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Radiation Effects in Ordered Fe - Al Alloys: Mössbauer Experiments Following Neutron Capture [*]

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Abstract:

Mössbauer effect with Fe^{57} following neutron capture in Fe^{56} has been investigated in Fe - Al alloys in the concentration range from about 26 to 52 at. % Al. In ordered alloys of CsCl structure we observed line shifts due to recoil effects following neutron capture. In a Fe_3Al alloy of DO_3 type order additional changes in the internal magnetic fields have been found. Both effects are discussed in terms of possible final positions for the primary recoiled iron atom in the lattice.

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[**] Guest scientist from the Oak Ridge National Laboratory, 1967 - 1968

1. Introduction

One of the open problems in radiation damage theory concerns the final position of primary recoil atoms. The question has been studied extensively from the theoretical side [1,2], but direct experimental evidence has hardly been obtained. The possibility of observing the Mössbauer effect following neutron capture [3,4,5] offers a means for direct experimental investigation.

Neutron capture leads to the population of a highly excited level (6 to 8 MeV) of the product nucleus. The capture level is de-excited by emission of gamma rays which impart recoil energies of a few hundred eV to this nucleus. The nucleus is displaced from its lattice position, and will come to rest in the lattice within a time $\lesssim 10^{-13}$ sec. The dominant mechanism of energy loss in this energy range is by elastic collisions with other lattice atoms. These may in turn be displaced, and the final position of the recoiled atom is likely to be disturbed.

The capture level in Fe^{57} populated by neutron capture in Fe^{56} is at 7.643 MeV [6]. In about 50 % of the capture events the de-excitation leads to the 14.4 keV level in Fe^{57}, and the Mössbauer effect can be observed with 14.4 keV gamma rays emitted by these nuclei [5]. The recoil energies due to high energy gamma emission range between about 100 and 544 eV. The mean lifetime of the Mössbauer level ($1.4 \cdot 10^{-7}$ sec) is very long compared to the flight time of approximately 10^{-13} sec.

Earlier (n,γ)-Mössbauer experiments with pure iron targets [5] showed no detectable influence of the recoil event on the hyperfine interactions of the nuclei. Strong effects were found in ionic crystals. There, in addition to lattice disturbances, electronic after effects play an important role, and it is difficult to separate the influence of either type of disturbance on the hyperfine interactions. In metallic systems electronic after effects can be neglected, and if recoil effects on the hyperfine interactions are observed they can be ascribed uniquely to lattice disturbances.

It has been established that hyperfine interactions at an iron atom in Fe - Al alloys are strongly influenced by the distribution of iron and aluminium atoms in the first few neighbourhood shells of the sensing nucleus [7,8,9]. Therefore it could be expected that such alloys should be well suited for the observation of recoil effects.

2. Experiments

The experiments were performed at the Karlsruhe FR2 reactor with a bismuth filtered neutron beam of about $3 \cdot 10^7$ neutrons/cm^2 sec. The Mössbauer spectrometer consisted of a conventional loudspeaker system as velocity drive in connection with a multichannel analyzer operated in the time mode. For detection of the 14.4 keV gamma rays we used a proportional counter with an anticoincidence shield described previously [5]. The (n,γ)-Mössbauer spectra were taken with a single line absorber of $Na_4Fe(CN)_6 \cdot 10\ H_2O$ containing 1 mg/cm^2 Fe^{57}. With every sample we also took an absorption spectrum using a Co^{57} in Cu source and the alloy targets as absorbers. The comparison of these two spectra reveals directly the difference between the hyperfine interaction of the Fe^{57} nuclei in an undisturbed lattice position and in a position reached after recoil. For all spectra mentioned in this paper target and absorber were kept at room temperature.

Alloy samples were used in the composition range from about 26 to 52 at. % Al. The lattice structure of the alloys is body-centered cubic. The equilibrium order at room temperature is of Fe_3Al type (DO_3 superlattice) for alloys containing about 25 to 33 at. % Al, and of CsCl type (B 2 superlattice) in the range from ∼33 to ∼54 at. % Al [10] (Fig. 1).

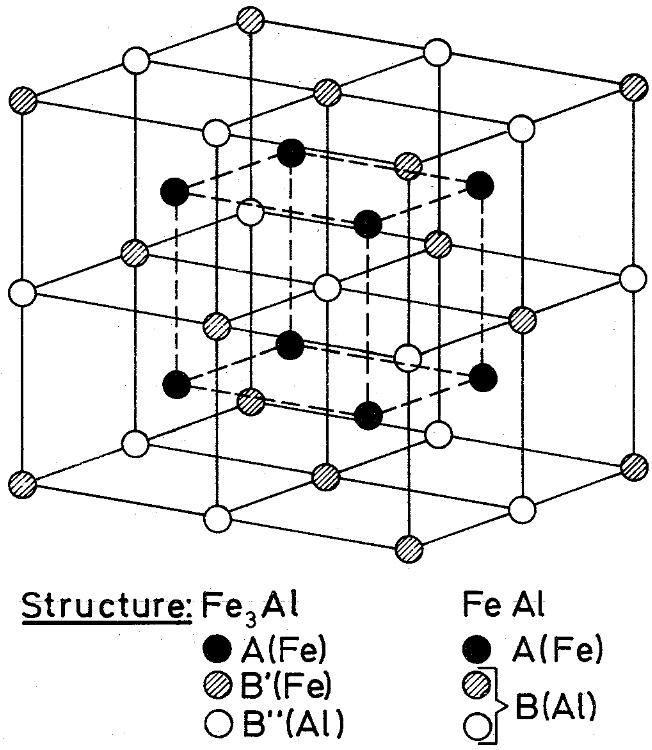

Fig. 1 Lattice structure and order of Fe - Al alloys (25 to 54 at. % Al)

At deviations from stoichiometric composition surplus atoms of one kind are randomly substituted in the sublattice of the other atoms.

We prepared samples with FeAl order (35 to 52 at. % Al) by vacuum melting a mixture of high purity iron and aluminium powder. After a homogenization treatment the alloys were crushed, and annealed at 700 °C for about 24 hours. All samples were examined by chemical and X-ray diffraction analysis. Fe_3Al samples and one FeAl specimen (35.5 at. % Al) were kindly provided by Professor V. Gerold (MPI Stuttgart). The preparation of these samples is identical to that described in Ref. [11].

3. Results

a) Alloys with FeAl structure (35 to 52 At. % Al).

These alloys are paramagnetic at room temperature, and the Mössbauer absorption spectrum taken with the Co^{57} in Cu source

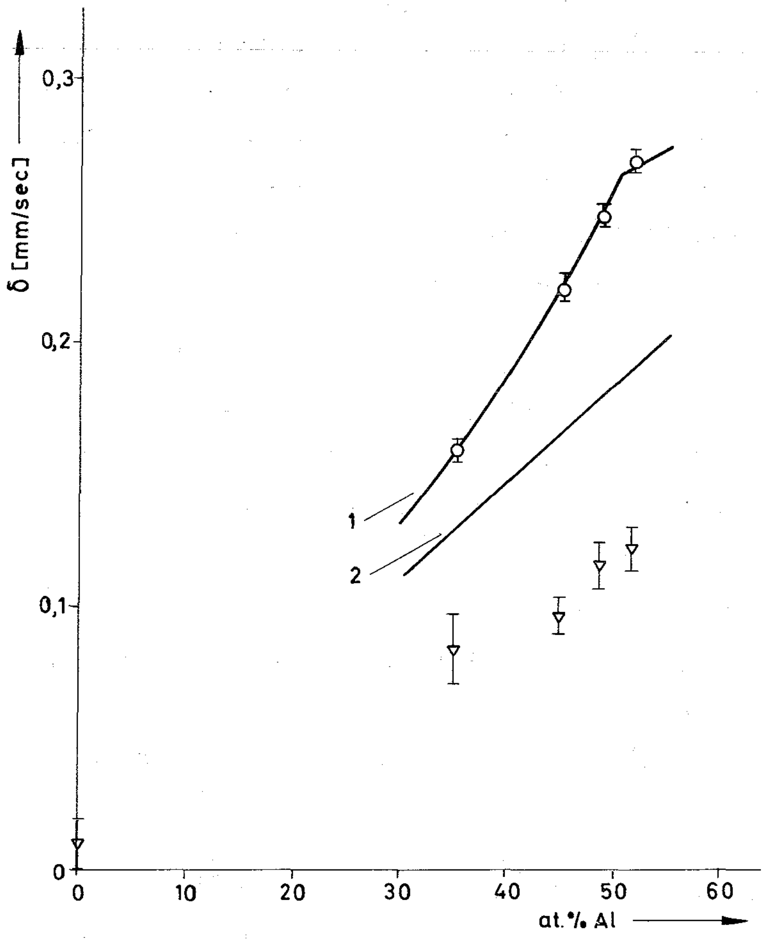

Fig. 2 Line shifts in Fe - Al alloys with CsCl type order relative to metallic iron

o absorber spectra

Δ (n,γ)-spectra

Curves 1 and 2 are explained in the text.

consists of one broadened line. We have fitted these spectra with a single line of Lorentz shape. The overall fit is not good, but the center of the line can be determined in this way with good accuracy. The resulting shifts relative to the center of a pure iron spectrum are shown in Fig. 2 as a function of aluminium concentration. (The positive sign of the shift means that the resonance is centered at a higher energy than in iron.)

The dependence of the observed shifts on aluminium concentration can be explained by assuming that the spectra consist of several overlapping lines, each shifted by an amount $S = n_1 \Delta I_1 + n_2 \Delta I_2 + n_3 \Delta I_3$, where n_k is the number of aluminium atoms in the k - th neighbourhood shell of the absorbing iron nucleus, and ΔI_k, the shift caused by an aluminium atom in the k - th shell, is independent of the aluminium concentration. We also assume that in our samples deviations from the order as specified in section 2 can be neglected (Fig. 2, curve 1).

The (n,γ)-Mössbauer spectra also show only one line. The center shifts relative to iron, determined in the same way as for the absorber spectra, are at a given Al concentration much smaller than those of the absorber lines (Fig. 2).

b) Fe_3Al sample (26.75 at. % Al)

This sample is ferromagnetic at room temperature. The Mössbauer absorption spectrum obtained with the Co^{57} in Cu source is shown in Fig. 3 a. Similar spectra have been reported by other groups [8].

The general interpretation of the spectra is that iron nuclei on lattice sites A (4 nearest Al neighbours) see a smaller hyperfine field than iron nuclei on sites B' (no Al atoms in the first neighbourhood shell).

The (n,γ)-Mössbauer spectrum for the same sample is displayed in Fig. 3 b. Comparison of the two spectra shows clearly that at most a small fraction of the recoiled nuclei can be in positions equivalent to normal iron lattice sites in Fe_3Al.

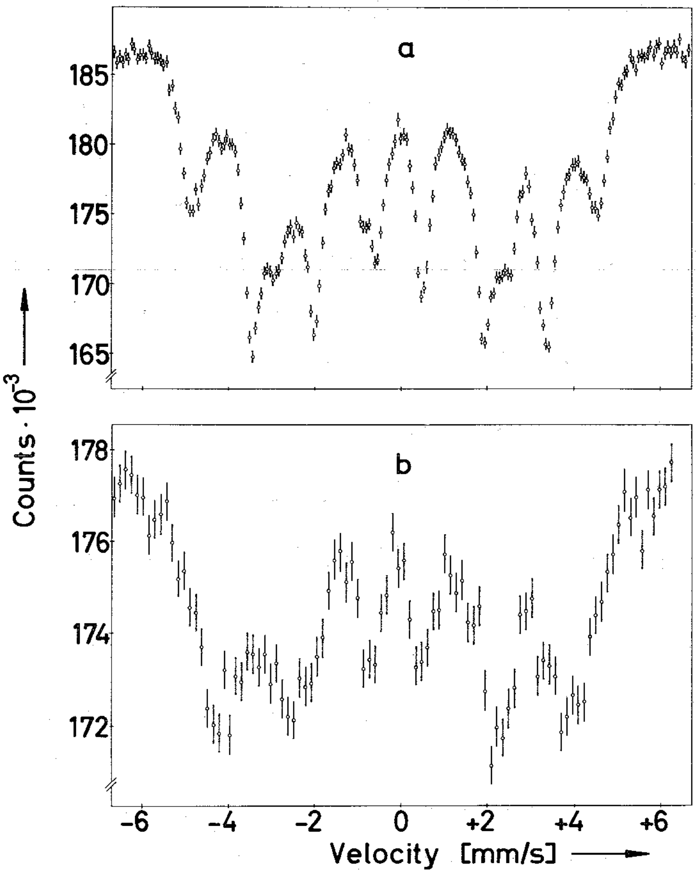

Fig. 3 Mössbauer spectra of Fe_3Al (26.75 at. % Al)

a) absorber spectrum

b) (n,γ)-spectrum

4. Discussion

For the interpretation of the observed effects we have considered the possibilities for the predominant final positions of the recoiled nuclei.

1.) Undisturbed lattice sites
    a) Fe site (may be further subdivided in A, B (or B') site)
    b) Al site
    c) Fe and Al sites (no distinction between the previous occupation of the site)
2.) Lattice site with disturbed order in its neighbourhood.
3.) Lattice site in close association with one or several defects (vacancies or interstitials). Order in the vicinity may be maintained or not.
4.) Interstitial position.

Regarding these possibilities we try to find a single explanation of the phenomena observed in all samples investigated.

Let us first discuss point 1 of the list:

Both the Fe_3Al (n,γ)-Mössbauer spectrum and the (n,γ)-line shifts in alloys with FeAl structure exclude Fe A positions and points 1 a and 1 c, but the shifts would allow predominant ($\gtrsim 90$ %) occupation of Al sites or of B sites (independent of previous occupation). However, both assumptions cannot explain the spectrum obtained with the Fe_3Al specimen. Thus we conclude that it is very unlikely that a large fraction of the recoiled nuclei comes to rest at an undisturbed lattice site.

The Fe_3Al (n,γ)-Mössbauer spectrum is similar to spectra obtained with disordered alloys [9], but the average hyperfine field derived from this spectrum (248 ± 3 k Oe) is much smaller than the average field expected for this specimen according to the data given in Ref. 9 (270 k Oe). The aluminium concentration in the neighbourhood of the final position would have to be substantially above the average concentration of the alloy to explain the reduced field.

Experimental data on isomer shifts of disordered alloys in the concentration range from 35 to 50 at. % Al have not been published. If we calculate shifts with the parameters ΔI_k deduced from the ordered absorbers, we obtain curve 2 in Fig. 2. The (n,γ)-shifts are far below this line. This would indicate a deficiency of Al atoms in the neighbourhood of the final position, in contradiction to the Fe_3Al result. Therefore we believe that the assumption of disorder alone cannot explain the Mössbauer spectra of the recoiled nuclei, but that defects do contribute to the hyperfine interactions at the final positions. With the present results we cannot decide whether the final position of the recoiled nuclei is an interstitial position or whether it is a lattice site in close association with interstitials or vacancies. Experiments at lower and higher temperatures are in progress which will give information about the stability of the final position or the defects associated with it, respectively.

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