WE WILL

NEVER STOP

ENABLING ADVANCEMENT IN THE SEMICONDUCTOR INDUSTRY

FUJIFILM

TORU FUJIMORI （藤森　亨）

2023. 10. 26

FUJIFILM Corporation

Electronic Materials Research Laboratories

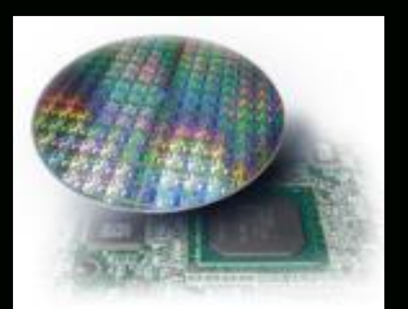
Electronic Materials Business Division

Biography

TORU FUJIMORI（藤森　亨）

現在、*Senior Expert*（资深专家）
（Deputy director of photo resist development group in FUJFILM Corporation.)

B.S. and M.S. degrees in organic chemistry from Saitama University
1991; Joined FUJIFILM Corporation (Synthetic Organic Chemistry Lab.)
Synthesized novel functional materials for photo films for 3 years,
and then for photo resist materials for 8 years.



2002; Moved Electronic Materials Research Lab.
Color resist for image sensor for 6 years,
and then for *photo resist (KrF, ArF, ArF immersion, EB and EUV)*
for 14 years and beyond !!
2006; Research manager
2014 - 2016; Senior researcher at EIDEC as an assignee from FUJIFILM
(EIDEC : EUVL Infrastructure Development Center (National project))
2021; Senior Expert
Remarks; Over 200 patents in this field.
Many papers, presentations, lectures and textbooks.
(CSTIC, IWAPS, SPIE, EUVL symposium, Photopolymer conference and so on.)

EUV lithography has come !!

In 2019（Reiwa 1 (令和 元年)） Anniversary for EUV enthusiasts !

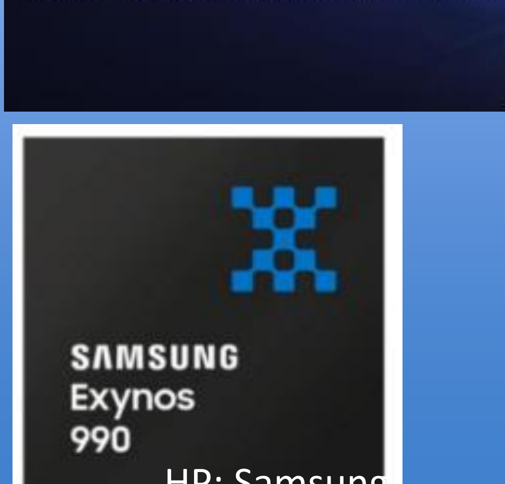
Finally, EUV generation has come in 2019 !!

7nm+ design rule was applied to HVM by using EUV lithography !!

Qualcomm

K-tai. watch. impress

HP; Huawei



HP; Samsung

Already installed to 5G smart phone in 2020

HP; Sharp

Resist materials development : " Never Stop "

Recent situation of our life

The smart phone is **the essential device** for our life.

Source:Apple

iPhone history

Higher performance year by year

Design rule ->

ASML continues to enable Logic process evolution

2007 iPhone	2009 iPhone 3G	2010 iPhone 4	2011 iPhone 4S	2012 iPhone 5	2013 iPhone 5S	2014 iPhone 6(+)	2015 iPhone 6S(+)	2016 iPhone 7 (+) (dual lens design)	2017 iPhone 8,8+,X	2018 iPhone XS,XR	2019 iphone 11/SE	2020 iphone 12/SE	2021 iphone 13/Pro/Max
APL0098 90nm	APL2298 65nm	A4 45nm	A5	A6 32nm	A7 28nm	A8 20nm	A9 14/16nm	A10 16nm+	A11 10nm	A12 7nm	A13 7nmP	A14 5nm	A15 5nm
XT:12X0 Shrink Add functions	XT:14X0 Shrink Add functions	XT:17X0i Shrink Add functions	Add functions	XT:19X0i Shrink Add functions	NXT:1950Ai Shrink Add functions	NXT:1960Bi Shrink Add functions	NXT:1970Ci Shrink Add functions	Add functions	NXT:1980Di Shrink Add functions	NXT:1980Di Shrink Add functions	NXT:2000i Shrink	NXE:3400C Shrink	NXE:3400C Shrink
72mm²	71.8mm²	53.3mm²	122.2mm²	97mm²	102mm²	87.4mm²	96-104mm²	125mm²	89mm²	83mm²	A13	A14	
16 bit 1 Core 1 GPU 412 MHz	32 bit 1 Core 1 GPU 412 MHz	32 bit 1 Core 1 GPU 0.8 GHz	32 bit 2 Core 2 GPU 0.8 GHz	32 bit 2 Core 3 GPU 1.2 GHz	64 bit 2 Core 4 GPU 1.3 GHz	64 bit 2 Core 4 GPU 1.4 GHz	64 bit 2 Core 6 GPU 1.85 GHz L14&N16FF	64 bit 4 Core 6 GPU 2.37 GHz N16FF+ InFO wafer-level pckg	4.3B transistors 2+4 Core CPU (4+1)GPU+NPU 2.4 GHz N10FF	6.9B transistors 2+4 Core CPU (6)GPU+8 NPU 2.5 GHz N7FF			

ASML Public Slide 21 20 March 2019

Source: ASML Market Research

ArF immersion lithography

EUV lithography

Source:anandtech

Source: ASML Market research

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ArF immersion lithography

EUV lithography

ASML Public Slide 21 20 March 2019

Source:Apple

Source:anandtech

Source: ASML Market research

*Index based on an aggregate demand of 100 in FY2001/3

Axis	Values
Y	0, 20, 40, 60, 80, 100
X	FY1994/3, FY2001/3, FY2011/3

Peak

Rapid Decline

2007

Source: FUJIFILM

Instead, the **photo film has declined…**

NEVER STOP

Recent situation of our life (FUJIFILM)

FUJIFILM business must be shifted.

Boldly diversified its business utilizing its high technological capabilities cultivated through photographic business

Recent situation of our life (FUJIFILM)

FUJIFILM business must be shifted.

FUJIFILM

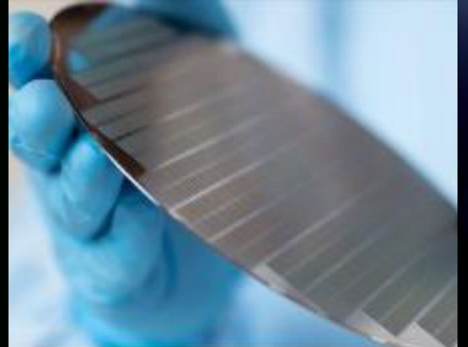
SUPPORTING THE WORLD'S DIGITAL TRANSFORMATION

Electronic Materials

Leveraging our expertise in nanotechnology and photosensitive material, Fujifilm diversified into the semiconductor industry more than 30 years ago.

Semiconductor Materials

We offer a broad portfolio of advanced high-purity chemicals & materials that enable global semiconductor manufacturers to develop the next generation devices that power our digital world.



NEVER STOP

Recent situation of our life (FUJIFILM)

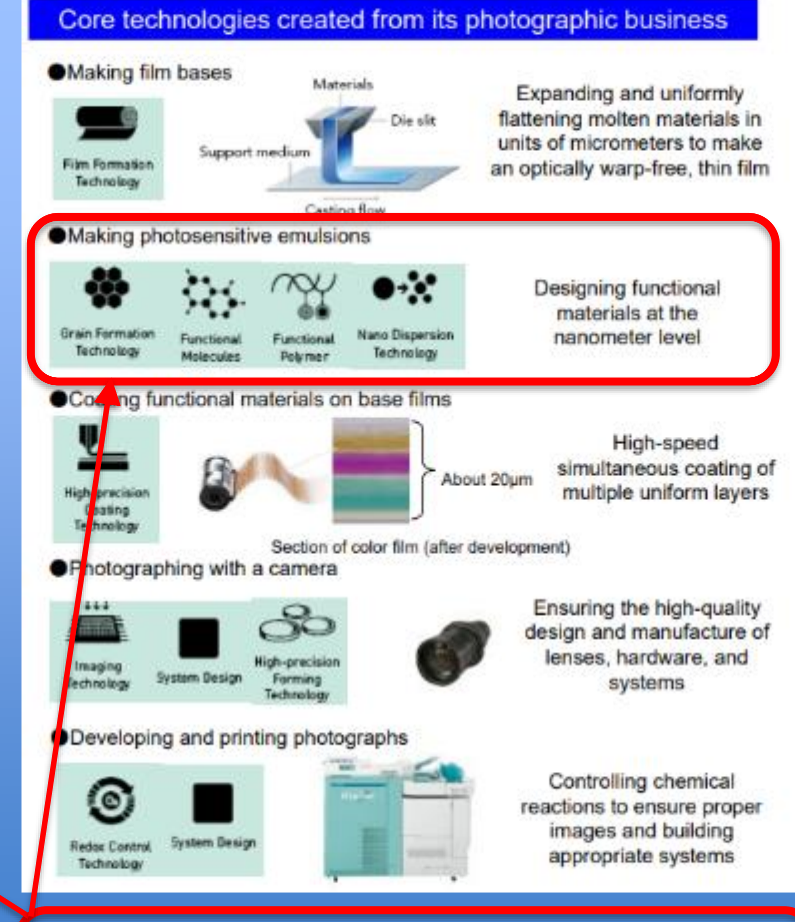
Business Fields of Fujifilm Group※

FUJIFILM

FY2021/3 Revenue ¥2,192.5 billion

- **Imaging** ¥285.2 billion (13%)
  - Professional Imaging 31%
  - Consumer imaging 69%
- **Healthcare** ¥579.4 billion (26%)
  - Medical Systems 60%
  - Bio CDMO 20%
  - Life Sciences 20%
- **Materials** ¥570.1 billion (26%)
  - Electronic Materials 21%
  - Display Materials 17%
  - Industrial Products, Fine Chemical, Recording Media 15%
  - Graphic Communication 47%
  - Advanced materials
- **Business Innovation** ¥757.8 billion (35%)
  - Business Solutions 33%
  - Office Solutions 67%

※This revenue composition ratio is provisional numbers for FY2020 after changing in segment from FY2021



We are here.
Electronic Materials Division

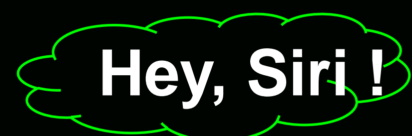
Source: FUJIFILM Group Business overview, Nov. 30, 2021

Recent situation of our life

=> Smart Social

Smart phone

HP: Apple.com



Smart home

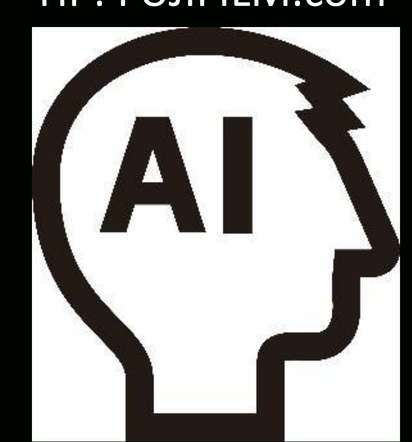
Smart car

TOYO Keizai

ChatGPT !!

"High speed"
"Big data"
"Energy saving"

HP: FUJIFILM.com



Recent situation of our life

Magic Eraser
by Google Pixel 7 Pro

Google HP

Remove the person, who recognized by AI.

NEVER STOP

Recent situation of our life

Semiconductor devices are indispensable to our life.

Computer

Smart phone

"High speed"
"Big data"
"Energy saving"

Game

Digital cameras

DRAM

Flash

Logic

System LSI

Image Sensor

Necessary to make design and manufacture of

' Small, high performance semiconductor devises '

The Pattern shrinkage history

Pattern shrinkage has been driven by shorter exposure wavelength.

g-line (436nm)

CD = 500nm

i-line (365nm)

350 nm

KrF (248nm)

150 nm

ArF (193nm)

90nm

ArFi (134nm)

40nm

EUV (13.5nm)

15nm

"The number of transistors and resistors on a chip doubles every 24 months" -Gordon Moore

Technology and Manufacturing day, intel

$$\text{Resolution Limit} = k1 \times \frac{\lambda}{NA}$$

Pattern Shrinkage : " Never Stop "

Resist materials development : " Never Stop "

NEVER STOP

The Pattern shrinkage for realizing 'Moore's low'.

However, there were a lot of happening in the history from KrF .

KrF (248nm)
150 nm

Successful ! Materials improvement !!

OH

ArF (193nm)
90nm

X Exposure tool
X Materials

F2 (157nm)
50nm

Surprise !! Game change ! tsmc !!

immersion technology Using water !!

ArFi (134nm)
40nm

Process improvement !!

ArFi
Multiple patterning
30nm

Surprise !! Game change ! FUJIFILM !!

Novel process !!

ArFi
NTD
(Negative-tone Development)
+ Multiple patterning
20nm

NGL (Next Generation Lithography)

EUV

Successful ! But taking long time

EUV (13.5nm)
15nm

~~EBDW (EB Direct Write)~~ X Wafer output
~~EB stepper~~ X Wafer output
~~DSA~~ X Defectivity
~~NIL~~ (Nanoimprint Lithography) X Defectivity

Nobody believed that

Immersion technology
Negative-tone Development
EUV lithography
How about next generation ??

Resist materials development : " Never Stop "

NEVER STOP

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Novel process !!

ArFi
NTD
(Negative-tone Development)
+ Multiple patterning
20nm

F2 (157nm)
50nm

X Exposure tool
X Materials

NGL (Next Generation Lithography)

EUV High NA /Hyper NA
Beyond EUV (6.7nm)
...

EUV

Successful ! But taking long time

EUV (13.5nm)
15nm

EBDW (EB Direct Write) — *X Wafer output*
EB stepper — *X Wafer output*
DSA — *X Defectivity*
NIL (Nanoimprint Lithography) — *X Defectivity*

Nobody believed that

Immersion technology
Negative-tone Development
EUV lithography
How about next generation ??

Re-born ?!

Resist materials development : " Never Stop "

NEVER STOP

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g-line (436nm)
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350 nm

KrF (248nm)
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40nm

EUV (13.5nm)
15nm

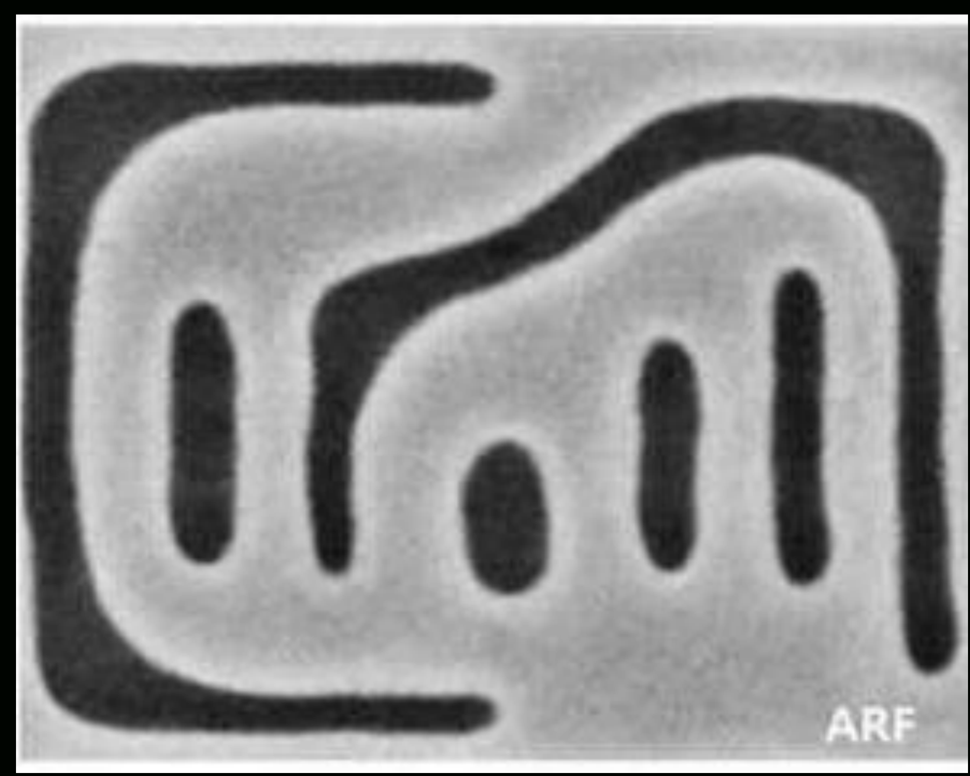
EUV lithography
Key technologies
to improve semiconductor manufacturing

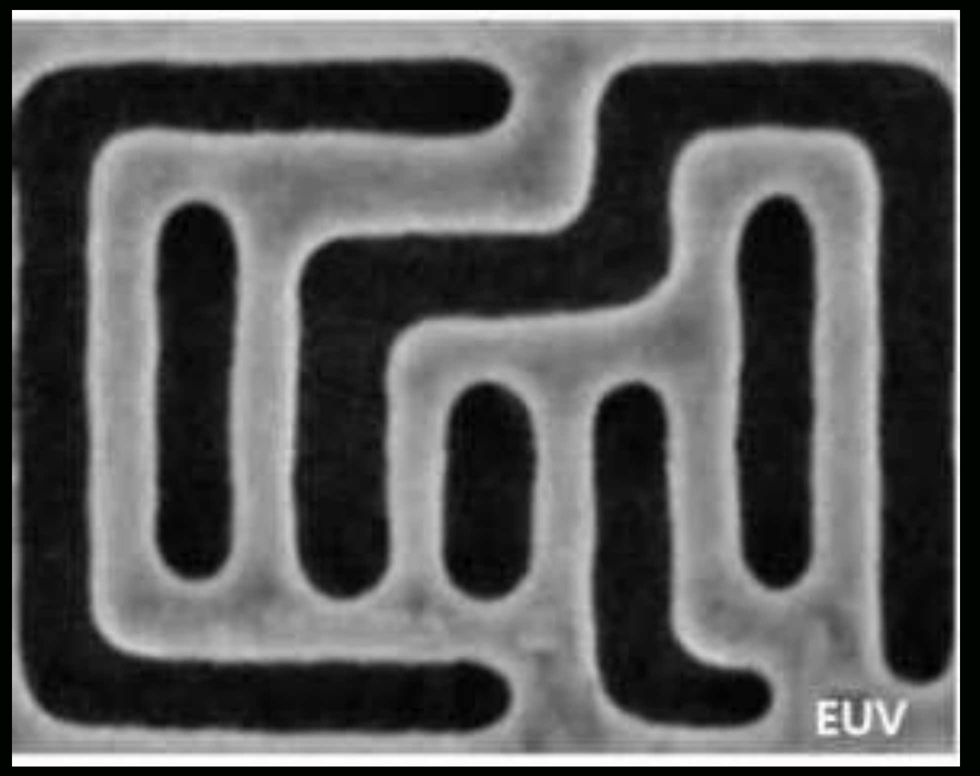
Pattern Shrinkage : " Never Stop "
Resist materials development : " Never Stop "

NEVER STOP

Why need EUV lithography ?

1st ; Pattern quality ; example





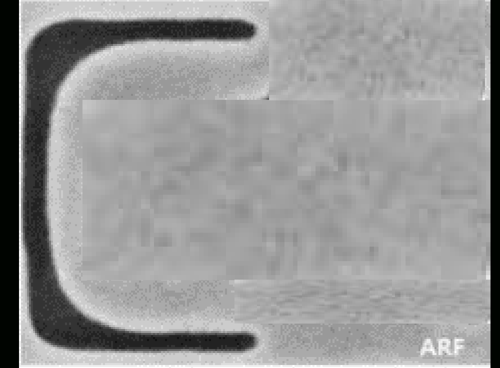
ArF i **EUV**

Source: Samsung

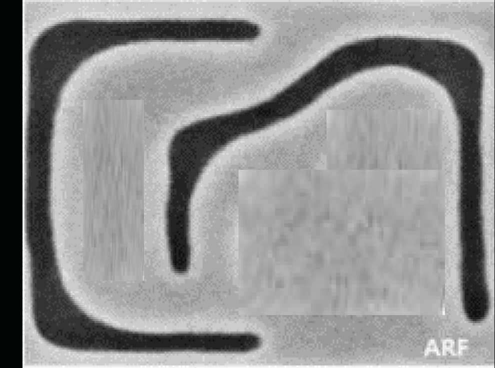
Resist materials development : “ Never Stop ”

Why need EUV lithography ?

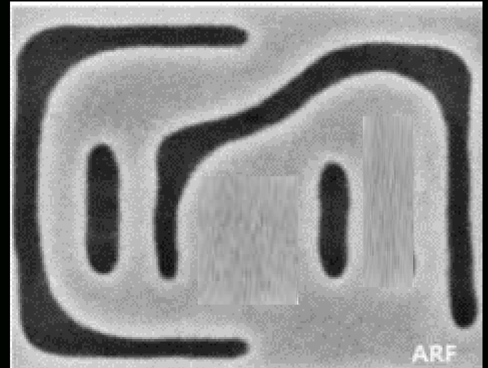
2nd ; Lithography steps ; example



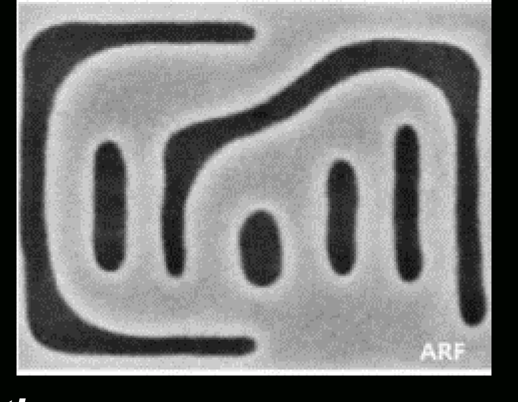
1st step



2nd step

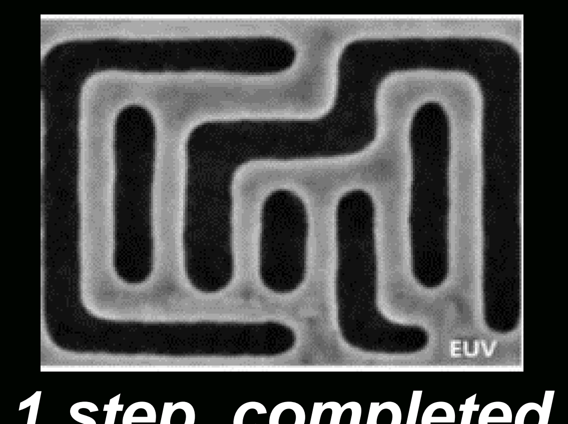


3rd step



4th step, completed

Many steps and masks needed…



1 step, completed

Source;
Pattern picture :Samsung
Lithography steps example :Prepared by author

Resist materials development : “ Never Stop ”

The history of EUV lithography

6

The first announcement on EUV lithography

tsmc
TSMC Property

Extended Abstracts (The 47th Autumn Meeting,1986) : → **1986**

The Japan Society of Applied Physics

35 years !!

28-ZF-15 Study on X-ray Reduction Projection Lithography

NTT ETL Hiroo Kinoshita, Ryuji Kaneko, et al.

Anthony Yen, EUV lithography: From the very Beginning to the eve of manufacturing (SPIE 2016)

It takes so long time to realize EUV lithography !

From now on, EUVL development will be accelerated !!

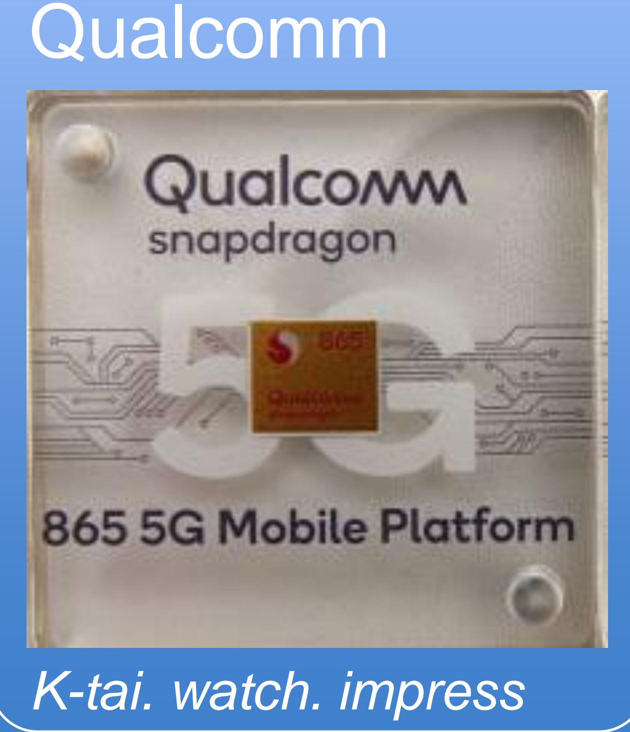
Resist materials development : " Never Stop "

EUV lithography has come !!

In 2019（Reiwa 1 (令和　元年)）Anniversary for EUV enthusiasts !

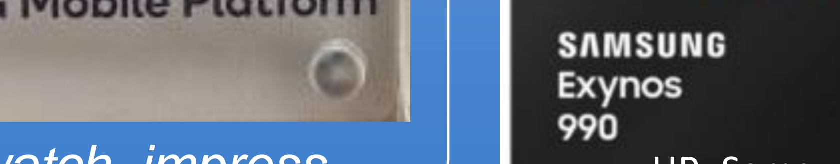
Finally, EUV generation has come in 2019 !!

7nm+ design rule was applied to HVM by using EUV lithography !!



K-tai. watch. impress

HP; Huawei



HP; Samsung

Already installed to 5G smart phone in 2020

HP; Sharp

Resist materials development : " Never Stop "

Recent advanced chips status

In 2020 **Already 5nm/5nm+ design rule …**

Qualcomm Snapdragon 888 5G Mobile Platform

Qualcomm HP

Samsung Exynos 2100

Samsung HP

5 nanometer process
11.8 billion Transistors
A14
16-core NEURAL ENGINE
11 trillion Operations per second

Apple.com
News.mynavi.jp

G2

Google HP

Google HP

Resist materials development : " Never Stop "

Recent advanced chips status

2018 **N7/N7+ design rule** 40nm ***20nm*** * Metal pitch

2020 **N5/N5+ design rule** 28nm ***14nm*** CD-size

（1:1 Line and Space as deffinition.）

2022 **N3 design rule** 22nm ***12nm***

2024 **N2 design rule** 21nm ***10nm***

2026 **A14 design rule** 18nm ***9nm***

2028 **A10 design rule** 16nm ***8nm***

2030 **A7 design rule** 16-14nm ***8-7nm***

2032 **A5 design rule** 16-12nm ***8-6nm***

2034 **A3 design rule** 16-12nm

2036 **A2 design rule** 16-12nm

Pattern Shrinkage : “ Never Stop ”

Resist materials development : “ Never Stop ”

NEVER STOP

3 key factors for EUVL realization

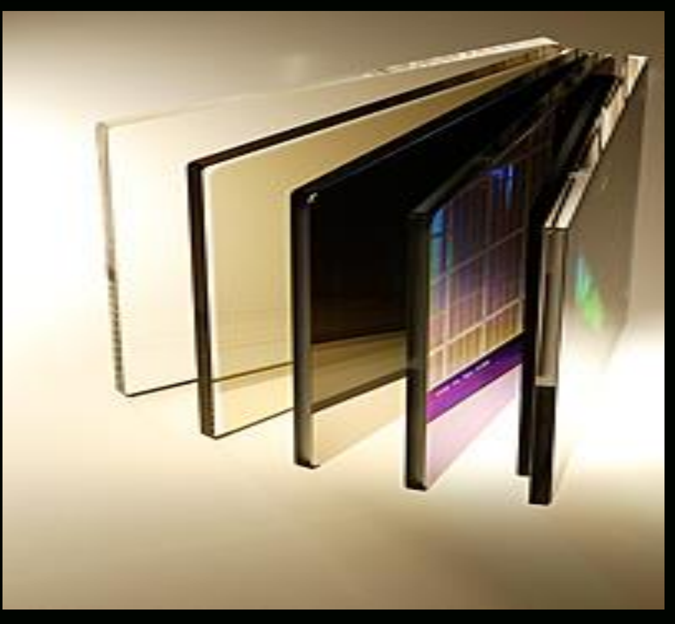
1) Light source

Light Source

2) MASK

Mask Defect

MASK Inspection



3) Resist

Resist

The 1st critical issue was 'Light Source' for a long time due to their lower source power until 2016 .

Resist materials development : " Never Stop "

3 key factors for EUVL realization

With recent rapid progress on source power improvement,
process and material explorations are more and more accelerated !

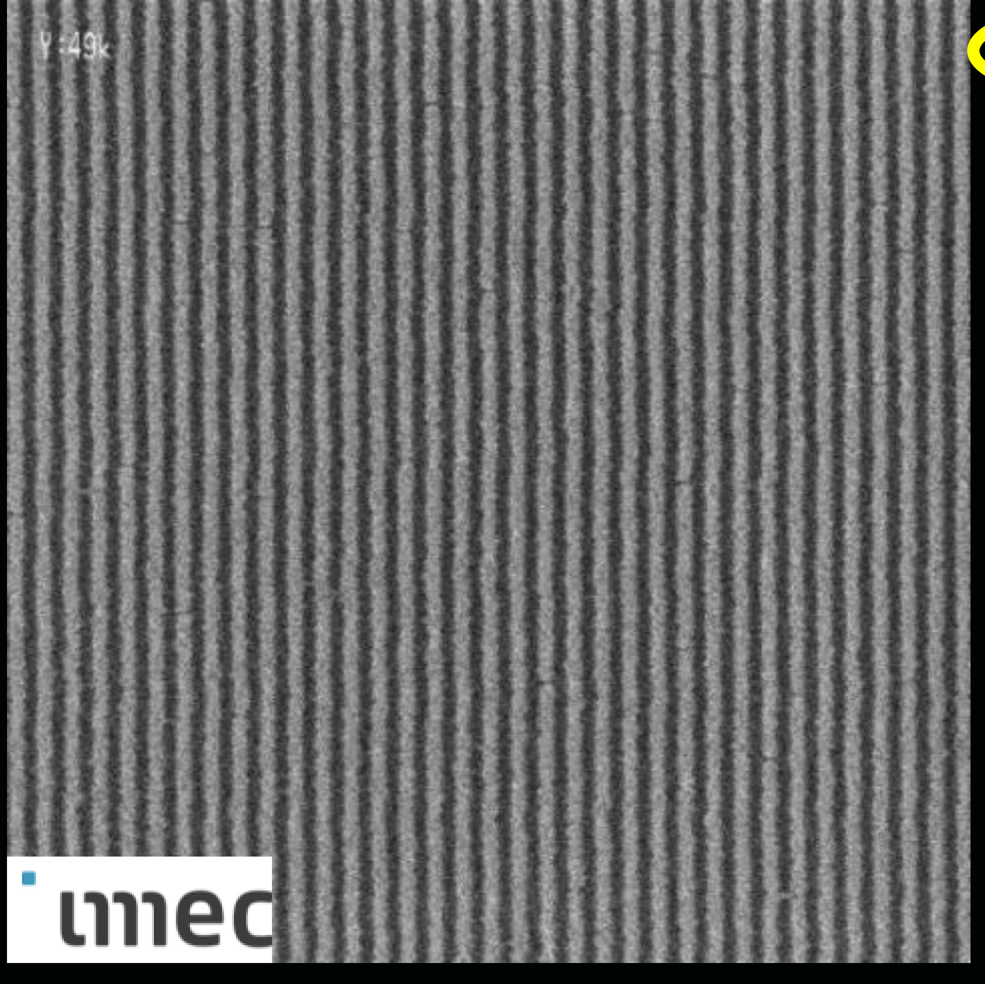
Focus Area: EUVL extension including High NA

2016	2017	2018	2019	2020
Light Source	Resist	Resist	Resist	Resist
Resist	Light Source	Mask Defect	Mask Defect	Mask Defect
Mask Defect	Mask Defect	Light Source	MASK Inspection	MASK Inspection
MASK Inspection	MASK Inspection	MASK Inspection	Light Source	Light Source

The Japan Society of Applied Physics imec EUREKA SPIE.

Resist materials development : " Never Stop "

Challenging of EUV resist



HP13nm

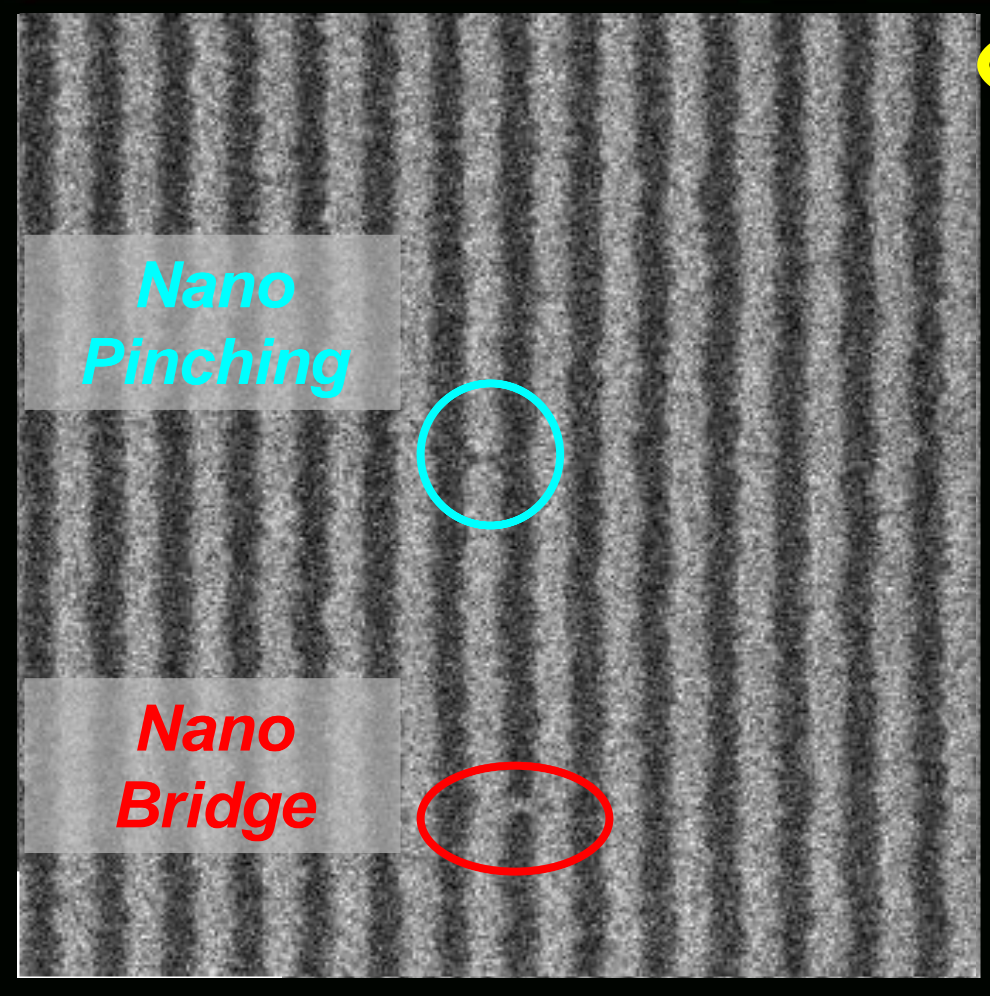
Eopt = 42 mJ/cm^2

LWR = 5.3 nm

The resolution looks good enough.

Resist materials development : " Never Stop " NEVER STOP

Challenging of EUV resist



HP13nm
Eopt = 42 mJ/cm^2
LWR = 5.3 nm

The resolution looks good enough.

However, 'stochastic error', "Nano-Bridge" and "Nano-Pinching" were observed.

=> Becomes an obstacle for HVM.

How to reduce 'stochastic error' ?

Resist materials development : " Never Stop "

Challenging of EUV resist

Have you ever heard "stochastic" ?

What is the stochastic issues ?

Why now ?? No issues before ?

Basically, it means...

隨機

Influence of pattern size shrinkage !

Let us imagine ;

64nm L/S	32nm L/S	16nm L/S	8 nm L/S
Previously	*Just past*	*Now !!*	*Future !!*

6nm x 6nm defect

No impact

6nm x 6nm defect

Huge impact

Resist materials development : " Never Stop "

Stochastic factors in lithography

Resist coating	*Exposure*	*Post exposure Bake*	*Development*
PAG Quencher Polymer	Photon	Diffusion De-protection	Dissolving
Materials location	Poor photon number Exposure locality	Location of reaction points	Dissolving locality Swelling behavior

Resist materials development : " Never Stop "

Stochastic factors in lithography

2 (two) kinds of stochastics in the lithography process.

1) Photon Stochastic

2) Chemical Stochastic

2)	1)	2)	2)
Materials location	Poor photon number Exposure locality	Location of reaction points	Dissolving locality Swelling behavior

Resist materials development : " Never Stop "

Stochastic factors in lithography

Resist coating	*Exposure*	*Post exposure Bake*	*Development*
PAG Quencher Polymer	Photon	Diffusion De-protection	Dissolving
Materials location	Poor photon number Exposure locality	Location of reaction points	Dissolving locality Swelling behavior

Resist materials development : " Never Stop "

Stochastic factors in lithography

Photon shot noise

Statistical simulation of resist at *EUV* and *ArF*

John J. Biafore[1], Mark D. Smith[1], Chris A. Mack[2]
James W. Thackeray[3], Roel Gronheid[4],
Stewart A. Robertson[1], Trey Graves[1], David Blankenship[1]

1. *KLA-Tencor*, FINLE, 8834 N. Capital of Texas Highway, Austin, TX, USA
2. Gentleman scientist, *lithoguru.com*, USA
3. *Rohm & Haas Electronic Materials*, 455 Forest St. Marlborough, MA, USA
4. *IMEC*, Kapeldreef 75, B-3001, Leuven, Belgium

SPIE. Event: SPIE Advanced Lithography, 2009, San Jose, California, United States

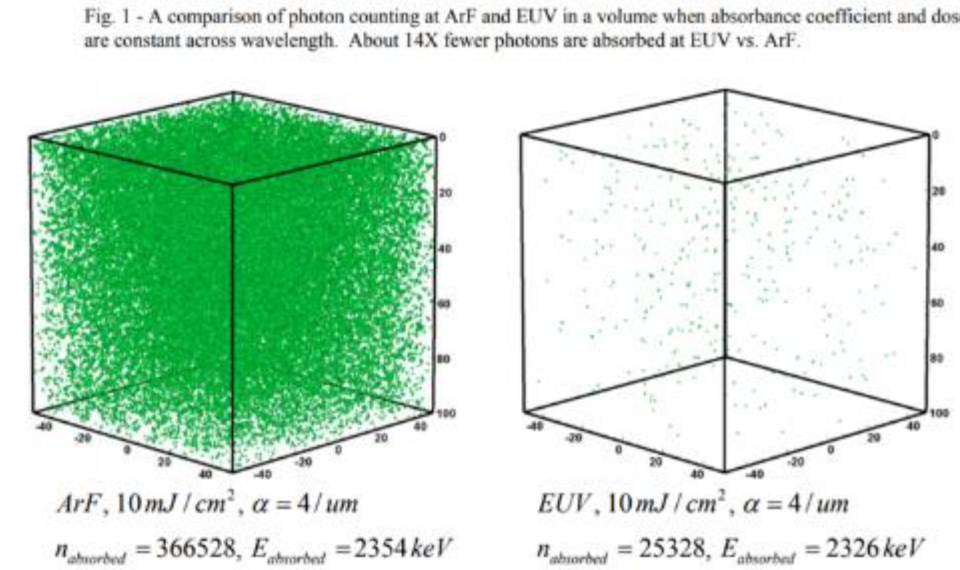


Fig. 1 - A comparison of photon counting at ArF and EUV in a volume when absorbance coefficient and dose are constant across wavelength. About 14X fewer photons are absorbed at EUV vs. ArF.

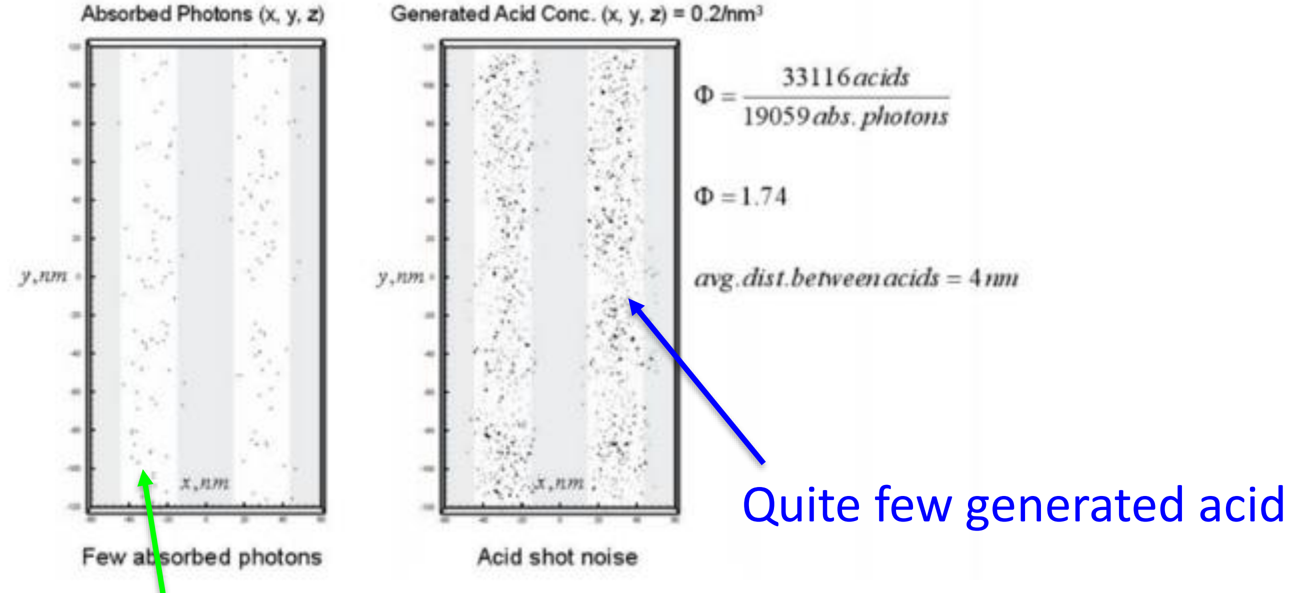


Fig. 7 - Simulation of photon absorption and the acid shot noise image, at EUV, 30 nm lines, 2-beam imaging, Esize. Simulated quantum efficiency is 1.74. Acid 'clumps' are visible about photon absorption sites.

About 14X fewer photons
are absorbed at EUV vs. ArF

Resist materials development : " Never Stop "

What is the photon shot noise ? Example) A photograph

Shot noise !

Evening scene

Dark (Insufficient light)

The total lunar eclipse
2022/11/8@Tokushima, taken by T. FUJIMORI

How to reduce the shot noise !

1) **Increase input light**
   Taking under daylight, or with stroboscope
2) **Using 'High sensitivity film'**

Resist materials development : " Never Stop "

NEVER STOP

What is the photon shot noise ?

How to reduce the shot noise !

1) Increase input light

   Taking under daylight, or with stroboscope

2) Using 'High sensitivity film'

Resist materials development : " Never Stop "

What is the photon shot noise ?

How to reduce the shot noise !

1) *Increase input light*

Taking under daylight, or with stroboscope

2) *Using 'High sensitivity film'*

In case of EUV lithography

Using 'High power scanner'
No…, under development.

Using 'High sensitivity resist'
⇒ Developed new materials !
'Catch more EUV light !'

Resist materials development : " Never Stop "

Reaction mechanism of CAR with EUV exposure

1st step

Catch the light

2nd step

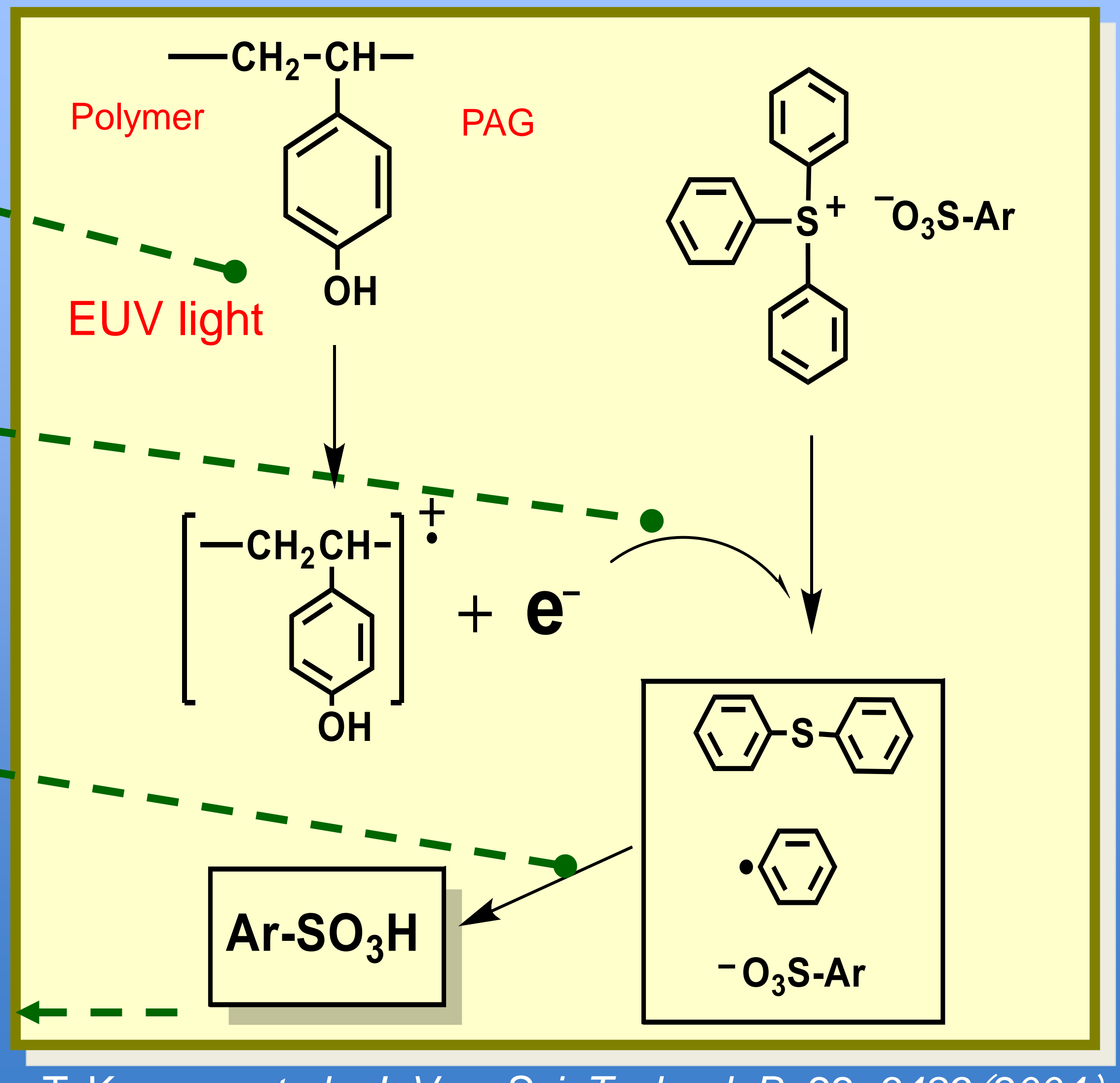
Generate the secondary electron

3rd step

Generate the acid

4th step

React with acid labile group



T. Kozawa *et al., J. Vac. Sci. Technol. B, 22, 3489* (2004)

1st /2nd step
Different
from previous lithography.

1st step
(Catch the light)
Very important to realize EUV lithography.

Resist materials development : " Never Stop "

Organic high EUV absorption materials

For catching more EUV photon, Newly designed and synthesized.

Standard

3x high abs.

Absorption coefficient (normalized by carbon)

H

B,C,N,O,F

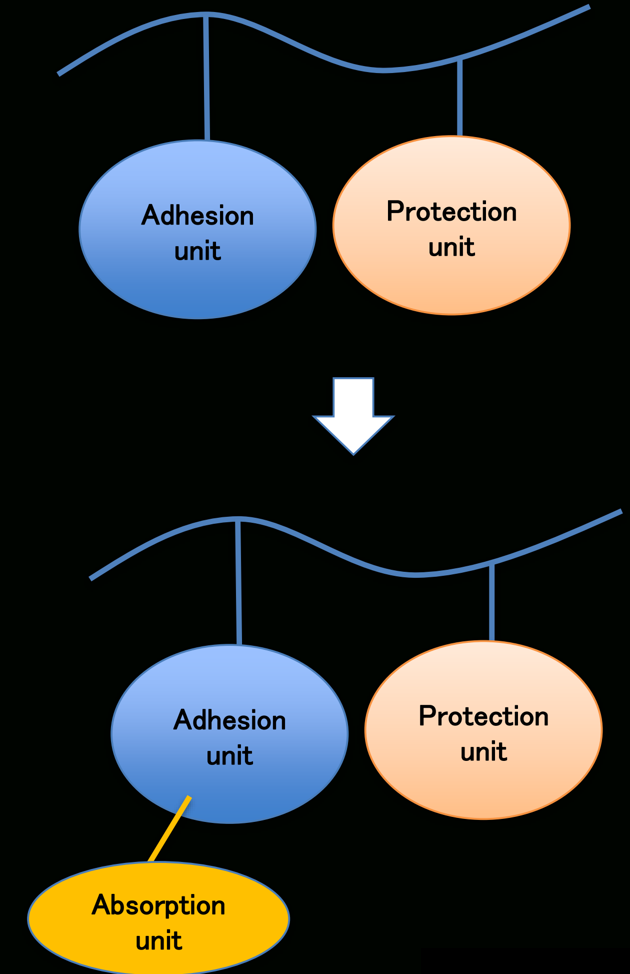
Si,P,S,Cl

Br

I

Atomic number

J.J. Yeh, et al. Atomic Data and Nuclear Data Tables, 32, 1−155 (1985)



Resist materials development : “ Never Stop ”

Litho performance of Organic high EUV absorption materials

imec

Standard type

NG 34.8 mJ 19.03 nm	NG 36.0 mJ 17.71 nm	NG 37.2 mJ 17.39 nm	OK 38.4 mJ 16.63 nm

High EUV absorption type

NG 27.6 mJ 18.52 nm	OK 28.7 mJ 17.73 nm	OK 29.8 mJ 17.09 nm	OK 30.9 mJ 16.45 nm

High EUV absorption type indicated excellent *bridging performance*.

Resist materials development : " Never Stop "

Stochastic factors in lithography

Resist coating	*Exposure*	*Post exposure Bake*	*Development*
PAG Quencher Polymer	Photon	Diffusion De-protection	Dissolving
Materials location	Poor photon number Exposure locality	Location of reaction points	Dissolving locality Swelling behavior

Resist materials development : " Never Stop "

What is Chemical stochastic ?

Resist coating

PAG
Quencher
Polymer
Blocking group

Materials location

Cause from resist materials and processes

Resist materials development : " Never Stop "

What is Chemical stochastic ?

PAG
Quencher
Polymer
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Materials location

Cause from resist materials and processes

Resist materials development : " Never Stop "

What is Chemical stochastic ?

Resist coating

PAG
Quencher
Polymer
Blocking group

Materials location

Cause from resist materials and processes

It's a kind of 'nature phenomena'.
' Chemical Stochastic !! '
Nobody can control

You can see the materials location randomess.

Resist materials development : " Never Stop "

What is Chemical stochastic ?

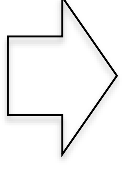
The image of 'Chemical stochastic'

Example) ' Dragon fruits'

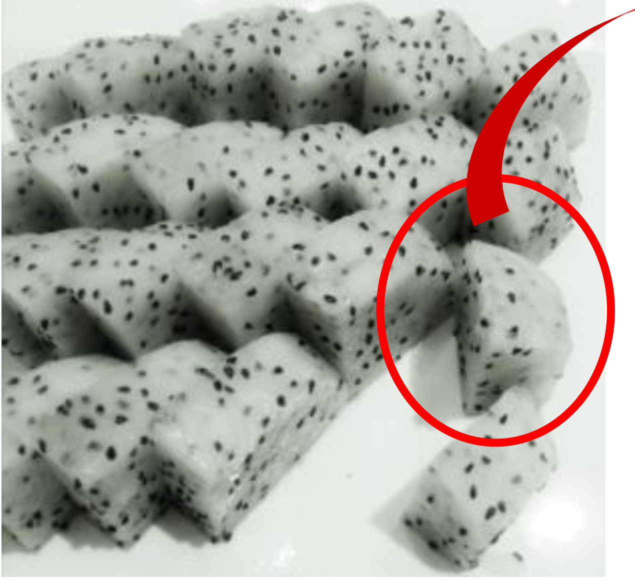
火龍果

Seeds : Chemicals

Fruits : Polymer matrix



Global CDU



Local CDU

Each piece has *own locality* of the seeds.

***The seeds has locality* in one piece.**

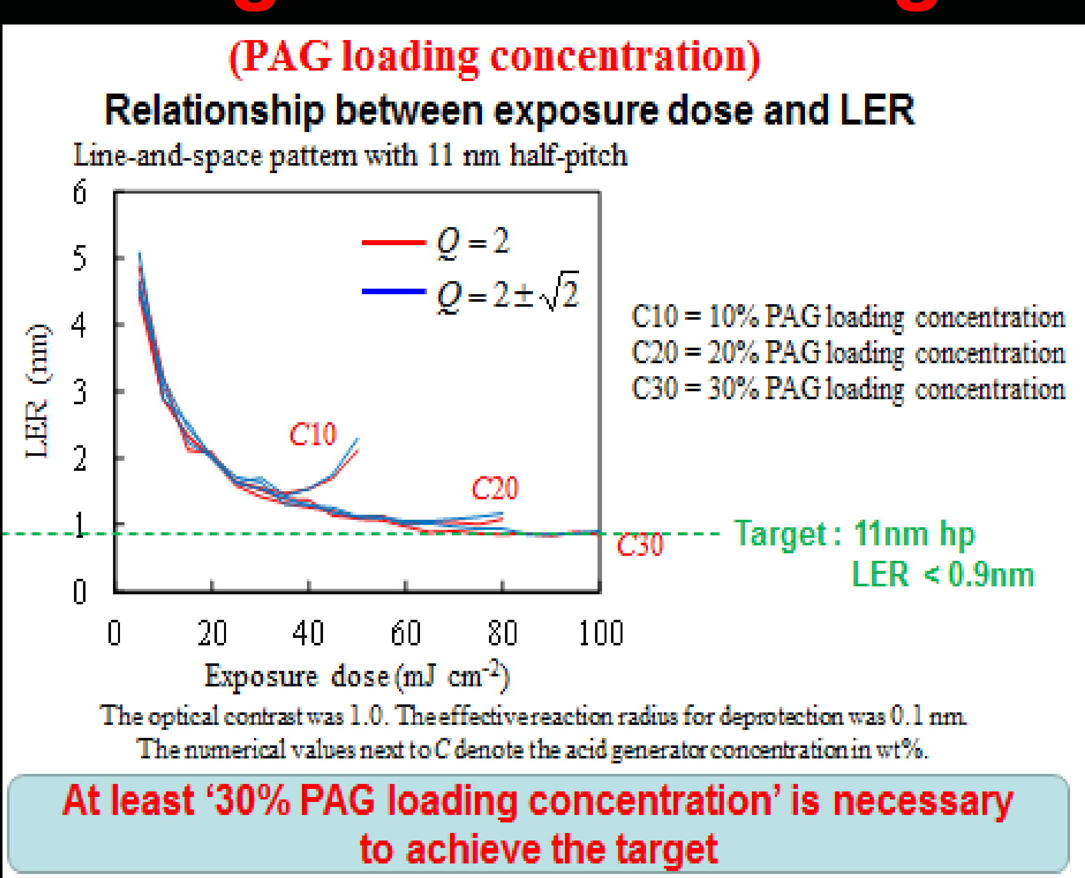
Nobody can control the position of the seeds.

Resist materials development : " Never Stop "

Improvement of chemical stochastic

One of the famous method to reduce 'Chemical stochastic'.

=> *Higher PAG loading* effect.



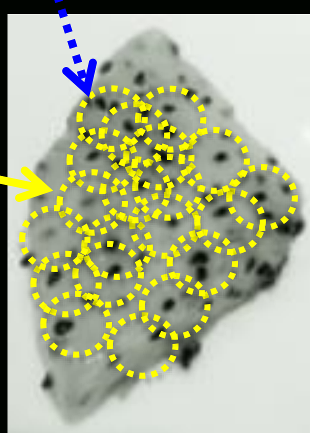
T. Kozawa et al., simulation study with EIDEC.

< Usual PAG loading >

Lots of locality part

< Higher PAG loading >

Better uniformity



Higher chance to generate acids.
Higher chance to react.
Improve
'Chemical stochastic' !

Excellent LER !!

Resist materials development : " Never Stop "

Improvement of chemical stochastic

Higher loading mitigated stochastic effect.

Higher loading

LWR
@same abs.& dose

4.5
4
3.5
3
2.5
2
1.5

Number of deprotection unit

6.99
6.28
6.85
7.07
6.44
7.23
7.36

0.03
0.04
0.05
0.06

Number of acid

Higher loading

imec

PAG higher loading =>

Deprotection unit
higher loading =>

Higher chance to generate acids.

Higher chance to react.

Improve
'Chemical stochastic' !

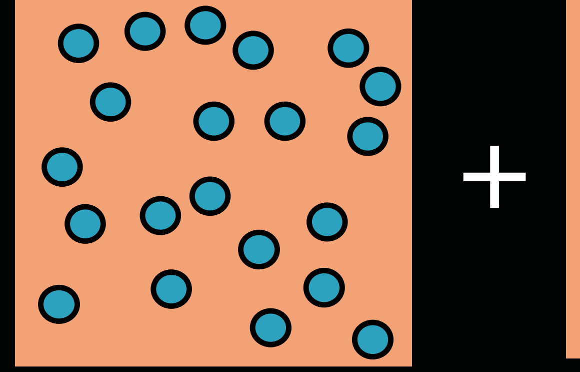
Indicated excellent LWR !!

Further improvement of chemical stochastic

Even, introduced *higher loading technology*, still remaining the fluctuation.

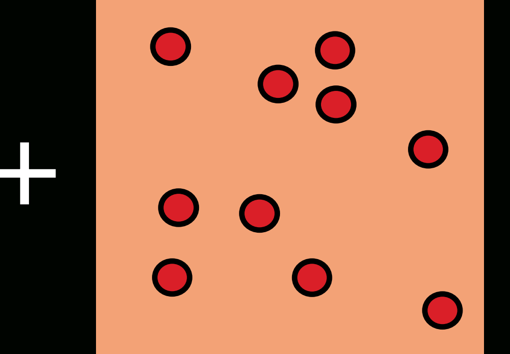
The 'materials fluctuation' can be controlled.

Current



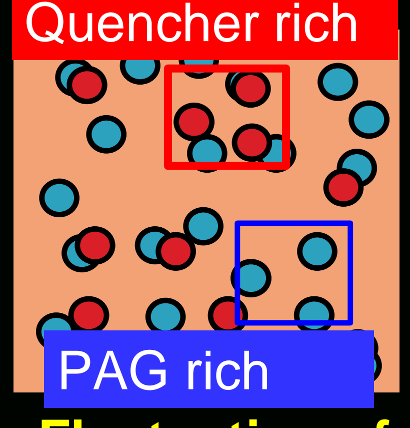
Fluctuation of PAG

+

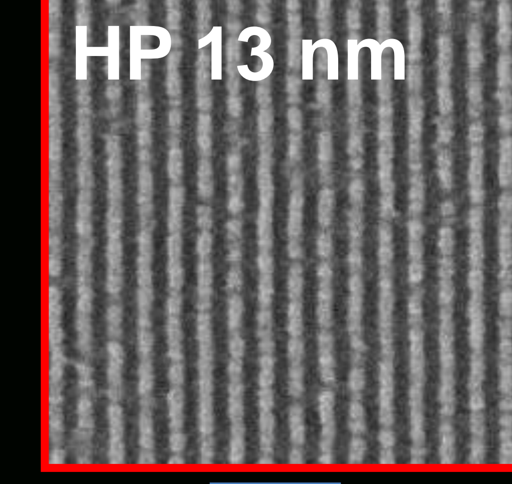


Fluctuation of Quencher

=



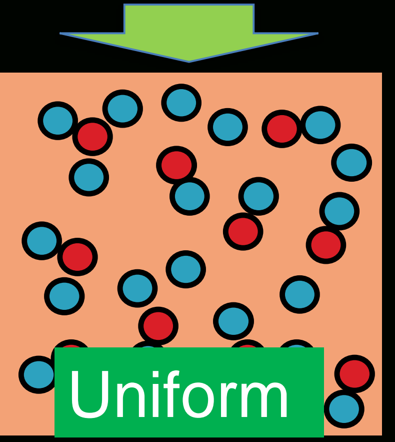
Fluctuation of Reaction

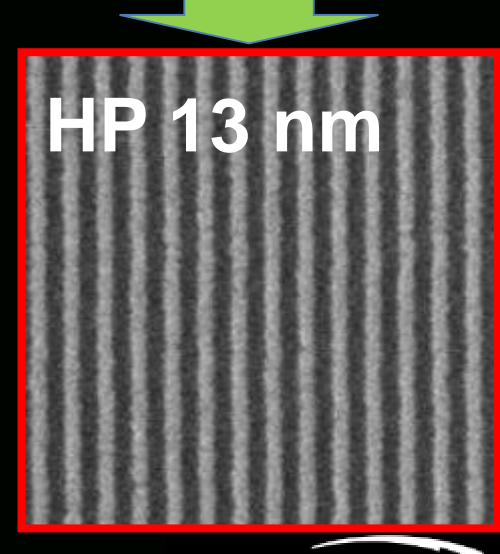


New technology

Uniform disperse in the film by using

Novel functionalized materials





Resist materials development : " Never Stop "

Stochastic factors in lithography

Resist coating	Exposure	Post exposure Bake	Development
PAG Quencher Polymer	Photon	Diffusion De-protection	Dissolving
Materials location	Poor photon number Exposure locality	Location of reaction points	Dissolving locality Swelling behavior

Resist materials development : " Never Stop "

How to reduce the stochastic factor ? NTI process

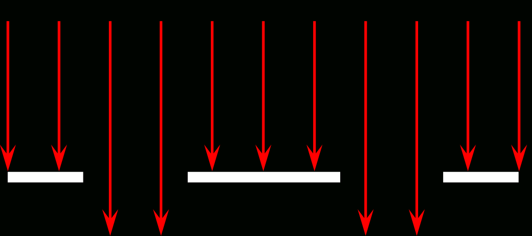
Key point : Developer (Alkali aqueous based or Organic solvent based)

Resist coating

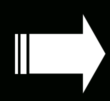
Exposure & Bake

Development

Conventional
TMAH aq.



PTI (Positive Tone Imaging)



Negative Tone Imaging process
is expected to reduce 'Chemical Stochastic'.
Organic compounds with organic solvent.
Dissolving smoothly ? Less Swelling ?

Organic
Solvent

NTI (Negative Tone Imaging)

Resist materials development : " Never Stop "

How to reduce the stochastic factor ? NTI process

In situ dissolving behavior evaluation by using High Speed AFM.

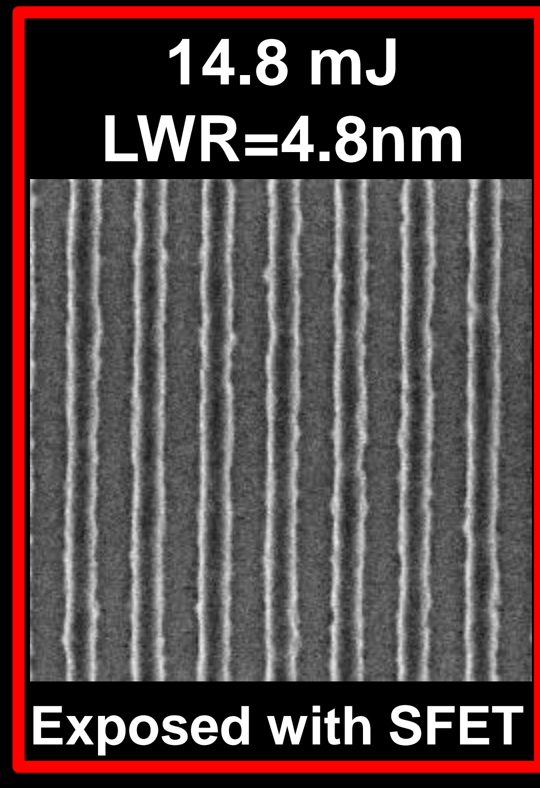
Development time

Positive-tone

Negative-tone

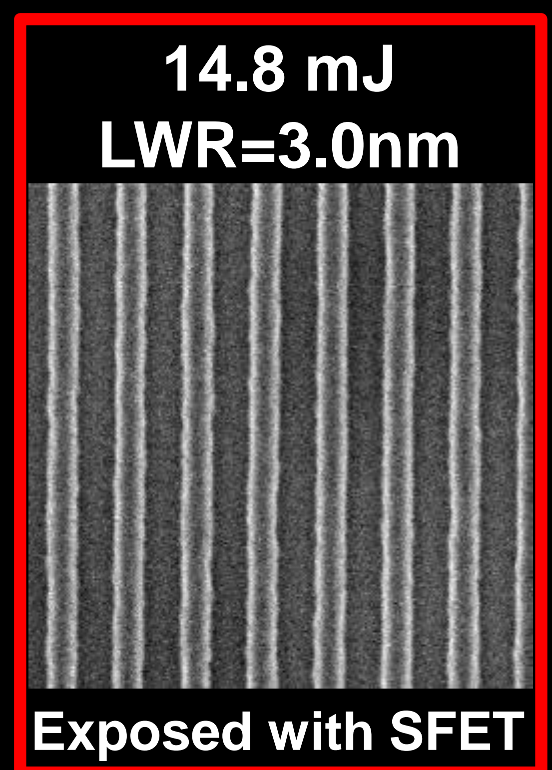
Negative-tone : extremely better LWR

14.8 mJ
LWR=4.8nm



Exposed with SFET

14.8 mJ
LWR=3.0nm



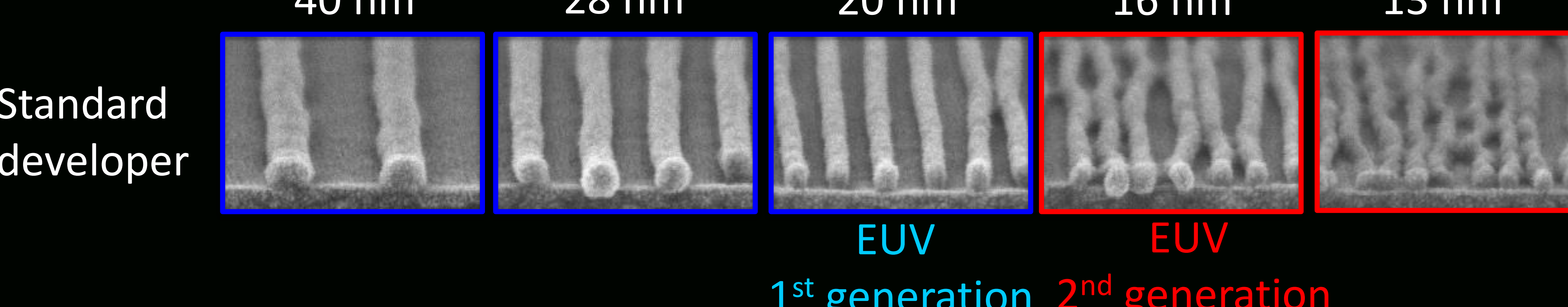
Exposed with SFET

Resist materials development : " Never Stop "

How to reduce the stochastic factor ? NTI process

Further improvement : New approach !

Feasibility study with EB lithography in 2015, Tsubaki et. al., SPIE



However, by using standard organic solvent developer process seems difficult to achieve the 2nd generation EUV lithography and beyond.

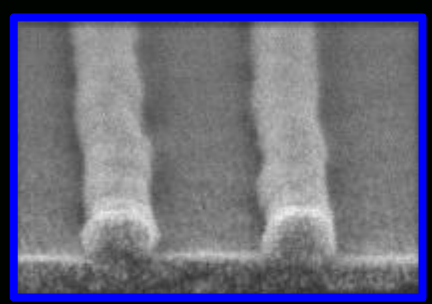
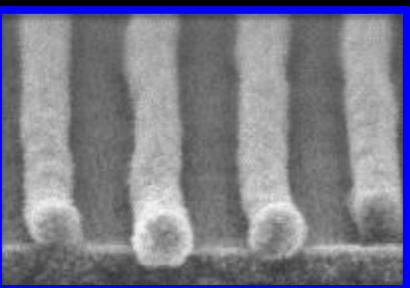
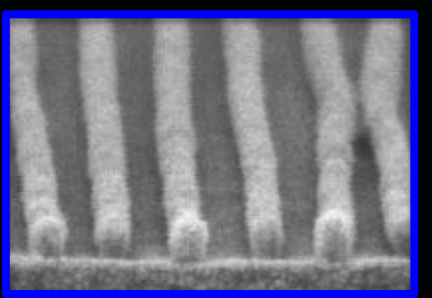
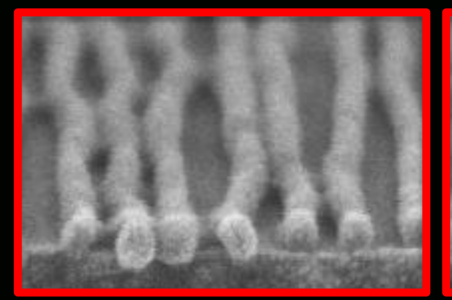
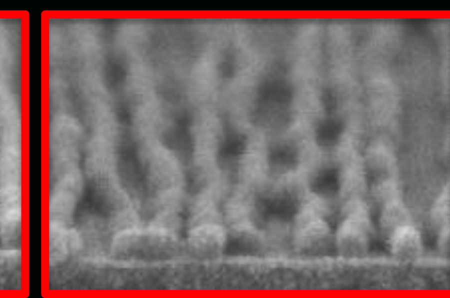
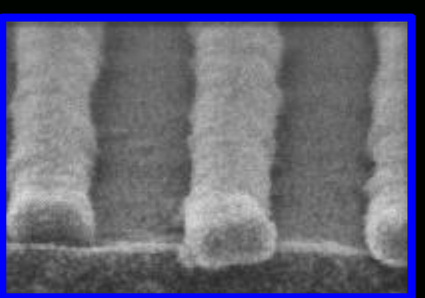
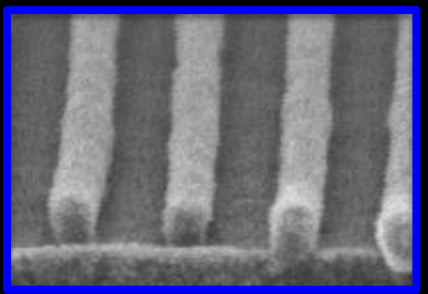
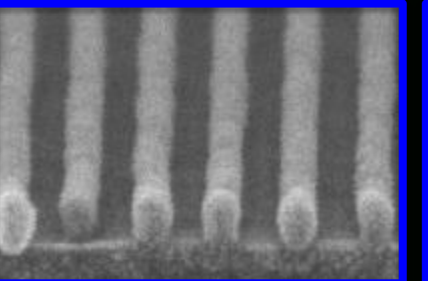
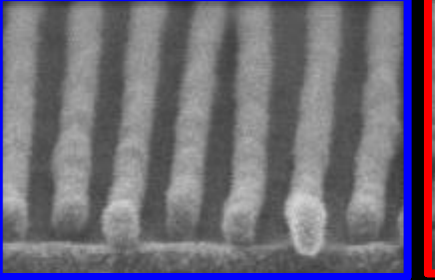
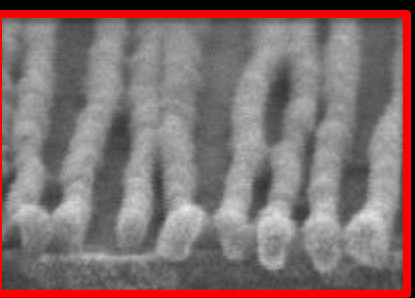
The standard developer may not be suitable for patterning small feature size.
It seems to be too soluble.

Resist materials development : " Never Stop " NEVER STOP

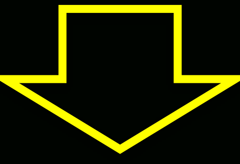
How to reduce the stochastic factor ? NTI process

Further improvement : New approach ! ***= > New developer***

Feasibility study with EB lithography in 2015, Tsubaki et. al., SPIE

	40 nm	28 nm	20 nm	16 nm	13 nm
Standard developer					
New developer					

Applied new developer seems very effective for improving resolution.



Resist materials development : " Never Stop "

How to reduce the stochastic factor ? NTI process

Further improvement : New approach ! => New developer

Feasibility study with EUV exposure by using SFET in 2015, Tsubaki et. al., EUVL symposium

HP20 HP19 HP18 HP17 HP16 HP15

Std. dev. 52 mJ/cm²

20.2 nm

New dev. 39 mJ/cm²

19.8 nm 18.9 nm 18.2 nm 17.2 nm

New NTD process improve the contrast /resolution

resist thickness (nm)

dose (mJ/cm2)

Std. dev.

New dev.

Resist materials development : “ Never Stop ”

How to reduce the stochastic factor ? NTI process

Further improvement : New approach ! **=> New developer**

Feasibility study with EUV exposure by using SFET in 2015, Tsubaki et. al., EUVL symposium

Now under developing ' New developer ' !!

New NTD process improve the contrast /resolution

20.2 nm

New dev. 39 mJ/cm²

19.8 nm

18.9 nm

18.2 nm

17.2 nm

resist thickness (nm)

dose (mJ/cm2)

Std. dev.

New dev.

Resist materials development : " Never Stop "

Summary

Excellent improving the litho performance due to reduce the stochastic issue.

PTI (Positive-tone)	L/S 16nm hp	C/H 32nm CH	Trench 16nm slit
NTI (Negative-tone)	L/S 16nm hp	Pillar 16nm Dot	Line 16nm line

Resist materials development : " Never Stop "

Summary

Key technologies of reducing the 'stochastic issues'.

'***Organic EUV high absorption materials***'

'***Novel functionalized materials***'

'***Negative-tone imaging***' with EUV exposure (EUV-NTI)

The most critical issue is how to apply each item.

Expected to apply the real EUV lithography HVM.

If you have any questions, comments, or would like to communicate with me, please let me know.
You can send me an e-mail.

toru.fujimori@fujifilm.com

Resist materials development : " Never Stop "

Thank you for your kind attention !!