighbor.

The result for ^{57}Co is almost similar with that of ^{60}Co in the features described above. The difference in the gyromagnetic ratio μ/I where μ is the nuclear magnetic moment and I is the spin, between the two probe nuclei shifts the ^{57}Co spectrum to 1.77 times higher frequency than that of ^{60}Co. A better resolution of possible fine structure in each line is expected for ^{57}Co spectrum because of its presence at higher frequency.

The effect of the magnetization direction, which is one of the main purpose of the present experiment, will be described next. For ^{60}Co, the width of R1 is broader for the <100> magnetization than the <111>. On the other hand R2 shows the reverse trend. This behavior is in accord with the expected ones from the dipolar effect of Si atoms which has been concluded as the cause of the resonance line width change in Mössbauer spectrum for Fe-Si alloy single crystal.[3] Namely for the R2, the 8 Si site in the first neighbor split into 2 and 6 in energetically different sites for <111> field thus giving rise to the broadening. For R2, the contribution of the Si atom in the 2nd neighbor (<100> defect) overlaps to it, namely without Hf reduction thus giving rise to the dipolar contribution in <100> field but not in <111> field. If this interpretation of R1, R2 is correct the R3 is most likely as due to the Si atom in the 3rd neighbor site. This interpretation is in accord with the ones proposed by Cranshaw et al.[5] based on their Mössbauer spectroscopy results or several others for pulsed NMR results for Fe alloys.[6]

More detailed fine structures are present in a ^{57}Co result as expected. Namely, the R1 width is almost independent on the magnetization direction and yet R2 and R3 show different fine structures with the different magnetization direction. Namely R2 splits into three sub-resonances for <111> field from two for <100> field. The R3 splits into two for <111> field from one for <100> field. Before discussing the origin of these possible fine structures, more experiments will be necessary to examine their reproducibility. Further experiments are now in progress.

Part of this work has been reported at the VIIIth international conference on hyperfine interactions (Prague, Aug. 1989).

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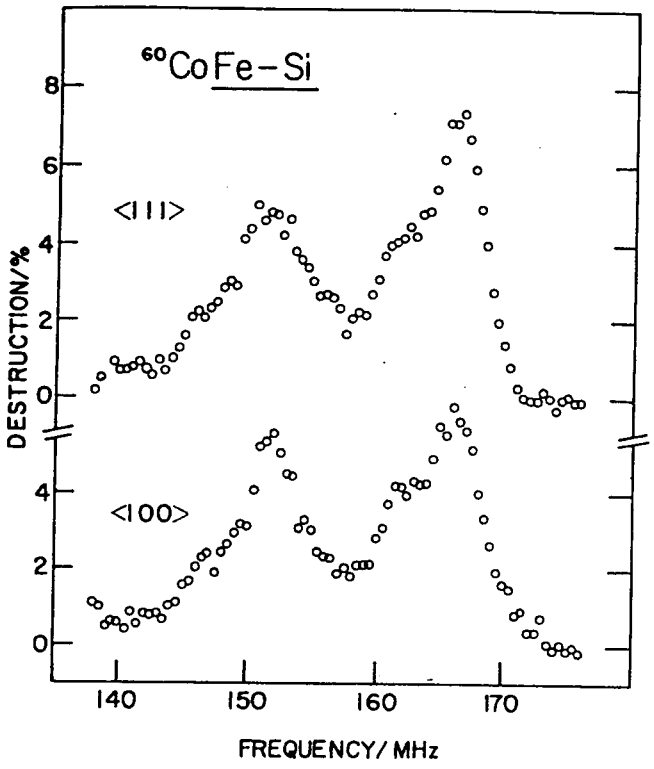

Fig. 1. NMR/ON spectrum for a Fe-Si (6 at %) single crystal alloy. ^{60}Co is used as the probe nuclei. Upper: Magnetization along <111>. Lower: along <100>. 0.5 MHz step sweep.