STUDY OF THE REPLACEMENT COLLISION PROBABILITY IN THE CASE OF Co IMPLANTED IN Al AT 4.2 K

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ABSTRACT

Earlier Mössbauer Effect experiments[1] on ^{57}Co diffused in Al show basically two different positions for the Co-atoms after irradiation of the sample with fast neutrons : substitutional Co and interstitial Co. In order to study the landing dynamics of Co in Al we have implanted ^{57}Co in Al at 4.2 K, at which temperature the Al-interstitials are still immobile. The total implanted dose was 10^{14} at/cm^2, at [illegible] energy 85 keV. The Mössbauer spectrum shows two lines : one line corresponds to substitutional Co-atoms, the other one comes from interstitial Co-atoms (in fact the <100> Co-Al mixed dumbbell position).

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+ Work supported by the U.S. Department of Energy DOE under contract DE-AC04-76-DP00789

The population of the substitutional site, found to be 0.46 ± 0.02, can give information on the interaction potentials between Co-Al and Al-Al when the results are interpreted on the basis of a replacement collision model modified by taking into account a spontaneous trapping volume for the lattice atom recoil, around substitutional Co.

1. INTRODUCTION

From the hyperfine interaction studies of the behaviour of impurities implanted in metals a great deal of information can be obtained. The interaction of an impurity with the defects produced in its own damage cascade is observed if during the implantation the defects are immobile and if the dose is kept sufficiently low[1] : one then sees the substitutional and/or the interstitial landing of the impurities as well as the association of the impurities with vacancies or interstitials. As a function of temperature, the annealing behaviour of damaged metals can be investigated by examining how the populations of the different non-equivalent sites change. To study phenomena related to vacancies in metals it is in several cases sufficient to perform the implantations at room temperature. In several

metals vacancies are indeed still immobile at room temperature. As an example, we found the vacancy annealing stage in Mo and W by Mössbauer effect measurements on ^{133}Xe implanted in these metals at room temperature[2].

The study of phenomena which are related to interstitials requires much lower temperatures[3-6]. Very few experiments have been performed where a direct landing into an interstitial site was observed[7].

By means of the technique of the Mössbauer Effect (ME), we have investigated the behaviour of ^{57}Co implanted in Al at liquid helium temperature. From the following it will turn out that a direct interstitial landing of the cobalt atoms in aluminium at liquid helium temperature is possible, which, to the authors' knowledge, never has been observed to date in such a direct way.

2. EXPERIMENTAL

2.1 On the Mössbauer Effect

The Mössbauer Effect (ME) has been used to study the implantation behaviour of Co in Al.

Basically the ME is the recoilless emission and absorption of nuclear γ-radiation which can occur when the emitting and absorbing nuclei are embedded in a crystal at a sufficiently low temperature.

Due to the interaction between the nuclear moments and the electromagnetic fields produced by the surroundings of the decaying nuclei (the hyperfine interaction), the resonant absorption can be destroyed by shifting the γ-energy over a distance which is typically of the order of 10^{-8}eV. This shift can be measured by scanning such an energy range - using the Doppler effect - by moving the source relative to the absorber. The γ-ray intensity transmitted through the absorber and recorded as a function of the relative velocity between source and absorber yields the ME spectrum.

An absorption curve can be characterized by a limited number of parameters which we will mention briefly.

The (scalar) Coulomb interaction of the (electrical) charge distribution of the nucleus with the s-electrons which have a non-zero wave function at the nucleus, will change the energy of the γ-transition. If the nuclei of the source have a different Coulomb interaction than

those of the absorber, the centre of the absorption line will be shifted relative to the zero velocity (the isomer shift). Different non-equivalent sites in the source (or the absorber) can have different isomer shifts. The isomer shift is thus sensitive to the surrounding of the impurity.

Due to the (vector) interaction between a magnetic field and the magnetic dipole moment of the nucleus, the nuclear energy levels can be splitted. In the experiment that we have performed, we have used a natural Fe-absorber. The magnetic hyperfine field which Fe-nuclei experience in a Fe foil splits the nuclear energy levels and the whole Mössbauer spectrum (full velocity range) consists of six lines, at least when the source is unsplitted. If the source is splitted (e.g. when two or more non-equivalent sites occur for the nuclei in the source), each of the six lines will be splitted into two or more components (in our case into two as will turn out).

The charge distribution around an impurity in a lattice can produce an electrical field gradient which interacts with the nuclear quadrupole moment. This (tensor) interaction can again produce a splitting of the nuclear energy levels. The quadrupole interaction

occurs only when the surrounding of the nucleus is non-cubic. A substitutional impurity in a cubic lattice has therefore no quadrupole interaction. A defect associated with an impurity in a metal can disturb the cubic symmetry thus producing an electric field gradient. The splitting caused by the quadrupole interaction will often be so small that it may give rise to only a line broadening.

Another parameter which can characterize a Mössbauer line is the area under the absorption spectrum. This area is proportional to the Debye-Waller factor or recoilless fraction f.

$$f = e^{-\bar{k}^2 \langle x^2 \rangle} \qquad (1)$$

where $\bar{k}$ is the wave vector of the γ-radiation, $\langle x^2 \rangle$ the mean quadratic vibration amplitude of the emitting nucleus. f will be, via $\langle x^2 \rangle$, a function of the immediate surrounding of the emitting nucleus. For more information on all this see, e.g., ref.8.

The analysis of a ME spectrum thus can give a great deal of information about the atomistic environment of the Mössbauer nucleus. The scheme of a ME experiment is presented in fig. 1.

2.2 Results on CoAl, 4.2 K implantation

^{57}Co decaying to ^{57}Fe (the 14.4 keV γ-radiation of which is a suitable Mössbauer transition), has been implanted with the Leuven Isotope Separator. The implantation energy was 65 keV, the total implanted dose was 10^{14} at/cm^2. The 14.4 keV was detected by means of a NaI-crystal. The absorber was a natural Fe foil of 12.5 μm thickness. The counting rate of the 14.4 γ-radiation was 300 Hz. The target material was very pure (6 N) polycrystalline Al mounted in a cryostat which can be connected to the isotope separator. The cryostat facility contains the whole Mössbauer set-up. It is thus possible to perform the implantation with the target at 4.2 K and to do ME experiments after the implantation without warming up the sample. The complete facility is to be described elsewhere[9].

From several experiments on neutron and electron irradiated ^{57}CoAl[10] one knows that at least two sites can occur for the cobalt-atoms : substitutional Co is possible as well as Co which has formed a "mixed dumbbell" with an Al-interstitial. The velocity range was set in such

a way that in the ME-spectrum only the two inner lines (of the six lines caused by the magnetical splitting in the absorber) would be visible ; these two lines can split in a "substitutional" and a "defect" component (as found by Mansel et al[10]). The ME-spectrum recorded directly after the implantation is shown in figure 2. It could be fitted satisfactorily with two times two single lines (two 'doublets'). One doublet coming from Co on a substitutional site, has an isomer shift $\delta = 0.60 \pm 0.01$ mm/s relative to a Fe-absorber. The second doublet is shifted relative to the first by 0.40 mm/s. The linewidth of the second component is 0.47 ± 0.02 mm/s which is high compared to the value 0.27 ± 0.01 mm/s found for the substitutional component. Therefore it is reasonable to fit each line of the second doublet with a quadrupole doublet, similarly to the procedure of Mansel and Vogl [11, 12]. The features of the two components are summarized in table I.

The parameters which characterize the second component agree well with those found by Mansel and Vogl for their "defect-line". Thus we are led to identify our "defect-site" with the Co-Al <100> mixed dumbbell : an Al-interstitial is associated to an otherwise substitutional Co.

The site population of the defect site is calculated following

$$\delta = \frac{2\Gamma_2 I_2/f_2}{\Gamma_1 I_1/f_1 + 2\Gamma_2 I_2/f_2} \qquad (2)$$

Γ_1 and Γ_2 are the linewidths of respectively the first and the second component, I_1 and I_2 are their absorption depths, f_1 and f_2 their recoilless fractions.

$f_1 = f_2$ when the measurement is done at 4.2 K (12-14).

3. DISCUSSION

Although interstitials in Al are immobile[3,4] during the implantation at 4.2 K, a considerable fraction of the Co atoms are already associated with an Al-interstitial. To explain this, several mechanisms could be invoked.

First of all, there can be a direct landing in the interstitial position. One of us has studied theoretically the replacement collision probabilities for an arbitrary ion-target combination [15]. The replacement collision probability for Al as a target material is shown in figure 3. As can be seen on the figure, Co has a probability of

0.46 to have a replacement collision with an aluminium atom. 0.54 is then the probability for landing interstitially. These values are not very strongly potential dependent. They are exactly the values we have found from the analysis of our Mössbauer spectrum. With this model alone the behaviour of Co implanted in Al can be understood, even quantitatively. A second phenomenon - additional to the first one - is the following. A Co atom suffering a replacement collision with an Al atom has produced an Al-interstitial. When the energy transfer to this interstitial is too low, it will be trapped spontaneously - thus athermally - by the substitutional Co atom. This effect can be calculated more quantitatively.

It is straigthforward to show that for a projectile with initial energy E_o the probability $P(E_o, R)$ that the recoiling target atom comes to rest within a radius R of a replacement collision is given by

$$P(E_o,R) = \int_0^{E_o} dE \int_{T_a(E)}^{T_m(E)} Q(E,T)\, X(T,R)\, dT/S(E) \qquad (3)$$

In (3) S(E) is the stopping power at energy E, K(T,R) is the probability that a recoil with energy T will come to rest within a radius R of its point of origin, and Q(E,T) dEdT/S(E) is the probability that the incident projectile will suffer a replacement collision when its energy is in the interval (E,E + dE) and will simultaneously create a recoil with kinetic energy in the interval (T,T + dT). The lower limit on the T-integral, $T_a(E)$, is defined in reference 15 and $T_m = 4M_1M_2E/(M_1 + M_2)^2$ where M_1 and M_2 are projectile and target atom masses, respectively.

The function K(T,R) is obtained by integrating the spatial distribution function for a target atom with energy T over a sphere of radius R.

The function Q(E,T) is given by

$$Q(E,T) = N\sigma(E,T) \exp\left\{-\int_E^{E_0} dE' \int_{T_a(E')}^{T_m(E')} N\sigma(E',T')dT'/S(E')\right\} \quad (4)$$

where N is the target atomic density, and $\sigma(E,T)$ is the elastic scattering cross section for energy transfer T to a recoiling target atom by a projectile of energy E.

The above expressions have been evaluated for Co incident on Al using both the Thomas-Fermi and Lenz-Jensen scattering cross sections. The

displacement threshold energy was assumed to be 25 eV. A three dimensional gaussian distribution was used for the recoil spatial distribution in calculating K(T,R). Further, the capture volume for the recoiling target atom was taken to be 200 atomic volumes[10]. The results are concisely expressed in terms of f, the fraction of replacement events in which the target atom recoil is captured, where $f = P(E_o,R)/P(E_o,\infty)$. For the T-F potential we obtain f(TF) = 0.96, while the Lenz-Jensen potential yields f(LJ) = 0.20. Thus, the Lenz-Jensen potential would be favoured by the experimental results of Table I.

A third mechanism for the association of Al-interstitials with Co atoms is the production of the interstitials within the spontaneous trapping volume around substitutional Co. This physical process is described elsewhere[1]. Measurements on the evolution of the site population as a function of implanted dose can give information about the values for the spontaneous trapping volumes.

To separate the influence of the first and second mechanisms from the third one, low dose experiments should be performed (dose $< 10^{12}$ at/cm^2) which are very difficult because of the low γ-ray counting rate. If

the influence of the third mechanism is already appreciable in our dose region, the conclusions about the interatomic potentials become ever more firm.

4. CONCLUSIONS

The present work has shown the use of ion implantation with the target at a low temperature, combined with Mössbauer Effect measurements to study the landing dynamics of impurities in a solid.

For the first time an interstitial landing of a heavy atom implanted in a metal at liquid helium temperature has been observed : Co implanted in Al at 4.2 K lands interstitially for more than 50 %. It can be explained very well by a replacement collision model. Furthermore a modification of that model, taking into account a spontaneous trapping volume for interstitials around a substitutional impurity, gives information about the potentials between the slowing down particles at the end of their track. In that respect, we have shown that the Lenz-Jensen potential fits our results better than the Thomas-Fermi potential.

ACKNOWLEDGEMENTS

The authors wish to thank the "Interuniversitair Institut voor Kern-wetenschappen" (IIKW) for financial support. We thank R. Van Autgaerden

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TABLE I

	Isomer shift relative to a Fe absorber	Linewidth	Quadrupole splitting	Site population
substitutional Co	-0.50 ± 0.01 mm/s	0.27 ± 0.01 mm/s	-	0.46 ± 0.02
Co-Al mixed dumbbell	-0.20 ± 0.01 mm/s	0.33 ± 0.01 mm/s	0.16 ±0.01 mm/s	0.54 ± 0.02

FIGURE CAPTIONS

Fig. 1 Visualisation of the Mössbauer experiment.

Fig. 2 Mössbauer spectrum of ^{57}Co implanted in Al at 4.2 K; the absorber is a Fe-foil.

Fig. 3 Replacement collision probabilities as a function of the atomic number of the incoming atoms ; targetmaterial is Al.

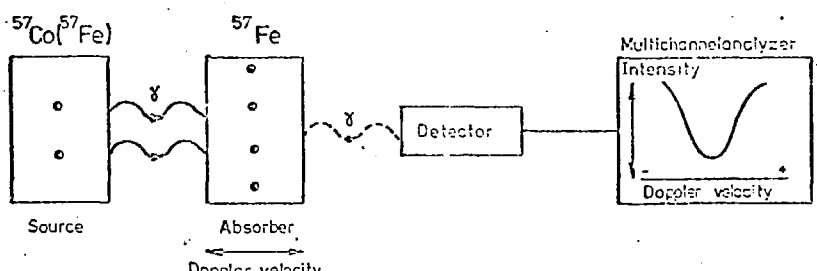

$^{57}Co(^{57}Fe)$
^{57}Fe
γ
γ
Detector
Multichannelanalyzer
Intensity
Doppler velocity
Source
Absorber
Doppler velocity

Fig. 1

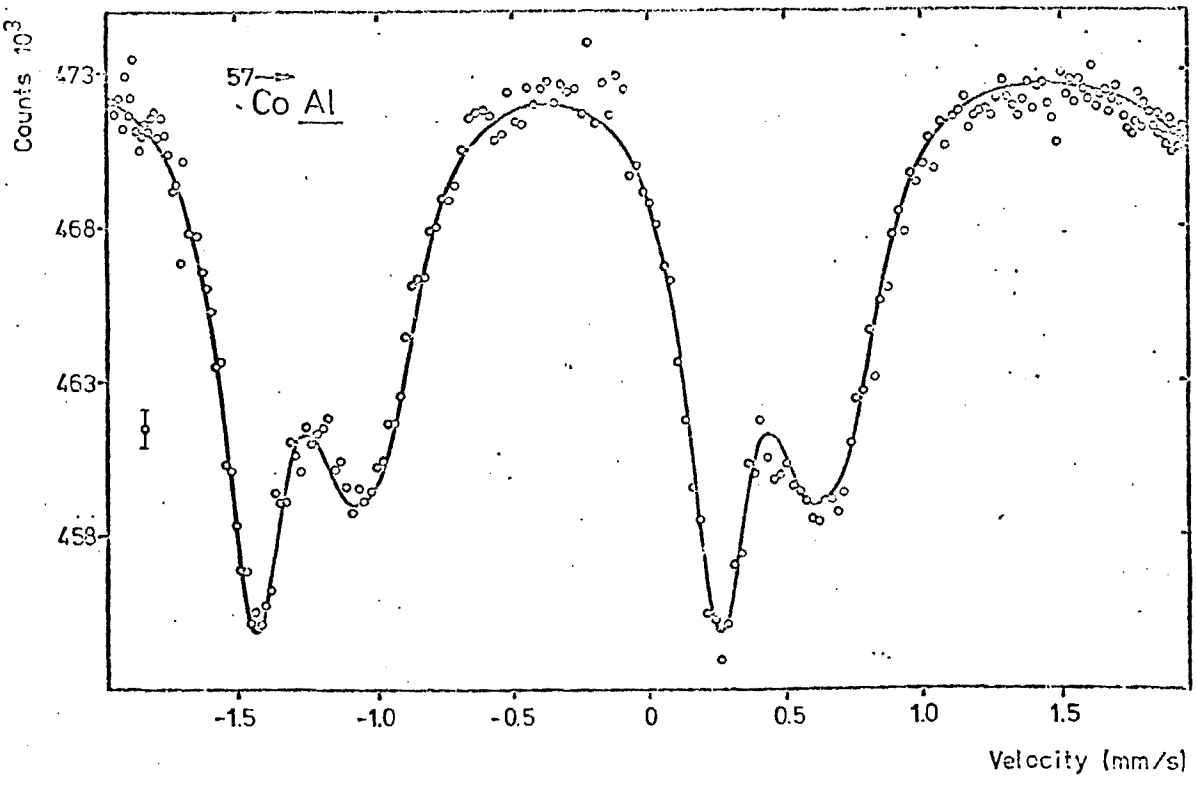
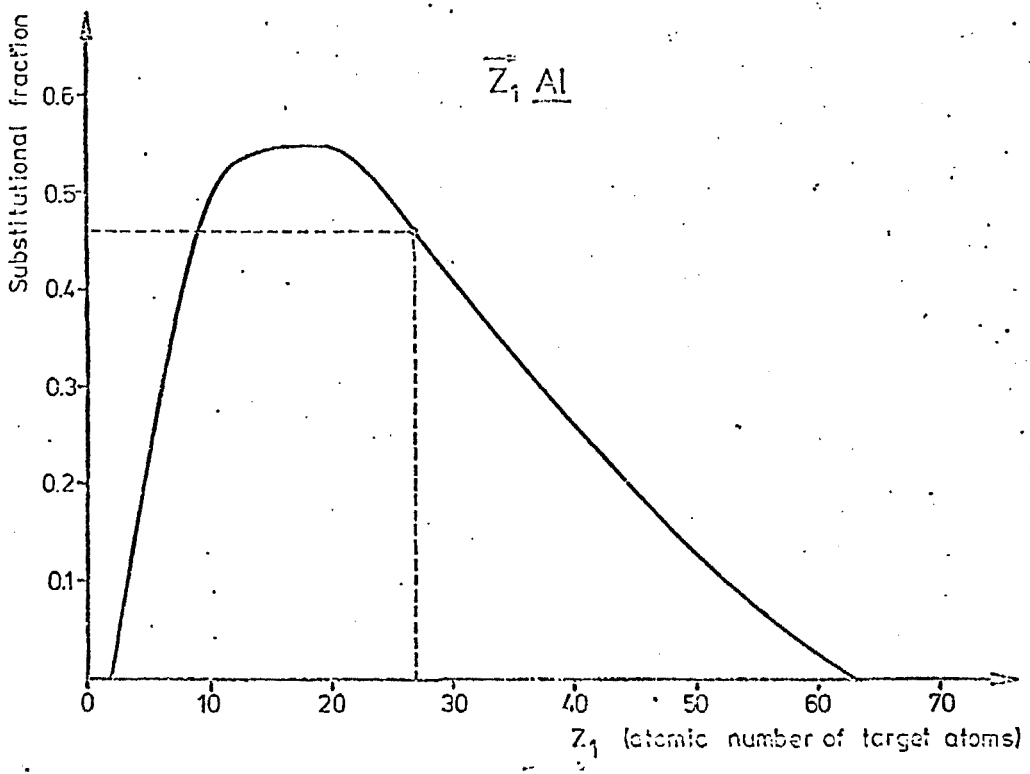

Counts 10³
473
468
463
458
57→
Co Al
-1.5
-1.0
-0.5
0
0.5
1.0
1.5
Velocity (mm/s)

Fig. -


Substitutional fraction
0.6
0.5
0.4
0.3
0.2
0.1
$\overline{Z_1}$ Al
0
10
20
30
40
50
60
70
Z_1 (atomic number of target atoms)