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On the development of the (001) texture of gold leaf fabricated by hammering

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The texture development of gold leaf fabricated by the traditional pack and hammering method, in which sheets of metal and paper were alternately packed and beaten by a hammer, was investigated. Inspection of the results revealed that the (011) texture once developed was replaced by (001) texture at the final stage of hammering. The development of (001) texture was considered to be attributed to cross slip due to severe deformation caused by hammering.

1. Introduction
Metallic leaf is produced by the traditional hammering method and used for ornamental purposes in Japan. Now, very thin leaf gold, silver, and aluminium and brass are produced in domestic industries. Metallic leaf can be produced as thin as 0.1–0.5 μm by a pack and hammering method, in which sheets of metal and paper are alternately packed and then beaten by a hammer.

This working method is very different from other modern methods such as rolling. We expected the leaf texture to be different from that of cold rolled sheets of metals and reported the textures of aluminium [1] and gold leaf [2] to be (001). This texture cannot be expected from the simple compressive deformation theory.

Many studies have been performed for many metals on both drawing and rolling textures, however, there have been none for hammering texture. On the basis of the theories explaining the compressive, drawing and rolling textures, the development of the texture of gold leaf fabricated by hammering was investigated.

2. Experimental details
The starting material was a rolled sheet of gold having a thickness of 40 μm and containing 4.90 wt % Ag and 0.66 wt % Cu. The sheets were annealed twice at 400° C for 1 h during the last stage of rolling. Three hammering processes were employed to produce the final thin foils. In the first process, the rolled sheets, 55 × 55 mm^2, were packed alternately with sheets of brown craft paper, 150 × 150 mm^2 which were pretreated to induce good lubrication and prevent adhesion between the gold and paper sheets. Approximately 200 sheets of this assembly were sandwiched by two leather skins as shown in Fig. 1. This pack was beaten at a rate of 400 strokes min^{-1} by a hammer with a diameter of 80 mm. After about 6000 strokes of hammering, the thickness of the gold sheet was reduced to about 5 μm. In the second process, the sheets, cut to 75 × 75 mm^2, were packed alternately with another set of rice paper. This pack was beaten under the same hammering conditions as in the first process until the thickness of gold sheet was reduced to about 1 μm. The total number of strokes was about 24 000 in this stage. In the third stage, 1600 sheets of gold foil of 55 × 55 mm^2, were packed alternately with high quality rice papers. The pack was beaten at a rate of 700 strokes min^{-1} by a hammer with a diameter of 40 mm. Excess heating over about 100° C induced by heavy hammering was avoided by interrupting the hammering. The thickness of the final leaf was approximately 0.1 μm. The total number of strokes was about 100 000 at this stage. The thickness of the sample was measured by a micrometer or X-ray absorption method.

Inverse pole figures were obtained to discuss the texture. Mo-$K\alpha$ radiation was used in order to detect as many peaks as possible. The X-ray intensity was corrected by the following formula

$$I^* = I/[1 - \exp(-2\mu t/\sin\theta)]$$

where I is the measured intensity and I^* the corrected one, θ is Bragg angle, t is thickness of the leaf and μ is the linear absorption coefficient.

3. Results
Fig. 2a shows the (200) pole figures of the rolled sheet (40 μm thick). The characteristic features of the rolling texture has not been developed. This is probably because intermediate annealing was employed during rolling. The hammering process results in a marked change in texture. Fig. 2b is the (200) pole figure of the final leaf (0.11 μm thick). The intensity maxima occur in the centre and at the periphery. It is seen that the directionality with respect to the rolling direction disappeared almost completely. The normal axisymmetry to the leaf plane is now noted. Particularly from Fig. 2b, we can describe the leaf texture as being based on (100) planes becoming parallel to the plane of the heavily hammered leaf.

Fig. 3 shows the relationship between the reduction percentage of thickness and the relative diffraction intensity of the important crystal plane. The intensity

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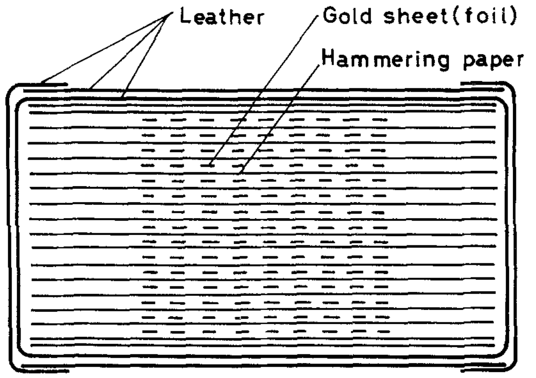

Figure 1 Schematic illustration of a hammering pack in which gold sheet and paper are placed alternately.

of the (011) plane increases rapidly with the increase of reduction percentage. However, it decreases over about 80% reduction (about 10 μm thick). It is noted that the intensity of this plane of final leaf (about 0.1 μm thick) is nearly zero. On the other hand, the intensity of the (001) plane decreases slightly with an increase of reduction percentage. However, as soon as the thickness of the leaf is reduced to less than about 1 μm, it increases rapidly with the increase of the reduction of the thickness. The intensity of the final thin leaf is 17 times higher than that of the starting material of rolled sheet. In other components without (001) and (011), intensity peaks are not seen.

Fig. 4 shows inverse pole figures of rolled sheet, hammered foil and hammered leaf. As shown in Fig. 4b, hammering causes a big change in the texture. There is a strong intensity maximum near (011). However, the concentration of contour lines near (011) disappeared in the following stage (Fig. 4c) and was replaced by a completely new one at the final stage of hammering. Fig. 4d shows that the intensity maximum occurs at (001). However, the texture is not so completely developed as to describe the (001) texture and thus should be called quasi (001) texture.

4. Discussion

There have been several attempts to explain the deformation behaviour of polycrystalline aggregates in terms of the slip mechanisms operative when single crystals are deformed, but no wholly satisfactory treatment has been devised. The treatment of Taylor [3], mathematically more rigorous, is based on the assumption of perfectly homogeneous deformation of the aggregate. This demands the simultaneous operation of five special slip systems. Taylor has derived the systems and corresponding shears which conform to the principle of virtual work. The magnitudes and directions of the crystal rotations may be deduced from the shears selected. Good agreement with the experimentally determined tensile stress–elongation curve for a polycrystalline aggregate and the main features of the tension and compression textures for f.c.c. metals is obtained. According to this treatment, compression texture for f.c.c. metals is described to be (011).

In our study, rolled sheet, 40 μm thick, was hammered as a starting material. When the thickness of the sheet was reduced to about 10 μm, the development of (011) texture was observed. Therefore, the texture of this stage is in good agreement with Taylor's treatment. However, we cannot explain the quasi (001) texture of final thin leaf reduced to 0.1 μm thick. Now, we review the studies concerning texture development in order to explain the (001) texture of the leaf.

The texture of gold leaf somewhat resembles the cube texture observed in cold rolled and recrystallized sheets of aluminium [4], copper [5] and gold [6]. If recrystallization occurs after heavy hammering and hammering–recrystallization cycles are repeated, the present (001) plane texture of gold leaf might be explained on the basis of the cube texture formation. However, this is very unlikely. First, the temperature endured by the pack of the leaf–paper assembly was at most about 100° C. Second, a heavy tangled cell structure of dislocations was actually observed in the leaf

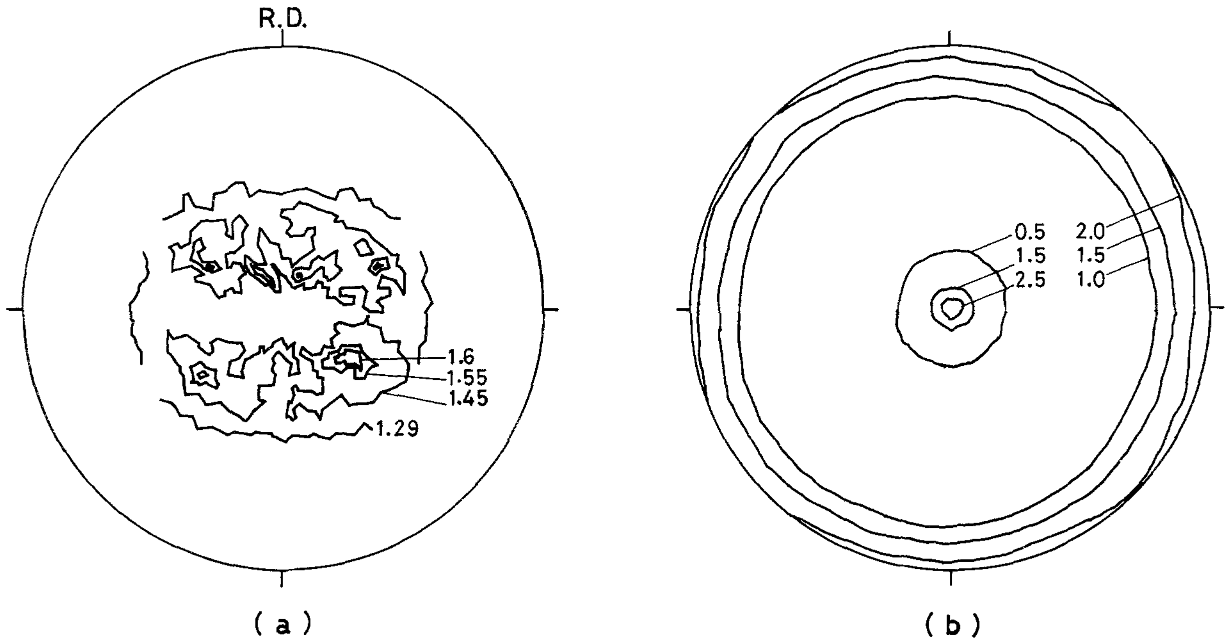

Figure 2 (200) pole figures of (a) rolled sheet (40 μm) and (b) hammered leaf (0.11 μm).

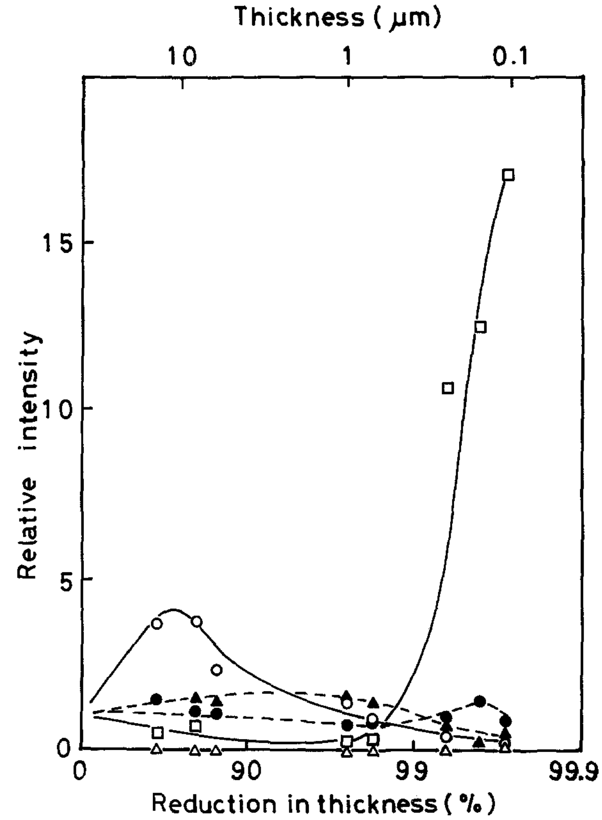

Figure 3 Variation of X-ray diffraction intensity from important crystal planes with reduction percentage by hammering. (□ (001), ○ (011), △ (111), ▲ (012), ● (013))

as shown in Fig. 5. Thus, we exclude the possibility that the gold leaf underwent recrystallization during hammering.

The following results should be noted to consider the development of the (001) texture of gold leaf. Vargha and Wassermann [7] reported that surface texture, (001)[110], was developed in the surface layer of cold rolled aluminium strips. Moreover, the surface texture of the 2S aluminium strip rolled in reversed pass also corresponds to (001)[110] orientation [8]. Kamijo *et al.* [9] have studied the factors that affect the formation of surface textures: friction between roll and sheet, one-pass reduction, total reduction and the rolling sequence. They concluded that under conditions of very high friction and low one-pass reduction, the (001)[110] surface texture is markedly developed with an increase of total rolling reduction, whereas the (001)[110] component was only to some extent formed for high one-pass reduction. The reason why the surface texture developed prominently in aluminium was attributed not only to the facility of cross slip due to high stacking fault energy (SFE) of aluminium but to the high friction between roll and aluminium sheet. The results indicate that the (001)[110] surface texture develops when metals with high SFE are subjected to both compressive and shear stresses. Here, we would like to point out that the leaf texture is very similar, in nature, to the surface texture of aluminium strip sheets produced by severe rolling passes or under a condition of heavy friction between the roll and sheet. This is understandable because both hammered leaf and the surface layer of a heavy rolled sheet are deformed under a constrained condition imposed by strong friction. The difference is only in the directionality: a rolled sheet should have a directionality described by the rolling direction, while hammered leaf should have symmetry around the normal to the leaf plane.

There are many reports concerned with the wire texture of f.c.c. metals. It seems to be significant to discuss the development of leaf texture in comparison with wire texture. Wire textures of cold-drawn f.c.c. metals and alloys are described as some combination of ⟨100⟩ and ⟨111⟩ orientations. The relative amounts of each, however, are quite variable from one alloy to

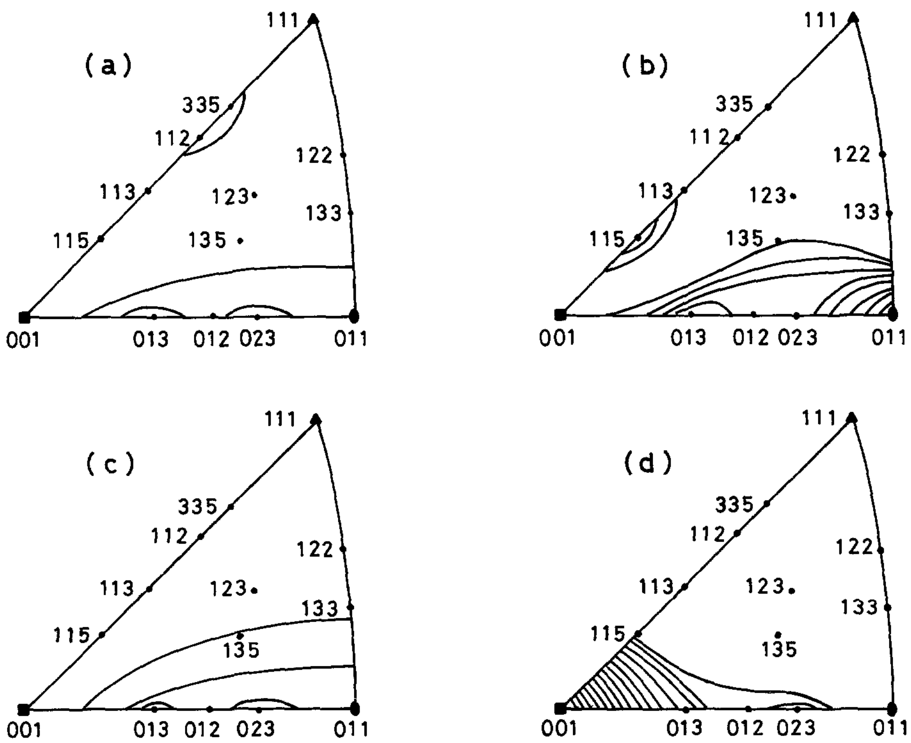

Figure 4 Inverse pole figures of (a) rolled sheet (40 μm), (b) hammered foil (8 μm), (c) hammered foil (0.7 μm) and (d) hammered leaf (0.11 μm).

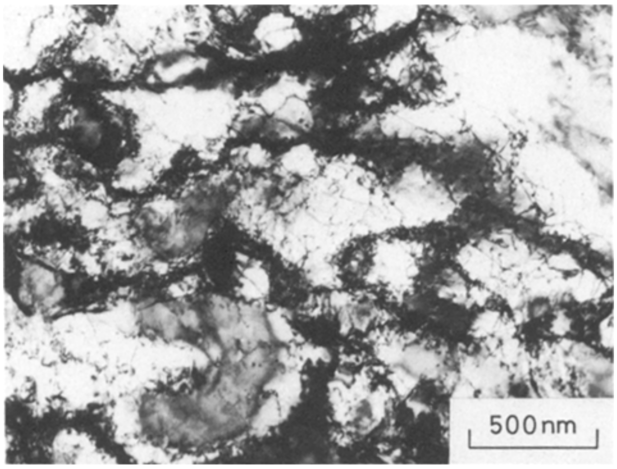

Figure 5 Transmission electron micrograph showing tangled cell structure of dislocations in hammered leaf.

another. Using an argument relating texture to the ease of cross slip, Brown [10] has proposed that the proportion of ⟨100⟩ increases as SFE decreases. English and Chin [11] studied the wire textures of various f.c.c. metals and alloys, as a function of the parameter γ/Gb. Here γ is the SFE, G the shear modulus and b the Burgers vector. The most important conclusion from the results is that the general trend toward a larger proportion of ⟨100⟩ with reduced SFE is reversed for the lowest values of γ/Gb. He indicated that this reversal was not easy to explain on the basis of any single mechanism, such as the influence of cross slip proposed by Brown. However, this reversal is not found in rolling textures, which vary monotonically from brass to copper type with increasing SFE [12].

Kamijo [13] studied the drawing textures, assuming that the stress system in drawing is a multiaxial one, i.e. a tensile stress in the direction of drawing together with a circular array of compressive stress perpendicular to this direction. He considered that the resulting rotation of the wire axis could be assumed to be described by combining the tension rotation vector with the compression rotation vector. It was found that the wire axis of f.c.c. metals tended to rotate towards the [111] or [001] orientation. Applying the idea to the compression texture, the possibility of [011] rotation was predicted.

There has not been a theory explaining the hammering texture whereas there have been several for drawing and rolling textures. It may be difficult to explain wholly the hammering texture of thin gold leaf because there are several unknown factors, i.e. elastic properties of paper, mutual interaction between paper and leaf etc. However, we try to discuss the texture development of gold leaf on the basis of theories explaining the compressive, drawing and rolling textures.

Standard stereographic projection with Thompson's notation is shown in Fig. 6. Let us consider a crystal grain whose compressive axis is oriented in the triangle [001]–[011]–[111]. The primary slip system β-AC activates and the compressive axis of the grain tends to rotate to $[\bar{1}11]$ orientation when the sheet of gold is subjected to compressive stress by hammering. When the compressive axis coincides with that on the [001]–[011] line after the rotation, the Schmid factor (SF) of the conjugate slip system α-DB becomes equal to β-AC, but it cannot operate immediately because of latent hardening of β-AC. After the over shooting of the compressive axis into the triangle [001]–[011]–$[\bar{1}11]$, α-DB will soon operate. The compressive axis will rotate and become closer to [011] by the activation of both slip systems. As soon as the compressive axis coincides with the [011] orientation, the slip systems of α-CB and β-AD are expected to activate simultaneously, because these slip systems become equivalent to β-AC and α-DB. However, the dislocations of these slip systems react and the product dislocation provide a strong barrier to dislocation glide. This product is the so-called Lomer–Cottrell barrier. In many studies previously performed on deformation texture, the degree of deformation is not

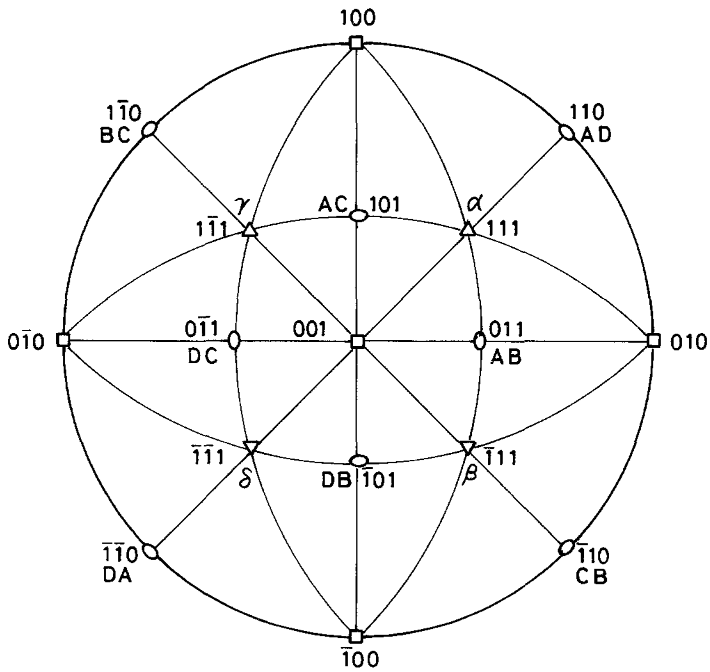

Figure 6 Standard stereographic projection with notation by Thompson.

TABLE I X-ray intensity ratios, $I_{(011)}/I_{(001)}$, of several kinds of metallic leaf

Leaf material	$I_{(011)}/I_{(001)}$	SFE (J^{-2}) × 10^{-7}	References
Aluminium	0	280 ± 50	[14]
Gold	0.01	32 ± 5	[15]
Silver	0.02	16.3 ± 1.7	[16]
Brass (12 wt % Zn)	1.32	5 ± 1	[17]

as large as in the case of leaf production. That is, the leaf is the product subjected to extremely heavy deformation. Here, it is noted that cross slip plays an important role in the deformation of crystal. Because of the high dislocation density and strong barrier, the dislocations on the slip systems α-DB and β-AC cannot be easily activated. However, the situation can be overcome by the activation of cross slip. The cross slip systems of β-AC and α-DB are δ-AC and γ-DB, respectively. The activation of these cross slip systems result in the axis rotation of the crystal to the normal of these plane orientation, $[\bar{1}\bar{1}1]$ and $[1\bar{1}1]$, as shown in Fig. 6. That is, if these cross slip systems δ-AC and γ-DB activate simultaneously, the compressive axis situated near [011] will shift to [001] along the [001]–[011] line. Since facility of cross slip depends on SFE of metals, completeness of (001) texture of the leaf is expected to be different between the metals. That is, the (001) texture of the metal with high SFE is considered to be more complete than that with low SFE.

By using the cross slip mechanism we can explain the transition from the (011) to the (001) texture of the leaf. In order to obtain the experimental evidence which supports the cross slip mechanism, discussion was made on another metallic leaf with different SFE. The intensity ratio of $I_{(011)}/I_{(001)}$ was obtained for aluminium (99.5 wt %), silver (99.99 wt %), and brass (12.3 wt % Zn) leaves whose SFEs were higher than, comparable with and lower than that of gold leaf respectively. Here $I_{(011)}$ and $I_{(001)}$ are the X-ray diffraction intensities from the (011) and the (001) plane parallel to the leaf plane, respectively. The leaf was fabricated by almost the same method as for the gold leaf. The thicknesses of the aluminium, silver and brass leaves were 0.45, 0.34 and 0.3 μm, respectively. The results are shown with the values of SFE [14–17] in Table I. It is noted that the (011) texture is not seen in the aluminium leaf, while it is predominant in the brass leaf. On the other hand, the (011) texture remains in limited amounts in both the gold and silver leaves. These experimental results agree well with expectation based on the cross slip mechanism. Thus, the development of the quasi (001) texture of gold leaf can be attributed mainly to cross slip due to severe condition by hammering.

5. Conclusions

The texture development of gold leaf produced by a pack and hammering method, in which rolled sheets of metal and paper were alternately packed and beaten by a hammer, was studied.

(1) The final texture of gold leaf reduced to 0.1 μm thick was quasi (001) texture, but it could not simply be developed from the rolling texture. At the intermediate stage of hammering, (011) texture was developed once. However as the deformation proceeded, the (011) texture was replaced by (001) texture.

(2) The development of the (001) texture was attributed to cross slip due to severe deformation condition by hammering.

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