PHYSICAL REVIEW B VOLUME 19, NUMBER 12 15 JUNE 1979

Anisotropic scattering of conduction electrons on defects in quenched and irradiated Al

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(Received 20 December 1977)

We have measured scattering rates of electrons on lattice defects for a number of points on the Al Fermi surface using surface Landau-level resonance. Large, annealing-dependent, scattering anisotropy is observed which we explain by near-Bragg-reflection scattering on defect agglomerates in quenched and irradiated samples. The data indicate that vacancy agglomeration occurs at temperatures near 240 K.

I. INTRODUCTION

The understanding of the anisotropic scattering of conduction electrons on lattice self-defects has remained rather limited compared to our knowledge of scattering on impurities. The self-defects, such as quenched-in vacancies, radiation-induced Frenkel defects (vacancy and self-intersitial), and their agglomerates, have themselves been the subject of numerous studies. A wealth of information on the structure and kinetics of self-defects exists for Al, a metal with a relatively simple and well understood electronic band structure. Here we report a study of the anisotropic scattering of electrons on self-defects using surface Landau-level resonance (SLLR). It is the first direct measurement of local scattering rates on defects in Al, and on Frenkel defects in any metal.[1] An analysis of the scattering anisotropy as a function of annealing temperature reveals a previously unidentified defect restructuring, which we ascribe to agglomeration of vacancies.

II. EXPERIMENT

The SLL resonances are microwave transitions between surface bound states in low magnetic fields H, which correspond classically to electrons skipping along the sample surface, reaching a depth of approximately 10^{-5} cm.[2] The states that contribute to a given resonance are localized on the Fermi surface (FS). The relative width $\Delta H/H$ of the resonance is therefore related to the scattering rate of a very small group of electrons. Practically all scattering events, including small angle scattering, are effective, and contribute additively to the measured scattering rate.

Measurements of electron scattering rates were made for nearly-free-electron points A and B, which refer to centers of the spherical portions of the second zone FS, and for point c, which lies on a strongly curved portion of the third zone FS, near the Brillouin zone boundary[3] [Fig. 1(c)]. Apart from these three principal points on the FS, data will be given for point a, which has yet not been unambiguously identified on the FS, but can be assumed to also lie on a strongly curved portion.[3]

The Al samples used were discs (20-mm-diam × 5 mm), cut from a high-purity single-crystal boule.[4] One of the plane faces, oriented to be parallel to a (110) or to a (100) plane, was electropolished to permit observation of SLLR. The quenching was done by moving the samples in air from an oven at 473 K into a cold acetone bath at 180 K. The neutron irradiation (total fluence 7.2×10^{15} cm^{-2}), and the electron irradiation (4×10^{17} cm^{-2}), were performed below 10 K, and were followed by a rapid transfer to 77 K. Samples were stored at 77 K, measured at 4.2 K, and annealed at a number of temperatures for a holding time of 15 min.

Figure 1 shows the relative widths of the resonances measured during the isochronal annealing program for a quenched and a neutron-irradiated sample [both (110)]. Changes in the relative width $\Delta H/H$ are directly proportional to changes in the electron scattering rate Γ.[2] At our microwave frequency (35 GHz) one obtains $\partial\Gamma/\partial(\Delta H/H) = 1.5 \times 10^{11}$ sec^{-1}. It is surprising to see the rapid and large decrease and increase of $\Delta H/H$ in the annealing curves of the quenched sample for points c and a at temperatures below 240 K [Fig. 1(a)]. Equally striking is the large anisotropy involved, as points A and B show no corresponding changes. Similar behavior can be detected in the recovery after neutron irradiation [Fig. 1(b)]; the annealing curves for points A and B have constant slopes between 210 and 250 K, contrary to the sharp changes visible for points c and a.

These features have been seen in samples with both (110) and (100) polished surfaces, and with various irradiation doses, as is shown in Fig. 2 where the annealing curves for point c are given. Also included are the c-point data for an electron-irradiated sample. The unusual increase of scattering as a result of annealing can be seen for a neutron-irradiated sample [Fig. 2(f)] which

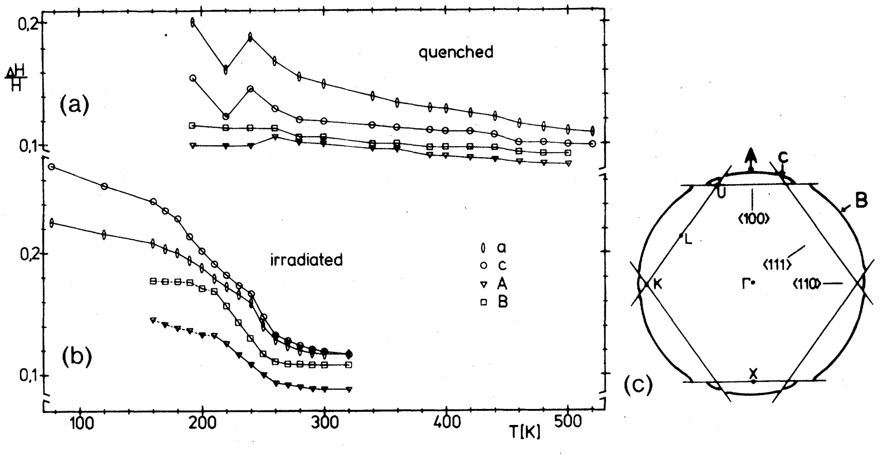

FIG. 1. Relative widths of surface Landau-level resonances for four points on the FS of Al vs isochronal annealing temperature for quenching from 473 K (a) and neutron irradiation at 4.6 K (b). Induced defect concentrations are approximately 1 ppm. The values of $\Delta H/H$ at the highest temperatures represent complete recovery. Experimental uncertainties are smaller than symbols; the broken line indicates a slightly larger error resulting from uncertainty in line-shape analysis. The locations of three points are shown on the central (110) slice of the Al-FS (c).

warmed up very slowly between 230 and 270 K. A similar shift to higher temperatures of this scattering peak was observed in a quenched sample for which only one annealing step was made below 250 K [Fig. 2(a)].

III. DISCUSSION

New information is contained in the scattering anisotropy. The SLLR method can differentiate between defect structures provided they cause distinguishable scattering anisotropies. All scattering events contribute additively to the scattering rates; scattering on a particular defect structure contributes in a varying degree to the scattering rates of various points on the FS. This scattering anisotropy is most clearly apparent when, as a result of annealing, this defect structure is removed. The ratio $\Delta\Gamma_i/\Delta\Gamma_j$ of the resulting changes in scattering rates for two points i,j on the FS will be referred to as an anisotropy ratio; it is representative of the defect structures that change or anneal out at a particular temperature.

As an example of such an anisotropy ratio we show in Fig. 3 $\Delta\Gamma_a/\Delta\Gamma_c$ for three samples. Vacancies in quenched samples appear to be characterized by an anisotropy ratio of approximately 1.2 over a temperature range in which the scattering changes drastically. In neutron-irradiated samples the increase in $\Delta\Gamma_a/\Delta\Gamma_c$ above 200 K towards a value of 1 can be explained by assuming that also here a process involving vacancies contributes to the changes in scattering rates. The same applies for the electron-irradiated sample; however, the process only begins above 230 K. This is consistent with the higher local density of vacancies in neutron-irradiation cascades.[5] The data of Figs. 1, 2, and 3 imply that in both neutron-irradiated and in quenched samples the vacancies undergo a process for which the scattering rate first decreases, then increases, then

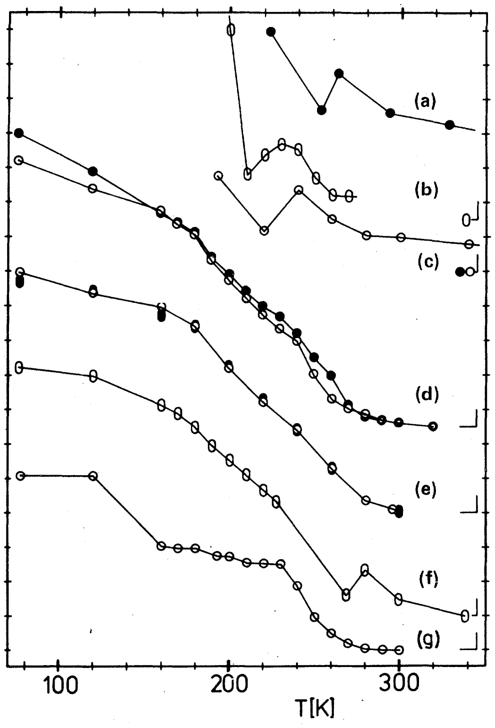

FIG. 2. Annealing curves, point c, for samples that were quenched (a)–(c), neutron irradiated (d)–(f), and electron irradiated (g). The sample of Fig. 1(b) was irradiated with the full neutron dose (d○), and with a half dose (d●, curve scaled by factor 2). Two samples with different orientations are compared in (e) [○(110), ●(100)]. To the far right the fully annealed values are shown.

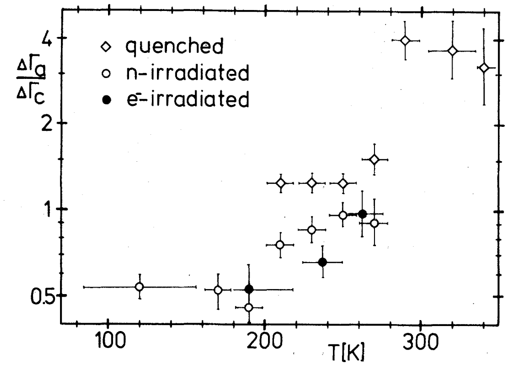

FIG. 3. Anisotropy ratio for two points on the FS near Brillouin zone boundaries, in the quenched sample of Fig. 1(a), and in irradiated samples. An average was taken from three neutron irradiations.

decreases again with annealing. In neutron-irradiated samples this scattering peak appears superimposed on a smoothly decreasing scattering rate, such as those of points A and B. In the following we will discuss a model in which the smooth decrease of scattering is due to recombination of vacancies with interstitials, and the scattering peak results from vacancy agglomeration. The presence in these samples of small vacancy clusters, such as mobile divacancies, below 200 K is a supposition of such a model.

A description of how agglomeration can cause large changes of highly anisotropic scattering rates requires a discussion of electron scattering. The basis for the different scattering rates of electrons are the differences in the electron wave functions; the actual anisotropy depends on the scattering potential and on the displacement of lattice ions due to the defect. A single vacancy causes hardly any lattice relaxation, and using the Al pseudopotential to describe it, one can show that all anisotropy ratios are close to 1. If several vacancies are clustered together, scattering with a scattering vector q very close to 0 or to a reciprocal-lattice vector G_i is enhanced above other scattering,[6] in a way that is closely analogous to small angle diffuse x-ray scattering.[7] In an integral over all possible q's, scattering for $q \sim G_i$ will be particularly effective, and will dominate the scattering rates for points on the FS near Brillouin zone boundaries, such as a and c. With progressing agglomeration, the vacancy cluster becomes restructured into a vacancy loop, and sizable lattice relaxation develops. As a result the small angle scattering on the vacancy loop will be sharply reduced, while the Huang scattering on the lattice displacement field will increase. Since destructive interference between the two scattering components can cause many of the scattering matrix elements to become very small, the scattering rate can go through a sharp minimum [e.g., Fig. 2(b) at 210 K], before increasing with further lattice relaxation. As loop size increases, the Huang scattering will become restricted to very-near-Bragg reflections. As a result, the enhanced scattering rates will decrease again, and their remaining changes will be strongly anisotropic. Also, the inversion of the anisotropy ratio for large vacancy loops ($\Delta\Gamma_a/\Delta\Gamma_c > 1$ at 300 K) compared to large interstitial loops ($\Delta\Gamma_a/\Delta\Gamma_c < 1$ at 120 K) as seen in Fig. 3, is analogous to the defect-asymmetric character of Huang scattering.[7] For points A and B which are far from Brillouin zone boundaries Huang scattering is of little importance; therefore, the scattering rates will decrease monotonically, primarily due to the restructuring of clusters into loops.

IV. SUMMARY

The agglomeration of vacancies into loops around 250 K in quenched Al is well known; its earlier identification in neutron-irradiated samples, however, has been hindered by the simultaneous defect recombination. The influence of defect agglomeration and restructuring on the resistivity cannot be distinguished from the effect of removing defects. This might be the reason why so many questions have arisen in explaining the recovery of the resistivity in irradiated and quenched Al.[5] The close correspondence of the scattering anisotropy in electron- and neutron-irradiated samples with that of quenched samples allows the specific agglomeration of vacancies to be identified. The anisotropy of scattering also demonstrates for the first time the direct analogy between electron scattering and small angle or Huang scattering of x rays. A detailed comparison of the experiments with a theoretical calculation is in progress.[6] The measurement of highly anisotropic scattering on agglomerates of self-defects is an example of the new information the SLLR method promises.

ACKNOWLEDGMENTS

We would like to express our gratitude to J. F. Koch, who initiated the project. We are indebted to R. E. Doezema, K. Böning, and K. Pfändner for their advice regarding experimental techniques, and for many useful discussions. The work was supported by the Bundesministerium für Forschung und Technologie within the project "Nukleare Festkörperforschung."

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*Based on a doctoral thesis to be submitted in partial fulfillment of the requirements for the degree Dr. rer. nat (Ph.D).

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