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Inelastic Neutron Scattering in Sendust Alloy in 8–295 K Temperature Range

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The effects concerning magnetic and nuclear excitations in a single crystal of Sendust alloy (73.5 at.% Fe, 9.5 at.% Al, 17 at.% Si) are discussed. The excitations have been investigated by means of elastic and inelastic neutron scattering with the use of triple axis and small angle spectrometer. A few potential causes of spin wave damping in Sendust, like interactions of excitations or small-size precipitations of different compositions or directions of magnetization were disproved. It is suggested that one- and two-phonon scattering can explain observed strong increase of incoherent inelastic scattering with temperature.

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1. Introduction

Sendust alloy ($Fe_{2.94}Si_{0.68}Al_{0.38}$) is very soft ferromagnet. Its DO_3-type of structure comprises four fcc sublattices located in the following positions in the unit cell: [0, 0, 0], [1/4, 1/4, 1/4], [1/2, 1/2, 1/2], [3/4, 3/4, 3/4], named A, B, C, and D. In the stoichiometric composition (Fe_3Al, Fe_3Si-like) Fe atoms locate in the positions A, B, and C, while the other element locates in position D. An important feature of Sendust is that position D is occupied by two types of atoms: either Si or Al. In the investigated Sendust sample (73.5 at.% Fe, 9.5 at.% Al, 17 at.% Si) there is also a slight excess of Al and Si atoms in comparison to the stoichiometric alloy composition.

Previous experiments at room temperature revealed strong spin wave damping in the sample [1, 2]. The aim of the present investigation was to determine the cause of this damping.

There are three potential sources of spin wave damping worthy of consideration:

- mutual interaction of magnons or magnons with other excitations,
- chemical and magnetic disorder within the lattice,
- existence of precipitations that differ either in the composition or in magnetization direction. The precipitations must be of supposedly little size allowing them to break the continuity of a single crystal from perspective of a short spin wave.

Magnon–magnon interaction is well known to cause the spin-wave energy broadening, which for small magnon wave-vector $\boldsymbol{q}$ and low temperature is proportional to q^3 and $T^{5/2}$ [3]. On the other hand, the magnon–magnon interaction is also well known to influence the magnetic stiffness constant giving its temperature dependence: $D(T) = D(0)(1 - BT^{5/2})$.

For the Heisenberg model of amorphous system of localized spins, local differences in exchange interaction yield spin-wave energy broadening proportional to q^5 independent of temperature [4]. The same q^5-dependence was obtained for the Heisenberg model of random binary ferromagnetic alloy [5].

For the itinerant electron model local differences of the Coulomb potential without spin-flipping scattering gives spin-wave damping proportional to q^4, independent of temperature [6]. Spin flipping scattering gives the additional term proportional to q^2 [7].

To check the causes of the strong damping the inelastic scattering of neutrons was performed on Sendust single crystal in 8 K to 295 K temperature range. In addition, in order to verify the possibility of the damping on precipitations of different composition or magnetic direction, small angle neutron scattering on the sample was measured.

2. Coherent inelastic neutron scattering

The coherent inelastic scattering of neutrons of fixed final energies of 14.8 meV and 20.5 meV was measured to investigate the temperature dependence of the spin wave damping. The preliminary results of inelastic neutron scattering together with detailed analysis of the $\boldsymbol{q}$-dependence of spin-wave peaks are to be published elsewhere [8]. The scattered neutron intensities were measured for a fixed energy transfer and selected range of

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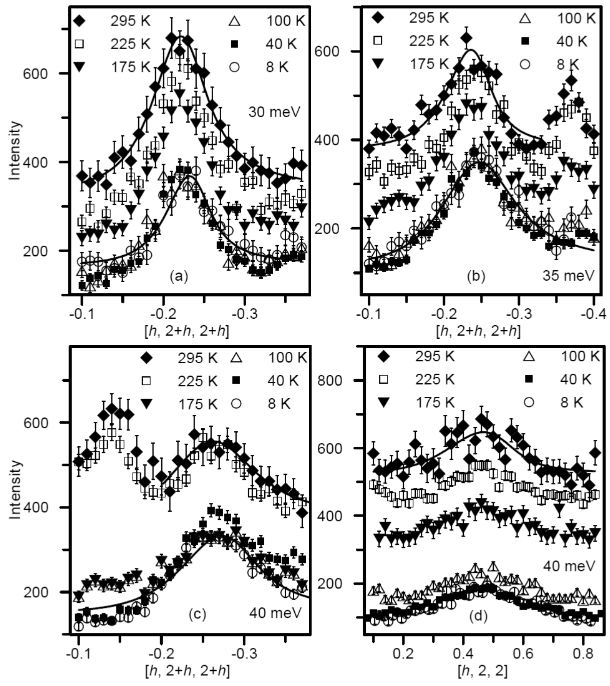

Fig. 1. Inelastic scattering intensity distributions for a given neutron energy transfer and a varying scattering wave-vector at a few temperatures. The scattering vectors are measured along directions shown on horizontal axes. Solid lines are fits for 8 K and 295 K.

spin wave-vectors. The temperature dependence of the four selected distributions is presented in Fig. 1.

The crucial parameters characterizing spin waves in given ferromagnetic material are spin stiffness constant D and damping parameter A. They are defined by the following q-dependent expressions for dispersion relation (Eq. (1)) and energy broadening (Eq. (2)):

$$\hbar\omega_q = Dq^2, \tag{1}$$

$$\hbar\Gamma_q = A_n q^n. \tag{2}$$

The quoted expression for dispersion relation $h\omega_q$ is typical for ferromagnets with negligible anisotropy and for small q values. The general formula for energy broadening $\hbar\Gamma_q$ has been used with both $n = 2$ and $n = 3$, according to the earlier results [1]. Equations (1) and (2) served as a basis for fitting the distributions of scattered neutron intensity with the function $I(Q,\omega)$:

$$I(Q,\omega) = I_0 \int \mathrm{d}Q'\,\mathrm{d}\omega' R(Q,\omega,Q',\omega') \times \frac{\mathrm{d}^2\sigma(Q',\omega')}{\mathrm{d}\Omega'\,\mathrm{d}\omega'} + B. \tag{3}$$

The intensity of scattered neutrons $I(Q,\omega)$ depends on the intensity I_0 of the incoming neutrons and is a convolution of spectrometer resolution function $R(Q,\omega,Q',\omega')$ with neutron scattering cross-section of the form

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}\omega} \propto \frac{k_\mathrm{f}}{k_\mathrm{i}} f^2(\boldsymbol{Q})[n_\mathrm{B}(\omega)+1]\left[\frac{\hbar\Gamma_q}{(\hbar\omega-\hbar\omega_q)^2+(\hbar\Gamma_q)^2} - \frac{\hbar\Gamma_q}{(\hbar\omega+\hbar\omega_q)^2+(\hbar\Gamma_q)^2}\right], \tag{4}$$

which characterizes the neutron scattering on the spin wave with energy $h\omega_q$ and broadening $\hbar\Gamma_q$. B is the background of the distribution. The parameters (D_2, D_3, A_2, A_3) obtained as the result of fitting procedures have the superscript 2 or 3 used for the identification of fitting procedures made for $n = 2$ or $n = 3$ according to Eq. (2).

Some exemplary dependences of stiffness constant D_3 are shown in Fig. 2. Every point in temperature dependence below comes from single distribution (shown in Fig. 1) measured for fixed spin wave energy and varying spin wave vector.

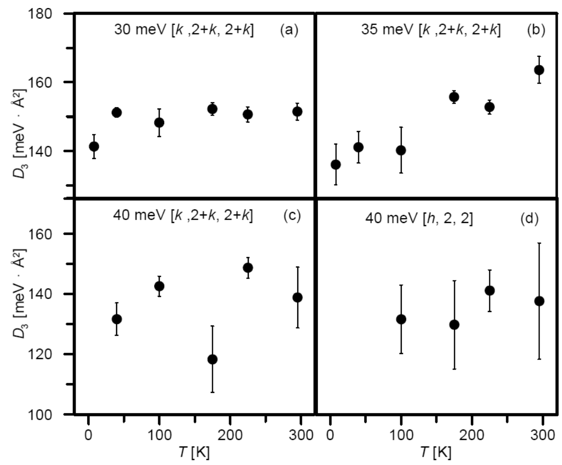

Fig. 2. The temperature dependence of spin stiffness constant in Sendust alloy obtained for exemplary individual scans. The neutron energy transfer and the scattering vector are given in each figure.

Damping parameter A_3 dependence on temperature is shown in Fig. 3.

The stiffness constants and energy broadenings obtained for $n = 2$ or $n = 3$ are the same within the experimental uncertainty, and do not show any directional dependence as suspected in Ref. [1]. The parameters D and A reveal no change with temperature larger than the experimental error. Nevertheless, the slight tendency of spin stiffness constant to increase with temperature can be noticed which is against its expected decrease caused by magnon–magnon scattering. The energy broadening does not show the expected increase, so the interactions of excitations may be excluded as the source of strong damping.

In Fig. 4 the spin wave damping $\hbar\Gamma_q$ dependence on wave-vector length q for a few selected temperatures is presented.

As it can be seen from Fig. 4, the spin wave damping can be well described by $\hbar\Gamma_q = A_n q^n$ with $n = 2$

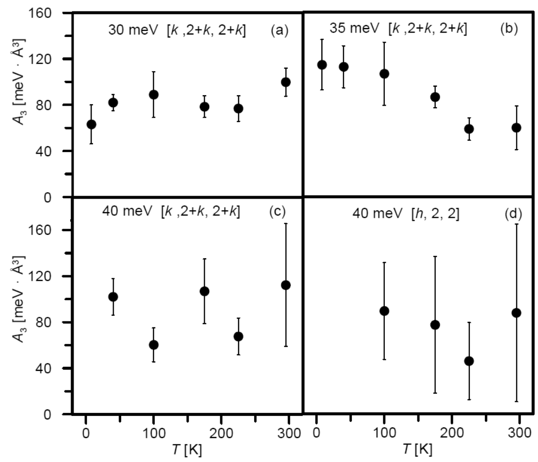

Fig. 3. The temperature dependence of parameters characterizing spin wave damping in Sendust alloy obtained for exemplary individual scans. The neutron energy transfer and the scattering vector are given in each figure.

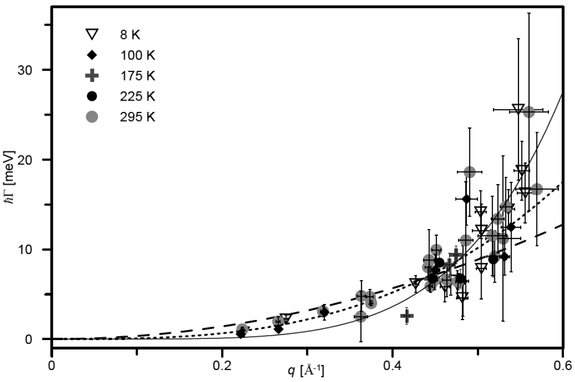

Fig. 4. The spin wave energy broadening versus wave vector for five selected temperatures. The lines represent the fits to the data for 8 K: dashed line — the Aq^2, dotted line — the Aq^3, and solid line — the Aq^5 relation.

or 3 at small $\boldsymbol{q}$-vectors. This description is certainly not adequate for $q > 0.45$ Å^{-1}. In this range of q, the Aq^5 broadening form presented as a solid line in Fig. 4 fits the results quite well. Therefore, one can suspect that such situation may arise from presence of the sample disorder coming from the randomness of the occupation of D site in unit cell. Indeed, recent electronic structure calculations of $Fe_3Al_{1-x}Si_x$ alloys [9] show that Fe–Al and Fe–Si bonds have different lengths. Locally different numbers of Al and Si neighbours may cause local changes in the Fe–Fe distances leading to chaotic variations of exchange integrals. According to [4, 5] the disorder of the exchange integrals should cause temperature-independent spin-wave energy broadening proportional to q^5. However, possible influence of hypothetical nanocrystallites cannot be excluded as a source of this peculiar situation. Therefore the measurements of the small angle scattering have been carried out.

3. Small angle neutron scattering

Were the damping caused by a number of discontinuities of the crystal, it would weakly affect long spin waves. Short spin waves of the size comparable to the size of different-phase precipitations in the crystal would, however, become perturbed to a considerable extent. It is possible that the sample contains precipitations of the composition different from the average or of the differing magnetization direction, of the size suitable to produce the damping of spin waves with $q > 0.45$ Å^{-1}.

The small angle neutron scattering was performed in order to test the size of possible grains. The investigations were conducted in magnetic field H of the intensity of 0.3 T in two setups — with the scattering vector parallel and perpendicular to the direction of the applied field to check whether the grains are of magnetic character.

Figure 5 reveals the results of measurements aiming to detect an existence of some potential inhomogeneities in the sample. As seen, there is no doubt that some inhomogeneities exist in the sample. However, there is no difference in the intensity distribution obtained for two magnetic field directions, so possible precipitations cannot be of magnetic character. Thus, one can wonder whether the precipitations may not differ in their individual content of elements (Fe, Si, Al).

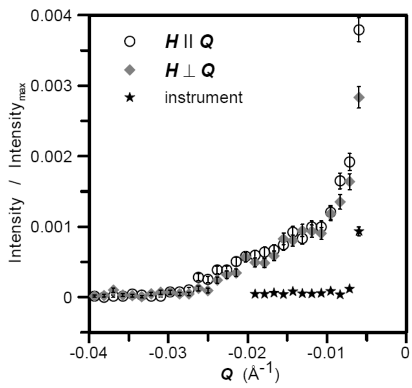

Fig. 5. The normalized intensity distributions of small angle neutron scattering performed for the magnetic field direction parallel and perpendicular to the scattering vector. The intensity observed for empty instrument without the sample is also given.

The grain sizes estimated from the observed distributions are of the order of 280–360 Å which accounts for approximately 50–63 unit cells. They are much larger than the spin-wave lengths for which the damping increase is strong, i.e. shorter than 14 Å ($q > 0.45$ Å^{-1}). Therefore, whatever is the character of the observed precipitations, they should not cause the observed increase of damping.

4. Incoherent inelastic neutron scattering

It can be seen from Fig. 1 that the background B of the distributions of inelastic neutron scattering increases significantly with temperature. The neutron background coming from the reactor hall is independent of temperature. Therefore, the observed distributions contain primarily large contribution of incoherent inelastic scattering of neutrons, henceforth called B. The supposition was that it comes from the incoherent inelastic scattering on phonons which are governed by the Bose statistics. In order to check this suspicion we analyzed the temperature dependence of the intensity of incoherent inelastic scattering for various energy transfers. The analysis was made for the bare intensity and the intensity divided by the factor $n_{\mathrm{B}}+1$. The temperature dependences of the both mentioned quantities for chosen energy transfers are shown in Fig. 6.

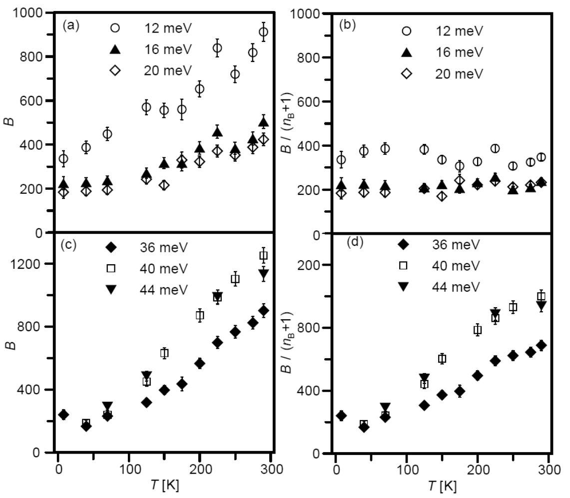

Fig. 6. The temperature dependence of the incoherent INS for energy transfers smaller than 24 meV (a, b) and energy transfers greater than 24 meV (c, d): B — unaltered, and $B/(n_{\mathrm{B}}+1)$ — divided by $n_{\mathrm{B}}+1$ factor.

It follows from Fig. 6 that for small energy transfers (12–20 meV) the division by $n_{\mathrm{B}}+1$ factor results in the quantity which does not change with temperature. Therefore, the incoherent one-phonon scattering may explain the observed increase of intensity for those energy transfers. The same procedure performed for larger energy transfer shows that one should search for still different cause of the abrupt raise of background intensity, two-phonon scattering for example. The temperature dependence of the two-phonon contribution to the incoherent inelastic scattering cross-section is given by [3]:

$$\left(\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E}\right)^{\mathrm{2ph}}_{\mathrm{inc}} = \frac{N\sigma_{\mathrm{i}}}{8\pi\hbar}\frac{k_{\mathrm{f}}}{k_{\mathrm{i}}}\exp(-2W(Q))\left(\frac{\hbar Q^2}{2M}\right)^2$$

$$\times\int_{-\infty}^{\infty}\mathrm{d}\omega_1\frac{Z(\omega_1)Z(\omega-\omega_1)}{\omega_1(\omega-\omega_1)}n_{\mathrm{B}}(\omega_1)n_{\mathrm{B}}(\omega-\omega_1), \quad (5)$$

where $\exp(-2W(Q))$ is the Debye–Waller factor, M is the mean atomic mass and $Z(\omega)$ is the phonon density of states. Other symbols have their common meaning. Temperature dependence was calculated using the Debye approximation for the density of states: $Z(\omega)=C\omega^2$ and normalization fitted to the experimental data.

The comparison of the calculations according to the formula (5) with experimental results is presented in Fig. 7 for neutron energy transfer 36 meV.

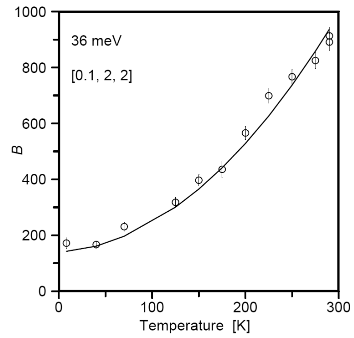

Fig. 7. The temperature dependence of the calculated cross-section of the incoherent two-phonon neutron scattering with normalization fitted to the experimental data (solid line) and intensity of incoherent inelastic scattering measured at (0.1, 2, 2) (open circles), both for energy transfer 36 meV.

The temperature behavior of calculated cross-section of the incoherent two-phonon neutron scattering is consistent with the experimental results. However, the question is why the two-phonon contribution should be so intense? According to calculations for vanadium [10] the contribution of two-phonon scattering in the total scattering cross-section is negligible compared to the contribution of one-phonon scattering for the energy of incident neutrons up to 40 meV and is still much smaller than one--phonon contribution up to 1000 meV. In our experiment the incident neutron energy does not exceed ≈ 60 meV and two-phonon contribution should be small.

5. Conclusions

The incoherent inelastic neutron scattering on Sendust have been analyzed in order to find the origin of the strong spin waves damping obtained in previous investigations [1, 2]. The present results revealed that the damping is independent of temperature and especially strong for wave-vector values q greater than 0.45 Å^{-1}. The value of spin wave energy linewidth (of a given wave vector) in the whole q range is a few times smaller than spin wave energy value.

The analysis of the small angle neutron scattering suggests that the sample contains precipitations of differing composition but they are too large in comparison with the spin-wave wavelengths to produce the observed strong damping.

The lack of the temperature dependence of spin wave damping and its proportionality to the larger power of q

(maybe q^5) for $q > 0.45$ Å^{-1} shows that in contrast to original expectations, the interactions of excitations (governed by temperature dependent Bose statistics) are not the source of the damping. The source of the strong damping for $q > 0.45$ Å^{-1} may be the sample disorder, namely, the randomness of the occupation of the D position by Al and Si atoms and inexact stoichiometry of the alloy composition in comparison to pure Fe_3Si or Fe_3Al. According to electronic structure calculations by Ma et al. [9], they produce local differences of the distance of Fe atoms, which locally affect the Coulomb forces between them and the exchange interaction. Spin-wave damping caused by dispersion of the exchange integral was shown to be proportional to q^5 within the Heisenberg model [4, 5]. Within the itinerant electron model, dispersion of the Coulomb interaction yields spin wave damping proportional to q^4 [6] and spin-flipping impurity interaction produces damping proportional to q^2 [7]. In both localized and itinerant model spin wave damping caused by disorder is temperature independent. Perhaps observed $\boldsymbol{q}$-dependence of the linewidth, Fig. 4, contains two contributions, e.g. q^3 and q^5.

The other interesting feature of the material is large increase of the incoherent neutron inelastic scattering with temperature, especially at energy transfers higher than 30 meV. The increase of the incoherent scattering intensity with temperature is larger for these energies than that of magnon occupation factor corresponding with the increase of spin wave peak intensity. The increase for lower energies is consistent with the magnon occupation factor. The origin of the incoherent inelastic scattering growth may be the one-phonon scattering for lower energies and the two-phonon scattering for higher energies although the strength of the latter can hardly be understood at this stage of investigations.

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