Thermal stability of Pb-alloy Josephson junction electrode materials. II. Effects of SiO coating on Pb-In-Au base electrode

Masanori Murakami

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Thermal stability of Pb-alloy Josephson junction electrode materials: II. Effects of SiO coating on Pb-In-Au base electrode

Masanori Murakami

IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

The preceding paper pertains to studies which were carried out for Pb-In-Au films without overlying SiO layers. Because use is made of configurations in Josephson junction integrated circuits presently under investigation in which Pb-In-Au base-electrode films are coated by overlying layers of SiO, a study was undertaken of the effects of such layers on the strain behavior of Pb-In-Au films using an x-ray diffraction technique. Pb-12 wt. % In-4 wt. % Au films were prepared at 298 or 77 K to obtain large or fine grains, respectively, and then thermally cycled between 300 and 4.2 K and between 300 and 350 K. In large-grained Pb-In-Au films, the strain relaxation, upon thermal cycling, by dislocation glide or by grain-boundary diffusion creep, was observed to be significantly inhibited by overlying SiO layers. However, in fine-grained films, that was not observed. For junctions in which the tunnel oxide is formed within an opening in an overlying SiO layer, it is believed, e.g., that upon heating, a compressive strain gradient develops in the large-grained base-electrode films between the junction area and its surrounding, leading to base-electrode deformation inside the opening that could rupture the tunnel oxide. This study indicates that the strain gradient should be reduced by reducing the average grain size of the base-electrode films. These results agree well with the previous experimental results in which no hillocks were detected by scanning electron microscopy inside similar SiO-layer openings above fine-grained Pb-In-Au films during repeated thermal cycling between 300 and 4.2 K or upon heating from 300 to 343 K.

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I.INTRODUCTION

In previous paper,[1] to reduce the strain relaxation of the base-electrode material during low-temperture thermal cycling, *in situ* strain measurement and strain-relaxation-mechanism analysis were carried out for Pb-12% In-4% Au films that were deposited at 298 or 77 K and then repeatedly thermally cycled between 300 and 4.2 K. It was found that grain size and film thickness were the key factors affecting strain relaxation. By reducing grain size and/or film thickness, the strain relaxation was reduced to near zero and a significant improvement in the device cyclability between 300 and 4.2 K was obtained in junctions fabricated using a thinner or fine-grained base electrode.[2]

The previous paper[1] pertains to studies which were carried out for Pb-In-Au films without overlying SiO layers. In interferometer devices, 0.2-μm-thick base-electrode films are coated by 0.4-μm-thick SiO layers to insulate the electrodes from the counter electrodes except at the "windows" (opening down to 2.5-μm diam.)[3] at which the junctions are defined.

The main purpose of the present study is to examine the effects of such SiO layers on the strain behavior during thermal cycling between 300 and 4.2 K or between 300 and 350 K of Pb-12% In-4 % Au films. The films were 0.2-μm thick, a thickness similar to that used in Pb-alloy Josephson junction base electrodes. The effects were studied by an x-ray diffraction technique for large-grained Pb-In-Au films prepared by deposition at room temperature and for fine-grained films prepared by deposition at 77-K substrate temperature.[1,2] The present results were compared with a previous hillock-formation experiment which was carried out by (SEM) scanning electron microscopy.[2]

II. EXPERIMENTAL PROCEDURES

The preparation procedures of Pb-In-Au films on oxidized Si substrates are the same as those described in previous papers.[1,2] Two substrate temperatures T_s of 298 and 77 K were used to obtain the films with different average grain sizes g. The grain sizes of 0.2-μm-thick films were $g = 0.18$ μm for films prepared at $T_s = 298$ K and $g = 0.08 \mu$m for films prepared at $T_s = 77$ K.[1] The thickness of each layer was measured by a calibrated quartz thickness monitor during film deposition. The total thickness of each film was 0.2μm, and the nominal concentration was 4wt % Au, 12wt. % In, and 84wt % Pb. After the Pb-In-Au films were prepared, the SiO films were deposited in a vacuum of $\sim 2 \times 10^{-5}$ Torr using a resistance-heated source. The depositions were carried out at a substrate temperature of room temperature using Ga backing. The deposition rates were ~ 1 nm/s. The SiO films prepared in this manner were reported to have low stress levels ($\sim 2 \times 10^8$ dyn/cm^2) upon deposition,[4] and their intrinsic stress was believed not to influence the present experimental results. The SiO layer thicknesses were in the range of 0.05 to 0.5 μm.

Thermal treatments of specimens were carried out using a cold or hot stage attached to the x-ray diffractometer. The enviroment of a specimen was evacuated to below 1×10^{-6} Torr. Before further thermal treatments, the samples were kept at 300 (± 0.5) K in the cold or hot stage for several days to permit equilibrium to be established. The

cooling time from 300 to 4.2 K was about 5 min and the heating time from 4.2 to 300 K was ~20 min. The heating time from 300 to 350 K was about 5 min and the cooling time from 350 to 300 K was about 6 min.

X-ray diffraction intensity measurements were made by a computer-controlled x-ray diffractometer. The CuKα radiation (operated at 40 kV and 20 mA) was used, and monochromatic diffraction intensities were obtained by using a singly-bent graphite monochromator. The diffracted intensities were step scanned for sufficient times at an interval of $2\theta = 0.01°$ for a predetermined diffraction angle range.

III. EXPERIMENTAL RESULTS
A. Thermal cycling between 300 and 4.2 K

Effects of SiO coating on the strain behavior of Pb-In-Au films during thermal cycling between 300 and 4.2 K were studied by measuring (333) interplanar spacing D in a Pb-In-Au film normal to the film surface by an x-ray diffraction technique. The calculated and measured D values are shown in Fig. 1. In this figure the D^0 denotes the D values of the strain-free film, which are calculated using the reported thermal-expansion coefficients of Pb (Ref. 5); and D_{cool}^{max} denotes the D values of the maximum-strained film, which are calculated for (111) oriented grains using a biaxial strain model[6,7] and using the values of thermal-expansion coefficients and elastic constants of Pb and Si.[8] The calculation method is described in a previous paper.[6] It is noted that a film is in tension in a planar direction upon cooling, because its thermal-expansion coefficients are larger than those of Si. The D

FIG. 1. (111) interplanar spacing change during thermal cycling between 300 and 4.2 K for 0.2-μm-thick Pb-In-Au films prepared at 298 K with or without a 0.4-μm-thick overlying SiO layer.

values of 0.2-μm-thick Pb-In-Au films with or without 0.4-μm-thick SiO layer which were deposited at 298 K are shown by squares and circles, respectively. Although all samples were annealed at 300 K for several days before further thermal treatments, slightly different D values were obtained in different specimens at 300 K. Thus the D values obtained at 300 K from (333) diffraction peaks for different specimens were normalized to be 0.094734 nm. It is seen that the D values of both films deviate from the D_{cool}^{max} upon cooling and that the D values at 300 K after cycling are larger than those before cycling. The results indicate that strain relaxation occurs upon cooling to 4.2 K, and that compressive strain is introduced upon subsequent heating to 300 K. Note that the D value of the film is affected by the SiO layer; smaller and larger D values were observed at 4.2 and 300 K, respectively, in the film with an overlying SiO layer compared with the D values of the film without an overlying SiO layer.

The strain at 4.2 K normal to the film surface was obtained from the (111) interplaner spacings by

$$\epsilon'_{33} = (D - D^0)/D^0, \tag{1}$$

where D is the measured interplanar spacing and D^0 is the spacing of the strain-free film at 4.2 K. The strain values are shown in Fig. 2(a) for 0.2-μm-thick Pb-In-Au films which were deposited at 298 or 77 K followed by overlying SiO layers of various thicknesses. The expected maximum strain ϵ'_{max} is shown by a dashed dotted line; it was calculated to be ~0.67% assuming that no strain relaxation occurred upon cooling.[6] The strain of the film which was prepared at T_s = 298 K and was not covered with SiO is relaxed upon cooling; the relaxed strain is seen to decrease with increasing the overlying SiO-layer thickness. The strain supported by the films is increased by an amount of ~0.13% when the SiO layers are thicker than 0.2 μm. For films prepared at T_s = 77 K, the strain in the film without the overlying SiO layer present (dashed line) is believed to be almost completely supported elastically upon cooling, even though the strain value is smaller than the calculated ϵ'_{max}, as explained in previous papers.[6,9] Thus the strain values of these films did not appear to be influenced by the SiO layers.

As seen in Fig. 1, compressive strain is introduced into a Pb-In-Au film during the heating process from 4.2 to 300 K when the strain is initially relaxed upon cooling to 4.2 K. The compressive strain supported in the film is calculated using Eq. (1). In this case, D is the measured interplanar spacing at 300 K after thermal cycling, and $D°$ is the value of the strain-free film before thermal cycling. The strain values calculated from Eq. (1) are shown in Fig. 2 (b) for films prepared at T_s = 298 or 77 K. Note that the strain values of the films prepared at T_s = 298 K increase with increasing the SiO-layer thicknesses, indicating that the strain relaxation is retarded by the SiO layers. For films prepared at T_s = 77 K, the compressive strain is ~0.02%. These small or essentially zero strains are believed to be due to the absence of relaxation occurring upon cooling to 4.2 K. Again, the overlying SiO layers did not affect the strain behavior after heating to 300 K for these fine-grained films.
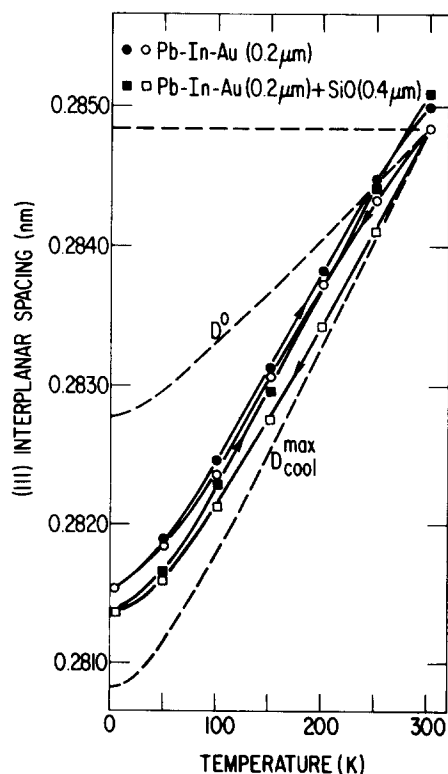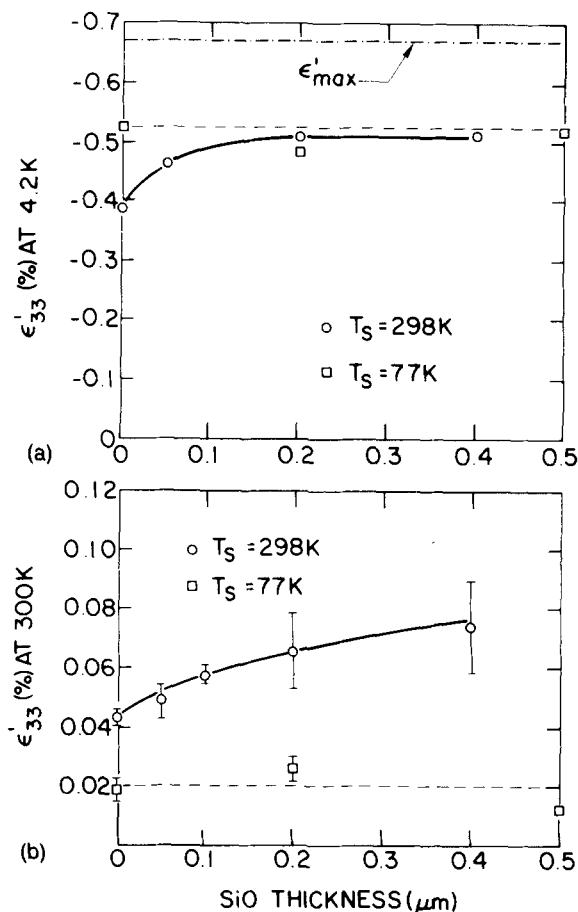
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FIG. 2. Average elastic strain (a) 4.2 K and (b)300 K supported by Pb-In-Au films deposited at 298 or 77 K with various thicknesses of overlying SiO layers.

FIG. 3. (111) interplanar spacing change during thermal cycling between 300 and 350 K for 0.2-μm-thick Pb-In-Au films prepared at 298 K with or without a 0.4-μm-thick overlying SiO layer.

B. Thermal cycling between 300 and 350 K

The (111) interplanar spacings D normal to the film surfaces were measured from (333) diffraction peaks for films which were thermally cycled between 300 and 350 K at an interval of 10 K. The D values measured at each temperature for films with different film thickness are shown in Fig. 3. It took about 15 min to stabilize the temperature and to take the diffraction peak at that temperature. The $D°$ line shown in Fig. 3 denotes the calculated strain-free values of the Pb-In-Au film, which were calculated using the reported thermal-expansion coefficients of Pb. The D_{heat}^{max} denotes the calculated D values of the maximum strain for the Pb-alloy on the Si substrate. The values were calculated for (111) oriented grains using the biaxial-strain model, assuming no strain relaxation occuring upon heating.[10] In the calculation, the temperature dependence of elastic constants C_{ij} were obtained from the literature:[8] $\partial C_{11}/\partial T = -2.128$, $\partial C_{44}/\partial T = -1.571$, and $\partial C_{12}/\partial T = -1.114$ in the unit of 10^8 dyn/cm^2 deg.

The typical experimental D values are shown in Fig. 3 by different symbols for films with or without an overlying SiO layer. The D values of a 5-μm-thick film were observed to be very close to the $D°$ values upon heating (the data are not shown in Fig. 3). This is interpreted to indicate that the compressive strain (we give the sense of the strain parallel to the sustrate unless other wise specified) at the film surface
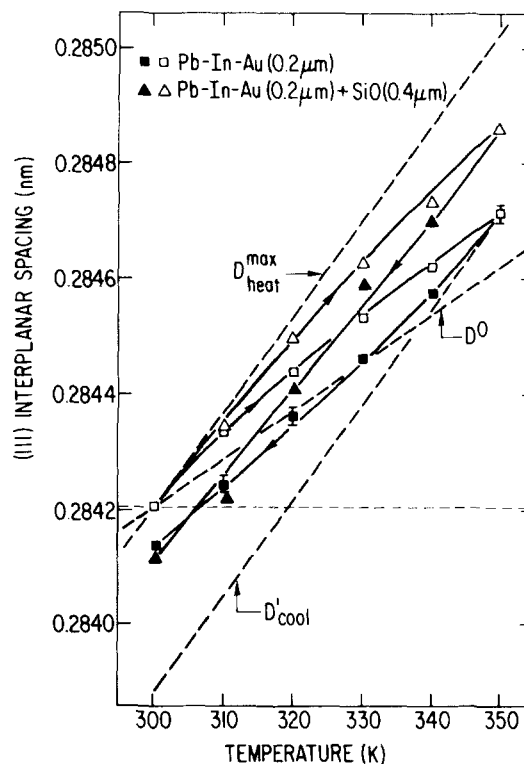
within the x-ray-penetration distance of $\sim$ 1 μm is relaxed almost completely upon heating. However, a tensile strain was observed upon cooling from 350 to 300 K. For a 0.2-μm-thick film without an SiO layer (square), a small amount of strain relaxation was detected in 15 min at 310 K and more relaxation was observed at temperatures above 310 K. The dashed curve denoted as D'_{cool} indicates the calculated D values of the 0.2-μm-thick film when no strain relaxation was assumed upon cooling from 350 to 300 K. The deviation of the experimental data from the curve indicates that part of the strain is also relaxed upon cooling. The D values of the 0.2-μm-thick Pb-In-Au film with a 0.4-μm-thick SiO layer are shown by triangles. It is noted that the D value at 350 K is much larger than the one of the 0.2-μm-thick film without SiO layer, indicating that the strain relaxation at 350 K is prevented by the 0.4-μm-thick SiO layer. The D values of the film with or without the SiO layer were found to be almost the same values at 300 K after thermal cycling.

Strains supported by films at 350 or 300 K after thermal cycling were obtained for the 0.2-μm-thick films with or without SiO layers from the interplanar spacings by using Eq. (1), where $D°$ is the interplanar spacing of the strain-free film and D is the spacing measured at 350 or 300 K. The calculated ϵ'_{33} values at 350 and 300 K are shown in Figs. 4(a) and 4(b), respectively, as a function of thicknesses of the SiO layer. The ϵ'_{max} shown in Fig. 4(a) is the maximum strain expected in the film upon heating from 300 to 350 K when no strain relaxation was assumed during the heating process. The ϵ'_{max} was calculated to be 0.146% for (111) oriented grains.[10] For a film without an SiO layer prepared at T_s
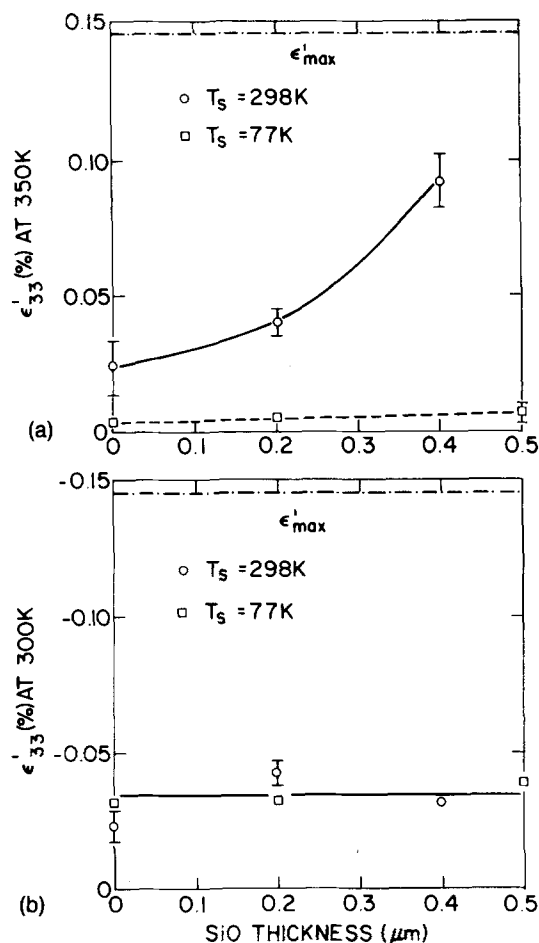
FIG. 4. Average elastic strain at (a) 350 K and (b) 300 K supported by Pb-In-Au films prepared at 298 or 77 K with various thicknesses of overlying SiO layers.

= 298 K, the strain value of ~0.025% was obtained at 350 K. The values were observed to increase with increasing thicknesses of the SiO layers, which indicates that the strain relaxation is retarded by the SiO layers. For a film without

SiO layer prepared at $T_s = 77$ K, the strain was near zero at 350 K and no change in the ϵ'_{33} values was observed by use of an SiO coating. Thus the SiO-coating effect was reduced by reducing the average grain size. Tensile strain is expected to be introduced into a Pb-In-Au film upon cooling from 350 to 300 K when the strain is previously relaxed upon heating to 350 K. The strains induced upon cooling to 300 K are shown in Fig. 4(b). It is noted that the strains of the films prepared at $T_s = 298$ or 77 K are almost constant, and that the strain values are not influenced by the SiO coating. The reasons why the strain values are not affected by SiO layers are not presently understood.

C. Isothermal annealing at 350 or 300 K

In order to study the effects of grain size and SiO coating on strain relaxation rates, the ϵ'_{33} values during isothermal annealing at 350 K were measured for 0.2-μm-thick films which were deposited at 298 or 77 K and then coated by SiO layers with various thicknesses. The changes of the ϵ'_{33} values are shown in Figs. 5(a) and 5(b). For films prepared at $T_s = 298$ K, most of the strain was relaxed concurrently with heating to 350 K. (This rapid strain-relaxation process is referred to as the primary relaxation process.) The ϵ'_{33} values after 30 min at 350 K was about 0.03% for the film without SiO coating. The strain relaxing during the primary relaxation process was observed to decrease when the thickness of the SiO layer was increased. Thus the primary strain-relaxation process was significantly inhibited by SiO layers with thickness greater than 0.2 μm. During subsequent isothermal annealing the strain continued to relax (this slow process is referred to as the secondary strain-relaxation process). The strain did not come to zero level even after 1000 min at 350 K. For films deposited at 77 K [Fig. 5(b)] the strain value was close to zero immediately after heating up to 350 K. The strain is believed to be relaxed completly during the primary strain-relaxation process. The strain did not change during isothermal annealing, and the SiO layer did not influence the strain behavior of these fine-grained films
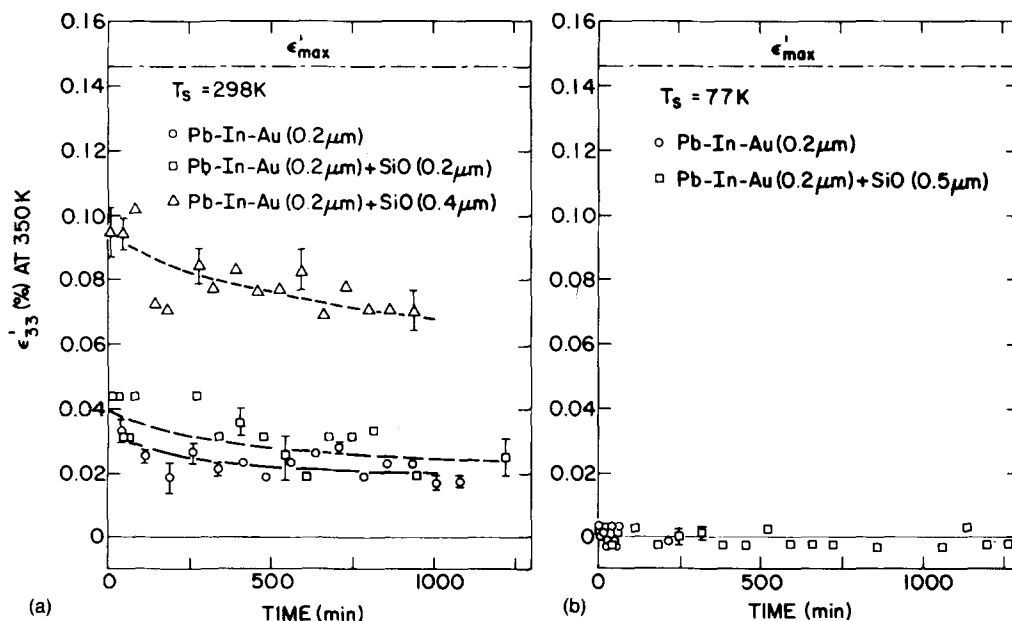

FIG. 5. Changes of average elastic strain during isothermal annealing at 350 K supported by Pb-In-Au films prepared at (a) 298 K or (b) 77 K with or without an overlying SiO layer.
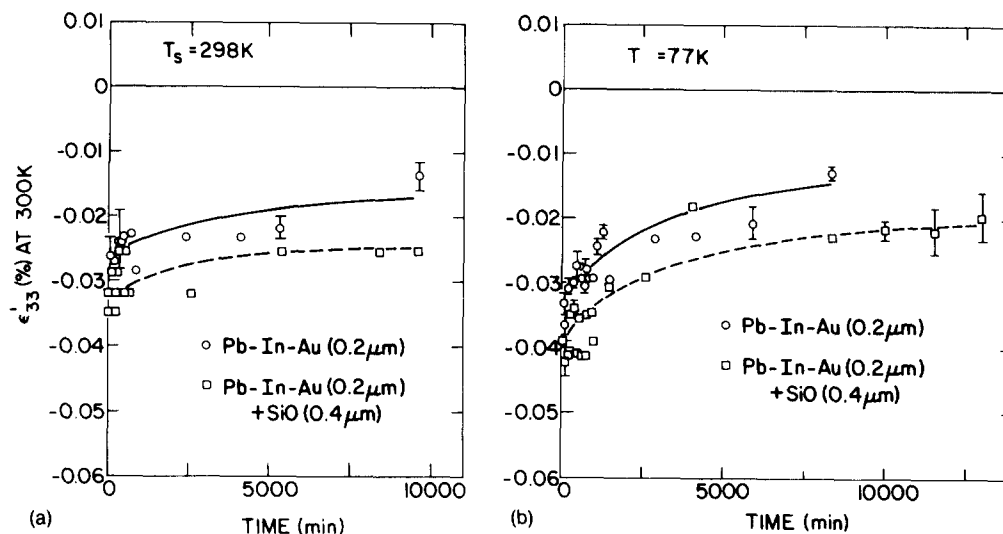
FIG. 6. Changes of average elastic strain during isothermal annealing at 300 K after cooling from 350 K supported by Pb-In-Au films prepared at (a) 298 K or (b) 77 K with or without an overlying SiO layer.

during both primary and secondary relaxation processes.

Upon cooling from 350 to 300 K, tensile strain is observed to be introduced into a film when the strain is relaxed upon previous heating from 300 to 350 K (Fig. 3.). The strain relaxation during isothermal annealing at 300 K was followed for films prepared at $T_s = 298$ or 77 K, and the changes in the strain are shown in Figs. 6(a) and 6(b), respectively. For either films prepared at $T_s = 298$ or 77 K with or without SiO layer, the primary and secondary relaxation processes were observed. More than half of the expected strain was relaxed during the primary relaxation process. The primary relaxation process was not significantly influenced by the SiO coating. During the secondary relaxation process, the amounts of strain relaxed for ~1 week are 0.01% and 0.02% for films prepared at $T_s = 298$ K and 77 K, respectively. Faster relaxation rates for films prepared at $T_s = 77$ K are considered to be due to fine grains.

IV. DISCUSSION
A. Strain relaxation mechanisms

From a deformation-mechanism map which was constructed for Pb (Ref. 11)-and Pb-In-Au-alloy films,[12] it was predicted that the dominant strain-relaxation mechanism is dislocation glide at temperatures well below room temperature and grain-boundary diffusion creep at temperatures above room temperature. It has appeared likely that these mechanisms occur although direct experimental evidence for their occurrence has not yet been obtained.

The secondary relaxation, which would not have appeared in the deformation-mechanism map, was observed during isothermal annealing at 350 and 300 K in Figs. 5 and 6, respectively. In previous experiment with pure Pb films[10] the secondary strain relaxation was analyzed using the Nabarro-Herring diffusion-creep model.[13,14] The diffusion coefficients obtained using this model at a temperature range of 300 to 350 K were found to be very close to values of the lattice diffusion coefficients obtained by extrapolating from high temperatures to temperatures below 350 K. Thus it was concluded that the secondary relaxation mechanism was lat-

tice diffusion creep.

The analogous method was used for Pb-In-Au films to determine the secondary strain-relaxation mechanism observed at 350 and 300 K. In the Nabarro-Herring diffusion-creep model it was assumed that excess atoms diffuse from grain boundaries to the film surface and to the film substrate interface through a grain when the compressive strain is applied to the film. The atoms diffuse in the opposite direction when the tensile strain is applied. The strain-relaxation rate $\dot{\epsilon}'_{11}$ (parallel to the film surface) as given by Gibbs[15] for a thin film which is uniaxially strained is

$$\dot{\epsilon}'_{11} = B_0(\sigma'_{11}\Omega/gh\,kT)(D_1/f), \qquad (2)$$

where $B_0 (= 7.98)$ is a geometrical factor, σ'_{11} is the applied stress in the film plane, Ω is the atomic volume, $f(= 0.781)$ is the correlation factor, D_1 is the lattice diffusion coefficient of Pb-alloy, g is the grain size of the film, h is the film thickness, k is the Boltzmann's constant, and T is temperature. Equation (2) can be integrated to obtain

$$ln\epsilon'_{11} = ln\dot{\epsilon}_{11} + (B_0Y\Omega/ghkT)(D_1/f)t \qquad (3)$$

where ϵ'_{11} is the strain at time zero, t is the annealing time, and Y is Young's modulus ($\sigma'_1 = Y\epsilon'_{11}$). Equation (3) was derived for a uniaxially strained film. The present Pb-alloy films are biaxially strained. However, the strain-rate equation for biaxially strained films is extremely complicated and several uncertain assumptions have to be made to derive a rate equation equivalent to Eq. (3). In addition, Pb-In-Au has a strong anisotropy in elastic character: the value of σ'_{11} or Y is dependent on the crystallographic directions, although the value of ϵ'_{11} is constant in any directions in the film plane. The maximum stress is about a factor of 2.8 larger than the minimum value. The atoms in the films would diffuse most rapidly along a direction in which the maximum stress is applied, i.e., the behavior is "quasi-uniaxial." Thus it should be reasonable to treat the diffusion problem of the present Pb-alloy films using the uniaxial-strain model rather than a biaxial-strain model which involves several uncertain assumptions.

Equations (3) shows that the slope of a plot in $\ln \epsilon'_{11}$ vs t
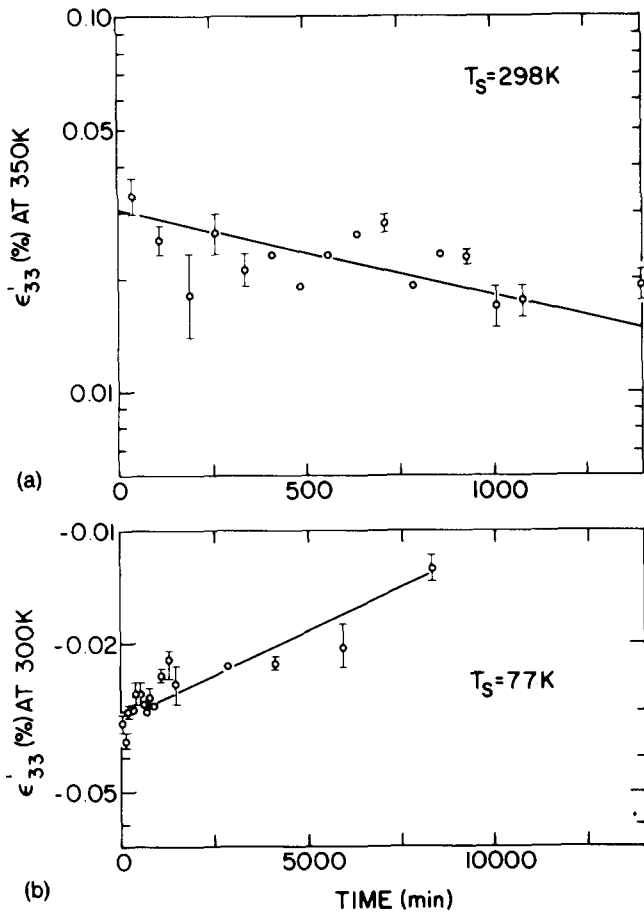
FIG. 7. Plots of log ϵ'_{33} vs t for Pb-In- Au films prepared at (a) 298 K or (b) 77 K.

is proportional to the latice diffusion coefficient D_1. For examples, the isothermal changes in ϵ'_{33} of Pb-In-Au films prepared at T_s = 298 or 77 K without SiO layers shown in Figs. 5(a) and 6(b) are replotted on a logarithm scale in Figs. 7 (a) and 7(b), respectively. Linearities were obtained in both plots. From slopes of these plots, D_1 values were obtained using Eq. (3). The D_1 values obtained from slopes of Fig. 7 are plotted in Fig. 8. The Y values[10] calculated in Pb were used to determine the D_1 values of the Pb-In-Au films. In this figure the D_1 values at 350 and 300 K are shown for films with various deposition temperatures and with various SiO layer thicknesses. In Fig. 8, for reference, the D values of Pb lattice diffusion which were calculated using an equation given by Mehrer and Seeger[16] are shown by a dashed line and the values determined for Pb thin films using the present

technique are shown by circles.[10] Agreement between the extrapolated and experimentally determined D values is seen. The D values determined for the Pb-In-Au films are about 2–3 orders of magnitude smaller than those of Pb films. The physical meaning of D values determined in the present ternary alloy using the Nabano-Heering creep equation is not understood, but it is concluded that the secondary relaxation rate of Pb films by the diffusion creep was reduced at least two orders of magnitude by the addition of In and Au to Pb films. It is noted that the SiO coating did not influence the secondary strain-relaxation rates. The SiO layers on the Pb-alloy films are believed to be deformed uniformly to relax the strain during the secondary relaxation process.

B. Effects of SiO layers on strain relaxation

The effects of SiO coating on strain relaxation which were observed in the present experiment are summarized in Table I and discussed here.

When the strain is relaxed by dislocation glide, dislocations are introduced from grain boundaries into grains.[17] The dislocation glide motion was observed to be prevented by a thin layer of the native surface oxide which was grown on the Pb film, if the shear modulus of the oxide is larger than the one of the film.[17] Since the shear modulus of the SiO is more than a factor of 5 larger than the one of Pb, the dislocation motion is believed to be prevented by the SiO layer coated onto the Pb-alloy film like a case of the native oxide. Thus more strain is considered to be supported in the films at 4.2 K with SiO layers as seen in Fig. 2.

The strain relaxation by grain-boundary diffusion creep was observed to be prevented by SiO coating only when the compressive strain is introduced into the large-grained films upon heating from 4.2 to 300 K or from 300 to 350 K. No effect was observed in fine-grained films prepared at T_s = 77 K. This may be explained by the difference in the number of atoms which would be protruded locally on the strain-free film surface through grain boundaries. For the films prepared at T_s = 77 K, the average grain size is less than half of the film thickness. In these fine-grained films, the excess atoms could be distributed over the high dense grain boundaries, lifting the SiO layer uniformly. However, in the large-grained films, a large number of atoms could be protruded locally on grain boundaries. The protrusion of atoms could be prevented by the SiO layer, resulting in the inhibition of strain relaxation by grain-boundary diffusion creep.

The strain relaxation by lattice diffusion creep was not found to be influenced by the overlying SiO layers, as seen in

TABLE I. Effects of SiO layers on strain relaxation in Pb-In-Au films.

| Strain-relaxation mechanism	Strain	Effect of SiO coating	Related figures
Dislocation glide	Compressive	Relaxation is prevented	Fig. 2(b)
	Tensile	Relaxation is prevented	Fig. 2(a)
Grain-boundary diffusion creep	Compressive	Relaxation is prevented for large-grained films	Fig. 4(a)
	Tensile	No effect	Fig. 4(b)
Lattice diffusion creep	Compressive	No effect	Fig. 8
	Tensile	No effect	Fig. 8
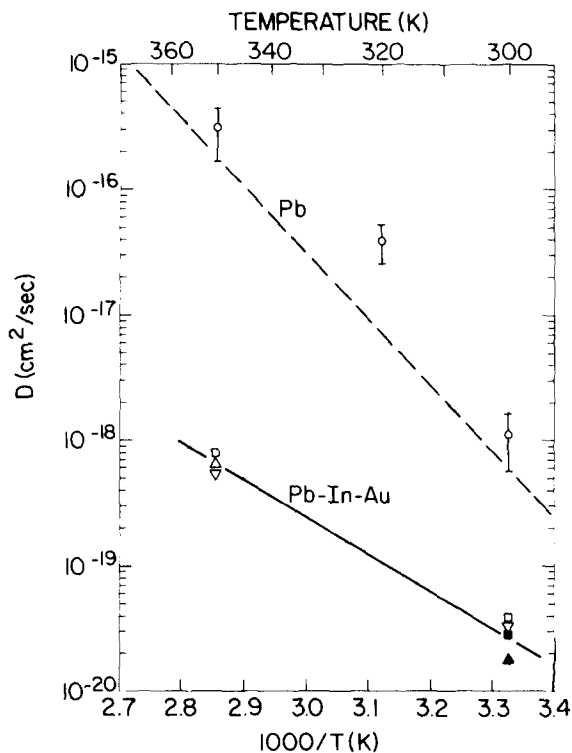
FIG. 8. Diffusion coefficients of Pb-In-Au films determined by the present experiment: □ Pb-In-Au ($T_s = 298$ K); △ Pb-In-Au ($T_s = 298$ K) + SiO (0.2 μm); ▽ Pb-In-Au ($T_s = 298$ K) + SiO (0.4 μm); ■ Pb-In-Au ($T_s = 77$ K); ▲ Pb-In-Au ($T_s = 77$ K) + SiO (0.5 μm). The coefficients of Pb films which were determined previously[10] are shown for reference.

Fig. 8. Thus it is believed that the Pb-alloy films are not stressed by the SiO coating and that it is free to move up and down uniformly.

C. Hillock formation inside an SiO window

Huang et al.[2] studied hillock formation using SEM upon heating 0.2μm-thick Pb-In-Au films which are covered by the SiO layer except for $\sim 5\mu$m × 5-μm opening. The SiO layer prevents hillocks from forming beneath it, but enhances the size of hillocks that form in the SiO opening in proportion to the amounts of Pb alloy enclosed by the SiO. Hillocks were observed inside the SiO opening within 30 min after heating at 343 K for large-grained films prepared at T_s = 297 K and any changes visible by SEM at a resolution of ~ 20 nm were not found in fine-grained films prepared at T_s = 77 K.

The strain-relaxation processes expected for large or fine-grained films with or without SiO coating are schematically illustrated in Fig. 9. At the very early stage of the relaxation process ($t < t_1$) a big difference in the strain-relaxation rates in films with or without SiO layer would be expected in large-grained films. The difference in the relaxation rates at this stage would produce a sharp strain gradient between the films inside the opening and the film beneath the SiO layer. The gradient causes the diffusion of atoms toward the opening, forming hillocks. In fine-grained films [Fig. 9 (b)], no difference in the strain-relaxation process at the later stages was detected by the present x-ray diffraction technique. From this result it is expected that the relaxation rates in the
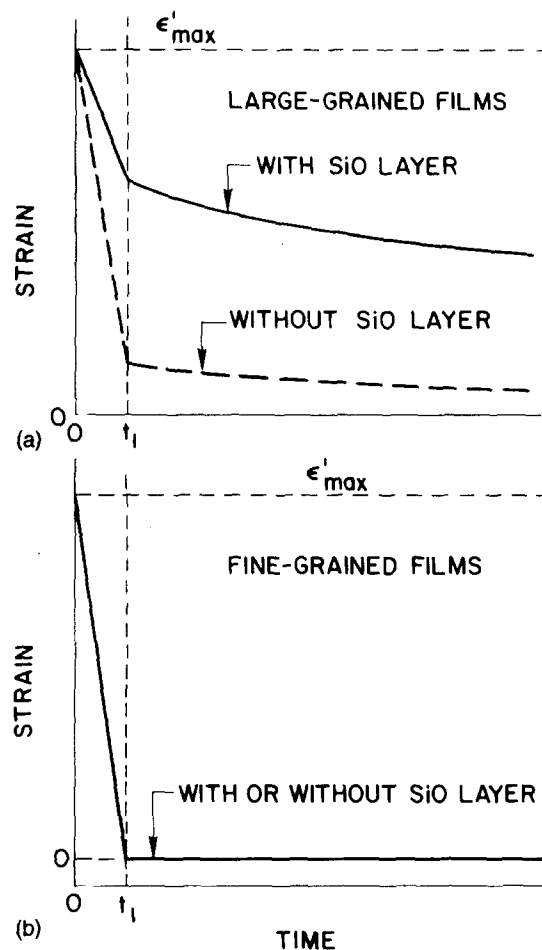

FIG. 9. Schematic illustrations of strain relaxation processes during isothermal annealing of Pb-In-Au films with (a) large or (b) fine grains with or without overlying SiO layers.

films with or without SiO coating at the early stages would be the same. Thus no driving force of atom diffusion toward the opening should exist in fine-grained films.

V. SUMMARY

Effects of overlying SiO layers on the strain behavior of Pb–12wt. % In–4wt % Au films which were deposited onto Si wafers at 298 or 77 K and then thermally cycled between 300 and 4.2 K and between 300 and 350 K were studied by an x-ray diffraction technique. For large-grained films the strain relaxation by dislocation glide or by grain-boundary diffusion creep was inhibited by the SiO layers. For fine-grained films, no effects on the strain behavior by the SiO layers were detected by the present x-ray diffraction technique. These results were consistent with a previous SEM experiment in which hillocks were observed in SiO-layer openings above large-grained films and not above fine-grained films during repeated thermal cycling between 300 and 4.2 K and upon heating from 300 to 343 K. Thus this is a further indication that thermal stability of junctions formed on a Pb-In-Au base electrode should improve upon reduction of the average grain size of the electrode.[1]

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