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MÖSSBAUER EFFECT OF ^{61}Ni IN SPINEL TYPE MAGNETIC OXIDES

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Résumé. — La spectroscopie Mössbauer appliquée au ^{61}Ni a été utilisée pour étudier les champs magnétiques internes d'oxydes contenant du Nickel, de structure spinelle ou spinelle déformée. Les champs magnétiques internes dans des oxydes contenant des ions de Nickel en site octaédrique, ont été trouvés de l'ordre de 70 à 100 kOe. Dans NiCr_2O_4 , où les ions de Nickel se trouvent sur des sites tétraédriques, nous avons mesuré un champ magnétique interne voisin de 450 kOe à la température de l'Hélium liquide. Cette valeur semble être la plus élevée jamais trouvée pour le ^{61}Ni .

Abstract. — The Mössbauer spectroscopy of ^{61}Ni was effectively utilized to investigate the internal magnetic fields in Ni-containing oxides with spinel or distorted spinel structure. The internal magnetic fields in oxides with the nickel ions in octahedral site were observed to be 70-100 kOe. In NiCr_2O_4 , in which nickel ions are in tetrahedral site, an internal magnetic field amounting to 450 kOe was observed at liquid He temperature. This value seems to be the largest one ever observed for ^{61}Ni .

I. Introduction. — In spite of many experimentally undesirable nuclear properties, ^{61}Ni can be a powerful probe in solid state physics as the only Mössbauer nuclide other than ^{57}Fe in the iron group transition elements [1, 2, 3, 4], especially in the studies of internal magnetic fields.

The purpose of the present paper is to discuss the Mössbauer effect of ^{61}Ni in various Ni-containing oxides with spinel (or distorted spinel) structure.

II. Experimental. — The nuclear reactions,

and $^{58}\text{Ni}(\alpha, n)^{61}\text{Zn} \xrightarrow{\beta^+, \text{EC}} ^{61}\text{Cu}$ were utilized to produce the source nuclide ^{61}Cu . A thin Ni-V (84-16) alloy disc of about 20 mm in diameter was irradiated for several hours with 21 MeV α -particles from the 160 cm IPCR Cyclotron of this Institute, and was used as the source without annealing. The Ni-V (84-16) alloy crystallizes in f. c. c. structure and paramagnetic in nature. The specimens of various oxides were prepared by the convenient ceramic method, using nickel enriched with ^{61}Ni up to 80-90 %. An NaI(Tl) scintillation counter was employed to detect the transmitted radiation. The Mössbauer spectrometer made by Elron or the one by Hitachi was operated in constant acceleration mode with a 400 channel pulse height analyzer.

Because of the low resolution of the ^{61}Ni Mössbauer spectra, computer fitting of them to theoretical curves was an indispensable procedure to extract various parameters from the obtained spectra. Concerning the quadrupole moment of the excited state of ^{61}Ni , the rough estimation of -2 for the ratio $Q_{\text{ex}}/Q_{\text{gr}}$ due to Erich [3], had been adopted tentatively in the analysis. Fortunately, the quadrupolar interactions in the spinels investigated turned out to be almost negligible compared with the magnetic interactions, except for a few cases mentioned in the text. The values of the isomer shift of the obtained spectra were also found to be of the order of the experimental uncertainty. Consequently, only the internal field is discussed in this paper, and the numerical values of the quadrupolar interaction and the isomer shift will not be presented.

III. Results and discussion. — The two groups of oxides with spinel (or distorted spinel) structure were

investigated: the first group with the nickel ions in the octahedral site (NiFe_2O_4 , NiCo_2O_4 , NiMn_2O_4 and GeNi_2O_4), and the second group with the nickel ions in the tetrahedral site (NiCr_2O_4 and NiRh_2O_4).

1. THE SPINELS WITH NICKEL IONS IN THE OCTAHEDRAL SITE. — NiFe_2O_4 : The Mössbauer spectrum at 80 °K is shown in figure 1. It is an almost symmetric pattern.

FIG. 1. — The ^{61}Ni -Mössbauer spectra of NiFe_2O_4 , NiCo_2O_4 , and NiMn_2O_4 .

with noticeable peaks on the top, characteristic to a case with appreciable internal magnetic field H_i and negligible electric field gradient (EFG) effect. The value of H_i was found to be 94 kOe. This coincided with the value reported in [1], but the appreciable EFG effect reported therein had not been observed.

NiCo_2O_4 : The absorption pattern at 80 °K is shown in figure 1. The values of H_i were 73 kOe and 87 kOe at 80 °K and at liquid He temperature, respectively.

NiMn_2O_4 : According to the recent report by Boucher et al. [5], this spinel is incompletely inverse depending on the heat treatment. The fitting was made under the assumption of completely inverted ionic distribution, but the fitting at liquid He temperature was not very unsatisfactory with H_i of 97 kOe as can be seen in figure 1, and at 80 °K the best fit was obtained with H_i of 72 kOe.

GeNi_2O_4 : The pattern at 80 °K, which is far above its reported Néel temperature of 16 °K, was slightly asymmetric suggesting that the EFG effect is working.

2. THE SPINELS WITH NICKEL IONS IN TETRAHEDRAL SITE. — NiCr_2O_4 : As is well known, this oxide has cubic spinel structure at temperatures higher than 310 °K, below which it becomes tetragonal due to the Jahn-Teller effect of the Ni^{2+} ions in the tetrahedral sites. The ferrimagnetic Curie temperature is reported to be about 80 °K. According to Prince [6], the spin structure investigated by neutron diffraction is very complicated but the detailed spin structure is still unknown. At 80 °K, the spectrum seems to be slightly asymmetric with a width wider than the single line cases, as can be seen in the upper part of figure 2.

The most remarkable feature of this spinel is the widening of the pattern on cooling down to the liquid He temperature, as shown in the lower part of the same figure. The value of the internal magnetic field determined by the fitting amounted to 450 kOe, which seems to be the largest one ever reported for ^{61}Ni . Without data with an applied external magnetic field, detailed discussion of the possible origin of such a large field should be premature. However, contribution from the only incompletely quenched orbital moment of the Jahn-Teller distorted Ni^{2+} ions might be a possible mechanism to explain it.

NiRh_2O_4 : The transition temperature from cubic to tetragonal symmetry is 380 °K, and the antiferromagnetic Néel temperature is reported to be 18 °K. The spectrum taken at 80 °K showed a noticeable asymmetry.

FIG. 2. — The ^{61}Ni -Mössbauer spectra of NiCr_2O_4 .

IV. Conclusion. — A striking difference in magnitude of the internal magnetic field at ^{61}Ni nuclei has been observed between the spinel oxides with nickel ions in the octahedral site and NiCr_2O_4 with its nickel ions in the tetrahedral site. The former shows an internal magnetic field of the order of 70-100 kOe and a negligible EFG effect. The latter shows an internal magnetic field as high as 450 kOe, the origin of which is not yet established.

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