Journal of Magnetism and Magnetic Materials 333 (2013) 13–17

Contents lists available at SciVerse ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Effect of 50-keV proton irradiation on the magnetism of a $Fe_{66}Ni_{34}$ Invar alloy

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ARTICLE INFO

Article history:
Received 6 November 2012
Received in revised form
21 December 2012
Available online 31 December 2012

Keywords:
Fe–Ni
Invar alloy
Curie temperature
Ion irradiation

ABSTRACT

The magnetism of Fe–Ni Invar alloys is very sensitive to the lattice constant, stress, and the number of nearest-neighbor Fe–Fe atomic pairs. Ion irradiation is a useful tool to alter the local atomic structure of a given material. Therefore, the effects of low-energy and light-ion irradiation on the magnetism of a $Fe_{66}Ni_{34}$ Invar alloy were investigated in this study. The $Fe_{66}Ni_{34}$ Invar alloy was irradiated with 50-keV protons at a fluence of 1×10^{15} ions/cm^2 at room temperature. The Curie temperature was found to increase from 465 K (before irradiation) to 535 K (after irradiation). The X-ray absorption analysis of the fine structure of the alloy revealed that irradiation had no effect on the atomic structures surrounding Fe and Ni.

1. Introduction

$Fe_{1-x}Ni_x$ alloys with $32 < x < 42$ at% show characteristic magnetic properties, such as near-zero thermal expansion coefficients below the Curie temperature T_C (commonly referred to as the "Invar effect"), magnetic moment behavior that deviates from the Slater–Pauling curve, and anomalously large magneto-volume effects [1]. The Invar effect is a well-known problem in magnetism, and many theoretical and experimental studies that deal with this effect have been published over the years. Calculations that are based on first principles have shown that various magnetic states exist in Fe–Ni Invar alloys, and that the energy and volume barriers between each state are very low. Thus, some mixings of the magnetic states, high-spin and low-spin states, are expected to occur with increasing temperature and pressure (e.g., Refs. [2–4]). These calculations indicate that the characteristic spin fluctuations, caused by the competition between the magnetic states, are at the origin of the Invar effect. Experimental studies, performed under high pressure conditions, have demonstrated that there is a continuous magnetic transition and the collapse of the ferromagnetism with increasing pressure, which is consistent with the results of previous theoretical studies [5–9].

According to the experimental studies investigating the effects of high-energy milling on the magnetism of a $Fe_{64}Ni_{36}$ alloy, the T_C increases to 150 K, and the lattice parameter slightly increases [10,11]. Gorria et al. reported that stresses could make the high-spin state stable; hence, an increase in T_C occurs due to high-energy milling. Further Invar effects, such as low thermal expansion, can be shown to occur below the increased T_C following high-energy milling [10,11].

Irradiation by ion beams can generate changes at the atomic level (e.g., atomic mixing and defects) at selected locations in a solid. The effects of ion irradiation can be controlled not only over the effective area of a solid but also along its depth, because the irradiated ions release energy at their stopping points. Therefore, if magnetic properties can be controlled by ion-beam irradiation, it may be possible to fabricate controlled three-dimensional magnetic materials [12].

The effects of mega- and giga-electronvolt ion irradiation on the magnetism of Fe–Ni Invar alloys have been previously investigated [13–15]. The T_C of the alloys increased as a result of the irradiation by ion beams. The increase in T_C was reported to be lower than 100 K, and it was strongly dependent on the ion fluence [13]. Furthermore, according to 3.71-GeV Ta-ion irradiation studies, T_C increases only in the ion-stopping region [14]. This means that the interaction between ion irradiation and magnetism occurs in the ion-stopping area, which is consistent with the calculated ion energy-loss behavior. In terms of the effect on T_C, irradiation is similar to high-energy milling.

However, according to recent research on the effect of irradiation on the thermal expansion of the Fe–Ni Invar alloy, the thermal expansion coefficients increase following ion irradiation [16]. This is clearly different from the effect of high-energy milling.

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0304-8853/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jmmm.2012.12.043

Previous ion irradiation experiments have been concerned with the ion irradiation effect above the mega-electronvolt level of ion energy [13–15]. In the present study, we investigated the low-energy and light-element irradiation effects. A $Fe_{66}Ni_{34}$ film was irradiated with 50-keV protons, and the effects of this irradiation on the magnetism and structure of the film were investigated. The proton energy was much lower than that used in previous studies [13–15]. Control of T_C by low-energy and light-ion irradiation offers the possibility to expand the applications of Fe–Ni alloys.

The thickness of the Fe–Ni Invar alloy used in previous experiments was either larger or smaller than the range of ion penetration into the solid ion; this precluded the effect of ion irradiation from being observed [13–15]. To facilitate the study of the ion irradiation effects, the thickness of a Fe–Ni invar alloy film must be consistent with the projection range of 50-keV protons.

2. Experimental methods

Fe–Ni films, measuring 400 nm in thickness (as confirmed by X-ray fluorescence spectroscopy), were fabricated using magnetron sputtering on non-heated MgO substrates. The sputtered samples were annealed in an evacuated silica tube at 1273 K for 2 h, and then quenched. The Fe–Ni films had a face-centered cubic (FCC) crystal structure with a lattice constant of 3.58 , and their chemical composition was confirmed by energy dispersive spectroscopy. The structure and the lattice constant were the same as those reported in previous studies for $Fe_{66}Ni_{34}$ [1]. The films were irradiated with 50-keV protons at a fluence of 1×10^{15} ions/cm^2 at room temperature. Monte Carlo simulations that used the transport ions in matter (TRIM) code, reported in Ref. [17], were performed to calculate the stopping range of the 50-keV protons in $Fe_{66}Ni_{34}$; the results are shown in Fig. 1. The ion penetration depth roughly corresponded to the sample thickness.

The magnetic properties of the films were investigated by analyzing the temperature dependence of the alternating current (AC) susceptibility and magnetization.

Structural variations, such as variations in the distance between atoms and chemical reactions, were investigated by obtaining exit-angle-resolved X-ray absorption fine structure (XAFS) spectra using synchrotron radiation at the SPring-8 BL01 beam-line (Proposal no. 2010B1404). Exit-angle-resolved XAFS was performed for the Fe and Ni *K*-edges. The synchrotron radiation beam passed through a Si (111) monochromator, and was reduced to a cross-sectional area of 3×0.8 mm using slits.

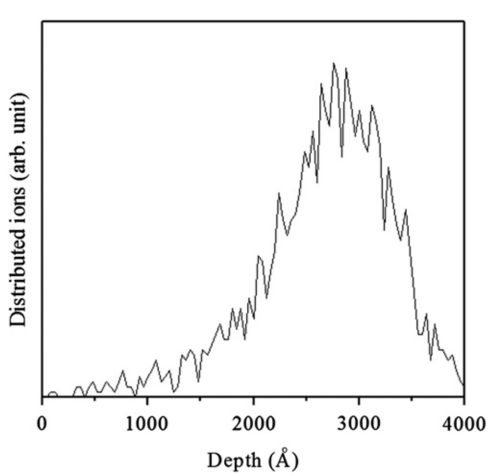

Fig. 1. Distribution of the stopping range of 50-keV proton ions in $Fe_{66}Ni_{34}$, calculated using the TRIM code.

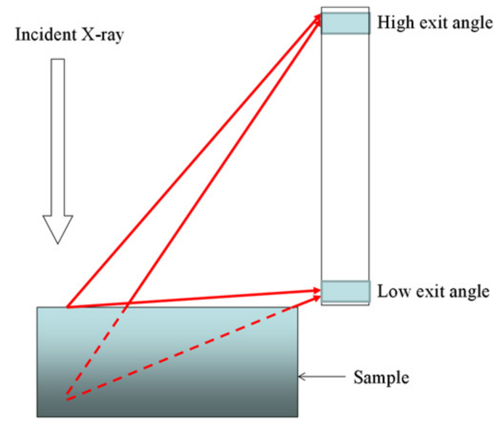

Fig. 2. Schematic diagram of the exit-angle-resolved XAFS. Fluence of the dotted line path is reduced by self-absorption.

The sample was inclined 6° relative to the incident beam, and a two-dimensional pixel array detector (Pilatus, [18]) was set parallel to the sample to detect the emitted fluorescence. The width of one pixel in the array was 172 μm, and the number of pixels was 194. The distance between the surface of a sample and the detector was set to 230 mm. A schematic presentation of the measurement system is shown in Fig. 2. For low exit angles, the contribution of fluorescence from atoms near the surface is relatively high, because the self-absorption loss of the emitted fluorescence from within the sample is larger than that from the surface of the sample. For larger exit angles, the self-absorption loss of the emitted fluorescence from within the sample decreases, and the contribution of the sub-surface fluorescence increases.

3. Results and discussion

In Fig. 3, the variation in the AC susceptibilities of the irradiated and non-irradiated $Fe_{66}Ni_{34}$ alloys is shown as a function of temperature. A 1 kHz AC field was used in these experiments. For both samples, the AC susceptibility (χ) decreased when the temperature (T) increased above 463 K. The χ of the non-irradiated $Fe_{66}Ni_{34}$ alloy decreases sharply to zero; this behavior is typical of ferromagnetic materials. The χ of irradiated $Fe_{66}Ni_{34}$ alloy shows an anomalous two-step decrease. The χ shows a slow decrease for temperatures ranging from 463 to 527 K. For higher temperatures, the susceptibility rapidly approached zero. As shown in Fig. 3, in this study, the temperature at which the maximal slope of the χ–T curve intersected the temperature axis was defined as Curie temperature (T_C). Therefore, the T_C of non-irradiated and irradiated $Fe_{66}Ni_{34}$ alloys were defined as 465 and 535 K, respectively. This change in the shape of the χ–T curve indicates that T_C increased due to proton irradiation across the entire $Fe_{66}Ni_{34}$ film. Considering both the shape of χ–T curves and the results of TRIM calculation shown in Fig. 1, the effect of ion irradiation on the magnetism could be determined as being variable along the depth dimension, which would lead to an inhomogeneous increase in T_C. The increase in the magnitude of T_C due to the 50-keV proton irradiation is similar to that reported for irradiation with ions in the mega- and giga-electronvolt energy range [13,14].

The magnetization (M) of Invar alloys, such as Fe–Ni and Fe–Pt Invar alloys, was previously discussed as a function of T/T_C [1,19]. For temperatures that satisfy as $T/T_C > 0.6$, the magnetization of Invar alloys decrease more rapidly than that of Ni, which suggests the existence of a hidden excitation in Invar alloys. This hidden