

LONG RANGE EFFECT OF ION IRRADIATION ON DIFFUSION COEFFICIENT OF Ni IN SINGLE CRYSTAL AND POLYCRYSTALLINE COPPER

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Abstract. 1 μm Ni films were deposited on both single crystal and polycrystalline Cu disks by magnetron sputtering. Diffusion at 420 ± 3 °C was performed on the Ni/Cu couples with Ar ion irradiation at the Cu side of samples. Diffusion of samples without irradiation were performed for comparison. The Ni concentration distribution profiles analyzed by glow discharge optical emission spectroscopy (GDOES) revealed an enhanced Ni diffusion in single crystal Cu substrate 1 mm away from the irradiated surface, which can be explained by a mechanism of nonlinear lattice vibration. No diffusion enhancement identified in polycrystalline Cu indicates the experimental observation of grain boundary attenuation on the nonlinear lattice vibration modes.

1. INTRODUCTION

All condensed matter processes involving high energy density or transport of energy can be impacted by vibrational energy localization in extended lattice [1], such as deformation, crack propagation, irradiation and diffusion. It is generally postulated intrinsically localized vibrational modes (ILMs) or so called discrete breathers (DBs) in lattice is induced by the matching of the nonlinear atomic force and the discreteness of the lattice [2]. Besides the intense theoretical interests, which were reviewed in [3] and [4], experimental results of spectroscopical studies in PtCl based crystal [1], α -helix [5], bcc ⁴He [6] and *etc.* showed the existence of ILMs in crystal lattices.

Both stochastic fluctuation of atomic motion and incident energetic particle irradiation can excite nonlinear interatomic force and then ILMs, but with

some differences. The incident particles introduce extra energy and momentum which can be adjusted, and the irradiation induced ILMs shows higher mobility [7]. Molecular dynamic simulation in layered crystals revealed a directional transfer of ILMs without attenuation [8]. Research works in atomic disorder in radiated single crystal silicon [9] and ejection of kalium atoms in single crystal mica [10] showed long range effect by irradiation, which is in accordance with the transport behavior of ILMs. Ar ion irradiation enhanced Ni diffusion in single crystal Cu beyond the irradiation effect zone in classical theory was found and the enhanced diffusion effect attenuates along with the distance between the diffusion zone and the irradiated Cu surface [11]. It is speculated the ILMs may be scattered by extended defects such as grain boundaries [12] but no experimental evidence has been found.

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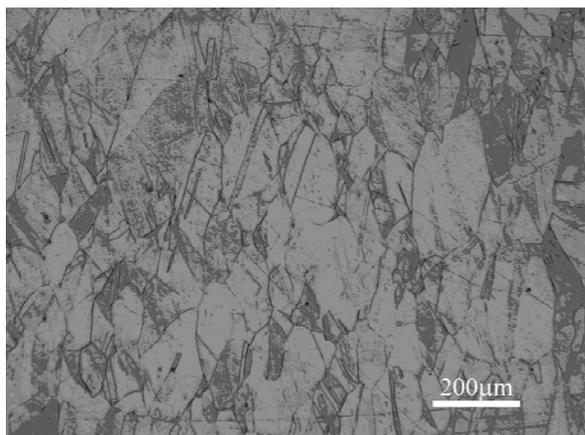


Fig. 1. The structure of polycrystalline Cu.

We employ in this work polycrystalline Cu in the pathway of the ILMs induced by ion irradiation to investigate the influence of grain boundaries on the transport of the ILMs.

2. EXPERIMENTAL SECTION

1.0 mm thick discs, 8 mm in diameter, were cut from commercial T3 copper rods and directional solidified single crystal Cu rods, respectively. Fig. 1 shows the optical microstructure of T3 Cu along the axial direction. The mean grain size of is 150 μm .

The disks were mechanically polished to a final roughness of ~ 15 nm, dilute sulfuric acid immersed, distilled water flushed and then ultrasonic cleaned in acetone. 1 μm thick Ni film was deposited on one side of the Cu disks by magnetron sputtering at room temperature after 500 eV argon ion sputtering for 10 min.

The detailed setup for the diffusion experiments can be seen in Fig. 2. Ar ion irradiation was performed by a Kaufman-type ion source of 3 cm diameter on the ceiling of the vacuum chamber. A sample holder of $\phi 35 \times 6$ mm with a small hole for setting thermocouple was below the ion source. Two Cu-Ni couples were stacked on the sample holder. The copper side of the upper sample faced to the ion source and endured Ar ion irradiation. The other one in thickness of 1 mm was annealed as control sample, whose copper side faced to the holder. A 0.5 mm thick graphite barrier was inserted between two samples to prevent diffusion between Ni films. Samples were heated by resistance wire around by thermal radiation. The diffusion temperature was 420 ± 3 °C and the diffusion time was 20 hrs for

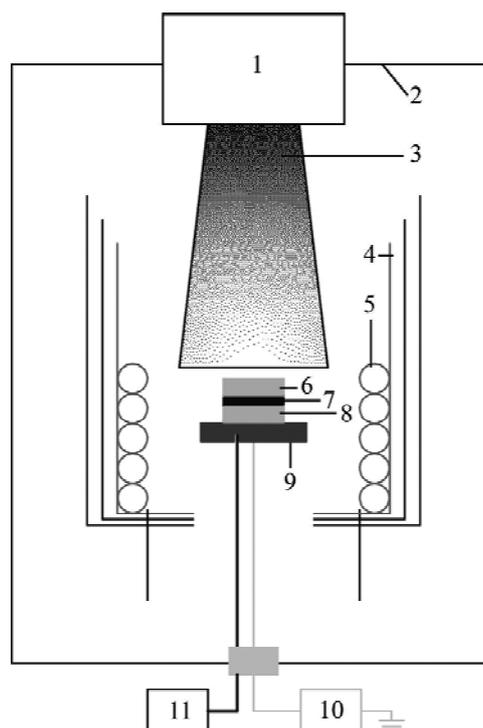


Fig. 2. The setup for the diffusion experiments. 1 - ion source; 2 - vacuum chamber; 3 - ion beam; 4 - radiation shields; 5 - resistance wire in insulating tubes; 6 - sample with ion irradiation; 7 - graphite barrier; 8 - annealed sample; 9 - sample holder; 10 - milliammeter; 11 - thermocouple.

polycrystal Cu substrate samples and 18 hrs for single crystal Cu substrate samples, respectively. The base pressure was 5×10^{-4} Pa, and the working pressure was 3×10^{-2} Pa. The Ar ion energy was 1500 eV and the ion current density was 0.4 mA/cm².

Simulation shows the temperature of sample with irradiation is no more than 4 °C higher than those without irradiation. Ni concentration depth profiles were measured by glow discharge optical emission spectroscopy (GDOES).

3. RESULTS AND DISCUSSION

The Ni concentration depth profiles of annealed and irradiated polycrystal Cu substrate samples after diffusion at 420 ± 3 °C for 20 hrs are indicated in Fig. 3a. The Ni concentration depth profiles of annealed and irradiated single crystal Cu substrate samples after diffusion at 420 ± 3 °C for 18 hrs are indicated in Fig. 3b.

An adaptation of Hall *et al.* model [13] was used to determine the diffusion coefficient of Ni in Cu. In this model, the average concentration C in the plane parallel to the surface can be expressed as

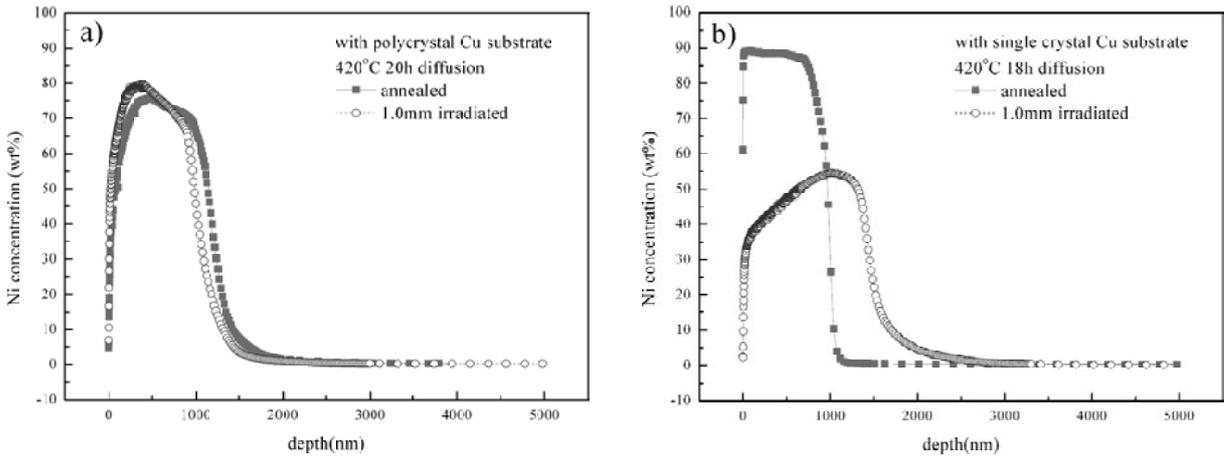


Fig. 3. Ni concentration profiles in Cu-Ni couples after diffusion at 420 °C employing polycrystal Cu for 20 hrs (a) and single crystal Cu for 18 hrs (b), respectively.

$$\frac{C}{C_0} = \frac{8(Dt)^{1/2}}{L\pi^{1/2}} C + \operatorname{erf} \left[\frac{x}{2(Dt)^{1/2}} \right] \times \left[1 - \frac{8(Dt)^{1/2}}{2\pi^{1/2}} \right], \quad (1)$$

where C_0 is the composition at the interface (assumed to be independent of time), D is the diffusion coefficient, t is the diffusion time, L is the grain size and x is the distance to the interface.

Considering the Cu side of the diffusion couple, the grain size of polycrystal Cu, 100 μm , is much bigger than the diffusion distance $(Dt)^{1/2}$, which can be seen in Fig. 3a. As a consequence, Eq. 1 can be simplified to be an error function. Considering the temperature accuracy and the temperature nonuniformity between samples with and without irradiation, the errors of diffusion coefficients were evaluated by the Arrhenius equation

$$D = D_0 \exp(-Q/kT). \quad (2)$$

The activation energy Q and the pre-exponential factor D_0 of Ni diffusion in Cu were employed from [14-18]. The calculated diffusion coefficients were listed in Table 1.

The higher Ni diffusion coefficients after annealing in polycrystalline Cu than in single crystal Cu is attributed to the grain boundaries in polycrystal Cu as fast diffusion path.

The enhanced Ni diffusion in single crystal Cu after diffusion with backside irradiation is in accordance with the results obtained at lower temperature [11], the mechanism of which was the nonlinear vibrational modes induced by the ion irradiation propagated to the rare surface and decreased the local vacancy migration energy.

There is no evident Ni diffusion enhancement in polycrystalline Cu induced by backside irradiation, which can be explained by a mechanism based on the interaction between the ion irradiation induced ILMs and the structural defects. The ILMs induced by irradiation can transport in a large scale without energy attenuation because their frequencies are higher than and do not resonate with normal, harmonic lattice vibrational modes [7,12,19]. The periodical condition is compromised at grain boundaries and the vibrational modes are associated with the local structure of grain boundaries. The ILMs could be scattered by such modes because of similar frequency, lost its energy to create lattice defects or phonons and eventually vanished, leading to the absence of enhanced diffusion.

Table 1. Calculated diffusion coefficient of Ni in Cu in the process of treatment.

Sample No.	Structure	Diffusion time [h]	Irradiation	Lower limit of D[cm ² /s]	Calculated D [cm ² /s]	Upper limit of D[cm ² /s]
1	polycrystal	20	annealed	2.9×10 ⁻¹⁵	3.4×10 ⁻¹⁵	4.0×10 ⁻¹⁵
2	polycrystal	20	irradiated	2.8×10 ⁻¹⁵	4.2×10 ⁻¹⁵	5.0×10 ⁻¹⁵
3	single crystal	18	annealed	2.3×10 ⁻¹⁶	2.7×10 ⁻¹⁶	3.2×10 ⁻¹⁶
4	single crystal	18	irradiated	4.3×10 ⁻¹⁵	6.4×10 ⁻¹⁵	7.6×10 ⁻¹⁵

4. SUMMARY

The Ni film diffusion behavior into both polycrystalline and single crystal Cu substrates under backside Ar ion irradiation were investigated. The enhanced diffusion of Ni in single crystal Cu can be attributed to the long range propagation of nonlinear vibrational modes induced by ion irradiation, while the normal diffusion in polycrystalline Cu due to the attenuation of the nonlinear modes by the grain boundaries.

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