Nuclear gamma resonance on Fe^{57} nuclei on copper grain boundaries

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A fine structure was observed in the spectrum of the nuclear gamma resonance (NGR) on Fe^{57} nuclei located on the grain boundaries of pure copper. The character of the spectrum indicates that the Co^{57} occupy several different types of sites in the large-angle grain boundaries of copper.

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The known possibilities afforded by NGR were successfully used for the study of point defects and their groups.[1-3] This method, however, has not been used so far for the study of dislocations and grain boundaries, although isolated attempts of such investigations were in fact undertaken.[4,5]

We describe in this paper the first results of an investigation of large-angle grain boundaries of pure copper by the NGR method. Samples in the form of polycrystalline foils of copper 10×10^{-4} cm thick with average grain dimension $(2-4)\times10^{-4}$ cm were obtained by thermal evaporation of cathode copper of $4N$ purity. The Co^{57} (the parent nucleus of the Mössbauer Fe^{57} nucleus) was introduced into the grain boundaries by preferred intercrystalline diffusion[6] (temperature 385 °C, time 15 min). The depth of volume penetration ($(D_{\rm vol}t)^{1/2}$) was 5×10^{-8} cm, and the depth of boundary penetration was of the order of the sample thickness. After annealing, the remnants of the diffusion source and the volume diffusion zone were removed by etching. The quality of this stage of preparation of the samples was monitored by autoradiography. The standard used was an analogous metal foil in which the diffusion saturation of the Co^{57} was carried out at a temperature 780 °C for 2 h. Under these conditions $(D_{\rm vol}t)^{1/2}$ is of the order of the sample thickness and consequently practically all the Co^{57} nuclei were

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located in defect-free regions of the sample and occupied the crystal lattice sites. The emission NGR spectra were plotted with an YaGRS-4 spectrograph at constant velocity and at room temperature. The absorber was potassium ferro-cyanide enriched with Fe^{57}. The statistical measurement error did not exceed 0.1%.

Figure 1 shows the standard (volume) and grain-boundary NGR spectra measured under identical conditions. The standard NGR spectrum is a single Lorentz-shaped line whose parameters agree well with the published data.[7] In the grain-boundary NGR spectrum one can see a weakly resolved fine structure. The central and more intense part of the spectrum is close in its position to the line of the volume spectrum. Naturally, some of the Co^{57} atoms that occupy the lattice sites in the region of volume withdrawal $[(D_{vol} t)^{1/2} = 5 \times 10^{-8}$ cm] or places in the grain boundary that are close in

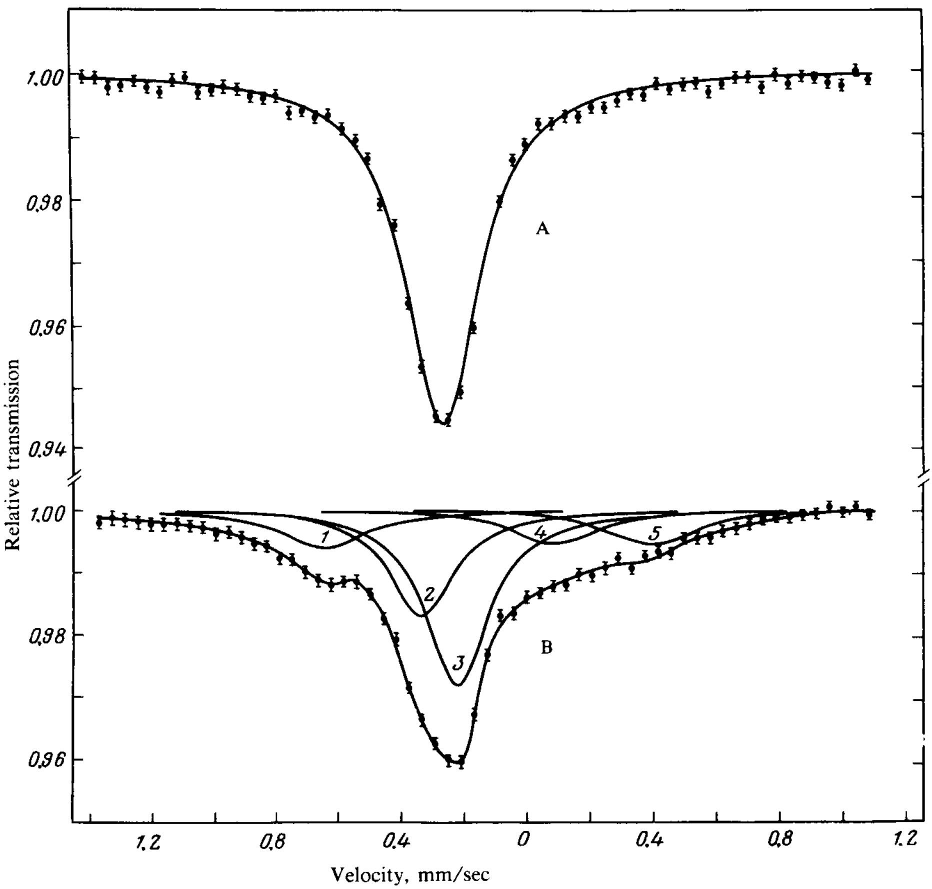

FIG. 1. Volume (A) and grain-boundary (B) NGR spectra in copper.

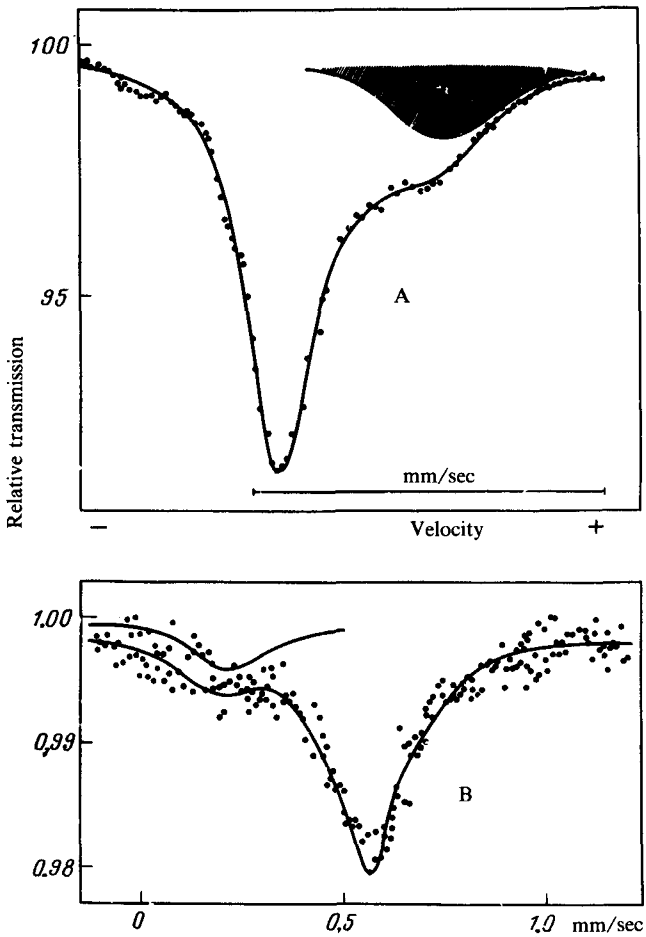

FIG. 2. NGR spectrum of Co^{57} in aluminum after bombardment by neutrons at 4.6 K and annealing at 41 K (A) (Ref. 2); NGR spectrum of Fe^{57} in aluminum quenched from a temperature 893 K (Ref. 3) (B).

symmetry and properties to the sites of the regular lattice, give rise to the most intense component of this spectrum–line 3.

Separation of this line from the grain-boundary NGR spectrum makes it possible to obtain four additional components. Lines 4 and 5 have identical intensity, this being a characteristic feature of the quadrupole doublet. The isomeric shift of the center of gravity of the doublet relative to the standard line amounts to $\Delta\delta^{b}_{4,5} = 0.5$ mm/sec, and the quadrupole splitting is $E^{b}_{4,5} = 0.3$ mm/sec. The positive isomeric shift indicates that the electron density at the Co^{57} nuclei in these positions is higher than in the sites of the regular lattice, this being apparently the consequence of the decreased atomic volume of the resonant nucleus, as revealed by experiments on the effect of high pressure on the isomeric shifts of the lines of the NGR volume spectra in copper.[8] Consequently the quadrupole doublet with positive isomeric shift is due to the Co^{57} nuclei located in the region of compression of large-angle grain boundaries.

The properties of such regions should be close to those of the interstices in the regular lattice, and the parameters of the corresponding NGR spectra should practically coincide. In fact, as shown in Ref. 2, in the NGR spectrum of a neutron-irradiated solid solution of Co^{57} in aluminum there appeared an additional "defect" line [Fig. 2) with an isomeric shift $\Delta\delta^{irr} = 0.4$ mm/sec and a quadrupole splitting $E^{irr} = 0.2$ mm/sec. The "defect" line, as shown in Ref. 2, is due to the Co^{57} nuclei produced by a mixed interstitial "dumbbell" with intrinsic inclusions.

Lines 1 and 2, which are located in the region of negative velocities, have different intensities and apparently reflect the presence of two other states of the Co^{57} nuclei in the boundary. Negative isomeric shifts ($\Delta\delta_1^b = -0.4$ mm/sec, $\Delta\delta_2^b = -0.1$ mm/sec), indicate that the electron density at the Co^{57} nuclei is lower in these positions than at the sites of the regular lattice, so that in these positions the atomic volume is larger than in the lattice sites. The properties of such grain-boundary regions can be close to the properties of clusters (small groups) of vacancies. As seen from Fig. 2, the NGR spectrum[3] of the quenched solid solution Fe^{57} in aluminum there appears a "defect" line with isomeric shift $\Delta\delta^{qu} = -0.35$ mm/sec. Since the quenching temperature in these experiments was close to the melting temperature, it follows that the predominant role in the quenched state was played not by single vacancies but by vacancy clusters produced on the residual impurities.[9]

The described qualitative treatment of the grain-boundary NGR spectrum is so far only one of several possibilities. Measurements of the Debye–Waller factor of all the components of the grain-boundary NGR spectrum, which are being carried out at the present time, yield more detailed information on the nature of the individual components of the grain-boundary NGR spectrum.

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[1]G. Volg, W. Mansel, and W. Vogl, J. Phys. F **4**, 2321 (1974).
[2]W. Mansel and G. Volg, J. Phys. F **7**, 253 (1977).
[3]H. Jchinose, K. Sassa, Y. Ishida, and M. Kato, Scripta Metal. **11**, 539 (1977).
[4]B.S. Bokshteĭn, Yu. V. Voĭtkovskiĭ, A.A. Zhukhovitskiĭ, and G.N. Nautkina, Fiz. Tverd. Tela (Leningrad) **10**, 3699 (1968) [Sov. Phys. Solid State **10**, 2940 (1968)].
[5]G. Martin, Phys. Status Solidi A **18**, 683 (1973).
[6]S.M. Klotsmann, A.N. Timofeev, and I. Sh. Trakhtenberg, Tr. Inst. Fiz. Met. Akad. Nauk SSSR No. 27, 275 (168).
[7]E. Fluck, W. Kerler, and W. Neuwirth, Angew. Chem. **75**, 461 (1963).
[8]G.M. Kalvius, U.F. Klein, and G. Wortmann, J. Phys. (Paris) Coll. **35**, 139 (1974).
[9]J. Levi, J.M. Lanore, and J. Hillairet, Philos. Mag. **28**, 373 (1973).