tra susceptibility. This effect can also not be produced by a peculiar field dependence in either of the two measurements. The total susceptibility and the local susceptibility are both linear functions of the external field for $H_{ext} \leq 60$ kG. The polarization in the electron gas obtained in this experiment may have a different cause from that which comes out of the analysis of Heeger.[2,5] Besides the difference in sign as deduced in Refs. 2 and 5, the field and temperature dependences seem to be different; the NMR data give lower values of these variables for the destruction of the polarization cloud although the data on the field dependence of Potts and Welsh[8] are not inconsistent with the present ones if one extrapolates them to very low concentrations. The present values ($H \approx 100$ kG; $T \approx 30$°K) have the advantage of being equal in terms of energy, taking the value of $\mu = 2.8\mu_B$ for Fe:*Cu*.

In conclusion, we have shown that the local susceptibility of the Kondo system *Cu*:Fe follows a temperature dependence as expected on theories following the model of Anderson and Yuval.[11] In addition, we find a small antiferromagnetic polarization of the electron gas in the Kondo state.

One of us (S.H.) wants to thank Professor Götze for a very useful conversation. This work was supported by the Bundesministerium für Forschung und Technologie.

[1]J. Kondo, *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1969), Vol. 23, p. 183.
[2]A. J. Heeger, *Solid State Physics*, edited H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1969), Vol. 23, p. 283.
[3]R. B. Frankel, N. A. Blum, B. B. Schwartz, and D. J. Kim, Phys. Rev. Lett. 18, 1050 (1967).
[4]T. A. Kitchens, W. A. Steyert, and R. D. Taylor, Phys. Rev. 138, A467 (1965).
[5]D. C. Golibersuch and A. J. Heeger, Phys. Rev. 182, 584 (1969).
[6]J. L. Tholence and R. Tournier, Phys. Rev. Lett. 25, 867 (1970).
[7]C. Stassis and C. G. Shull, Phys. Rev. B 5, 1040 (1972).
[8]J. E. Potts and L. B. Welsh, Phys. Rev. B 5, 3421 (1972).
[9]D. V. Lang, J. B. Boyce, D. C. Lo, and C. P. Slichter, Phys. Rev. Lett. 29, 776 (1972).
[10]R. Tournier and A. Blandin, Phys. Rev. Lett. 24, 397 (1970).
[11]P. W. Anderson and G. Yuval, Phys. Rev. Lett. 23, 89 (1969).
[12]K. D. Schotte and U. Schotte, Phys. Rev. B 4, 2228 (1971).
[13]W. Götze and P. Schlottmann, to be published.
[14]C. M. Hurd, J. Phys. Chem. Solids 28, 1345 (1967).
[15]A. Narath, Solid State Commun. 10, 521 (1972).
[16]A. Narath, CRC Crit. Rev. Solid State Sci. 3, 1 (1972).

Direct Evidence for Interstitial-Atom Trapping by Co^{57} Impurities in Aluminum from Mössbauer-Effect Measurements after Low-Temperature Neutron Irradiation

W. Mansel, G. Vogl, and W. Koch
Physik-Department, Technische Universität München, München, Germany
(Received 25 May 1973)

The Mössbauer effect was used to study the effects of neutron irradiation on aluminum doped with Co^{57}. Annealing between 32 and 70 K after irradiation at 4.6 K induces in the previously single-line Mössbauer source spectrum a new component with small quadrupole splitting, which has an isomer shift of +0.40 mm/sec against the original single line. The new component disappears irreversibly on further annealing between 180 and 250 K. This component is interpreted as being due to interstitials trapped in the immediate proximity of the Co^{57} impurity atoms.

Up to now most studies of radiation damage in metals have been carried out by observing the change in macroscopic solid state properties, such as the electrical resistivity. The Mössbauer effect promises the advantage for such studies in that it gives detailed information about the microscopic properties of defects in the vicinity of Mössbauer atoms. Particular sensitivity for such defects occurs in two special cases: (a) When the excited level of the Mössbauer atom is populated via a nuclear reaction such as thermal-neutron capture or Coulomb excitation, then the atom receives a recoil from the nuclear reaction which can produce lattice defects in its immediate vicinity.[1-4] Such neighboring lattice defects are also produced when the Mössbauer atom is recoil-implanted into a lattice.[5] (b) Alternatively defects can be introduced into the lat-

tice by irradiation and be allowed to diffuse at the proper temperature. If the Mössbauer atom is an impurity atom, the migrating defects can be trapped in the elastic stress field caused by size differences between the host atoms and the impurity atoms.[6,7]

We have used this latter method to study the interaction of irradiation-induced point defects in aluminum with Co^{57} impurities. Since the Co^{57} decays by K capture to Fe^{57}, and since the Mössbauer effect measurements are actually done by the 14.4-keV γ rays of Fe^{57}, these impurities will in the following be written as $Co^{57}(Fe^{57})$. We believe this to be the first successful investigation of lattice defects in this way, although a Mössbauer investigation of this type was attempted some time ago by Gonser.[8] Up to now investigations on trapping effects by impurities in aluminum and other fcc metals have mostly been done with resistivity measurements.[9-13] In the following we present the first results of our measurements.

Our Mössbauer source was prepared by drying an aqueous solution of carrier-free $Co^{57}Cl_2$ on an aluminum foil. Afterwards the sample was reduced in an argon-hydrogen atmosphere at 550°C for 5 h, and then diffused for 2 h at 630°C.[14] The resulting sample with an activity of 1.3 mCi showed a single-line Mössbauer spectrum. The measured linewidth and isomer shift, respectively, against a 0.000 25-in.-thick natural-iron foil at room temperature were $\Gamma = 0.235 \pm 0.010$ mm/sec and $\delta = -0.425 \pm 0.005$ mm/sec for the sample at room temperature and $\delta = -0.575 \pm 0.010$ mm/sec for the sample at 4.2 K. These values are in good agreement with values of other authors[15-18] for Co^{57} or Fe^{57} in aluminum. For absorber and sample at 4.2 K the isomer shift was $\delta = -0.465 \pm 0.005$ mm/sec.

To produce lattice defects in the Mössbauer source, it was irradiated with fast neutrons at 4.6 K in the liquid helium neutron-irradiation facility of the Munich research reactor. The irradiation dose was 2.4×10^{18} fast neutrons ($E > 0.1$ MeV) per cm^2. The irradiation temperature of 4.6 K was used since point defects in aluminum begin to migrate above 20 K. After irradiation the sample was brought into the Mössbauer cryostat without warming up (the maximum sample temperature during transfer was 15 K). Details of this procedure and of the cryostat have been given by Rosner, Vogl, and Vogl.[19] Mössbauer measurements with the sample at 4.2 K were performed immediately after the irradiation as well as after various subsequent isochronal (10-min) annealing steps with annealing temperatures of 20, 25, 30, 42, 50, 60, 70, 85, 103, 120, 140, 160, 180, 200, 225, 250, and 300 K. After each 10-min anneal period the sample was recooled to 4.2 K, the recooling lasting between 0.5 and 10 min depending on the height of the annealing temperature.

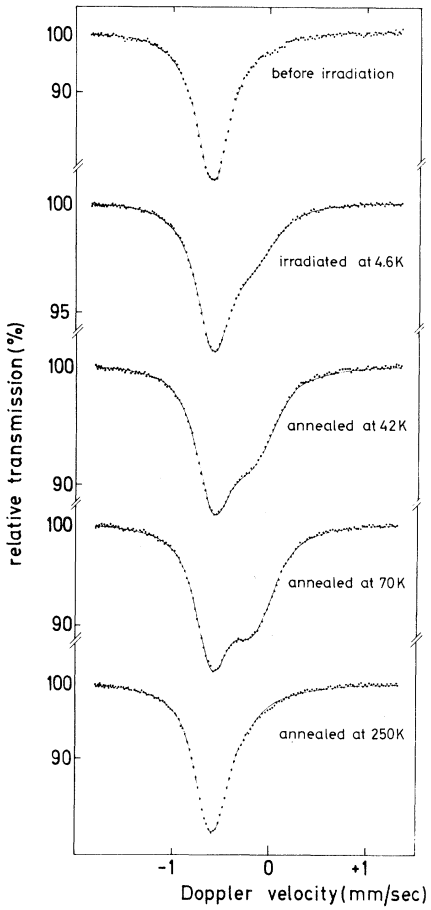

FIG. 1. Characteristic Mössbauer spectra of Co^{57} as impurity in aluminum before and after fast-neutron irradiation at 4.6 K and after subsequent annealing. Absorber: $Na_4Fe(CN)_6 \cdot 10H_2O$ (1.0 mg Fe^{57}/cm^2). Positive velocity means absorber is approaching source.

The moving Mössbauer absorber was $Na_4Fe(CN)_6 \cdot 10H_2O$ with 1 mg Fe^{57}/cm^2, which leads to a linewidth of $\Gamma = 0.43 \pm 0.01$ mm/sec with the unirradiated source of Co^{57} in aluminum. This thickness was used to shorten the total measuring time.

Five characteristic Mössbauer spectra are shown in Fig. 1. Before irradiation the spectrum consists of a single line. After irradiation a new peak appears in the spectrum which begins to increase during the annealing at 30 K, and increas-

TABLE I. Isomer shifts δ relative to a natural-iron absorber, quadrupole splittings ΔE_Q, and ratios R of the areas of the irradiation-induced new line and the single line in some characteristic Mössbauer spectra of $Co^{57}(Fe^{57})$ in aluminum as derived from computer fits.

		Irradiation-induced new line			Temperatures	
	δ (mm/sec)	δ (mm/sec)	ΔE_Q (mm/sec)	R (%)	Source (K)	Absorber (K)
Before irradiation	-0.425 ± 0.005	...	...	0	293	293
Before irradiation	-0.575 ± 0.010	...	...	0	4.2	293
Before irradiation	-0.465 ± 0.005	...	...	0	4.2	4.2
Irradiated at 4.6 K	-0.460 ± 0.005	-0.065 ± 0.005	0.100 ± 0.005	36 ± 2	4.2	4.2
Annealed at 42 K	-0.460 ± 0.005	-0.060 ± 0.005	0.105 ± 0.005	73 ± 2	4.2	4.2
Annealed at 70 K	-0.455 ± 0.005	-0.050 ± 0.005	0.095 ± 0.005	78 ± 1	4.2	4.2
Annealed at 250 K	-0.450 ± 0.005	...	...	0	4.2	4.2

es strongly up to 42 K and slightly further up to 70 K where it reaches its maximum intensity. In the annealing range from 70 to 180 K there is no change in the spectrum. During the 200-K annealing the peak decreases, and it completely disappears during the 250-K annealing.

The spectra after irradiation were least-squares fitted with a single line and a quadrupole doublet, all of Lorentzian shape. Though a fit of the new peak with a broadened single line instead of the quadrupole doublet worked out equally well, we believe the fit with a quadrupole doublet to be reasonable, since lattice defects in the vicinity of a $Co^{57}(Fe^{57})$ atom might be expected to distort the cubic symmetry, thus giving rise to an electric field gradient. The main point in this Letter is not the nature of the new peak, but the fact that it increases during annealing between 30 and 70 K and that it disappears between 180 and 250 K. The best fits with single line and doublet gave values of the linewidths nearly equal to that of the single line before irradiation (0.43 mm/sec), and an isomer shift for the single line also equal to that before irradiation. Thus all measured spectra after irradiation were ultimately fitted with a fixed linewidth of 0.43 mm/sec. The calculated fit parameters of some characteristic spectra are given in Table I. The other spectra yielded consistent results. The new line has a quadrupole splitting of $\Delta E_Q = 0.10\pm0.02$ mm/sec and an isomer shift of $\delta = -0.06\pm0.01$ mm/sec with respect to a natural iron absorber at 4.2 K.

We interpret our data as follows: Defects are created by the irradiation at 4.6 K but are frozen into place. Between 30 and 70 K certain defects migrate and are trapped by the Co^{57} impurity atoms. Above 180 K these defects anneal. The annealing model for lattice defects in pure and impurity-doped fcc metals has been discussed previously, e.g., in the review papers by Schilling *et al.*[20] According to this model for aluminum, mobile interstitial atoms are trapped at the $Co^{57}(Fe^{57})$ impurity atoms mainly during the so-called annealing substages I_D and I_E. The main increase of the defect line in our spectra between 30 and 42 K corresponds to this trapping; this temperature range agrees with substages I_D and I_E. The difference between the effective radius (as defined by Dimitrov and Dimitrov[21]) of a cobalt atom (1.056 Å) and an aluminum atom (1.429 Å) is 26%; thus the elastic distortion around a cobalt impurity in aluminum is large. Cobalt impurities are therefore expected to be strong trappers for interstitials, retaining them over the whole temperature range of the so-called annealing stage II (50 to 180 K). The trapped interstitials should anneal in annealing stage III between 180 and 250 K.[20] This model fits our experimental results very well.

The fact that already after low-temperature irradiation there is a certain number of Mössbauer atoms with defects in their neighborhood might be understood in the following way: The energy loss of fast neutrons during irradiation takes place in a displacement cascade. The atoms in the cascade have briefly a high mobility until they come to rest forming vacancies and interstitials. Some of these mobile defects can be trapped at impurities already during irradiation.

The detailed nature of the defect will be discussed in a subsequent paper, along with results of more extensive irradiation experiments; thus we shall now only briefly try to draw a few conclusions from the hyperfine parameters of the new Mössbauer line about the configuration of the complex of impurity and trapped interstitial.

The fact that we can fit all the spectra after

irradiation with a single line and only one additional doublet indicates that there is a well-defined configuration of $Co^{57}(Fe^{57})$ impurity atoms and radiation-induced lattice defects. The annealing treatments after irradiation do not change the isomer shift or the quadrupole splitting of the new component, implying that the defect configuration around a Mössbauer atom does not change until the defect completely anneals. The difference in the isomer shifts of the new line and the single line by $\Delta\delta = +0.40 \pm 0.01$ mm/sec corresponds to an increase in the electron density at the Fe^{57} nucleus. It is tempting to attribute this rather large increase to the volume compression in the region of the impurity-interstitial complex. Only an interstitial atom in close proximity to the $Co^{57}(Fe^{57})$ impurity atom can cause such a large change.

Finally, we can compare our results with those of Howard *et al.*[22] for Co^{57} in aluminum. These authors produced a single-line source by annealing their sample above 560°C followed by a rapid quench. When, on the other hand, they annealed the sample below 500°C they found a new quadrupole doublet isomer-shifted in the same direction as in our experiment, corresponding to a second site for Co atoms with an increased electron density. The authors could not easily explain the appearance of the new line, but in a similar work on Co^{57} in gold[23] where they also found a new isomer-shifted quadrupole doublet they interpreted it as due to Co^{57} dumbbell interstitials.

Irradiations with varying concentrations of radiation defects and impurity atoms as well as studies on lattice defects in other fcc metals like copper, gold, and silver are in progress.

We thank H. Vonach and G. M. Kalvius for their interest in and support of this work, Ursel Zahn for much advice regarding the source preparation, and W. Vogl for experimental help and advice. We would also like to thank D. Schroeer for discussions and for his careful reading of the manuscript.

[1]G. Czjzek and W. G. Berger, Phys. Rev. B 1, 957 (1970).

[2]W. Mansel, G. Vogl, W. Vogl, H. Wenzl, and D. Barb, Phys. Status Solidi (b) 40, 461 (1970).

[3]G. Vogl, A. Scaefer, W. Mansel, J. Prechtel, and W. Vogl, to be published.

[4]R. L. Lambe and D. Schroeer, Phys. Lett. 41A, 435 (1972).

[5]H. G. Devare and H. de Waard, Phys. Rev. B 5, 134 (1972).

[6]R. R. Hasiguti, J.Phys. Soc. Jap. 15, 1807 (1960).

[7]R. A. Masumura and G. Sines, J. Appl. Phys. 41, 3930 (1970).

[8]U. Gonser, in *Proceedings of the International Conference on Vacancies and Interstitials in Metals, Jülich, 1968,* edited by A. Seeger *et al.* (North-Holland, Amsterdam, 1970), p. 649.

[9]A. Sosin and K. R. Garr, Phys. Rev. 161, 664 (1967); K. R. Garr and A. Sosin, Phys. Rev. 162, 699, 681 (1967).

[10]P. B. Peters and P. E. Shearin, Phys. Rev. 174, 691 (1968).

[11]C. Dimitrov and O. Dimitrov, Phys. Status Solidi (b) 34, 545 (1969).

[12]K. Papathanassopoulos and P. Andronikos, Radiat. Eff. 14, 215 (1972).

[13]A. Kraut, F. Dworschak, and H. Wollenberger, Phys. Status Solidi (b) 44, 805 (1971); D. E. Becker, F. Dworschak, and H. Wollenberger, Phys. Status Solidi (b) 47, 171 (1971).

[14]W. Koch, Diplom thesis, Physik-Department, Technische Universität München, 1973 (unpublished).

[15]S. Nasu and Y. Murakami, Phys. Status Solidi (b) 46, 711 (1971).

[16]R. S. Preston and R. Gerlach, Phys. Rev. B 3, 1519 (1971).

[17]K. Sørensen and G. Trumpy, Phys. Rev. B 7, 1791 (1973).

[18]C. Janot and H. Gibert, Phil. Mag. 27, 545 (1973).

[19]P. Rosner, W. Vogl, and G. Vogl, Nucl. Instrum. Methods 105, 473 (1972).

[20]W. Schilling, G. Burger, K. Isebeck, and H. Wenzl, in *Proceedings of the International Conference on Vacancies and Interstitials in Metals, Jülich, 1968,* edited by A. Seeger *et al.* (North-Holland, Amsterdam, 1970), p. 255; W. Schilling and K. Sonnenberg, J. Phys. F: Metal Phys. 3, 322 (1973).

[21]C. Dimitrov and O. Dimitrov, C. R. Acad. Sci., Ser. C 266, 304 (1968).

[22]D. G. Howard, R. H. Nussbaum, C. F. Steen, and A. Venkatachar, Bull. Amer. Phys. Soc. 16, 835 (1971).

[23]C. F. Steen, D. G. Howard, and R. H. Nussbaum. Solid State Commun. 9, 865 (1971), and 10, 584(E) (1972).