

## LABORATORY OF SOLID STATE CHEMISTRY

Head: Dr. Toshio Takada

Under the supervision of Professor T. Takada, various inorganic materials have been synthesized and the magnetic properties have been studied. The abstracts of major research are as the following.

### I. Preparation of Transition Metal Oxides from Aqueous Solution

Iron oxides and hydroxides were formed in aqueous suspension media. The precipitates of ferromagnetic spinel ferrites,  $M_xFe_{3-x}O_4$  ( $M=Mg, Co, Mn, Zn\dots$ ) and barium ferrites were also prepared in aqueous solutions. The precipitated particles were crystallographically studied and their magnetic properties were elucidated. Recently the technique for the ferrite formation was proved to be very useful for excluding heavy metal ions from waste water.

### II. Crystal Growth from Vapor Phase

Single crystals of ferrites, vanadium oxides, titanium oxides and some other oxides were grown by close-tube chemical transport method and their magnetic and electric properties were clarified. In order to study the mechanism of crystal growth from vapor phase, molecular species in the vapor phase were examined by a devised mass-spectrometer.

### III. Mössbauer Effect Studies

Mössbauer effect has been utilized to elucidate the magnetic properties of many compounds and alloys. The electronic properties and magnetic ordering were studied in some  $Fe^{6+}$  compounds and the spin structures were estimated in  $KFe_3(OH)_6(SO_4)_2$ ,  $Fe(OH)_2$ , and  $Fe_3(PO_4)_2 \cdot 8H_2O$ . The cation distribution in ferrites was determined by applying an external field. The magnetic properties of ferromagnetic metal surface were studied by using  $Fe^{57}$  emission spectroscopy and also with vacuum evaporated thin films. Very low temperatures, such as 80 mK, were realized by a  $He^3$ -dilution refrigerator and a nuclear polarization of  $Co^{57}$  was observed in the Mössbauer emission spectrum.

### IV. Magnetochemical Study of Organic Magnetic Materials (Activities of Professor H. Takaki's Group)

Professor H. Takaki and his coworkers have studied the magnetic properties of organic magnetic materials. Organic magnetic materials have some unpaired electrons in a molecule and exhibit magnetic properties like inorganic transition-metal com-

pounds. In order to study the magnetic behaviors and the mechanism of phase transition, the measurements of magnetic susceptibility, magnetic heat content, electron spin resonance (ESR) and nuclear magnetic resonance (NMR) have been carried out. The electron-nuclear double resonance (ENDOR) was also utilized to study the dynamical properties of the unpaired electrons. Published papers on these studies are included in the last part of the following publication list. Professor H. Takaki was retired from Kyoto University in 1973.

The low temperature laboratory was built in 1970 for the convenience of experiments at liquid helium temperature. A helium liquifier (Philips Phe 210) is equipped and liquid helium is always stored in a 500 l storage vessel (Cryenco). The evaporated helium gas is collected through the piping system and compressed and purified by an apparatus (Osaka-Sanso OHE-30). Some measurements like Mössbauer effect and magnetic susceptibility are routinely made at helium temperature region. Much lower temperatures than liquid helium are available by using a He<sup>3</sup>-dilution refrigerator.

### Publications

(\* indicates an article published in Japanese)

1. H. Asano, Y. Bando, N. Nakanishi, and S. Kachi: Order-Disorder Transformation of Fe-Co Alloys in Fine Particles, *Trans. Japan Inst. Metals*, **8**, 180 (1967).
2. T. Takada: The Preparation and Physical Properties of Fine Particles, *Bussei*, **8**, 525 (1967).\*
3. T. Oda, T. Takada, and S. Kachi: Studies on Sintering Mechanism by Autoradiography, *Funtai Oyobi Funmatsu-Yakin* (*J. Japan Soc. Powder Met.*), **14**, 118 (1967).\*
4. T. Shinjo: Mössbauer Effect Study of Magnetism in Fine Particles, *ibid.*, **14**, 248 (1967).\*
5. H. Yasuoka, A. Hirai, T. Shinjo, M. Kiyama, Y. Bando, and T. Takada, NMR Determination of Metal Ion Distribution in Manganese Ferrite Prepared from Aqueous Solution, *J. Phys. Soc. Japan*, **22**, 174 (1967).
6. Y. Bando: The Effect of Impurities on Densification of Oxide During Sintering, *Funtai Oyobi Funmatsu-Yakin* (*J. Japan Soc. Powder Met.*), **14**, 378 (1967).\*
7. S. Nasu, T. Shinjo, Y. Nakamura, and Y. Murakami: Size Distribution of Co Particles Precipitated in Cu Determined by the Mössbauer Effect, *J. Phys. Soc. Japan*, **23**, 664 (1967).
8. H. Miyamoto, T. Shinjo, Y. Bando, and T. Takada: Mössbauer Effect of Fe<sup>57</sup> in Fe(OH)<sub>2</sub>, *ibid.*, **23**, 1421 (1967).
9. J. Sakurai and T. Shinjo: Neutron Diffraction of Manganese Ferrite Prepared from Aqueous Solution, *ibid.*, **23**, 1426 (1967).
10. N. Yamamoto: The Shift of the Spin Flip Temperature of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> Fine Particles, *ibid.*, **24**, 23 (1968).
11. K. Nagasawa, Y. Bando, and T. Takada: Epitaxial Growth of Nickel Ferrite on (111) and (110) Surface of MgO, *Japan. J. Appl. Phys.*, **7**, 174 (1968).
12. M. Takano, T. Shinjo, M. Kiyama, and T. Takada: Magnetic Properties of Jarosites, *J. Phys. Soc. Japan*, **25**, 902 (1968).
13. T. Shinjo: Introduction to Mössbauer Spectroscopy, *Hyomen*, **6**, 732 (1968).\*
14. S. Nasu, Y. Murakami, Y. Nakamura, and T. Shinjo: Mössbauer Effect Study in Cu-2%Co Alloy: The Transformation of Precipitates Induced by Cold Rolling, *Scripta Met.*, **2**, 647 (1968).
15. N. Yamamoto, T. Shinjo, M. Kiyama, Y. Bando, and T. Takada: Mössbauer Effect Study of  $\alpha$ -FeOOH and  $\beta$ -FeOOH: Making Use of Oriented Particles, *J. Phys. Soc. Japan*, **25**, 1267 (1968).
16. T. Shinjo, T. Ichida, and T. Takada: Internal Magnetic Field at Fe<sup>57</sup> in Hexavalent States, *ibid.*, **26**, 1547 (1969).
17. T. Takada, Y. Bando, N. Yamamoto, and K. Nagasawa: Growth of Co<sub>3</sub>O<sub>4</sub> Single Crystals and Its

- Reaction Mechanism, *Japan. J. Appl. Phys.*, **8**, 619 (1969).
18. Y. Bando, K. Nagasawa, Y. Kato, and T. Takada: Growth of VO<sub>2</sub> Single Crystals by Chemical Transport Reaction, *ibid.*, **8**, 633 (1969).
  19. Y. Bando: The Role of Impurity in Sintering of Oxides, *Funtai Oyobi Funmatsu-Yakin (J. Japan. Soc. Powder Met.)*, **15**, 414 (1969).\*
  20. T. Shinjo, T. Takada, and N. Tamagawa: Mössbauer Effect of Fe<sup>57</sup> in CrO<sub>2</sub>, *J. Phys. Soc. Japan*, **26**, 1404 (1969).
  21. Y. Endoh, Y. Ishikawa, and T. Shinjo: Iron Impurity in the Antiferromagnetic Manganese Copper Alloy, *Phys. Lett.*, **29A**, 310 (1969).
  22. T. Shinjo: Application of Mössbauer Effect, *Funtai Oyobi Funmatsu-Yakin (J. Japan. Soc. Powder Met.)*, **15**, 7 (1969).\*
  23. K. Nagasawa, Y. Bando, and T. Takada: Growth of V<sub>4</sub>O<sub>7</sub> and V<sub>7</sub>O<sub>13</sub> Single Crystals, *Japan. J. Appl. Phys.*, **8**, 1262 (1969).
  24. K. Nagasawa, Y. Bando, and T. Takada: Growth of V<sub>3</sub>O<sub>5</sub> and V<sub>6</sub>O<sub>11</sub> Single Crystals, *ibid.*, **8**, 1267 (1969).
  25. H. Okinaka, K. Nagasawa, K. Kosuge, Y. Bando, T. Takada, and S. Kachi: Electrical Properties of the V<sub>3</sub>O<sub>5</sub> Single Crystals, *J. Phys. Soc. Japan*, **27**, 1366 (1969).
  26. S. Nasu, M. Nishio, Y. Tsuchida, Y. Murakami, and T. Shinjo: Temperature Shift of the Fe<sup>57</sup> Mössbauer Line in the Quenched Al-0.01% Fe Alloy, *ibid.*, **27**, 1363 (1969).
  27. T. Takada: Formation and Physical Properties of Iron Oxides and Hydroxides, *Denki-Kagaku*, **37**, 328 (1969).\*
  28. M. Kiyama: Commentary Experiments on the Formation of Fe<sub>3</sub>O<sub>4</sub> Precipitated from Aqueous Solution, *Bull. Inst. Chem. Res., Kyoto Univ.*, **47**, 607 (1969).
  29. K. Nagasawa, Y. Bando, and T. Takada: Growth of V<sub>5</sub>O<sub>9</sub> Single Crystals, *Japan. J. Appl. Phys.*, **9**, 407 (1970).
  30. H. Okinaka, K. Nagasawa, K. Kosuge, Y. Bando, S. Kachi, and T. Takada: Electrical Properties on the V<sub>4</sub>O<sub>7</sub> Single Crystals, *J. Phys. Soc. Japan*, **28**, 798 (1970).
  31. M. Takano and T. Takada: Magnetic Properties of MTa<sub>2</sub>O<sub>6</sub>(M=Fe, Co or Ni), *Mat. Res. Bull.*, **5**, 449 (1970).
  32. H. Okinaka, K. Nagasawa, K. Kosuge, Y. Bando, S. Kachi, and T. Takada: Electrical Properties on the V<sub>5</sub>O<sub>9</sub> Single Crystals, *J. Phys. Soc. Japan*, **28**, 803 (1970).
  33. H. Okinaka, K. Nagasawa, K. Kosuge, Y. Bando, S. Kachi, and T. Takada: Electrical Properties of V<sub>6</sub>O<sub>11</sub> and V<sub>7</sub>O<sub>13</sub> Single Crystals, *ibid.*, **29**, 245 (1970).
  34. T. Shinjo, T. Ichida, and T. Takada: Fe<sup>57</sup> Mössbauer Effect and Magnetic Susceptibility of Hexavalent Iron Compounds, *ibid.*, **29**, 111 (1970).
  35. K. Nagasawa, Y. Bando, T. Takada, H. Horiuchi, M. Tokonami, and N. Morimoto: Growth and Magnetic Properties of V<sub>8</sub>O<sub>15</sub> Single Crystals, *Japan. J. Appl. Phys.*, **9**, 841 (1970).
  36. K. Kosuge, H. Okinaka, S. Kachi, K. Nagasawa, Y. Bando, and T. Takada: Electron Diffraction Study on the Shear Structure of V<sub>n</sub>O<sub>2n-1</sub>, *ibid.*, **9**, 1004 (1970).
  37. K. Nagasawa, Y. Kato, Y. Bando, and T. Takada: Properties of Ti<sub>4</sub>O<sub>7</sub> Single Crystals, *J. Phys. Soc. Japan*, **29**, 241 (1970).
  38. T. Ichida, T. Shinjo, Y. Bando, and T. Takada: Magnetic Properties of  $\alpha$ -NaFeO<sub>2</sub>, *J. Phys. Soc. Japan*, **29**, 795 (1970).
  39. T. Ichida, T. Shinjo, Y. Bando, and T. Takada: Mössbauer Effect Study of KFeO<sub>2</sub>, *ibid.*, **29**, 1109 (1970).
  40. Y. Bando, Y. Ikeda, T. Akashi, and T. Takada: Role of CaO and SiO<sub>2</sub> in Sintering of Manganese Zinc Ferrite, *Modern Development in Powder Metallurgy* **4**, 339 (1971).
  41. H. Okinaka, K. Kosuge, S. Kachi, K. Nagasawa, Y. Bando, and T. Takada: Electrical Properties of V<sub>8</sub>O<sub>15</sub> Single Crystals, *Phys. Lett.*, **33A**, 370 (1971).
  42. K. Nagasawa, Y. Bando, and T. Takada: The Chemical Transport Reaction of Spinel Type Oxides, *Bull. Chem. Soc. Japan*, **44**, 1577 (1971).
  43. S. Muranaka, T. Shinjo, Y. Bando, and T. Takada: Mössbauer Study of Fe<sub>2</sub>TiO<sub>5</sub> and FeTi<sub>2</sub>O<sub>5</sub>, *J. Phys. Soc. Japan*, **30**, 890 (1971).
  44. M. Takano, T. Shinjo, and T. Takada: On the Spin Arrangement in Kagome Lattice of Anti-ferromagnetic KFe<sub>5</sub>(OH)<sub>6</sub>(SO<sub>4</sub>)<sub>2</sub>, *ibid.*, **30**, 1049 (1971).

45. T. Takada, Y. Bando, M. Kiyama, and T. Shinjo: Ion Distribution of Manganese Ferrite Prepared by Wet Method, *Proc. Int'l Conf. Ferrite*, 29 (1971).
46. T. Takada and M. Kiyama: Preparation of Ferrites by Wet Method, *ibid.*, 69 (1971).
47. T. Takada, Y. Ikeda, H. Yoshinaga, and Y. Bando: A New Preparation Method of the Oriented Ferrite Magnets, *ibid.*, 275 (1971).
48. S. Kachi, T. Takada, Y. Bando, K. Kosuge, H. Okinaka, and K. Nagasawa: Metal Insulator Phase Transition in  $V_nO_{2n-1}$ , *ibid.*, 563 (1971).
49. T. Shinjo, T. Matsuzawa, T. Takada, S. Nasu, and Y. Murakami: Mössbauer Evidence Against the Existence of Magnetically Dead Layers, *Phys. Lett.*, **36A**, 489 (1971).
50. T. Shinjo and H. Forstat: Further Mössbauer Studies of Vivianite,  $Fe_3(PO_4)_2 \cdot 8H_2O$ , *J. Phys. Soc. Japan*, **31**, 1399 (1971).
51. K. Nagasawa, Y. Bando, and T. Takada: Crystallography of  $V_nO_{2n-1}$  ( $3 \leq n \leq 8$ ), *Mat. Res. Bull.*, **6**, 833 (1971).
52. K. Nagasawa: Crystal Growth of  $V_nO_{2n-1}$  ( $3 \leq n \leq 8$ ) by the Chemical Transport Reaction and Electrical Properties, *ibid.*, **6**, 853 (1971).
53. K. Iwauchi, S. Yamamoto, Y. Bando, T. Hanai, N. Koizumi, T. Takada, and S. Fukushima: Dielectric Properties of the Mixtures of  $\alpha$ - $Fe_2O_3$ ,  $TiO_2$ , and  $Fe_2TiO_5$ , *Japan. J. Appl. Phys.*, **10**, 1513 (1971).
54. T. Shinjo: Introduction to Mössbauer Spectroscopy, *Kagaku (Chemistry)*, **27**, 952 (1971).\*
55. H. Nishihara, T. Tsuda, A. Hirai, and T. Shinjo: Nuclear Magnetic Resonance of  $Cr^{53}$  in Ferromagnetic  $CrO_2$ , *J. Phys. Soc. Japan*, **32**, 85 (1972).
56. M. Kiyama and T. Takada: Iron Compounds Formed by the Aerial Oxidation of Ferrous Solutions, *Bull. Chem. Soc. Japan*, **45**, 1923 (1971).
57. M. Kiyama, T. Akita, Y. Tsutsumi, and T. Takada: Formation of Titanic Oxides of Anatase, Brookite and Rutile Types by Aerial Oxidation of Titanous Solutions, *Chem. Lett.*, 21, (1972).
58. M. Kiyama, T. Akita, S. Shimizu, Y. Okuda, and T. Takada: Conditions Favorable for the Formation of  $\gamma$ - $FeOOH$  by Aerial Oxidation in an Acidic Suspension of Iron Metal Powder, *Bull. Chem. Soc. Japan*, **45**, 3422 (1972).
59. H. Okinaka, K. Kosuge, S. Kachi, T. Takano, and T. Takada: Mössbauer Effect Study of  $Fe^{57}$  Doped in the  $V_nO_{2n-1}$  System, *J. Phys. Soc. Japan*, **32**, 1148 (1972).
60. M. Takano: Mössbauer Study on the Magnetism of Trinuclear Complex Salts, *ibid.*, **33**, 1313 (1972).
61. N. Yamamoto, K. Nagasawa, Y. Bando, and T. Takada: Growth of  $Mn_3O_4$  Single Crystals by Chemical Transport Method, *Japan. J. Appl. Phys.*, **11**, 1754 (1972).
62. T. Shinjo: Mössbauer Spectroscopy, *Saishin no Bunseki Kagaku (Recent Analytical Chemistry)*, **23**, 151 (1972).\*
63. Y. Bando: Topotaxy in Solid State Reaction, *Kagaku (Chemistry)*, **27**, 995 (1972).\*
64. K. Nagasawa: Crystal Growth of Vanadium Oxides by Chemical Transport, *J. Cryst. Growth*, **17**, 143 (1972).
65. T. Ichida: Mössbauer Study of the Thermal Decomposition Products of  $K_2FeO_4$ , *Bull. Chem. Soc. Japan*, **46**, 79 (1973).
66. M. Kiyama, N. Jikuhara, and T. Takada: The Effect of the Addition of  $\gamma$ - $FeOOH$  Nuclei at the Initial Stage of Oxidation on the Formation of  $\gamma$ - $FeOOH$ , *Bull. Chem. Soc. Japan*, **46**, 323 (1973).
67. T. Ichida: Mössbauer Study of the Thermal Decomposition Products of  $SrFeO_4$ , *ibid.*, **46**, 1591 (1973).
68. M. Kiyama and T. Takada: The Hydrolysis of Ferrite Complexes. Magnetic and Spectrophotometric Studies of Aqueous Solutions of Ferric Salts, *Bull. Chem. Soc. Japan*, **46**, 1680 (1973).
69. M. Takano, T. Shinjo, M. Kiyama, and T. Takada: A Mössbauer Study of Magnetic Hyperfine Fields in  $Fe^{3+}$ -Antiferromagnetic Oxides, *J. Phys. Soc. Japan*, **35**, 53 (1973).
70. F. Kanamaru, M. Shimada, M. Koizumi, M. Takano, and T. Takada: Mössbauer Effect of  $FeOCl$ -Pyridine Complex, *J. Solid State Chem.* **7**, 297 (1973).
71. S. Muranaka: Magnetic Properties of  $FeTi_2S_4$ , *J. Phys. Soc. Japan*, **35**, 616 (1973).
72. S. Muranaka: Order-Disorder Transition of Vacancies in  $FeTi_2S_4$ , *Mat. Res. Bull.*, **8**, 679 (1973).
73. T. Shinjo, T. Matsuzawa, T. Takada, S. Nasu, and Y. Murakami: Surface State of Ferromagnetic Cobalt Metal by Mössbauer Spectroscopy, *J. Phys. Soc. Japan*, **35**, 1032 (1973).

74. K. Okada and T. Shinjo: A Simple He<sup>3</sup>-Dilution Refrigerator for Mössbauer Spectroscopy, *Japan. J. Appl. Phys.*, **12**, 1281 (1973).
75. Y. Bando: Preparation of Oxides by Chemical Vapor Deposition, *Erekutoronikku Seramikkus (Electronic Ceramics)*, **4**, no. 3, 29. (1973).\*
76. T. Bando and T. Takada: Role of Grain Boundary in Sintering and Electrical Properties of Ceramics, *Electroceramics*, **4**, no. 11, 24 (1973).\*
77. T. Shinjo: Mössbauer Effect and Electronic State, *Kagaku (Chemistry)*, **29**, 139 (1973).\*
78. S. Muranaka and T. Takada: Growth and Electronic Properties of FeMe<sub>2</sub>X<sub>4</sub> (Me = Ti or V; X = S or Se) Single Crystals, *Bull. Inst. Chem. Res., Kyoto Univ.*, **51**, 287 (1973).
79. T. Ichida, Y. Bando, T. Shinjo, and T. Takada: Mössbauer Study of Some Barium Orthoferrates, *ibid.*, **51**, 295 (1973).
80. N. Yamamoto, Y. Bando, and T. Takada: Single Crystal Growth of CuO by Chemical Transport Method, *Japan. J. Appl. Phys.*, **12**, 1115 (1973).
81. N. Yamamoto, M. Kiyama, and T. Takada: A New Preparation Method of Mn<sub>5</sub>O<sub>8</sub>, *ibid.*, **12**, 1827 (1973).
82. N. Yamamoto, S. Kawano, N. Achiwa, M. Kiyama, and T. Takada: New Preparation Method and Metal Ion Distribution in CoMn<sub>2</sub>O<sub>4</sub>, *ibid.*, **12**, 1830 (1973).
83. N. Yamamoto, T. Endo, M. Shimada, and T. Takada: Single Crystal Growth of  $\alpha$ -MnO<sub>2</sub>, *ibid.*, **13**, 723 (1974).
84. T. Ichida: Mössbauer Study of the Thermal Decomposition Products of BaFeO<sub>4</sub>, *J. Solid State Chem.*, **7**, 308 (1973).
85. T. Shinjo, T. Matsuzawa, T. Mizutani, and T. Takada: Mössbauer Study of Ferromagnetic Metal Surface, *Japan. J. Appl. Phys.* **13 sup. 2**, pt. 2, 729 (1974).
86. K. Okada and T. Shinjo: The Magnetic Hyperfine Field at Co<sup>67</sup> in Antiferromagnetic CoO: A Mössbauer Study at Very Low Temperatures, *J. Phys. Soc. Japan*, **36**, 1017 (1974).
87. K. Iwauchi, N. Koizumi, M. Kiyama, and Y. Bando: On the Dielectric Relaxation in Ferrites due to Electron Hopping at Low Temperatures, *Bull. Inst. Chem. Res., Kyoto Univ.*, **52**, 596 (1974).
88. M. Kiyama: Conditions for the Formation of Fe<sub>3</sub>O<sub>4</sub> by the Air Oxidation of Fe(OH)<sub>2</sub> Suspensions, *Bull. Chem. Soc. Japan*, **47**, 1646 (1974).
89. Y. Bando: Chemical Transport Reaction: *Kagaku (Chemistry)*, **30**, 152 (1974).\*
90. K. Kugimiya, E. Hirota, and Y. Bando: Magnetic Heads Made of a Crystal-Oriented Spinel Ferrite, *IEEE Trans. Magnetics*, **10**, 907 (1974).
91. T. Shinjo and K. Okada: Mössbauer Effect at mK Region, *Butsuri*, **29**, 854 (1974).\*
92. T. Takada: Topotactic Reaction of Ferrite, *Erekutoronikku Seramikkus (Electronic Ceramics)*, **6**, 53 (1975).\*
93. V. K. Garg, P. G. David, T. Matsuzawa, and T. Shinjo: Mössbauer and Magnetic Study of Mono-nuclear and Oxo-bridged Binuclear Iron (III) Complexes of 1,10-Phenanthroline and 2,2'-Bipyridine, *Bull. Chem. Soc. Japan*, **48**, 1933 (1975).
94. S. Muranaka and T. Takada: Magnetic Susceptibility Measurements of FeV<sub>2</sub>S<sub>4</sub>, FeV<sub>2</sub>Se<sub>4</sub>, and FeTi<sub>2</sub>Se<sub>4</sub>, *J. Solid State Chem.*, **14**, 291 (1975).
95. M. Takano, Y. Takeda, M. Shimada, T. Matsuzawa, T. Shinjo, and T. Takada: Mössbauer Study of Supertransferred Hyperfine Field of <sup>119</sup>Sn (Sn<sup>4+</sup>) in Ca<sub>1-x</sub> Sr<sub>x</sub>MnO<sub>3</sub>, *J. Phys. Soc. Japan*, **39**, 656 (1975).
96. Y. Bando: Heterogeneous Nucleation and Epitaxy, "Reactivity of Solids", ed. Chem. Soc. Japan, Univ. Tokyo Press, (1975) p. 73.
97. Y. Bando: Chemical Transport Reaction (I), "Reactivity of Solids", ed. Chem. Soc. Japan, Univ. Tokyo Press, (1975) p. 136.
98. T. Takada: Topotaxy and Nucleation, "Reactivity of Solids", ed. Chem. Soc. Japan, Univ. Tokyo Press, (1975) p. 79.
99. T. Ohtani, K. Kosuge, S. Kachi, M. Takano, and T. Takada: Interpretation of Mössbauer Spectra of <sup>57</sup>Fe Doped Ni<sub>1-x</sub> S, *Mat. Res. Bull.*, **10**, 709 (1975).
100. Y. Bando: Application of Mass Spectrometer to Ceramic Material Research, *Ceramics*, **11**, 53 (1976).\*
101. M. Kiyama: Chemistry of Fe(II) and (III) Hydroxides, *Funtai Oyobi Funmatsu-Yakin (J. Japan Soc. Powder Met.)*, **23**, 77 (1976).\*

102. T. Shinjo: Mössbauer Spectroscopy in Ferromagnetic Metal Surfaces, *IEEE Trans. Magnetics*, **12**, 86 (1976).
103. H. Miyamoto: The Magnetic Properties of Fe(OH)<sub>2</sub>, *Mat. Res. Bull.* **11**, 329 (1976).
104. H. Miyamoto: The Magnetic Properties of Ultrafine Particles of Ni(OH)<sub>2</sub>, *ibid.*, **11**, 599 (1976).
105. M. Kiyama, K. Murakami, T. Takada, I. Sugano, and T. Tsuji: Formation and Solubility of Basic Lead Chlorides at Different pH Values, *Chem. Lett.*, **23**, (1976).
106. M. Kiyama: Conditions for the Formation of Compounds Consisting of BaO and Fe<sub>2</sub>O<sub>3</sub> from Aqueous Suspensions, *Bull. Chem. Soc. Japan*, **49**, 1855 (1976).
107. T. Takada: Removal of Heavy Metal Ions from Waste Water by Ferritization, *Sekiyu-Gakkaishi* (*J. Japan. Petro Inst.*), **19**, 275 (1976).\*
108. T. Mizutani, T. Shinjo, and T. Takada: Mössbauer Study of Ni-Fe Hydrides, *J. Phys. Soc. Japan*, **41**, 794 (1976).

### Publications of Professor H. Takaki's Group

1. J. Yamauchi, H. Nishiguchi, K. Mukai, Y. Deguchi, and H. Takaki: Electron Spin Resonance Studies of Diphenyl Nitric Oxide Derivatives, *Bull. Chem. Soc. Japan*, **40**, 2512 (1967).
2. J. Yamauchi, T. Fujito, E. Ando, H. Nishiguchi, and Y. Deguchi: Magnetic Properties of Organic Stable Radicals. I 2,2,6,6-Tetramethyl-4-hydroxypiperidine-1-oxy, *J. Phys. Soc. Japan*, **25**, 1558 (1968).
3. T. Fujito, H. Nishiguchi, Y. Deguchi, and J. Yamauchi: Magnetic Properties of Organic Stable Radicals. II Porphyrin, *Bull. Chem. Soc. Japan*, **42**, 3334 (1969).
4. K. Watanabe, J. Yamauchi, H. Takaki, H. Nishiguchi, and Y. Deguchi: Synthesis of Stable Free Radicals: 2,2,6,6-Tetramethyl-4-Hydroxypiperidine-1-Oxy-4-Derivatives (I), *Bull. Inst. Chem. Res., Kyoto Univ.*, **48**, 88 (1970).
5. J. Yamauchi, T. Fujito, H. Nishiguchi, and Y. Deguchi: Linear Heisenberg Interaction in Stable Organic Free Radicals, *Proc. 12th Int'l. Conf. Low Temp. Phys. (Kyoto)*, 805 (1970).
6. K. Watanabe, J. Yamauchi, H. Takaki, H. Nishiguchi, and Y. Deguchi: ESR Studies of Stable Free Radicals: 2,2,6,6-Tetramethyl-4-Hydroxypiperidine-1-Oxy-4-Derivatives (II), *Bull. Inst. Chem. Res., Kyoto Univ.*, **48**, 264 (1970).
7. J. Yamauchi, T. Fujito, A. Nakajima, H. O.-Nishiguchi, and Y. Deguchi: Dipolar Interaction in TEMPAD Biradical, *Bull. Chem. Soc. Japan*, **44**, 2263 (1971).
8. J. Yamauchi: Linear Antiferromagnetic Interaction in Organic Free Radicals, *ibid.*, **44**, 2301 (1971).
9. O. Takizawa, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: Magnetic Properties of Organic Stable Radicals. III Diphenyl Nitric Oxide, *Bull. Chem. Soc. Japan*, **44**, 3188 (1971).
10. K. Watanabe, J. Yamauchi, H. O.-Nishiguchi, Y. Deguchi, and H. Takaki: Abnormal Hyperfine Splitting in the ESR Spectra of  $\alpha, \alpha, \gamma, \gamma$ -Tetraphenylallyl-type Organic Stable Free Radicals, *ibid.*, **45**, 371 (1972).
11. J. Yamauchi, K. Adachi, and Y. Deguchi: Heat Capacity Anomaly in the Crystalline Organic Free Radical, p-Cl-BDPA, *Chem. Lett.*, 733 (1972).
12. J. Yamauchi, K. Watanabe, H. Nishiguchi, and Y. Deguchi: Magnetic Properties of 2,2,6,6-Tetramethyl-4-Hydroxypiperidine-1-Oxy-4-Derivatives, *Bull. Inst. Chem. Res., Kyoto Univ.*, **50**, 483 (1972).
13. O. Takizawa, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: ESR Studies of Stable Free Radical Pairs in the Diamagnetic Matrix Crystal, *Bull. Chem. Soc. Japan*, **46**, 1991 (1973).
14. J. Yamauchi, K. Adachi, and Y. Deguchi: Magnetic Phase Transition of Organic Free Radical p-Cl-BDPA, *J. Phys. Soc. Japan*, **35**, 443 (1973).
15. N. Azuma, J. Yamauchi, K. Mukai, H. O.-Nishiguchi, and Y. Deguchi: The Anomalous Magnetic Behavior of Symmetrical Triphenylverdazyl, *Bull. Chem. Soc. Japan*, **46**, 2728 (1973).
16. J. Yamauchi: Organic Free Radicals in Diamagnetic Matrices, *Kagaku no Ryoiki*, **27**, 1090 (1973).\*
17. K. Watanabe, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: Studies of the Equilibria between Tetraarylallyl-type Radicals and Their Dimers, *Bull. Chem. Soc. Japan*, **47**, 274 (1974).
18. K. Uchino, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: Proton Magnetic Resonance in Organic Free Radicals, BDPA-Bz and p-Cl-BDPA, *ibid.*, **47**, 285 (1974).

19. K. Watanabe, J. Yamauchi, H. O.-Nishiguchi, Y. Deguchi, and K. Ishizu: Temperature-Dependent ENDOR Spectra of  $\alpha, \alpha, \gamma$ -Bisdiphenylene- $\beta$ -phenyl Allyl Radical and Its Derivatives, *Chem. Lett.*, 489 (1974).
20. H. Fujita, S. Kako, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: The ESR Spectra of Michler's Keton Anion Radicals, *Bull. Chem. Soc. Japan*, **47**, 1541 (1974).
21. J. Yamauchi: Antiferromagnetic Resonance of Organic Free Radical. Polycrystalline 1,3-Bisdiphenylene-2-(p-chlorophenyl)-allyl, *Chem. Lett.*, 1031 (1974).
22. N. Azuma, H. O.-Nishiguchi, J. Yamauchi, K. Mukai, and Y. Deguchi: The Magnetic Properties of Verbazyl Free Radicals. VI. ESR Studies of Pairs and Triads of the Symmetrical Triphenylverbazyl Radical in the 1,3,5-Triphenylbenzene Matrix, *Bull. Chem. Soc. Japan*, **47**, 2369 (1974).
23. T. Yoshioka, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: The Chlorine Nuclear Quadrupole Resonance in an Organic Free Radical, p-Cl-BDPA, *ibid.*, **48**, 335 (1975).
24. K. Watanabe: The Intramolecular Dynamic Behavior of Tetraaryl Allyl Radicals, *ibid.*, **48**, 1732 (1975).
25. K. Watanabe, J. Yamauchi, H. O.-Nishiguchi, Y. Deguchi, and K. Ishizu: ESR and ENDOR Spectra of Organic Stable Free Radicals:  $\alpha, \alpha, \gamma, \gamma$ -Bisdiphenylene- $\beta$ -phenyl Allyl and Its Derivatives, *Bull. Inst. Chem. Res., Kyoto Univ.*, **53**, 161 (1975).
26. H. Ozaki, H. O.-Nishiguchi, and J. Yamauchi: Magnetic Anisotropy of the Antiferromagnetically Ordered Free Radical, p-Cl-BDPA, *Phys. Lett.*, **54A**, 227 (1975).
27. T. Yoshioka, K. Watanabe, and H. O.-Nishiguchi: Proton Magnetic Resonance in Some Triarylaminium Salt Free Radicals, *Bull. Chem. Soc. Japan*, **48**, 2533 (1975).
28. A. Nakajima, J. Yamauchi, H. O.-Nishiguchi, and Y. Deguchi: Dipolar Interaction in TEMPAD Biradical. II, *ibid.*, **49**, 886 (1976).