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Annealing of defects in $AuIn_2$ after irradiation with electrons at low temperatures

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Abstract. $AuIn_2$ films were irradiated below 10 K with electrons of four different energies (0.65 to 2.5 MeV) up to an irradiation dose of 3.56×10^{19} e^- cm^{-2}. The irradiated samples were isochronally annealed and the recovery of the radiation damage was investigated by measuring the residual electrical resistivity. Characteristic for the recovery of the resistivity is a large narrow stage centred at 100 K of which the position is independent of electron dose and electron energy. Only the height of this stage increases with decreasing irradiation dose and electron energy.

1. Introduction

The experimental finding [1, 2] that crystalline $AuIn_2$ can be forced into the amorphous state by bombardment with light and heavy inert ions at 10 K and that the previously amorphized sample is transformed back into its crystalline phase after annealing to 150 K triggered the present investigation. Can initial stages of the amorphous state be observed after irradiation with energetic electrons at low temperatures and does the recrystallization occur at a temperature where point defects become mobile?

Annealing of $AuIn_2$ films, that were irradiated with 230 keV protons or 270 keV helium ions to such a low defect concentration that they became only partially amorphous, revealed a more or less continuous recovery of the electrical resistivity starting from 10 K with two broad transitions regions at 100 K and 200 K, respectively [3]. Broad recovery stages are expected in the case of ion irradiation since ions produce a spectrum of recoil energies so that the defect structure consists of a distribution of single point defects up to large defect cascades.

In the case of high energy electron irradiation the transferred energy is low enough to produce mainly single point defects. Such an initial defect state is an essential requirement for the study of the behaviour of single point defects. However, in case of intermetallic compounds the interpretation of the damage and its recovery can be still complicated due to the production of defects in two or more sublattices in addition to the production of antisite defects.

$AuIn_2$ crystallizes in the CaF_2 structure. In this structure each Au atom sits in the centre of a cube formed by the In atoms, but every second In cube is empty. Therefore, this compound is not densely packed.

2. Experiment

The sample material $AuIn_2$ was obtained by melting weighted amounts of high purity Au (99.999%) and In (99.9999%) in a graphite crucible for 10 min at 800 °C in an argon

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atmosphere. The composition of the compound $AuIn_2$ was confirmed by powder diffraction measurements of a ground piece of the ingot. Thin films of $AuIn_2$ were produced by flash-evaporating powdered $AuIn_2$ in a vacuum of 7×10^{-6} mbar on Si plates (4×15 mm^2), onto the ends of which 1 μm thick platinum pads had been sputtered. Before this process four 100 μm thick platinum wires for electrical resistance measurements were spotwelded onto these pads. Before the specimens were mounted into the irradiation and annealing cryostat, details of which are given in [4], they were annealed for two hours at 200 °C in a vacuum of 1×10^{-5} mbar. Diffraction measurements of the annealed specimen showed the same diffraction pattern [5] as that of polycrystalline $AuIn_2$.

The irradiated samples were isochronally pulse annealed in the upper part of the irradiation cryostat for fifteen minutes periods with temperature increments $\Delta T/T = 0.05$. The recovery of the radiation damage was investigated by electrical resistivity determinations performed at 4.2 K. Resistivities, ρ, were derived from the electrical resistances, R, by the relationship $R = k\rho$. The shape factor, k, was calculated from the geometric dimensions of the sample $k = L/wt$, with L the distance between the pads ($L = 10$ mm), w the width of the Si plates ($w = 4$ mm) and t the thickness of the films. Since t could not be determined sufficiently well either mechanically or by the evaluation of Rutherford backscattering measurements it was obtained by measuring and evaluating the temperature dependence of the electrical resistance, $R(T)$. According to Jan and Pearson [6] the resistivity increases linearly with temperature in the temperature interval 60 K to 300 K and amounts to $\mathrm{d}\rho(T)/\mathrm{d}T = 0.027$ $\mu\Omega$ cm K^{-1}. From the relationship

$$\frac{\mathrm{d}R(T)}{\mathrm{d}T} = \frac{L}{wt}\frac{\mathrm{d}\rho(T)}{\mathrm{d}T}$$

the thickness t can be calculated from a measurement of resistance versus temperature.

3. Experimental results

Four sets of samples were irradiated at temperatures below 10 K with 2.5 MeV electrons to different irradiation doses Φ and another three sets with electrons of three different lower electron energies E. (See table 1.) The radiation induced residual resistivity changes $\Delta\rho_{irr}$, residual resistivities ρ_0, thickness t and the maximum transferred energies T_{max} to Au and In, respectively, are also listed in table 1.

The electron energies are nominal values given by the terminal voltage of the accelerator. Due to multiple scattering of the electrons in the stainless-steel window and the cooling agent (liquid helium) the electrons lose energy. Only upper and lower limits of this energy loss can be calculated since the fraction of vaporized helium in the cooling agent is not known well enough. An upper limit of the energy loss of 0.2 MeV is obtained when the composition of the cooling agent is 100% liquid helium and the lower limit is 60 keV when the samples are cooled with 100% helium vapour at a temperature of 10 K. The irradiation dose Φ given in table 1 was obtained from the electron flux measured just before the beam entered the cryostat. Damage rates were not calculated since the electron flux at the position of the samples could not be measured, and corrections can be estimated only with a very large systematic error, especially for low energy electrons.

The isochronal recovery of the residual resistivity increase $\Delta\rho_{irr}$ induced by 2.5 MeV electron irradiation and its logarithmic derivative are shown in figures 1(a) and 1(b) as a function of annealing temperature for four different electron doses. From 10 K to 80 K $\Delta\rho_{irr}$ recovers gradually. The amount of the recovery in this temperature interval increases slightly with dose. The recovery of the samples irradiated to $\Phi = 0.54 \times 10^{19}$ e$^-$ cm^{-2}

Table 1. Typical parameters of irradiated samples.

E [MeV]	Au: T_{max} [eV]	In: T_{max} [eV]	Φ [10^{19} e^- cm^{-2}]	$\Delta\rho_{irr}$ [$\mu\Omega$ cm]	r_0 [$\mu\Omega$ cm]	t [nm]
2.5	88.8	152.2	3.56	5.645	1.964	121
2.5	88.8	152.2	1.58	3.413	1.721	142
2.5	88.8	152.2	0.54	1.420	4.325	51
2.5	88.8	152.2	0.134	0.348	1.837	222
1.0	20.3	34.8	1.10	1.538	1.097	246
0.8	14.6	25.0	1.83	1.795	1.024	284
0.65	10.8	18.6	0.71	0.356	1.794	199

is anomalous because it shows a much larger recovery up to 80 K than expected from the dose dependence of the other samples. This effect seems to be due to the extreme small thickness of the samples since it also occurs in other ultra-thin samples ($t \leqslant 50$ nm) irradiated to higher doses of 2.5 MeV electrons [7]. Unfortunately no thicker sample was irradiated to a dose of $\Phi = 0.54 \times 10^{19}$. No further conclusions can be drawn from this effect. Characteristic for the recovery of all samples is a large narrow stage centred at 100 K in which a large fraction of the defects anneal. The temperature position of this stage does not shift with dose as can be seen in figures 1(*b*). The amount of recovery of $\Delta\rho_{irr}$ lies between 37.5 and 62% and increases with the defect concentration. Two subsequent small recovery stages can be observed, one at 160 K and the second one at 210 K. Neither the position nor the height of these stages depend upon dose.

In figures 2(a) and 2(b) the isochronal recovery and its logarithmic derivative are shown for four different electron energies. The temperature dependencies of the annealing curves are qualitatively similar. The dependence of the recovery on the electron energy is somewhat modified by its dependence on the defect concentration, i.e. on $\Delta\rho_{irr}$ (for $E = 2.5$ MeV the recovery curve with the lowest $\Delta\rho_{irr}$ is plotted in figures 2(a) and 2(b)). With 0.8 MeV and 1.0 MeV electrons the samples were irradiated to higher defect concentrations than the other samples. Nevertheless, with decreasing electron energies the recovery of $\Delta\rho_{irr}$ occurs more and more in the stage centred at 100 K. The temperature position of this stage does not shift with electron energy as can be seen in figure 2(b). Only the height of this stage increases with decreasing electron energy, so that practically all of the radiation induced residual resistivity increase $\Delta\rho_{irr}$ anneals out in this stage in the case of an irradiation with 0.65 MeV electrons.

4. Discussion

An atomistic interpretation of the recovery of the radiation induced residual resistivity increase $\Delta\rho_{irr}$ in $AuIn_2$ is not possible because several types of vacancy and interstitial with different spatial distributions in addition to antisite defects can be produced in the Au and the In sublattice by the electron irradiation. Therefore, it is remarkable that $\Delta\rho_{irr}$ recovers mainly in one single stage. Assuming that different types of Frenkel defect are characterized by different migration energies and hence by different recovery stages we can conclude that electron irradiation produces mainly one type of Frenkel defect in $AuIn_2$ recovering at 100 K. Since the peak position of this stage does not depend on the initial concentration of defects the annealing of these defects can be described by a first order process. In analogy to the recovery in pure metals, it seems plausible to assign the 100 K stage to the recovery of a close Frenkel pair restoring antisite defects at the same time.

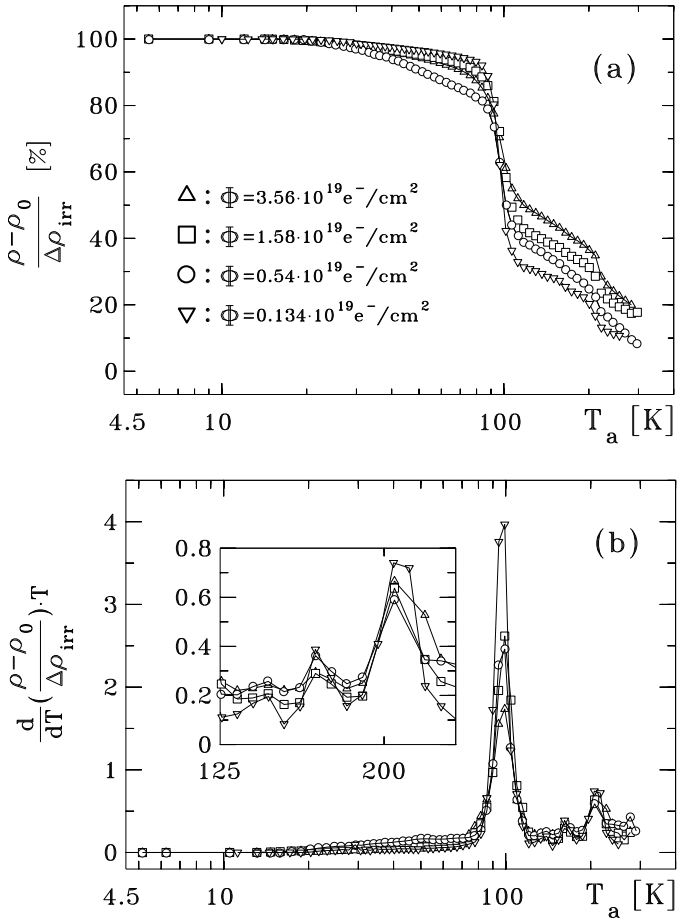

Figure 1. Isochronal resistivity recovery of $AuIn_2$ samples irradiated with 2.5 MeV electrons (a) and logarithmic derivative curves (b) as a function of annealing temperature for four different electron doses.

With 2.5 MeV electrons additional defect species are produced that anneal at higher temperatures. There is no indication for a free migration of these defects since the peak positions of the stages at 160 K and 210 K do not depend on the initial concentration of defects.

The open lattice structure, every second In cube being empty, and the mass differences between Au and In atoms could be the reasons for short displacement collision sequences and as a consequence for the dominant production of close Frenkel pairs in $AuIn_2$.

The maximum value $\Delta\rho_{irr}$ which has been reached (see table 1) amounted to only 6% of the resisitivity of the amorphous phase of $AuIn_2$ [8]. Therefore, without increasing the irradiation dose at least by a factor of ten, it cannot be decided whether irradiation with energetic electrons at low temperatures produces the amorphous state or not.

5. Conclusion

Electron irradiation of thin polycrystalline $AuIn_2$ films creates predominantly one type of Frenkel defect that anneals in a recovery stage centred at 100 K which does not shift with

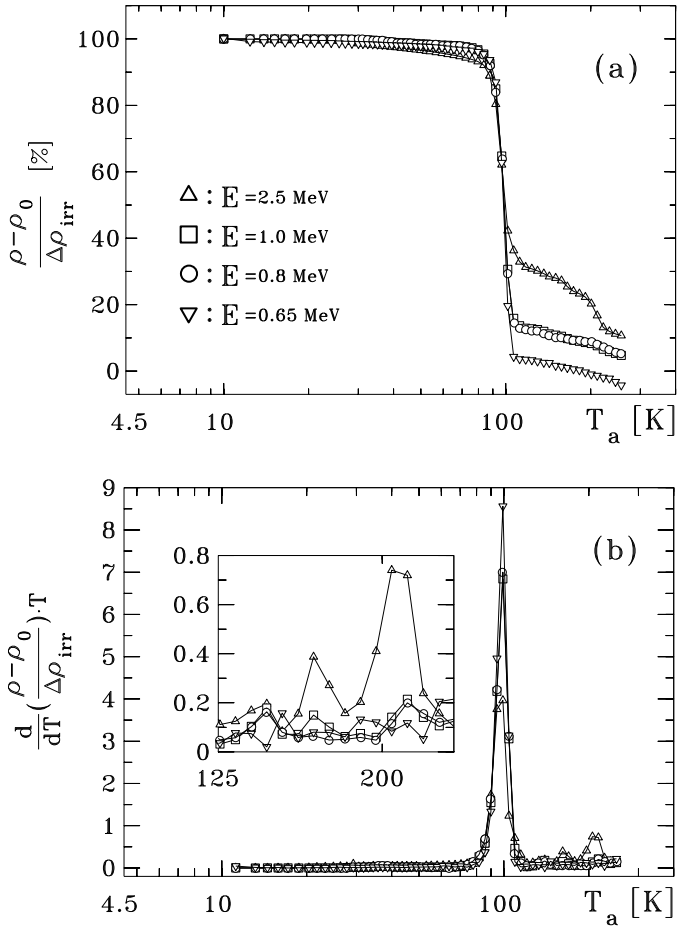

Figure 2. Isochronal resistivity recovery (a) and its logarithmic derivative (b) as a function of annealing temperature of $AuIn_2$ samples irradiated with four different electron energies.

electron dose. In analogy to the recovery in pure metals we can assign this stage to the recovery of a close Frenkel pair. Only the height of this stage increases with decreasing irradiation dose and decreasing electron energy. There is no evidence for a freely migrating defect.

References

[1] Miehle W, Plewnia A and Ziemann P 1991 *Nucl. Instrum. Methods* B **59/60** 410
[2] Ziemann P, Miehle W and Plewnia A 1993 *Nucl. Instrum. Methods* B **80/81** 370
[3] Miehle W 1992 *Dissertation* University of Konstanz
[4] Schroeder H and Schilling W 1976 *Radiat. Effects* **30** 243
[5] Zintl E, Harder A and Haucke W 1937 *Z. Phys. Chem.* B **35** 354
[6] Jan J P and Pearson W 1963 *Phil. Mag.* **8** 279
[7] Eppler I 1994 Private communication
[8] Häussler P 1983 *Z. Phys.* B **53** 15