Crystal Orientation of a Thin Film Evaporated in a High Magnetic Field

Masahiro Tahashi, Kensuke Sassa and Shigeo Asai

Dept.of Materials Processing Eng. Nagoya Univ., Furo-cho, Chikusa-ku, Nagoya, 464-8603, Japan Fax: 81-052-789-3247, e-mail: h982226m@mbox.media.nagoya-u.ac.

The aim of this study is to investigate the effects of a magnetic field on the crystal orientation in films deposited by use of a laser ablation method. Zinc and bismuth as a nonmagnetic substance were deposited on a glass substrate under a high magnetic field of 5T. The substrate was set in a container kept in 10⁻⁵Torr and submerged in parallel to the magnetic field line. When the magnetic field was applied, bismuth deposited to be in c-plane orientation. This experimental result agrees with the theoretical estimation based on the magnetization energy. The orientation index showing the degree of the c-plane orientation increased with increase of the distance between a target and a substrate. It is suggested that a radiation heat supplied from a laser heat source interrupted the crystal orientation in the vicinity of the target. On the other hand, the orientation of zinc which has smaller absolute magnetic susceptibility than that of bismuth, was not clearly appeared in the magnetic field of 5T.

Key words: Electromagnetic processing of materials, Vapor-deposition, Crystal orientation, High magnetic field,

Nonmagnetic substance

1. Introduction

The crystal orientation is one of the most crucial factors to determine the electric, magnetic and mechanical properties so that the development of crystal alignment methods has been desired. The technical development of superconducting magnets has enabled to introduce a rather high magnetic field, say 10T in ordinary laboratories, which could influence the physical properties of not only ferromagnetic materials but also nonmagnetic ones. That is, under such a high magnetic field the magnetization force acting on nonmagnetic materials is not negligible any more ¹⁾⁻⁶⁾. Furthermore, a material has a different magnetic susceptibility depending on each orientation of a unit crystal. Taking account of this physical characteristic and the utilization of a high magnetic field, we now have a possibility to develop a new method to align crystals in a favorable orientation.

The aim of this study is to develop a new method to align crystals of nonmagnetic materials such as zinc

and bismuth in a vapor-deposition process.

2. Theory

Zinc and bismuth have a h.c.p. lattice which shows a magnetic anisotropy depending on the crystal orientation, where magnetic susceptibilities along a,baxis and c-axis are $\chi_{a,b}|_{Z_n} = -1.81 \times 10^{-5}$, $\chi_c|_{Z_n} = -1.33 \times 10^{-5}$ ^{7),8)} and $\chi_{a,b}|_{B_i} = -1.24 \times 10^{-4}$, $\chi_c|_{B_i} = -1.76 \times 10^{-4}$ ⁹⁾, respectively. The magnetization

energy given in Eq. $(1)^{10}$ determines the favorable crystal direction depending on the magnetic susceptibility of each crystal axis in a given magnetic field.

$$U = -\frac{\chi}{2\mu_0 (1 + N\chi)^2} B_{ex}^2$$
 (1)

, where χ is a magnetic susceptibility, μ_0 is a permeability in vacuum, N is a demagnetization factor

and B_{ex} is an applied magnetic field. When a crystal was set in a magnetic field, the crystal tends to align to the favorable crystal direction with a lower magnetization energy. Substituting values of the magnetic susceptibility of zinc and bismuth into Eq.(1), we get $U_c < U_{a,b}$ in the case of zinc and $U_{a,b} < U_c$ in the case of bismuth. This means that c-axis of zinc crystal and a,b-axis of bismuth are the favorable crystal direction parallel to the magnetic field. When the substrate was set in parallel to the magnetic field as shown in Fig.1, the a,b-axis orientation in zinc and the caxis orientation in bismuth can be expected to appear on the substrate.



Fig.1 Crystal orientation of zinc and bismuth

3. Experimental

Figure 2 shows a schematic view of the experimental apparatus where a vacuum chamber was set in a 90mm diameter bore of a superconducting magnet. A glass was used as a substrate which was prepared by cleaning in an ethanol bath with an ultrasonic cleaner to eliminate organic materials on the surface. The vacuum chamber was evacuated up to 10⁻⁵ Torr by use of a diffusion and a rotary pumps. A target was vaporized by use of a YAG laser beam operating at the wavelength of 1064nm. In order to control the temperature of a substrate, it was heated by the electric current passed in a tantalum wire which was placed behind a substrate holder. The temperature was measured by a thermocouple contacted to the substrate holder.

The crystal orientation of a deposited film on the substrate was examined by means of the X-ray diffraction analysis and the orientation index¹¹) was calculated from the intensity of the X-ray diffraction lines.



Fig.2 Schematic view of experimental apparatus

4. Results and Discussion

Zinc was deposited on the substrate($10 \times 10 \times$ 0.7mm³) placed at the point with the maximum strength of the magnetic field. The X-ray diffraction patterns of zinc are shown in Fig.3, where the difference between the cases with and without the magnetic field is not clearly seen. That is, the magnetic susceptibility of 10^{-5} order in zinc is not large enough to align crystals in the magnetic field of 5T. On the other hand, the X-ray diffraction patterns of bismuth given in Fig.4 show that the peak of (012) scarcely appeared in the case of 0T, but the peak of a c-plane increased in the magnetic field of 5T. This result agrees with the theoretical estimation mentioned before. That is, the magnetic field of 5T is strong enough to align crystals of bismuth with the magnetic susceptibility of 10^{-4} order.

Taking account into the distribution curve of the magnetic field intensity in the superconducting magnet, we placed the substrate in the comparatively uniform magnetic field region and vaporized bismuth. The orientation indexes of the c-plane (003) in bismuth obtained under the different magnetic intensities were plotted along the distance between the target and the substrate as shown in Fig.5. The broken line with the value of 1 shows the standard orientation index. In the case of 0T any specific orientation is not clearly observed at every place, but the orientation indexes obtained under the imposition of the magnetic field of 3T and 5T, surely increase with increase of the magnetic field intensity and the distance from the target.

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Fig.3 X-ray diffraction patterns of zinc films at B_{max} position



Fig.4 X-ray diffraction patterns of bismuth films at B_{max} position





The magnetization energy difference due to the magnetic susceptibility difference is given as Eq.(2) from Eq.(1).

$$\Delta U = \frac{\left|\Delta \chi\right|}{2\mu_0 \left(1 + N\chi\right)^2} B_{ex}^2$$

$$\approx \frac{\left|\Delta \chi\right|}{2\mu_0} B_{ex}^2 \quad (\because 0 < N < 1, \quad \chi << 1)$$
(2)

, where $|\Delta \chi|$ is an absolute value of magnetic susceptibility difference depending on a crystal orientation and the values of zinc and bismuth are $|\Delta \chi|_{z_1} = 4.8 \times 10^{-6}$ and $|\Delta \chi|_{B_i} = 5.2 \times 10^{-5}$,

respectively. The relationship between the orientation index and the magnetization energy is shown in Fig.6. The significant effect of the magnetic field on the crystal orientation clearly appeared in the region over 100J/m³.

For the crystal orientation by the imposition of a magnetic field, the magnetization energy should be larger than the thermal one. This condition is given in Eq.(3),

$$\frac{\left|\Delta\chi\right|}{2\mu_0}B_{ex}^2 V > kT\tag{3}$$

, where V is volume of a particle, k is the Boltzmann's constant and T is a Kelvin temperature. From Fig.5 it can be imaged that the orientation could be suppressed in the vicinity of the target due to the thermal energy introduced by a laser source. The mechanism of the crystal orientation in a vapor-deposition process can be imaged as followings. Molecules evaporated from the surface of the target are agglomerating to become a particle during travelling from the target to the substrate. When the particle exceeds to the certain volume which is calculated from Eq.(3), it feels the magnetization force rather than the thermal disturbance force and rotates to the favorable crystal orientation and then attaches the substrate.



orientation index(zinc:(100), bismuth:(003))

5. Conclusion

- The crystal of bismuth deposited under the magnetic field of 5T shows to the crystal orientation and its orientation index increased with increase of the magnetic field strength and the distance from the target.
- The orientation of zinc which has smaller absolute magnetic susceptibility than that of bismuth, was not clearly detected under the magnetic field of 5T.
- The magnetization energy over 100J/m³ was required for the crystal orientation in the vapor-deposition process.
- 4) The crystal orientation in a high magnetic field

becomes tangible when the magnetization energy stored in a particle exceeds the thermal energy.

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