A challenge to the low-cost production of highly functional optical elements

- Fabrication of sub-wavelength periodic structures via glass-imprinting process -

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Factors, such as production cost, which impede practical applications of "sub-wavelength optical elements" with periodic structures smaller than light wavelengths, were targeted for alleviation through a combination of advanced Japanese glass molding methods and a novel imprinting process. Collaboration between material manufacturers, consumer-electronics companies, universities, and AIST under a clear sharing of roles enabled the fabrication of functions such as polarization rotation effects and antireflection effects on glass surfaces.

Keywords : Optical element, periodic structure, imprinting, glass, microfabrication

1 Introduction

Semiconductor technology and optical technology contributed to the formation of advanced information society. Optical technology has hundreds of years of history, and still receives high expectations because of the following two reasons. The first reason is the extremely large information capacity and communication speed. The second reason is that humans receive over 80 % of information through sight. The expectation for optical technology grows every day, because of the technological innovations of various kinds of hardware such as display, storage, and imaging devices, as well as the dramatic progress of mutual information network. To respond to such expectation, the methodology in question now is how to establish secure "technology" from the results of optics accumulated as "science".

This paper focuses attention on optical elements which are expected to significantly affect the next generation technologies for information input and output (I/O). I shall describe the works to realize the industrial fabrication technologies of next generation optical components placed behind the current refraction and diffraction optics, by combining factors which are scientifically clarified and predicting future optical device technologies. Such technologies will be required in the fields of home electronics and information technologies in the next 10 to 20 years.

If the fabrication technology of next-generation optical elements is established for the future information I/O, users will be able to enjoy high-speed and effective imaging, storage, and replay a large-volume data including high quality images. On the other hand, manufactures will receive substantial advantages such as greatly simplified manufacturing process of optical elements without so much process energy, and also the reduction of number of optical parts, which will enable the production of advanced home electronic devices earlier than the neighboring countries.

2 Future direction and research objectives for industrialization

Optical elements and their production method have appeared since 1600 are shown schematically in Figure 1. Manufacturing in this field can be classified into "optical material" and "microfabrication". The development of materials with properties required through the design such as refraction index and dispersion, the precise processing following the characterization are continued repeatedly until they satisfy each other. During the period from 1600 to 1800, various theories were formulated in geometric optics and wave optics based on three optical elements: lens, prism, and diffraction grating. Such theory and optical design led the manufacturing. The manufacturing responded to the demands accurately, and in turn promoted further



Fig. 1 Optical elements that appeared after 1600 and fabrication methods that contributed to the industry

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advancement of theory and design. This process has been continued to this day.

2.1 Barrier of cost

Let me direct attention to the field called "resonance and sub-wavelength", for which the research was started around 1990. Computer simulations demonstrated that the highly functional elements could be actualized if the periodic structures comparable to wavelength of visible light or less could be fabricated precisely. Later, some basic studies on the fabrication of prototypes were carried out actively, alongside the progress of semiconductor microfabrication technology such as silicon^[1].

The wavelength discussed in this paper is from visible to near-infrared region (wavelength about 400 nm to 2000 nm). If the period of structure is 2n times (n = 0, 1, 2, 3...) of the wavelength of incident light, the optical diffraction will occur. Such structures will also cause strong reflection and light trapping due to resonance of light within the periodic structure. When the period of structure decreases, diffraction and resonance do not occur, and the refractive index of such periodic structure can be considered as the average of air and material. This is the principle of optical devices classified in the "resonance and sub-wavelength" domain.

Although several optical elements have been realized in resonance and sub-wavelength domain, their applications were limited. Namely they were not installed widely in commercial devices such as home electronics products, because the industrial arena required an extremely low production cost and a large production scale of several million elements or more per month.

2.2 Barrier of function

The molding process and injection process invented in 1980 enabled the productions of aspheric lenses and diffraction gratings, which used to be difficult to fabricate at low cost with conventional grinding and polishing methods. The fabrication technology of precise molds accelerated the massproduction of several optical elements with various forms including aspheric lenses. For example, the glass lenses fabricated by the molding are used in almost 100 % of zoom optics for digital still cameras. Hundreds of millions of lenses



Fig. 2 Schematic diagram of glass molding method and imprinting method

Heat resistant, ultra-hard material is used for glass molding, while glass or silicon is used for imprinting mold.

are produced each year in Japan and neighboring countries. However, new demands arose for comprehensive suppression of ghost, spherical aberration, and color aberration, in addition to high-resolution, downsizing, and weight reduction. On the other hand, since the blue-ray with 405 nm wavelength is used in next-generation optical disc drive, which is called Blu-ray, the new optical element compatible with 3 wavelengths including conventional CD (wavelength 785 nm) and DVD (wavelength 655 nm) is required. To fulfill the needs of such functional elements, new technology is necessary to incorporate the several functions such as refraction and diffraction, structural birefringence without wavelength dependence, and antireflection with little dependence both on wavelength and incident angle, into conventional optical elements such as lens.

2.3 Objective of this research

Conventional imprinting technology can be used only in resin because there is an upper limit in molding temperature. There was no report on the application of the imprinting process to glass that requires high temperature of several hundred degrees. Also, glass-molding process is used for the production of optical elements with flat surface such as lenses. Eventually, there was almost no approach to forming structures smaller than wavelength of visible light. Thus the objective of this study is to develop new fabrication technology of resonance and sub-wavelength optical elements, which have been fabricated using microfabrication technologies so far, through the development of glass imprinting process by combining the imprinting process for resin material used in the academia, and the glass molding process used in industry.

3 Scenario to achieve objective

The size required for the resonance and sub-wavelength optical elements is in the range of tens of nm to several µm. It is also necessary to fabricate microstructures on a large surface in a shorter time possible. It should be out of the range covered by the methods such as lithography and etching, laser process, or mechanical process, which are currently used in industry. Such microstructures should be formed mostly on the surface of lenses, prisms, or window materials. Therefore, it is advantageous to transfer such structures to the surface of the optical elements using the principle of imprinting process, if the thermally durable molds can be fabricated. The concepts of molding process and imprinting process are shown in Figure 2. Imprinting process was first reported by Chou et al. of Princeton University in the United States^[2, 3]. It is a method where the mold with nano-structure is pressed against the resin, and the structure is transferred using ultraviolet light or heat^[4]. So far, the products realized using the imprinting process are mainly based on the resin materials such as light

guide panels for liquid crystal displays. The technological level of Japanese molding process, on the other hand, is outstanding in the world. There are plenty of accumulated human resources, facilities, and knowledge. Unfortunately, the products based on the reliable glasses are only micro lens arrays and diffraction gratings with the period over 10 μ m. Therefore, the glass devices in the resonance and subwavelength range have been an unexplored territory.

In this study, several research objectives were set based on the scenario shown in Figure 3, which were fundamental or intermediate technologies to accelerate the development of sub-wavelength optical element using glass-imprinting process. AIST studied the fundamental technologies using the vast amount of research results from the past, such as the development of glass compositions. On the other hand, the three processes including mold fabrication, coating for demolding, and the precision molding process relied heavily on the knowledge of the home appliance companies. Therefore, such works were conducted carefully and strategically while AIST supported the home appliance companies.

4 Development of new optical components through integration of elemental technologies

A central laboratory was established in Kansai Center of AIST to integrate the research potentials of home appliance

companies, material companies and universities. The former companies install the optical elements fabricated by the molding process in the final products. The latter companies develop several glass materials appropriate for molding process. The universities conduct the advanced simulation research in optics and rheology. Here, it was important that the knowledge of microfabrication and characterization technologies for glasses and ceramics were accumulated within the AIST research group. An example of successful development of optical elements is described in the following sections.

4.1 Development of wave plate based on structural birefringence

Wave plate is used in the optical disc drive to separate the light traveling from the light source to the disc and the light reflected on the surface of a disc in order to detect the optical signal by the photo diode. The materials for the current wave plate are resin or crystal, which have different specifications depending on the operating wavelength. The next-generation optical disc drive, therefore, requires 3 wave plates depending on the wavelengths between blue and red, which is considered to be an obstruction factor for the downsizing of optical system and the reduction of production cost. Also, as wavelength of the light source shortens, sufficient light resistance is required for the optical elements. The glass wave plate based on the structural birefringence is a promising candidate for overcoming these issues.



Fig. 3 Scenario to realize the fabrication of sub-wavelength optical elements using molding method

When one-dimensional microstructure with a period smaller than the wavelength is formed on the surface of transparent material, so-called "structural birefringence" appears, where the refractive index is different depending on the direction of electric field of passing light^[5]. Even in optically isotropic glass, structural birefringence can be realized if anisotropic sub-wavelength structure is formed on the surface. Theoretical optimization of the structure is possible using the calculation methods such as the effective medium theory or the rigorous coupled wave analysis. Important parameters are period, groove width, and height of the periodic structure, and refractive index of the material. Especially, the optimization of groove width is an essential point in order to minimize the wavelength dependency of phase retardation. Moreover, height of structure can be lessened by increasing refractive index of the material. Glass material is advantageous to attain higher refractive index than resin, resulting in the lower height of structure. However, there was no report on whether such structure can be formed on the surface of glass or not, using the molding process.

In this study, as the first step, the period was fixed at 500 nm, and various heat resistant molds with various groove widths were fabricated to study the molding characteristics of glasses quantitatively. The results are shown in Figure 4. In the case of mold with groove width 330 nm, the structural height of the molded periodic structure reached 730 nm, which means an extremely high precision molding was possible. It is the first quantitative investigation on the correlation between the shapes of mold and the molded periodic structure obtained by imprinting. The most important point here was the demolding condition. When the mold was released from the glass at high temperature, microstructure formed on the glass surface was deformed by heat. When the demolding was carried out at low temperature, a mechanical damage was caused either in mold or glass or in both by the difference in thermal expansion coefficients between the mold and the glass. Optical glass that could be molded at relatively lower temperature than 500 °C was advantageous because the deterioration of the mold can be prevented.

However, the determination of demolding temperature became extremely difficult, because, in general, the viscosity of glass changed rapidly near molding temperature. To solve this issue, the researchers of companies, who have plenty of experience in the molding process, and the researchers of AIST, who are knowledgeable in material property and microfabrication, collaborated successfully. The demolding the periodic structure with the highest aspect ratio in the world was successfully accomplished in a short period. In this study, large surface area of 6 mm x 6 mm, as shown in Figure 5, was confirmed. The phase retardation attained by this structure was 0.1 $\lambda^{[6]}$, which was the first result to realize the phase retardation caused by the periodic structure via glass molding process. Molding of the structure with 300 nm period was also successfully achieved. The future goal is to attain the phase retardation of 0.25 λ in the wavelength region between 400 and 800 nm by optimizing the dimension of periodic structure, which is the requirement for practical use in the next generation optical disc drive.

4.2 Development of sub-wavelength antireflective structure

The improvement of the transmission efficiency of light by minimizing the unnecessary reflection is desired for glass optical components used in wide-ranging products such as home electronics, lighting, etc. Currently, antireflection film is coated on the optical elements for imaging and display panels. However, in principle, such films can not respond to demand for the antireflection independent against wavelength or incident angle. On the other hand, it was known that advanced antireflection could be achieved if periodic conical shape structure with the sub-wavelength period could be formed on the surface of elements.

Important point was to arrange the conical shape units twodimensionally with a period smaller than the wavelength. The antireflection effect could not be achieved when the conical structure with the period comparable to wavelength



Fig. 4 SEM photographs of one-dimensional periodic structure mold and molded glass



Fig. 5 One-dimensional periodic structure with large area showing retardation.

level cause the diffraction of incident light. In the case of the structures with the period sufficiently smaller than the wavelength, volume fraction of air was greater than the glass at the top area of the structure, and their volume fractions gradually reversed as the incident light traveled further into the structure. Thus the interface of glass and air seemed nonexistent for the incident light coming into the structure from various angles. Also, two-dimensionally isotropic arrangement canceled out the polarization dependence.

Although several theoretical analyses and the fabrication researches have been published for sub-wavelength antireflection structures, most of them dealt with resins such as acrylic^[7], and never got beyond prototypes. There were some researches using electron beam lithography and dry etching for the fabrication of antireflection structure on the surface of glass ^[8, 9]. However, the mass production was difficult because of the prolonged fabrication time. Therefore, we decided to fabricate such antireflection structures using the glass molding process.

In this paper, I will describe the study in which silica glass was used as heat resistant mold. The metal thin-film coated on the silica substrate was patterned by electron beam lithography, and then the desired periodic structure was formed on the substrate by a dry etching process. The shape of mold was designed so as to minimize the reflection on the surface of glass. Figure 6(a) shows a two-dimensional periodic structure with period of 300 nm and the height about 550 nm. Using a vacuum coating process, a thin film was coated on the mold surface to prevent the thermal adhesion between the mold and the glass. A phosphate optical glass with refractive index of 1.6 was pressed at about 500 °C. Finally, an inverted periodic structure with height about 500 nm was successfully obtained as shown in Figure 6(b)^[10]. Here, the important point was the temperature of demolding. The reflectivity on the surface of optical glass with periodic structure was measured precisely using an integrating sphere, which was 0.56 % at the perpendicular incidence and at the wavelength 462 nm. This value was lower than that of a single antireflection thin film coated on the glass surface. Therefore, the performance of this antireflection structure was in the practical level. Figure 6(c) is a photograph of the prototype. The surface reflectivity decreased with optimization of molding condition, and the characters under



Fig. 6 SEM photographs of (a)mold and (b) molded antireflection structure. (c) External shot of molded glass.

the glass plate could be read clearly from a tilted angle. A current target of this research is the pickup lens for optical disc drive and digital still camera lens. The development of curved surface mold using ultrahard materials in place of silica is in progress. This research will open a possibility of overcoming the problems such as low cost and mass production that delayed the practical uses of antireflection structures.

5 Discussion

Vertical collaboration between companies, AIST and universities gathered from the three fields, i.e., materials, microfabrication, and devices, was effective beyond expectation. The optical glass technology in Japan is at the top level of the world. Actually, some glass materials suitable for imprinting are commercially available. However, many more hurdles must be cleared from the aspect of mass production. Glass companies are working efficiently on the three research issues of composition improvement, molding, and device characterization, with collaboration of AIST, mold material companies, and home appliance companies. However, since mold materials are not developed for the purpose to fabricate subwavelength structures with microfabrication process, the mold material and its production method must be optimized through close cooperation with manufacturers. If the results are adequately patented, these material technologies might be protected from the catch-up of neighboring countries. On the other hand, it is known that the glass materials at around the molding temperature range show viscoelastic behavior, but currently there are few data on such high temperature properties required for the molding simulation. In some cases, it is necessary to develop the measurement equipment of such properties, which will also be patented just like the materials.

On the other hand, three processes including the mold fabrication, the demolding thin film coating and the precision molding are faced with hard problems in terms of research strategy. Even if the technologies related to these processes could be patented, it would be extremely difficult to protect them from the catch-up of neighboring countries. Some believe that it may be of best interest to leave them as "black boxes". Recently, there are moves to commercialize the technical know-how on fabrication methods as a recipe packaged with the production instruments for processing, coating, molding, or others.

Actually, some recipes are packaged with the equipments used in our research project. Therefore, anyone can fabricate the conventional devices using such recipes, which are based on the knowledge accumulated in the collaborative work between the equipment companies and the optical device companies. In the future, it will be extremely important to achieve high performances in processing, coating, and molding equipments, in order to improve the mold fabrication and imprinting processes to a practical level. The optical device companies will be required to decide whether to continue the research and development entirely in a black box or to commercialize such knowledge as recipe by the collaboration with equipment companies.

6 Summary

The objective of the research described here is to develop the relatively small optical devices used in home electronic products such as imaging devices and optical disc drives. The technologies in this area are facing a fierce catch-up of neighboring countries, and are required to overcome the two issues of cost reduction and performance improvement. "Glass imprinting process" has potential to realize the reduction of component number, the manufacturing energy consumption and the cost. Therefore it is expected that this process becomes the core of next-generation fabrication process of optical components. For example, if the wavelength independent wave plate can be fabricated by this process, the number of wave plates necessary in nextgeneration CD/DVD drive is reduced to one-third. Also, the antireflection coating of the lens will become unnecessary.

Research and development for the sub-wavelength optical elements described in this paper will be applicable to large displays, lighting equipments, solar cells and so on. It is ultimately expected to reduce the energy consumed of information I/O devices with highly efficient control of optical signals. In order to realize such optical devices, the microfabrication technology and material technology must be fused efficiently. Until now, collaboration took place mainly between companies, and the universities and government institutes rarely participated. It was recognized that the technology was already completed not only in "Type 1 Basic Research" but in the field of "Type 2 Basic Research" as stated by AIST. However, after the efficient improvement of technology, we faced new technological barriers, that could not be solved by conventional disciplines. Therefore, researchers were required to return to both "Type 1" and "Type 2 Basic Researches" to extract issues and research subjects to be tackled, and to solve several problems efficiently and timely. Here the important point is the role of AIST assuming the missions of Type 2 Basic Research. AIST must maintain the potential to exhibit not only the research subjects directly linked to demand but also the research results obtained by the collaboration with companies and universities. Also AIST must centrally manage various materials and state-of-art infrastructures.

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References

- H. Kikuta and K. Iwata: Hacho yori komakana koshi kozo niyoru hikari seigyo (Optical control by grating structure finer than wavelength), *Kogaku*, 27, 12-17 (1998) (in Japanese).
- [2] S.Y.Chou, P.R.Krauss and P.J.Renstrom: Imprint of sub-25 nm vias and trenches in polymers, *Appl. Phys. Lett.*, 67, 3114-3116 (1995).
- [3] S.Y.Chou, P.R.Krauss, W.L.Guo and L.Zhuang: Sub-10 nm imprint lithography and applications, *J. Vac. Sci. Technol.*, B 15, 2897-2904 (1997).
- [4] T.Yoshikawa, T.Konichi, M.Nakajima, H.Kikuta, H.Kawata and Y.Hirai: Fabrication of 1/4 wave plate by nanocasting lithography, J. Vac. Sci. Technol., B 23, 2939-2943 (2005).
- [5] H. Miyakoshi, M. Morikawa, O. Masuda, M. Imae, M. Yamada, and K. Yoshida: Nano impurinto o riyo shita sabu hacho kozo kotai'iki hachoban no sakusei (Manufacture of sub-wavelength structure wide band wave plate using nanoimprinting), *Optics Japan 2005 Collection of Papers, 24pD5*, Tokyo (2005) (in Japanese).
- [6] T.Mori, K.Hasegawa, T.Hatano, H.Kasa, K.Kintaka and J.Nishii: Fabrication of sub-wavelength periodic structures upon high-refractive-index glasses by precision glass molding, *Proc. of 13th Micro Optics Conference*, B2, Takamatsu, Japan (2007).
- [7] S.J.Wilson and M.C.Hutley: The optical properties of 'moth eye' antireflection surfaces, *Optica Acta*, 29, 993-1009 (1982).
- [8] H.Toyota, K.Takahara, M.Okano, T.Yotsuya and H.Kikuta: Fabrication of microcone array for antireflection structured surface using metal dotted pattern, *Jpn. J. Appl. Phys.*, 40, L747-749 (2001).
- [9] J.Nishii, K.Kintaka, Y.Kawamoto, A.Mizutani and H.Kikuta: Two dimensional antireflection microstructure on silica glass, J. Ceram. Soc. Japan, 111, 24-27 (2003).
- [10] M. Umetani, K. Yamada, T. Tamura, Y. Tanaka, and J. Nishii: Garasu hyomen ni hansha boshi kozo o keisei suru seikei purosesu no kaihatsu (Development of molding process to form antireflection structure on glass surface), *The Japan Society for Precision Engineering Autumn 2007 Conference Lectures*, L33, Asahikawa (2007) (in Japanese).

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After completing masters degree at Graduate School of Engineering and Faculty of Engineering, Tokyo Metropolitan University in 1982, joined Nippon Sheet Glass Co., Ltd. Participated in project of Japan Key Technology Center, and obtained doctorate in engineering in 1990 by writing dissertation on work conducted during the stay. In 1993, joined Osaka National Industrial Research Institute, Agency of Industrial Science and Technology, and became Chief of Glass Structure Research in 1999. From 2002, Professor, Graduate School of Science and Technology, Kobe University. Project Leader of Next-Generation Light Wave Control Material and Element Creation Technology, NEDO from 2006. Visiting Professor, Graduate School of Engineering, Osaka Prefecture University in 2007.

Discussion with reviewers

1 Development of technology integration

Comment and Question (Naoto Kobayashi)

This research paper integrates "mold technology" and "nanoimprinting technology" for a breakthrough that allows practical application of high-performance optical element unavailable before, and in that sense it is extremely valuable. The realization of such integration represents true nature of Type 2 Basic Research.

Please comment on how we should think in terms of developing research and technology in combining technology where integration was considered impossible, as in this study, to use the experience of creating technology that never existed before, and to realize new values.

Answer (Junji Nishii)

It is clear that the reasons why "resonance and subwavelength element", which was actively pursued as academic disciplines, failed to become major industry because of "scale of manufacture" and "cost". I started working on this research thinking that the scenario for how to solve this matter was important. Here, I think it was important to consider the role of AIST that declares "Full Research" as its mission. I believe the research result should be directly reflected in people's lives or indirectly reflected in products of private companies. However, what is important here is the selection of subject for Full Research and the scenario beyond, and I think it's whether the research is in synch with policy set by government or whether the research can set the direction government should take. Subject selection for Full Research should not be by inspiration, on whim, or delusion, but it should be determined by whether it matches policy and demand.

As with other technologies, I think research and development for energy conservation is necessary in future optics technology. The case study discussed here should serve as foundation for "efficient use of light" in many optical devices.

2 Beginning of resonance and sub-wavelength optics research

Question (Naoto Kobayashi)

You mentioned that the research in the field called "resonance and sub-wavelength", which is the central issue of new technology described herein, started around 1990, but why was it started then (or in other words, why didn't it come to attention until then)? **Answer (Junji Nishii)**

Researchers of interference, polarization, and diffraction of light were certainly aware of the importance of optical elements in the region of "resonance and sub-wavelength". However, no method for designing or manufacturing such elements existed, so it could not be tackled as research subject until the latter half of 1900s. Advances in computer simulation and microfabrication technology triggered the research. In the same way, there is potential for new technology to emerge depending on what comes out of the semiconductor technology.

3 Technological points in integration

Question (Akira Ono)

In this research, you succeeded in the manufacture of structural birefringence wave plate and sub-wavelength antireflection structure using glass imprinting method. I understand that the factor of success was to realize the integration of three intermediate technologies including "precision mold", "demolding", and "microfabrication" by integrating the elemental technologies as shown in Figure 3. Please explain, as much as you are allowed, the methods of integration that were critical in obtaining the result.

Answer (Junji Nishii)

As mentioned in the latter part of Section 3 and the first part of Section 4, it was important to accurately extract research subjects for multiple elemental technologies necessary to achieve the goal, and to clarify the scenario for their integration. Although this was an ordinary process, I discussed again and again with researchers of universities, AIST, and companies that wished to use the research result in future products. As a result, I obtained agreement that the central lab method was preferable to maximize a limited research budget. However, "precision mold" and "demolding" technologies were corporate knowledge, and AIST and the universities decided not to step into those fields deeply, and decided to carefully and strategically support the companies. However, since this was a joint research conducted in the same place, we did share findings that we didn't know before. Please note that any more disclosure of information is not allowed at this point, but I am aware that public funds were invested, so things that could not be patented will be shared in the form of know-how recipe under the strict management.