Proceedings of the VI National Conference of Neutron Scattering, Chlewiska, Poland, June 14–18, 2009

Magnetic Properties of DO_3-Type Alloys Based on Fe_3Si and Fe_3Al

G. Parzych[a], J. Jankowska-Kisielinska[b] and L. Dobrzyński[a,c]

[a]The Soltan Institute for Nuclear Studies, 05-400 Otwock-Swierk, Poland

[b]Institute of Atomic Energy, 05-400 Otwock-Swierk, Poland

[c]Faculty of Physics, University of Białystok, Lipowa 41, 15-424 Białystok, Poland

The interest in magnetic properties of DO_3-type alloys stems from their ability to exhibit dependence of magnetic moments on local environment. It is shown that both, static and dynamic properties of the alloys based on Fe_3Si and Fe_3Al require further investigations. This concerns mainly the problem of selective substitution of chromium for iron, as well as spin dynamics, the latter problem is illustrated on the example of measured spin waves in Sendust.

PACS numbers: 75.30.Ds, 75.30.Et, 78.70.Nx

1. Introduction: preferential occupation of sites

The iron alloys exhibiting DO_3-type of structure are well-known in literature. The structure itself can be described by four interpenetrating fcc lattices with the origins at sites A$(0,0,0)$, B$(1/4,1/4,1/4)$, C$(1/2,1/2,1/2)$, and D$(3/3,3/3,3/3)$. In the case of Fe_3Si and Fe_3Al iron occupies the A, B and C sites, while D-site is occupied by silicon and aluminum. From the symmetry point of view, the sites A and C are equivalent and different from the site B. The nearest environment of all sites is presented in Table. It follows from this Table that the DO_3-type of structure is ideal for studying an influence of nearest neighbors on the formation of magnetic moments at given sites.

TABLE

Nearest-neighbors to (A, C), B and D-sites in DO_3-type of structure.

n	1	2	3	4	5	6	7	8
r_n/a	0.433	0.500	0.707	0.829	0.866	1.000	1.090	1.118
(A, C)	4B 4D	6A	12A	12B 12D	8A	6A	12B 12D	24A
B	8A	6D	12B	24A	8D	6B	24A	24D
D	8A	6B	12D	24A	8B	6D	24A	24B

In early papers [1–5] it was shown that when iron is substituted for another $3d$ transition metal this metal locates preferentially in one of the two iron positions. The suggested rule was rather simple: the metals to the left from iron in periodic table of elements locate at B-sites while the ones to the right of iron — at (A, C)-sites. This was convincingly proven for $Fe_{3-x}Mn_xSi$ for $x < 0.75$ and $Fe_{3-x}V_xSi$ for $x < 1$, as well as for Co and Ni impurities at low concentrations. Rather simple mechanism of such preferential occupation was first described by Swintendick [6] who showed that in order to form magnetic moments of iron 2.2 μ_B at B-sites and 1.2 μ_B at (A, C)-sites in Fe_3Si, one needs to transfer electrons from B to (A, C) sites, thus making the B-site electron-depleted. In such situation the impurities with fewer number of electrons than iron should prefer to occupy B-sites. This preference was also found in a number of theoretical papers, including the latest ones [7–11]. Unfortunately, there are experimental facts that contradict such a simple picture. The preference of Mn to occupation of B-sites is not so clear in Fe_3Al alloys [12, 13]. In the case of chromium substituting for iron the number of experiments [14–16] showed that chromium does not show any preference in Fe_3Si while there is certain preference to occupation of B-sites in Fe_3Al. One should immediately note that the case of Fe_3Al as a matrix poses a problem. It is very difficult to achieve full DO_3-type ordering, and part of Al atoms occupies B sites, while part of iron is occupying D-sites. This little B–D disorder, however, was included in the calculations [11] and shown to be not responsible for the lack of preferential occupation of sites in the case of chromium in Fe_3Si.

The spin dynamics of the considered alloys was shown [17, 18] to be strongly dependent on the lattice parameter. The measured spin wave dispersion relations brought some unexpected results. In the case of Fe_3Si the spin waves were measured in the whole Brillouin zone, and quite detailed analysis of these data were presented in the cited paper. It was shown that the exchange integrals depend strongly on the distance. In general, they exhibit oscillating behavior (see Fig. 1) as expected for Ruderman–Kittel–Kasuya–Yosida (RKKY)-type interac-

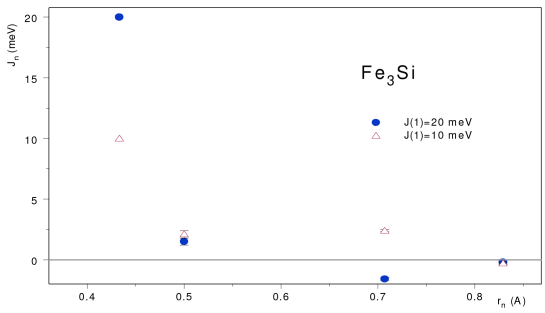

Fig. 1. Dependence of exchange integral on iron–iron distance in Fe_3Si for two limiting values of the nearest-neighbor exchange integral $J(1)$. The numerical data are taken from [18].

tions. In the case of Fe_3Al the measurements [19, 20] showed disappearance of the signal from spin wave scattering at relatively low energies. This was explained by low lying band of the Stoner excitations and raised an interest in studies of the spin dynamics of Sendust alloy with the formula $Fe_{2.96}Si_{0.68}Al_{0.38}$. In the spin-wave studies [21] carried out up to the spin wave energy of about 60 meV, the spin wave signals turned out to disappear above about 35 meV for spin wave measured along [100] direction and not along neither [110] nor [111] directions. Again, the low-lying Stoner modes could be suspected to be responsible for spin wave damping. Also a phonon–magnon interaction could be the cause, although the very weak magnetocrystalline anisotropy of Sendust makes such interpretation much less likely. In addition, the data [21] strongly indicated dependence of the spin-wave damping on the direction. Newly obtained results for the spin wave dispersion relation in Sendust along [100] direction are presented in this paper and compared with results of the paper [21].

2. Experimental

The measurements of spin wave scattering at room temperature have been carried out on TKSN-400 spectrometer installed at MARIA reactor at Świerk, Poland. The elastic and inelastic neutron scattering was measured by means of the classic 3-axis neutron spectrometer at MARIA reactor, IEA, Świerk. Pyrolytic graphite of mosaic spread 24′ was used as monochromator and analyzer. The horizontal collimations starting from the reactor core were: 44′–43′–43′–69′ and the vertical ones: 108′–83′–200′–360′. The single crystal of Sendust was same as used in the experiment [21].

The spin wave scattering was measured along [100] direction by ΔE-const technique mainly in the Brillouin zone centered at the (220) reciprocal lattice point. Only the measurements for the spin-wave energy range 6–17 meV were performed in the Brillouin zone centered at the (111) reciprocal lattice point with scattered neutron wavelength 2.35 Å. For the spin-wave energy range 20–40 meV the scattered neutron wavelength was 2.0 Å and below 1.9–1.65 eV above 40 meV.

The scattering was described by the cross-section of the form

$$F^t(\boldsymbol{q},\omega) = \frac{\Gamma_{\boldsymbol{q}}}{\hbar(\omega-\omega_{\boldsymbol{q}})^2+\Gamma_{\boldsymbol{q}}^2} - \frac{\Gamma_{\boldsymbol{q}}}{\hbar(\omega+\omega_{\boldsymbol{q}})^2+\Gamma_{\boldsymbol{q}}^2}\,. \quad (1)$$

In the data fitting it was checked that assumption of quadratic or cubic dependence of Γ_q on q resulted in similar goodness of fit and values of Γ_q. In what follows the results obtained assuming $\Gamma_q = Aq^2$ are presented.

3. Spin waves in Sendust

The measured spin wave dispersion relation is shown in Fig. 2. A typical magnon signal for the energy of 17 meV is shown in Fig. 3. The most important finding is that the magnon dispersion relation goes smoothly through the region close to 35 meV, and no anomalous behaviour is observed up to the energy 50 meV, where clear spin wave signals were observed. In the same figure the phonon dispersion relations measured by ΔQ-const technique are also shown and compared with the points known for Fe_3Si. It is seen that although the phonon branches cross the magnon branch, no anomalies connected with such crossings are seen. Therefore no hint of phonon-magnon interaction is observed, as expected. The spin-wave exchange stiffness constant calculated for [100] direction is 144.9 ± 1.8 meV Å^2 in rather good agreement with the data published in Ref. [21], 158.3 ± 1.0 meV Å^2. The difference between the values may arise from different form of the spectral function which in the case of paper [21] was taken as a sum of two Lorentzian peaks instead of their difference.

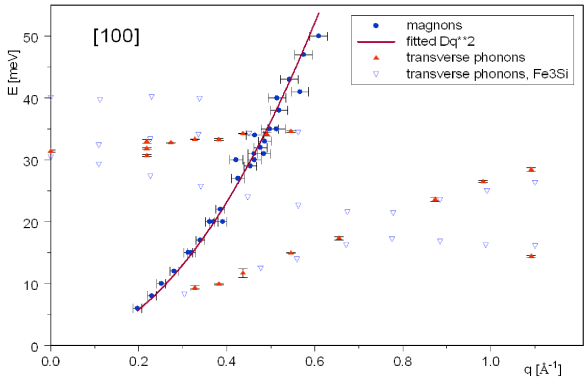

Fig. 2. Spin wave dispersion relation in Sendust measured along [100] direction. Full triangles show measured scattering from phonons. The open triangles are taken from paper [22] for Fe_3Si.

As observed in the experiment [21], the magnon line widths increase with magnon energy. This is presented in Fig. 4. The comparison of these widths measured along three high-symmetry directions is shown in Fig. 5, where the data of present paper are shown together with the

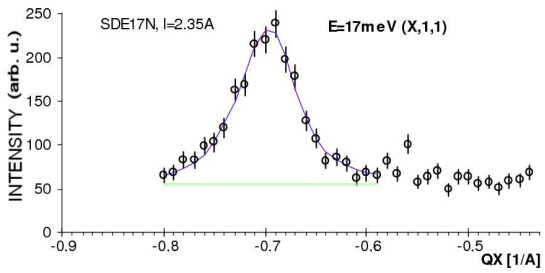

Fig. 3. Typical form of the spin-wave peak for the constant energy transfer of 17 meV.

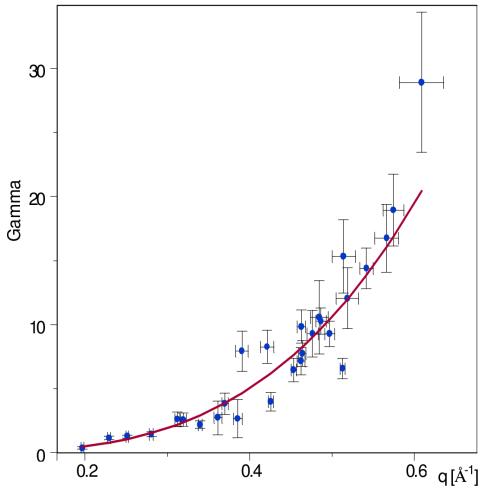

Fig. 4. Magnon line width vs. q.

data published in Ref. [21]. One should point out that the value of exchange stiffness constant as well as the type of dependence of magnon line width on magnon wave vector depend especially on the assumption concerning the latter form. The results presented in this paper were obtained under assumption of quadratic dependence, $\Gamma_q = Aq^2$. It has been checked that assumption

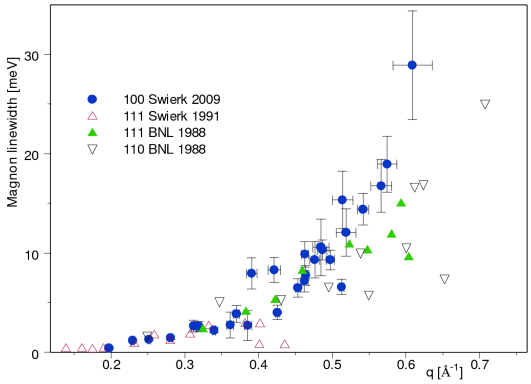

Fig. 5. Magnon line widths measured along three high-symmetry directions.

of cubic dependence, $\Gamma_q = Aq^3$, influences both values, D and A, however the differences are mainly observed at large wave vectors, for which the damping is strong. How well the formula used for the description of the scattering cross-section fits the reality remains the subject for separate studies.

4. Conclusions

The paper summarizes main findings concerned with static and dynamic properties of alloys based on Fe_3Si and Fe_3Al intermetallic alloys. It is shown that the question of preferential ordering of impurities is still open for theoretical investigation. In the case of spin dynamics, the paper shows spin-wave dispersion relation and the magnon line widths measured along [100] direction in Sendust. No anomalous damping of spin waves was observed up to 50 meV, which disproves the findings of [21]. The most likely explanation is that experimental problems like e.g. defocusing of the instrument appeared during measurements [21] at Brookhaven National Laboratory. There is general agreement with the findings concerning increased magnon line widths with quasi-momentum q, although numerical values are slightly different. There is clear dependence of the line widths on the spin-wave direction. While this is $(107 \pm 1)q^{3.3\pm0.2}$, the data from [21] indicate $(28 \pm 4)q^{1.95\pm0.15}$ for [110], and $(58 \pm 4)q^{2.83\pm0.06}$ for [111] directions. The reason for this directional dependence in otherwise isotropic system remains a mystery. Equally strange is huge line broadening which in case of all directions is much larger than can be calculated basing on theoretical calculations like [23, 24]. The problem of using formula (1) for spectral function must be separately considered. Therefore, detailed measurements along two other directions, under identical conditions as described in this paper, must be carried out. Such studies are underway.

References

[1] S. Pickart, T. Litrenta, T.J. Burch, J.I. Budnick, *Phys. Lett.A* **53**, 321 (1975).

[2] T.J. Burch, J.I. Budnick, V.A. Niculescu, K. Raj, T. Litrenta. *Phys. Rev. B* **24**, 3866 (1981).

[3] V. Niculescu, K. Raj, T.J. Burch, J.I. Budnick, *Phys. Rev. B* **13**, 3167 (1976).

[4] S. Yoon, J.G. Booth, *Phys. Lett. A* **48**, (1974) 381; *J. Phys. F, Met. Phys.* **7**, 1079 (1977).

[5] V. Niculescu, T.J. Burch, J.I. Budnick, *J. Magn. Magn. Mater.* **39**, 223 (1983).

[6] A.C. Swintendick, *Solid State Commun.* **19**, 511 (1976).

[7] E.J.D. Garba, R. Jacobs, *J. Phys. F* **16**, 1485 (1986).

[8] A. Go, M. Pugaczowa-Michalska, L. Dobrzyński, *Phys. Status Solidi B* **243**, 1241 (2006).

[9] A. Bansil, S. Kaprzyk, P.E. Mijnarends, J. Toboła, *Phys. Rev. B* **60**, 13396 (1999).

[10] A. Go, M. Pugaczowa-Michalska, L. Dobrzyński, *Eur. Phys. J. B* **59**, 1 (2007).

[11] A. Go, M. Pugaczowa-Michalska, L. Dobrzynski, *Acta Phys. Pol. A* **115**, 191 (2009).

[12] S. Mager, E. Wieser, T. Zemcik, O. Schneeweiss, P.N. Stetsenko, V.V. Surikov, *Phys. Status Solidi A* **52**, 249 (1979).

[13] T.V.S.M. Mohan, C. Babu, Bansal, *Phys. Status Solidi B* **193**, 167 (1996).

[14] J. Waliszewski, L. Dobrzyński, A. Malinowski, D. Satuła, K. Szymański, W. Prandl, Th. Bruckel, O. Scharpf, *J. Magn. Magn. Mater.* **132**, 349 (1994).

[15] D. Satuła, L. Dobrzyński, J. Waliszewski, K. Szymanski, K. Rećko, A. Malinowski, Th. Brückel, O. Schärpf, *J. Magn. Magn. Mater.* **169**, 240 (1997).

[16] D. Fuks, S. Dorfman, V. Liubich, L. Kutsenko, *Int. J. Quant. Chem.* **90**, 1478 (2002).

[17] L. Dobrzyński, K. Blinowski, S. Bednarski, H. Kepa, T. Giebultowicz, W. Minor, *Solid State Commun.* **46**, 217 (1983).

[18] M. Szymański, M. Jankowski, L. Dobrzynski, A. Wiśniewski, S. Bednarski, *J. Phys., Condens. Matter* **3**, 4005 (1991).

[19] L. Weiss, P. Urwank, *Proc. Inel. Scattering of Neutrons*, Vol. II IAEA, 1979, p. 197.

[20] L. Weiss, P. Urwank, *J. Phys. F* **12**, 765 (1982).

[21] L. Dobrzyński, A. Wisniewski, Y.J. Uemura, S.M. Shapiro, J.P. Wicksted, *Phys. Rev.* **37**, 7175 (1988).

[22] O.G. Randl, G. Vogl, W. Petry, B. Hennion, B. Sepiol, K. Nembach, *J. Phys., Condens. Matter* **7**, 5983 (1995).

[23] A. Brooks Harris, *Phys. Rev.* **175**, 674 (1968).

[24] R.B. Stinchcombe, T.L. Reinecke, *Phys. Rev. B* **9**, 3786 (1974).