Field-induced-moment nuclear Coupling for $^{59}$Co in a Heusler alloy Co$_2$TiGa

Y Furutani$^1$, H Nishihara$^1$, T Kanomata$^2$, K Kobayashi$^3$, R Kainuma$^4$, K Ishida$^3$, K Koyama$^5$, K Watanabe$^6$, T Goto$^6$

$^1$Faculty of Science and Technology, Ryukoku University, Otsu 520-2194, Japan
$^2$Faculty of Engineering, Tohoku Gakuin University, Tagajo 985-8537, Japan
$^3$Graduate School of Engineering, Tohoku University, Sendai 980-8579, Japan
$^4$Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Sendai 980-8577, Japan
$^5$Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
$^6$Faculty of Science and Technology, Sophia University, Tokyo 102-8554, Japan

E-mail: nishihara@rins.ryukoku.ac.jp

Abstract. The positive hyperfine field at Co nucleus in a Heusler alloy Co$_2$TiGa is discussed to be not dominated by the orbital hyperfine field from a quite different point of view from former studies. Field-induced-moment nuclear coupling for $^{59}$Co via spin-orbit interaction on Co atom in Co$_2$TiGa is discussed to be small from the observed high-field shift of the NMR of $^{59}$Co in the ferromagnetic state of only +0.83 % in contrast to the case of CoCl$_2$.2H$_2$O with +29%.

1. Introduction

Heusler-type ferromagnetic alloys have received renewed interests recently in relation to half-metallic ferromagnets, ferromagnetic shape memory alloys, or materials with large thermo-electric powers. One of them, Co$_2$TiGa is a ferromagnet with a Curie temperature of 130 K and a magnetic moment of 0.4 $\mu_B$ per cobalt atom [1]. Recent report suggests that the magnetic properties of Co$_2$TiGa are well described by the spin fluctuation theory for itinerant electron magnetism and the electronic state of Co$_2$TiGa lies between that of
very weak itinerant electron ferromagnetic limit such as Ni$_3$Al and that of the intermediate case such as Fe or Ni [2, 3]. Phase stability of B2 and L2$_1$ ordered phases in Co$_2$TiGa was reported [4]. In view of the hyperfine interaction, the most remarkable feature of the cobalt-based Heusler alloys is the positive hyperfine field at Co nucleus where the field is the same direction as the magnetization [5-9]. These positive fields are in contrast to the negative hyperfine fields observed in compounds or alloys of 3d transition atoms in which a typical value is a field of -100 kOe per 1 $\mu_B$ of the moment [10]. In an earlier stage, the orbital contribution was suggested to be dominant [7], while the contribution due to the polarization of 4s conduction electrons was discussed to be dominant deducing from the experimental result that the hyperfine field was proportional to the magnetic moment in the wide temperature range including both the ferromagnetic and the paramagnetic state in Co$_2$TiAl [9]. However, it is to be noted that the expectation value of the orbital angular momentum could have temperature dependence if low-lying excited orbital states exist, which induces temperature dependence of the orbital hyperfine field along with the total magnetic moment. Band structure calculations by the local-spin-density method reproduce the experimental hyperfine fields in semi-quantitative manner for Co$_2$TiAl and Co$_2$TiSn where the positive contribution from the valence s-electrons is dominant [11]. The aim of the present study is to estimate the orbital contribution of the hyperfine field at $^{59}$Co nucleus from quite different point of view, i.e. from the field-induced moment nuclear coupling. The effective gyromagnetic ratio in the frequency-field diagram in NMR of $^{59}$Co is strongly deviated from the known ratio by the coupling between $^{59}$Co and field-induced moment in Co atom with unquenched orbital angular momentum [12-15]. It was observed firstly in the case of $^{59}$Co in antiferromagnetic CoF$_2$ and discussed in detail for the cases of paramagnetic Co(NH$_4$)$_2$(SO$_4$)$_2$.6H$_2$O and metamagnetic CoCl$_2$.2H$_2$O [12-15].

2. Experimental Results and Discussion

The method of the sample preparation and the magnetic properties of Co$_2$TiGa were described in ref. 2. The lattice parameter is $a = 0.5857$ nm, the saturation moment at 5 K is 0.41 $\mu_B$ per Co atom and the Curie temperature determined from Arrott plots is 128 K [2]. They are in good agreement with values reported [1].

NMR measurements were performed for powdered samples using conventional phase-coherent pulsed NMR spectrometers and superconducting magnets up to field of 18 T with high homogeneities. A typical example of a field-swept spin-echo spectrum of $^{59}$Co in Co$_2$TiGa taken at 150 MHz and 8 K is shown in Fig.1. The width was found to be rather small in spite of powdered samples with a half width of 1.9 kOe in Fig.1 which is 1.4 % of the resonance field in consistent with the cubic environment around Co atom. The weak satellite signals in Fig.1 are probably due to Co atoms with atomic disorder in the neighbors. Such field-swept spectra were taken at various frequencies from 10 to 170 MHz and data of peak resonance fields are summarized in a frequency-field diagram at
4.2 K as shown in Fig.2. The data are well on a straight line with a fitted slope of 1.013
MHz/kOe. This value corresponds to the positive shift of +0.83 % to the value of 1.0054
MHz/kOe for a free $^{59}$Co nucleus. This shift is to be compared with a shift of +29 % in
the case of $^{59}$Co in an insulating, metamagnetic CoCl$_2$.2H$_2$O [15]. The ground state, $^4F$ of
the free ion Co$^{2+}$ splits into $^4T_1$, $^4T_2$ and $^4A_1$ by cubic crystal field. The lowest $^4T_1$ splits into six
Kramers doublets by the combined action of the tetragonal crystal field and the
spin-orbit coupling. The ground Kramers doublet has a large unquenched orbital angular
momentum which produces positive orbital hyperfine field of +650 kOe. The magnitude
of the orbital hyperfine field in Co$_2$TiGa is roughly estimated by the ionic model to be
about +20 kOe from the data that the high-field shift in Co$_2$TiGa is only 1/35 of that in
CoCl$_2$.2H$_2$O which cannot overcome the negative contribution of -40 kOe estimated from
the moment of 0.41 $\mu_B$ per Co atom. Thus, the positive hyperfine field at Co nucleus in
Co$_2$TiGa is concluded to be not dominated by the orbital hyperfine field. The transferred
hyperfine fields from neighboring atoms can also make positive contributions, but the

![Fig.1](image1.png)

Fig.1. Field-swept spin-echo spectrum of $^{59}$Co in Co$_2$TiGa at 150 MHz and 8 K.

![Fig.2](image2.png)

Fig.2. Frequency-field diagram for $^{59}$Co in Co$_2$TiGa at 4.2 K.
magnitude is considered to be less than the magnitude of the Fermi contact hyperfine field, 40 kOe, from the own atom. So, the dominant contribution to the positive hyperfine field is suggested to be due to the 4s conduction band electrons. This is consistent with the band calculations [11]. For more quantitative discussion, it will be necessary to compare the present experimental results with a band calculation including the spin-orbit interaction and induced-moment nuclear coupling.

In conclusion, the magnitude of the field-induced-moment nuclear coupling for $^{59}$Co via spin-orbit interaction on Co atom has been estimated from the observed high-field shift of the NMR of $^{59}$Co in the ferromagnetic state of a Heusler alloy Co$_2$TiGa. It is confirmed that the positive hyperfine field at Co nucleus can not be dominated by the orbital hyperfine field.

Acknowledgments
This work was partially supported by a grant based on the High-Tech Research Centre Programme for private universities from the Japan Ministry of Education, Culture, Sports, Science and Technology. This work was also supported in part by a research programme at the Institute for Materials Research, Tohoku University. One of the authors (H. N.) was helped by a sabbatical system at Ryukoku University.

References
[1] Webster P J and Ziebeck K R A 1973 J. Phys. Chem. Solids 34 1647
[2] Sasaki T, Kanomata T, Narita T, Nishihara H, Note R, Yoshida H and Kaneko T 2001 J. Alloys and Compounds 317-318 406
[3] Takahashi Y 1986 J. Phys. Soc. Jpn. 55 3553
[4] Kobayashi K, Ishikawa K, Umetsu R Y, Kainuma R, Aoki K and Ishida K 2007 J. Mag. Mag. Mater. 310 1794
[5] Endo K, Shinogi A and Vincze I 1976 J. Phys. Soc. Jpn. 40 674
[6] Vijayaraghavan R, Grover A K, Gupta L C, Nagarajan V, Itoh J, Shimizu K and Mizutani H 1977 J. Phys. Soc. Jpn. 42 1779
[7] Khi Le Dang, Veillet P and Campbell I A 1978 J. Phys. F: Metal Phys. 8 1811
[8] Yoshimura K, Miyazaki A, Vijayaraghavan R and Nakamura Y 1985 J. Mag. Mag. Mater. 53 189
[9] Shinogi A 1985 J. Phys. Soc. Jpn. 54 400
[10] Portis A M and Lindquist R H 1965 Magnetism, ed. Rado G T and Suhl H (Academic Press, New York) Vol. IIA 357
[11] Ishida S, Asano S and Ishida J 1984 J. Phys. Soc. Jpn. 53 2718
[12] Vincze J 1959 Phys. Rev. Letters 2 163
[13] Moriya T 1959 J. Phys. Chem. Solids 11 175
[14] Ho Choh Sung and Seidel G 1968 Phys. Rev. 174 385
[15] Nishihara H, Yasuoka H and Hirai A 1972 J. Phys. Soc. Jpn. 32 1135