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ArF Excimer Laser Projection Lithography Using Partially Achromatized Lens System

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To study the feasibility of ArF excimer laser lithography, four prototype projection lenses have been designed—a monochromatic lens, an achromatic lens with separate elements, an achromatic lens including cemented elements, and a partially achromatized lens. From the viewpoint of the current technology limit of bandwidth narrowing of ArF excimer laser and accuracy of optical surface fabrication, the partially achromatized lens is found to offer the best solution. 0.18- μ m resolution is confirmed using the fabricated prototype lens combined with the 10-pm bandwidth laser and Ag/Se-Ge inorganic resist. Quarter-micron lines-and-spaces pattern is obtained using a tri-layer resist system.

KEYWORDS: excimer laser, lithography, spectral narrowing, resist

§1. Introduction

ArF excimer laser projection lithography has attracted considerable attention due to its high resolution capability in the quarter-micron region. In a KrF excimer laser lithography system, an all-quartz monochromatic lens has been employed in combination with a spectral narrowed laser.¹⁾ A similar approach cannot be applied to ArF excimer laser lithography, because the transmissivity of currently available fused quartz at 193-nm is insufficient at present, and the spectral narrowing becomes much more difficult. One attempt to solve these problems by reducing the number of element lenses using an aspherical lens system has already been tried, and quarter-micron resolution has been reported.²⁾ We have adopted a different approach, in which a partially achromatized lens made of fused quartz and CaF₂ and a moderately spectral narrowed laser are used. In this work, we study the feasibility of ArF excimer laser lithography, through the fabrication of a prototype reduction projection lens. The fabricated lens is then employed in patterning experiments.

In §2 of this paper, various types of lens systems are compared in terms of design consideration. We conclude that the optimum lens system, available assuming current fabrication technologies and lens materials, is the partially achromatized one. In §3, properties of spectral narrowing using an etalon are described, based in optical absorption characteristics of dielectric films applied to the etalon. In §4, the resolution limit of the fabricated partially achromatized lens with 10-pm bandwidth laser is evaluated, and resolution limit of 0.18- μ m is demonstrated. Section 5 briefly describes resist systems suitable for ArF excimer laser lithography, and quartermicron tri-layer resist patterns obtained are shown.

§2. Optical System

In this study, we only employed a spherical lens for the element lens of the projection system, because the level of optical surface fabrication of this lens is higher than that of an aspherical lens. Four types of projection lenses were investigated by designing prototypes of each type with a 5×5 cm² field, a magnification of 1/10, and a numerical aperture of 0.47.

The transmissivity and refractive indexes of lens materials, and the precise central wavelength of ArF excimer laser are important value to design the projection lens system. Figure 1 shows the transmissivity characteristics of CaF_2 and quartz in the VUV region. The difference in transmissivity between the 5-mm-thick and 10-mmthick samples indicates the existence of their inner absorption. The critical wavelengths at which the inner absorption appear are 180-nm for CaF_2 , and almost the wavelength of an ArF excimer laser for quartz. We generally applied CaF_2 as the design material for the lenses.

The precise wavelength of the ArF excimer laser was determined to be 193.546-nm referring to the specific lights from low-pressure Hg lamp of 184.9497-nm and 194.2273-nm. The refractive indexes of CaF₂ and quartz at 23 °C were determined to be 1.4956 and 1.5598 respectively, by measuring the minimum deviation angles and the vertical angles of the prism.

The four lens types, along with the bandwidth (FWHM) and optical surface accuracies of the element lenses, they would require to achieve 40% of MTF for a 0.25-µm lines-and-spaces pattern, are summarized in Table I within an error of $\pm 10\%$. The bandwidth should be narrowed to 5-pm for the monochromatic CaF_2 lens. This degree of narrowing is difficult to attain as will be explained shortly. A bandwidth as wide as 200-pm satisfies the requirement of the two types of achromatic lenses consisting of fused quartz and CaF₂. However, the achromatic lens consisting of separate elements requires an extremely precise accuracy of optical surface fabrication of $\lambda/100$ ($\lambda = 632.8$ nm) as shown in Table I. Moreover, no currently available measurement method is capable of verifying accuracies as high as $\lambda / 100$. Application of the achromatic lens with the cemented elements calls for the development of low-absorption cement. The partially achromatized lens consisting of fused quartz and CaF₂ requires a bandwidth of 10-pm. The problems associated with achromatic lenses were not found in this type of lens. The partially achromatized lens type was



Fig. 1. Transmissivity of CaF_2 and quartz vs wavelength.

Table I. Comparison of lens characteristics.

Lens and material	Bandwidtl (pm)	Surface accuracy $(\lambda = 632.8 \text{ nm})$
Monochromatic lens	5	λ/50
(CaF ₂) Achromatic lens	200	λ/100
with separate elements $(CaF_2 and quartz)$	200	λ/50
Achromatic lens with cemented elements (CaF ₂ and guartz)	200	λ/ 30
Partially achromatized lens $(CaF_2 and quartz)$	10	λ/50



Fig. 2. MTF at the center of the field as a function of spatial frequency for three spectral bandwidth. The numerical aperture is 0.47. The coherence factor of a illumination system is 0.7.

judged to be most serviceable at present.

The trial partially achromatized lens consisting of 5 CaF₂ lenses, 4 quartz lenses and 1 quartz cover plate was fabricated. The cover plate prevents the bottom lens from being contaminated by the decomposed fragments of resist caused by the photo ablation. The numerical aperture is variable from 0.47 to 0.3. The magnification is 1/10, and the field is 5-mm square. However, the center field of the 500-µm square is focused to obtain a high resolution, while the field outside this area is not intentionally designed to obtain a high resolution. The simulated MTF is almost constant in the field of 1-mm square, and decreases by 95, 85, and 15% with increasing distances of 2.83, 3.54, and 7.07-mm, respectively, from the center. The illumination system was identical to the one we developed for KrF excimer laser lithography.³⁾ Three or four layer films consisting of NdF3 and MgF2 were used to suppress the surface reflection from the large or small curvature element lenses, respectively. The optical path from the laser to the wafer was purged with N_2 . The maximum power received by the reticle is 14.8% of emitted power from the laser. The transmissivity of the projection lens is 60%.

Figure 2 shows simulation results of the dependence of MTF on the spatial frequency. 40.3% of MTF is obtained at 2000 lines/mm, which corresponds to a 0.25- μ m lines-and-spaces pattern for a 10-pm bandwidth. Only 14.5% of MTF is obtained at 2000 lines/mm for the 20-pm bandwidth. Bandwidth narrowing is still important even for the partially achromatized lens.

§3. Spectral Narrowing

Several techniques are available for achieving spectral narrowing by inserting a dispersive element in the resonator such as a prism, a grating, or an etalon. However, the spectral bandwidth cannot be fully reduced

ArF Excimer Laser Projection Lithography

with the prism, because no prism material is available having a sufficient dispersion. Although both gratingand etalon-based techniques provide the same level of bandwidth narrowing, the grating technique is too susceptible to vibration of the grating.⁴⁾ Furthermore, the grating requires a lower beam divergence angle by two orders than that of the conventional excimer laser with the stable resonator. By default, therefore, we have focused on narrowing the spectral bandwidth by inserting an etalon in the resonator.

To maintain the lasing, transmissivity of the etalon must be maintained at a high level. This is directly related to the absorption of the dielectric multi-layers as a reflection film,⁵⁾ which consists of Al₂O₃ and SiO₂. Figure 3 shows the reflectivity and transmissivity of 241-nm-thick Al₂O₃ coated on a quartz plate. This thickness corresponds to 10 layers of a quarter-wavelength-thick Al₂O₃ film taking the refractive index into account. Deposition was done by vacuum evaporation at a substrate temperature of 250°C after evacuation to $\sim 10^{-6}$ Torr.

The optical energy incident on the film is consumed by three processes, i.e., the reflection, the transmission, and the absorption. The absorption of the film can be then



Fig. 3. Dependence of Al_2O_3 film reflectivity and transmissivity characteristics on deposition chamber environment. The thickness of Al_2O_3 film was 241-nm.

derived from Fig. 3 by subtracting the reflectivity and the transmissivity from 100%. The absorption of the Al₂O₃ film deposited in an O₂ environment (~ 10^{-5} Torr) at 193.5-nm is 4.85%. However, that without O₂ is 9.36%. Therefore, absorption of Al₂O₃ is recognized to be reduced by adding O₂ to the deposition chamber. Absorption of SiO₂ is hardly dependent on the presence of O₂ in the deposition chamber. The degrees of absorption of 241-nm-thick Al₂O₃ and SiO₂ are summarized in Table II.

A spectrum of the free running ArF excimer laser is shown in Fig. 4. The transmission property of the etalon required for the bandwidth narrowing is schematically superimposed in the figure. A free spectral range (FSR) above 0.4-nm is required. To reach this level, a finesse above 40 and 80 is also required to realize bandwidth narrowing to 10-pm and 5-pm, respectively. Figure 5 shows the calculated values of finesse as a function of reflectivity of the reflection film for three flatnesses as the parameter. In this calculation, two dielectric multi-layers are assumed to be perfectly parallel. Reflectivity of more than 90% and etalon flatness of less than $\lambda/100$ ($\lambda = 193.5$ nm) are necessary to achieve a finesse above 40.

The transmissivity of the etalon decreases as the reflectivity and absorption of the reflection film are increased and the flatness of the etalon is decreased.⁵⁾ For example, the transmissivity of an etalon consisting of reflection films with 70 and 80% reflectivity is 40 and 20%, respectively, for a reflection film absorption of 10% and etalon flatness of $\lambda/30$. High reflectivity of the reflection film causes low transmissivity of the etalon. A flatness as high as $\lambda/100$ is unobtainable. Thus, narrowing to even 10pm by one etalon is extremely difficult. In this experiment, therefore, spectral bandwidth is narrowed to 10pm by two etalons.

Table II. Dependence of absorption of 241-nm thick film on the environment in deposition chamber.

Film	Environment	Absorption (%)	
Al ₂ O ₃	with O ₂	4.85	
	without O_2	9.36	
SiO ₂	with O ₂	0.11	
	without O ₂	0.07	



Fig. 4. Spectrum of free running ArF excimer laser light. Dip is attributable to absorption by O_2 in the optical path. Superimposed scheme shows the transmissivity property of the etalon required.

Yoshiharu Ozaki, Yoshio Kawai and Akira Yoshikawa



Fig. 5. Finesse vs reflectivity of dielectric multi-layers at three flatness. λ is 193.5-nm. Both multi-layers in the etalon are assumed to be perfectly parallel.

Table III. Design parameters and realized performance of two etalons.

Etalon	Gap (µm)	Reflectivity (%)	FSR (nm)	FWHM (pm)	Finesse
#1	45	70	0.416	0.055	7.6
#2	310	70	0.059	0.010	5.9

The design parameters and achieved performance of the etalons are summarized in Table III. Taking both the finesse and transmissivity of the etalon into account, a reflectivity of 70% seemed to be adequate, and was realized by stacking 13 layers of Al_2O_3 and SiO_2 . The ArF excimer laser was successfully narrowed to 10-pm with 41% power of free running power. The use of three etalons would be required to narrow the bandwidth to 5-pm, but will not maintain lasing. The practical limit of narrowing is about 10-pm, at present.

§4. Resolution Performance

Evaluation of the resolution performance of the fabricated partially achromatized lens system combined with a spectral narrowed laser with 10-pm bandwidth was done through lines-and-spaces pattern replication. Among organic polymer resists, only a few are applicable at 193.5-nm. One such resist is PMMA.²⁾ Another candidate is the Ag/Se-Ge inorganic resist, especially promising because it has high resolution⁶⁾ and several times higher sensitivity in the VUV region.⁷⁾ This lead us to adopt an inorganic resist for the present resolution test experiments.

Figure 6 shows a SEM micrograph of a series of replicated lines-and-spaces patterns that vary in nominal width at the 0.01- μ m step. It can be seen that the resolution limit of this lens system is at around 0.18- μ m. Here, the term "resolution limit" means the minimum nominal width of the replicable lines-and-spaces pattern ignoring the pattern width accuracy. Assuming that the lens system is fabricated as designed, the MTF value of the 0.18- μ m lines-and-spaces pattern is only 20%. In other words, this will be the obtainable limit of the resist pattern. Some irregularity is observed on the line edge, for reasons not entirely clear at present. It might be attributed to temperature rise of the resist due to the very strong optical absorption ($\alpha > 10^6$ cm⁻¹) at 193.5-nm.

§5. Resist System for ArF Excimer Laser

As described in §4, pattern edge quality was somewhat deficient for the Ag/Se-Ge inorganic resist. Therefore, discussion in this section is restricted to organic polymer





resist. In a previous paper, we have clarified the relations between absorbance, dissolution rates of exposed and unexposed areas, and the pattern profile in 0.4- μ m-thick resist.⁸⁾ To obtain a sidewall angle of the negative resist pattern of 80–90°, an absorbance of 0.8–0.14 is required for a dissolution rate ratio of unexposed to exposed area of 30. The range of absorbance decreases as the ratio increases. To obtain the same angle in a positive resist pattern, absorbance is required to be lower than 0.4. Resist polymers with aromatic rings yield a large absorbance of more than 1.7 at 193-nm for 0.4- μ m-thick films.⁹⁾ Therefore, a steep profiled pattern cannot be obtained with these resist.

On the other hand, the initial absorbance of PMMA which has no aromatic ring, is 0.03 for a 0.4- μ m-thick film, so PMMA is a resist candidate for ArF excimer laser lithography. To clarify the development characteristics of this resist for ArF excimer laser exposure in greater detail, PMMA film was exposed employing a different exposure system with no imaging optics.

Figure 7 shows the thickness of PMMA film after exposure (denoted by solid marks) and after development (denoted by open marks) as a function of exposure dose. A mixture of IPA and MIBK with a volume ratio of 3:1 was used as the developer. Initial thickness was 1.25- μ m. The thickness was decreased during exposure by photoetching. The pulse energy density influenced the thickness, as well as exposure dose. This is because high pulse energy density caused a temperature rise in the PMMA film thus enhancing decomposition.

Figure 8 shows the reduction in thickness through dissolution by development for PMMA having initial thicknesses of 1.25- μ m and 0.66- μ m (i.e., the result of subtracting the thickness of the PMMA film after development from that after exposure). The dissolved thickness does not depend on the initial thickness, and saturates at 1- μ m. This is because the absorption coefficient increases during exposure, and bottom region under 1- μ m is not sufficiently exposed. Therefore, PMMA having an initial thickness in excess of 1- μ m is fully developed in association with photoetching.



Fig. 7. Dependence of PMMA film thickness on exposure dose just after exposure (solid marks) and after development (open marks). Exposure was done in N₂ environment. Developer was a mixture of 3 parts IPA and 1 part MIBK. Initial thickness of PMMA was 1.25- μ m.



Fig. 8. Reduction in thickness of PMMA during development as a function of exposure dose. Initial thickness of PMMA films are 1.25- μ m (solid marks) and 0.66- μ m (open marks). Developer was a mixture of 3 parts IPA and 1 part MIBK.

It is clear from Fig. 7, however, that photoetch depth saturates and $1.25 \ \mu m$ PMMA cannot be fully developed by exposure to pulses of 10 mJ/cm²/pulse. It is reported that a carbon-rich surface layer is formed on PMMA film in exposures using a low pulse energy density ArF excimer laser.⁹⁾ This surface layer disturbs photoetch and development. A pulse energy density as low as few mJ/cm²/pulse is only available at the wafer in the ArF excimer laser projection system. The thickness that can be fully developed will be much thinner than 1- μ m.



Fig. 9. SEM micrograph of 0.25- μ m lines-and-spaces pattern in PMMA(0.2- μ m)/amorphous-Si(0.05- μ m)/OFPR-800(0.4- μ m). Exposure dose was 1.6 mJ/cm²/pulse × 2695 pulses. PMMA was developed in pure MIBK for 2 min. Amorphous-Si and OFPR-800 were reactive ion etched with CF₄ and O₂, respectively.

7

8

Therefore, it is concluded that a steep-profiled pattern cannot be obtained in thick single-layer polymer resist in ArF excimer laser lithography.

Next, we examined the patterning performance using a tri-layer resist system. The top, intermediate, and bottom layers were 0.2- μ m-thick PMMA, 0.05- μ m-thick amorphous silicon, and 0.4- μ m-thick OFPR-800, respectively. Figure 9 shows a SEM micrograph of the 0.25- μ m lines-and-spaces pattern obtained with the fabricated prototype partially achromatized lens. Although the resolution area was restricted to 500- μ m in diameter and a certain amount of fluctuation in the line width was observed, 0.23- μ m lines-and-spaces pattern was also obtained.

Based on result obtained, we concluded that replication methods involving exposure of just the surface layer is the most suitable for ArF excimer laser lithography.

§6. Conclusion

With the aim of advancing the development of ArF excimer laser projection lithography, four types of projection lenses were investigated. A monochromatic lens required spectral bandwidth narrowing to 5-pm. The current limit of bandwidth narrowing is proved to be 10-pm (FWHM). Two types of achromatic lenses required an extremely high optical surface accuracy of the element lenses or the development of a low absorption cement. As a result, a partially achromatized lens is found to be offer the best solution. The resolution of 0.18- μ m is achieved using the fabricated partially achromatized lens

with 10-pm bandwidth laser. A steep-profiled pattern cannot be obtained in thick single-layer polymer resist in ArF excimer laser lithography. A quarter-micron pattern was replicated using a partially achromatized lens employing a tri-layer resist system.

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References

- V. Pol, J. H. Bennewitz, G. C. Escher, M. Feldman, V. A. Firtion, T. E. Jewell, B. E. Wilcomb and J. T. Clemens: *Proc. SPIE, Santa Clara, 1986*, 633 (1986) p. 6.
- N. Nomura, H. Nakagawa, Y. Tani, K. Koga, N. Araki, T. Sato and M. Sasago: *MICROCIRCUIT ENGINEERING 89*, ed. H. Ahmed *et al.* (Elsevier Science Publishers B. V., Amsterdam, 1990), p. 183.
- 3) Y. Ozaki, K. Takamoto and A. Yoshikawa: Proc. SPIE, Santa Clara, 1988, 922 (1988) p. 444.
- 4) T. J. McKee: Can. J. Phys. 63 (1985) 214.
- 5) R. Chabbal: J. Phys. Radium. 19 (1958) 295.
- 6) A. Yoshikawa, S. Hirota, O. Ochi, A. Takeda and Y. Mizushima: Jpn. J. Appl. Phys. 20 (1981) L81.
- 7) K. J. Polasko, D. J. Ehrlich, J. Y. Tsao, R. F. W. Pease and E. E. Marinero: IEEE Electron Device Lett. **EDL-5** (1984) 24.
- 8) Y. Kawai, A. Tanaka, Y. Ozaki, K. Takamoto and A. Yoshikawa: *Proc. SPIE, San Jose, 1989*, **1086** (1989) p. 173.
- 9) Y. Ozaki and K. Hirata: Extended Abst. of the 17th Conference on Solid State Devices and Materials, Tokyo, 1985 (The Japan Society of Applied Physics, Tokyo, 1985) p. 365.