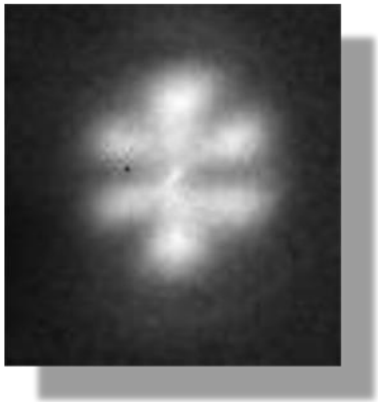
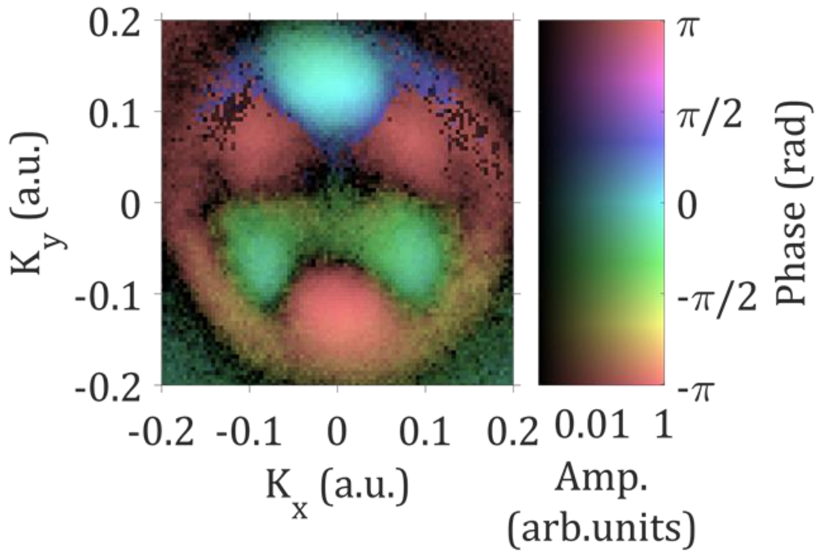
Visualization of a *complex* wavefunction by an attosecond laser pulse





The phase and amplitude distribution of photoelectrons are mapped in momentum space.

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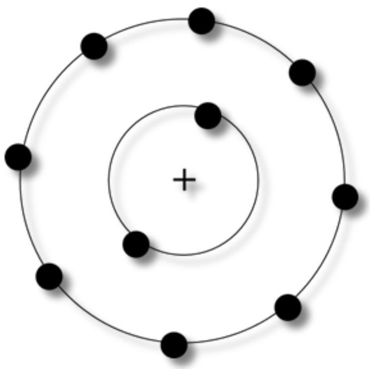
How to represent an "electron"

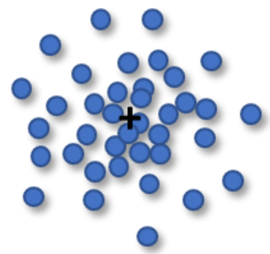
An electron is located in atoms, molecules and materials.

Bohr model (1913)



An electron "rotates" around the core.

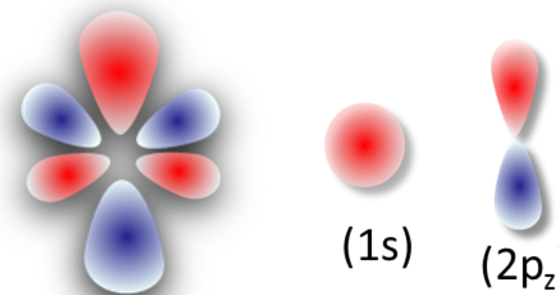
Electron "cloud"



An electron is statistically "distributed"

Schrödinger (1925)

Wavefunction Ψ model



***f*-orbital (m=0)**

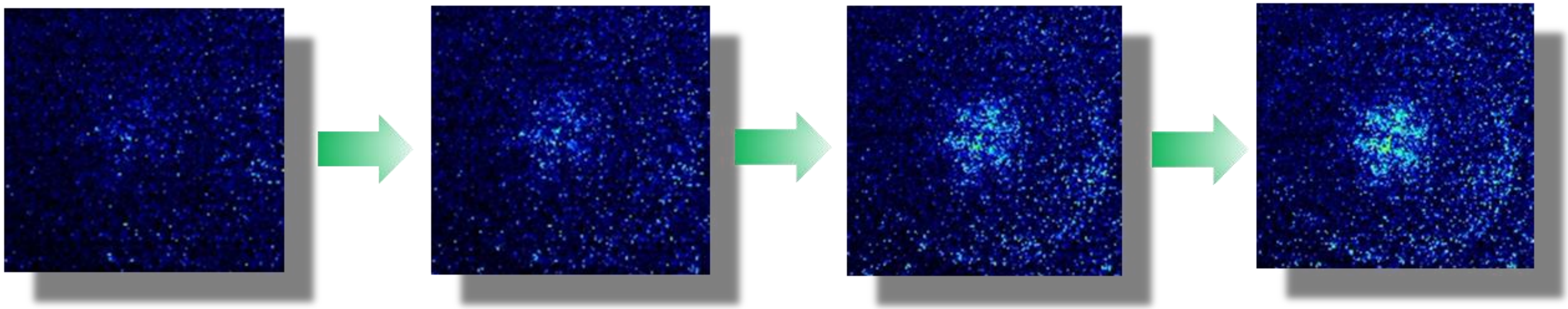
(1s)

($2p_z$)

statistical distribution
+ "**phase**"

- **An electron is statistically distributed in a space.**
- **In addition to that, an electron is characterized by "phase".**
- **The electron is represented by an "wavefunction".**

Watching an electron wavefunction $|\Psi|^2$

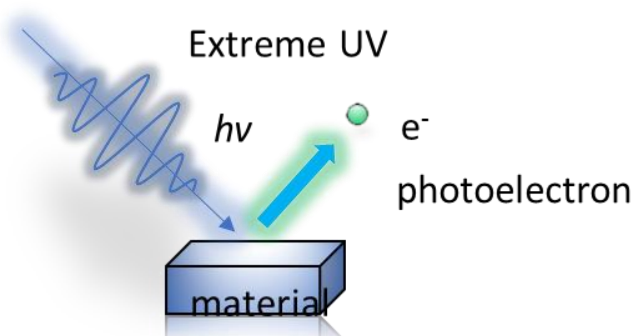


An electron is ejected by photoionization, and detected as a particle somewhere on the screen. Repeating the measurement many times, "the shape of the (square of) wavefunction, $|\Psi|^2$" can be seen.

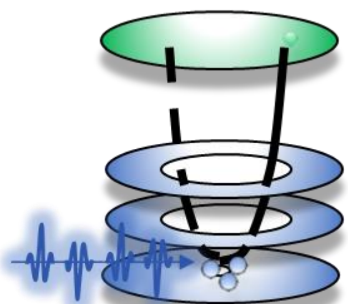
Max Born's statistical interpretation (1926) :

The square of wavefunction $|\Psi|^2$ represents the probability to find an electron.

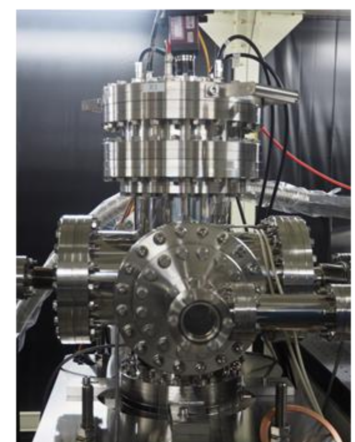
movie http://www.f.waseda.jp/niikura/en/d8.avi



Photoelectron effect



Record the energy and angular distribution of photoelectrons.



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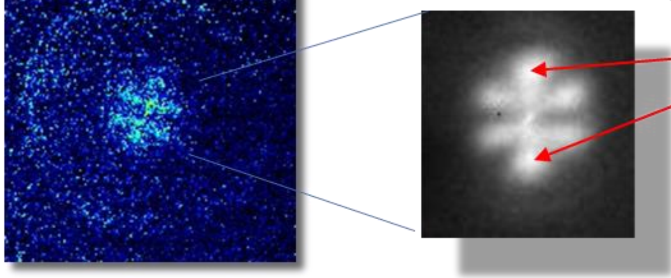
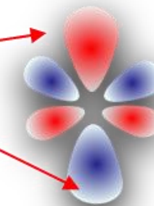
But ！

The “phase” information is disappeared when the electron hits on a “classical” detector”.

measured image

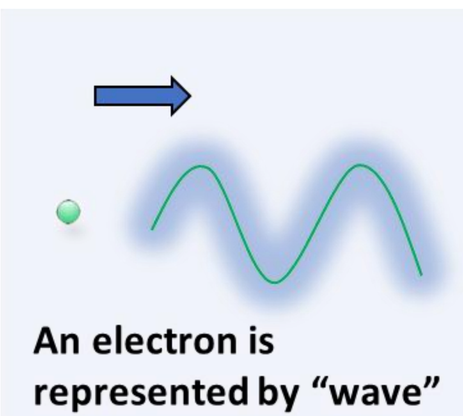


The phase (or sign) must be different, while it cannot be distinguished
by a conventional photoelectron spectroscopy.

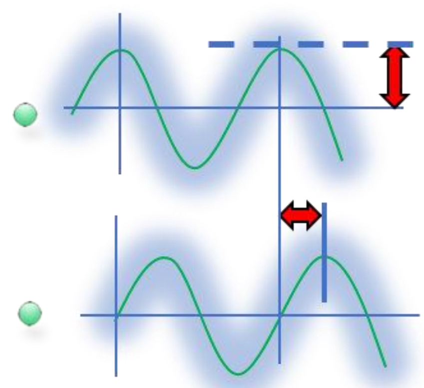


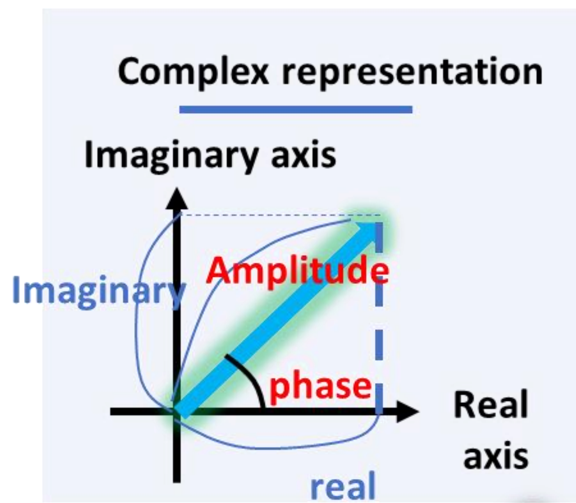
The square of wavefunction $|\Psi|^2$ can be measured,
but a complex wavefunction itself cannot be measured directly.

Wave nature of an electron



Amplitude and phase of wave





Both phase and amplitude, or real and imaginary values are required.

The “phase” measurement must be required.

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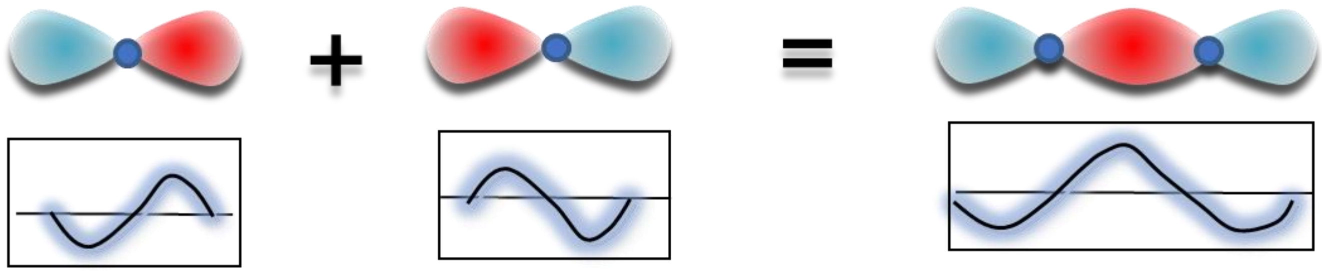
Why "phase" of wavefunction is important ?

(a) same phase

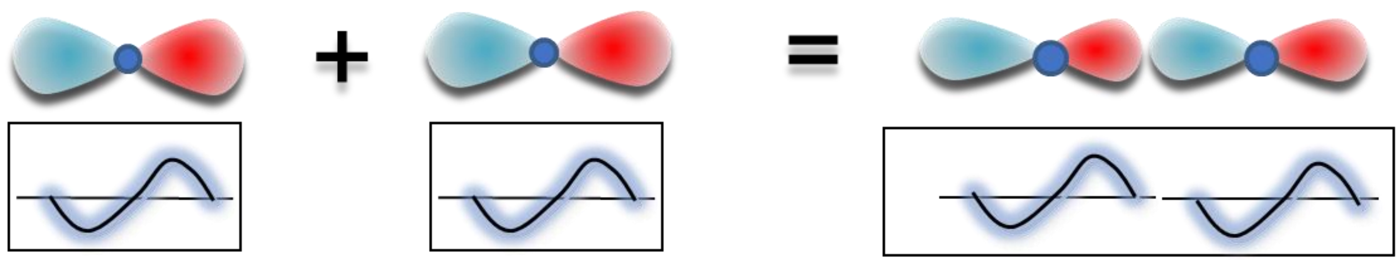


bonded

Two waves are overlapped **constructively.**

(b) opposite phase

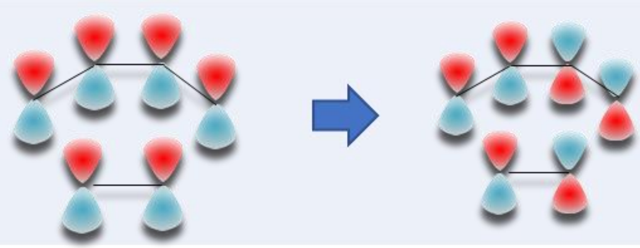
anti-bonded



Two waves are overlapped **destructively.**

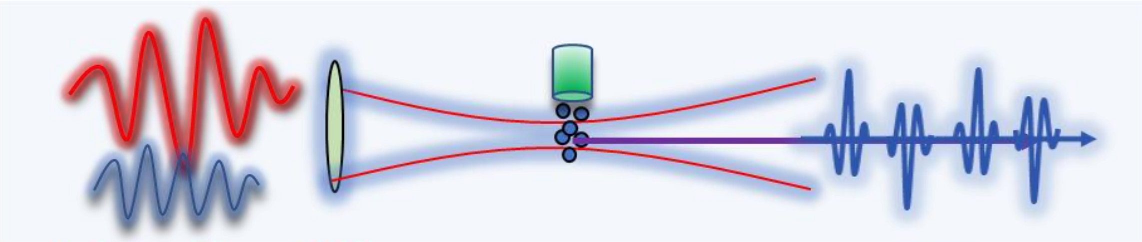
In order to understand chemical reaction, information on "a complex" wavefunction is needed.

Woodward-Hoffman rule
Frontier-orbital theory
（Nobel prize in chemistry 1981）



Attosecond laser pulse and measurement

(a) Generation of an attosecond laser pulse (high-harmonic generation, XUV)

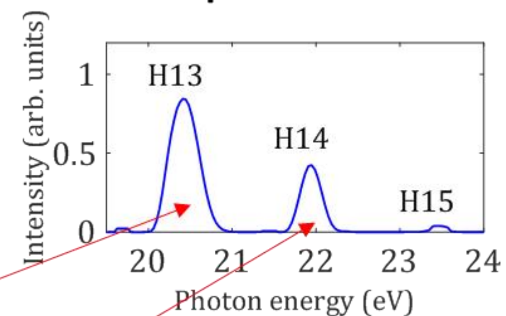


Infrared laser (IR)
(~790nm=1.57eV)
Second harmonic
(~395nm=3.14 eV)

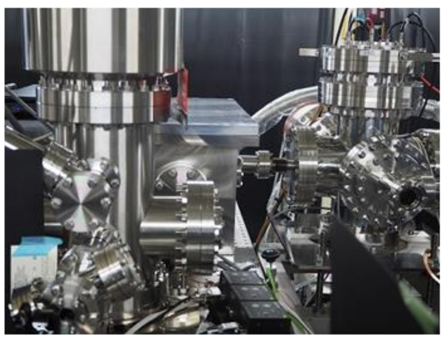
focus on
a gas-jet

XUV

Spectrum:



Harmonic 13 (H13): the wavelength is 1/13 of 790 nm (~61 nm)
Harmonic 14 (H14): the wavelength is 1/14 of 790 nm (~56 nm)



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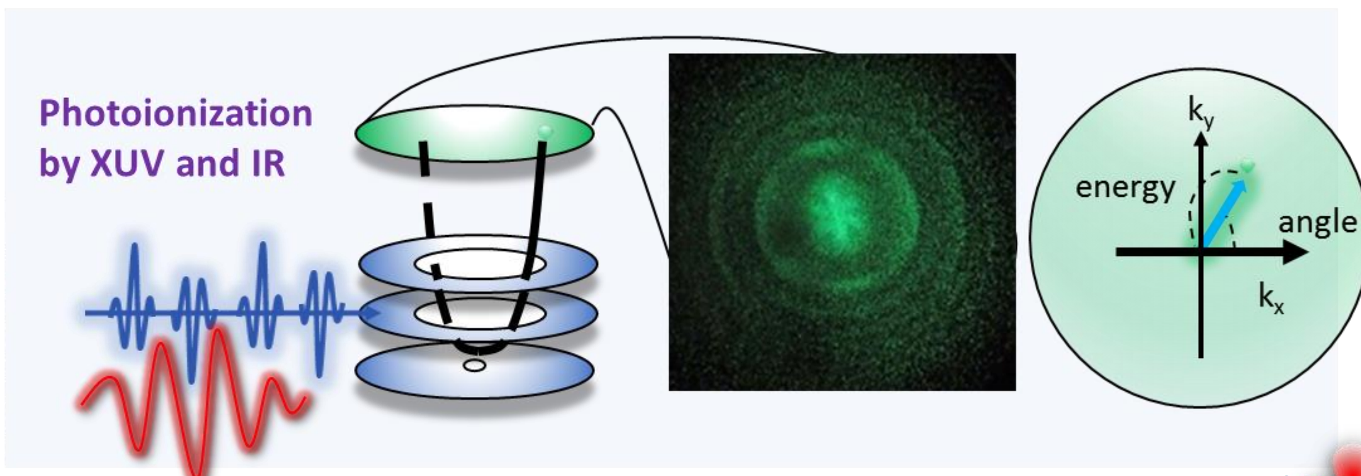
(b) Photoelectron momentum distribution recorded by velocity map imaging



An electron is ejected to a certain direction with a certain energy.
It is recorded on a two-dimensional momentum plane.

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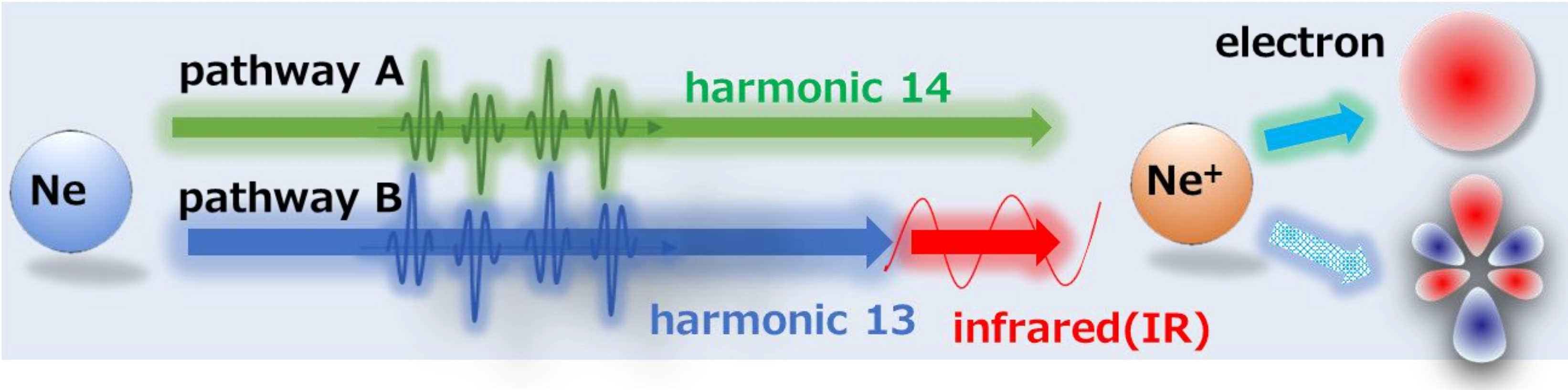
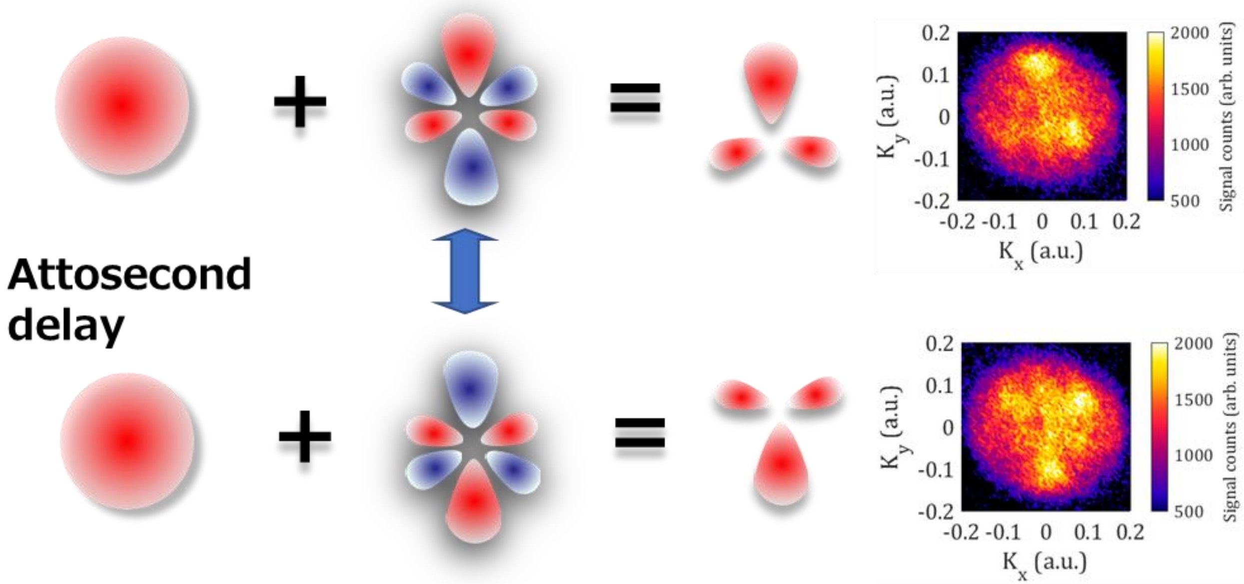
Phase measurements by an attosecond laser pulse

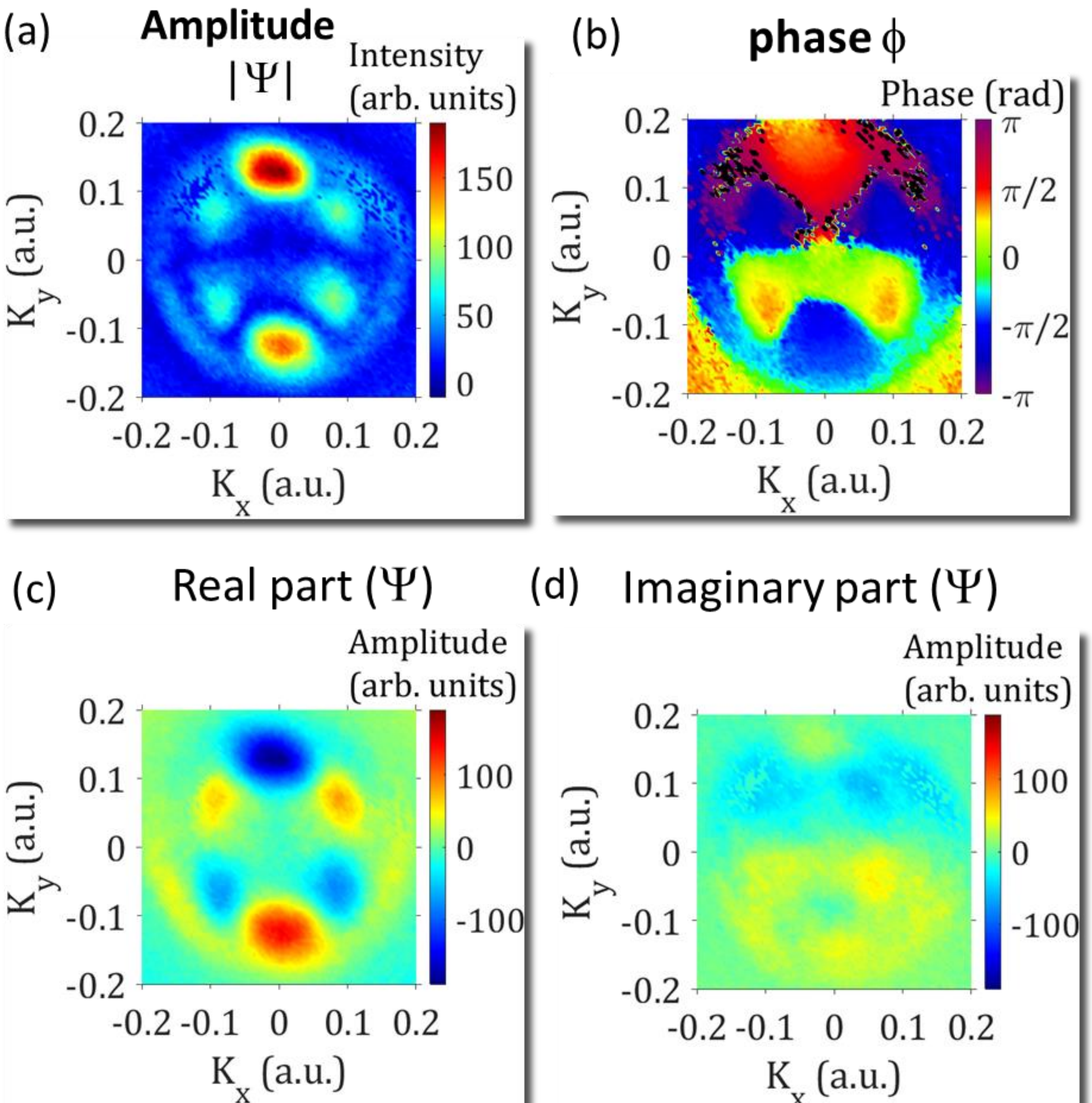


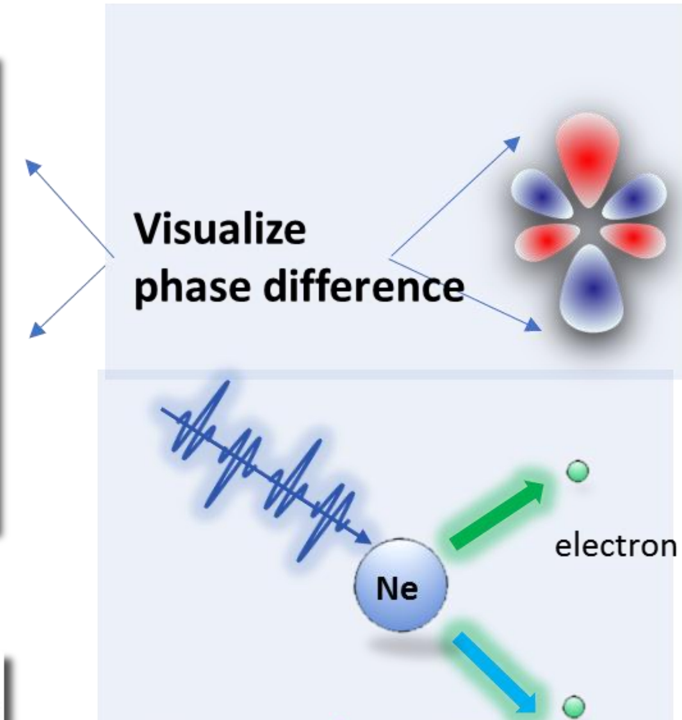
1. Add another ionization pathway (A) by harmonic 14.
2. Pathway A alone produces , pathway B alone produces
3. When both pathways can exist, coherent superposition of both wavefunctions is produced. From the interference, the phase can be retrieved.



Phase and amplitude are retrieved at every points.

Visualization of a complex wavefunction





Measure phase difference between photoelectrons ejected with different angle and energy (momentum).

a.u. : atomic unit

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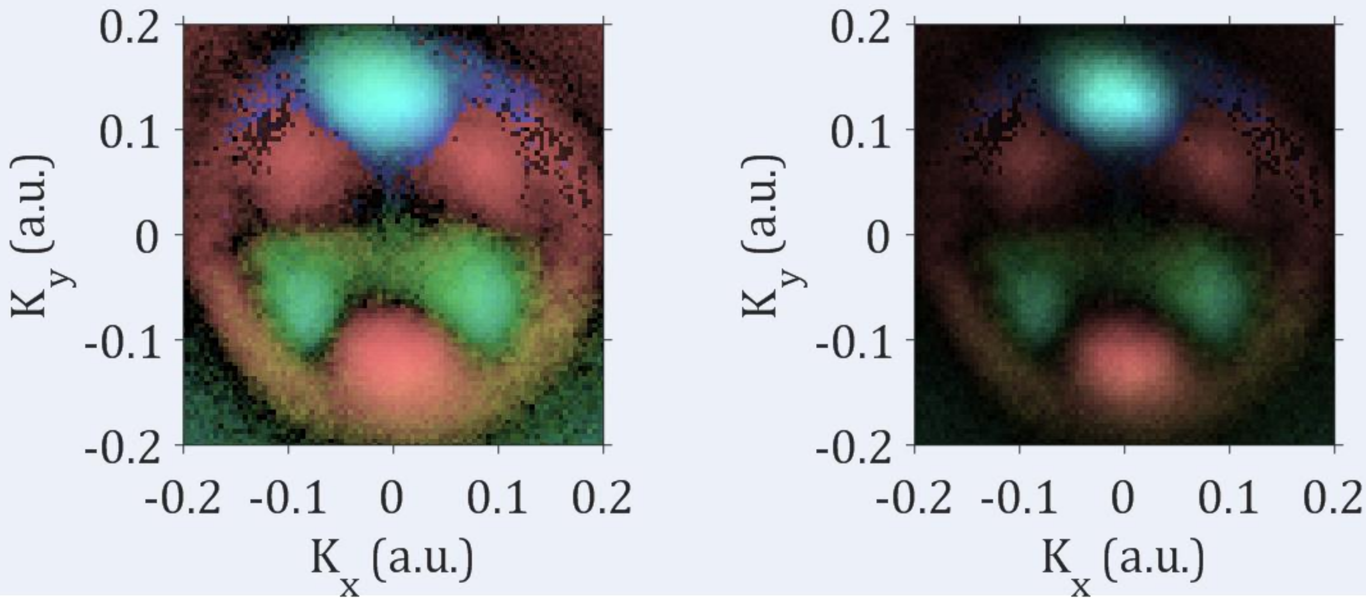
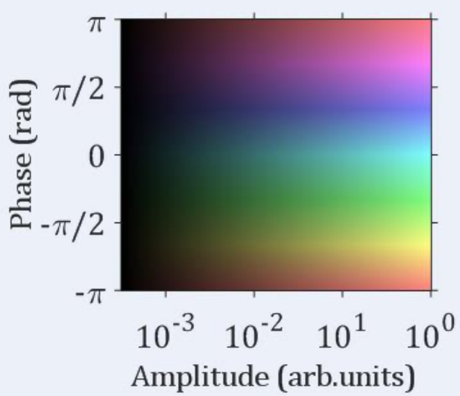
A complex wavefunction (HSV representation)



logarithmic scale

Linear scale

Visualization of both phase and amplitude distributions with one figure.



HSV ・・hue, saturation, value

phase・・・(color), hue
amplitude・・・(brightness), value
saturation=0.5

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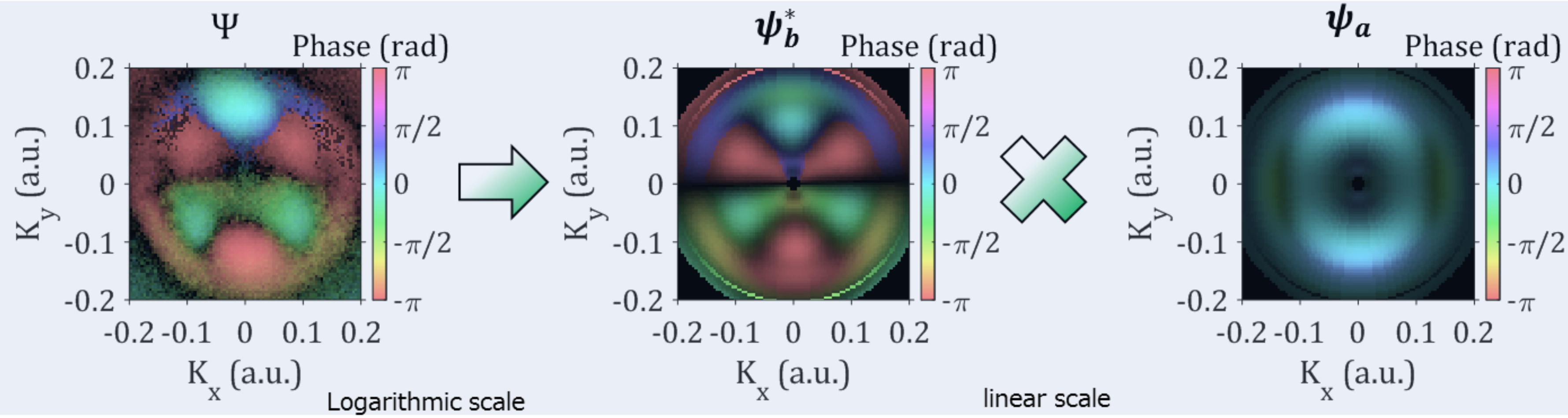
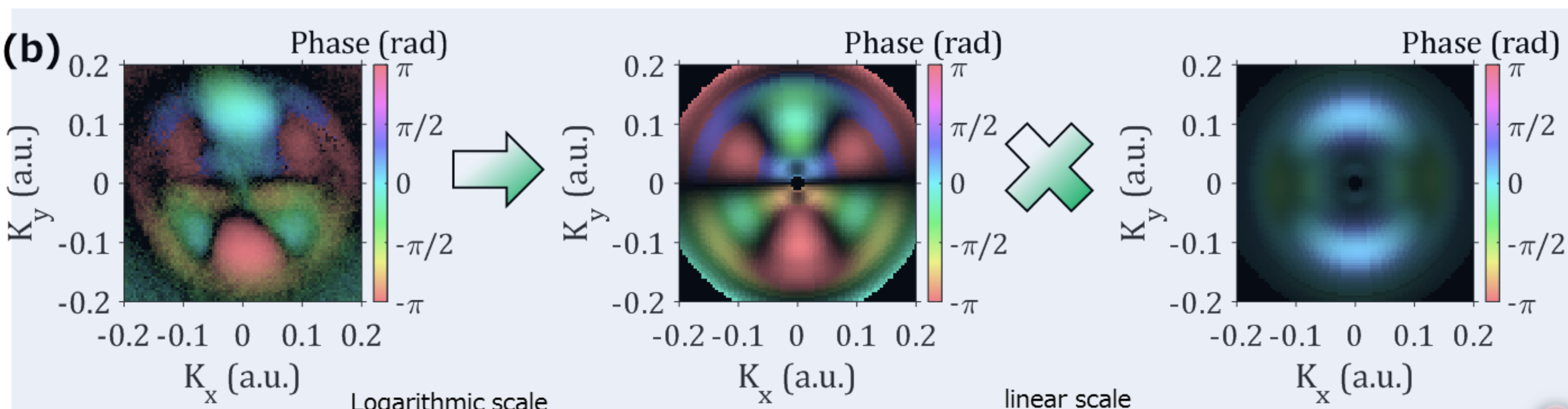
Visualize the wavefunctions produced by individual pathways

(a) **measured** **pathway B** **pathway A**



A detailed structure can be imaged.

(b)



$\Psi = \psi_b^* \psi_a$

We disentangle the measured wavefunction image into those produced by individual ionization pathways.

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Summary

An **electron** has both "**wave**" and "**particle**" nature (**duality**). The **wave nature** of an electron is characterized by both **phase** and **amplitude**. Thus, an electron is represented by a complex wavefunction.

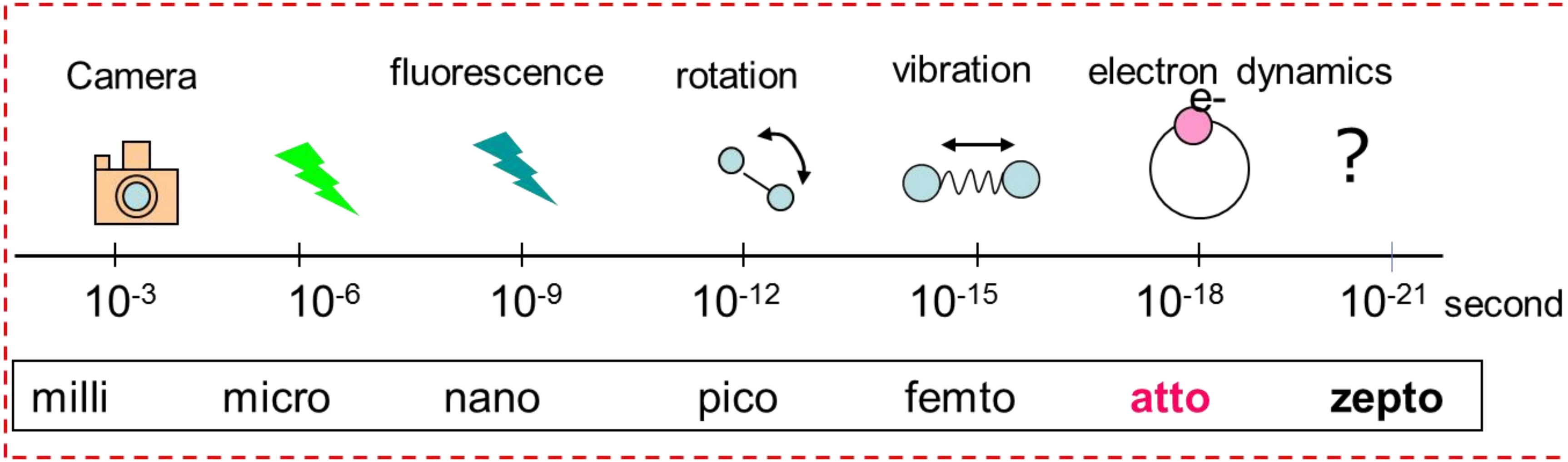
When an electron hits on a detector, it is observed as a **particle**. Repeating the measurement many times, a shape of "the square of wavefunction $|\Psi|^2$" can be seen. However, the "**phase**" information of the photoelectron is disappeared.

In this study, controlling the generation process of an attosecond laser pulse and utilizing a **two-path** interference, we measured both **phase** and **amplitude** distributions of photoelectrons. From both distributions, we **visualize the complex wave function, Ψ**. Furthermore, we visualize the wavefunctions produced by individual ionization pathways.

Using **phase** measurements, developments of a new photoelectron spectroscopy, understanding of a chemical reaction dynamics and contribution to produce a new material will be expected.

Appendix:

Attosecond science – watch "electrons"



Attosecond lab (Niikura lab) at Waseda university (Tokyo, Japan)

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Appendix:

- Characterization the sub-laser-cycle re-colliding electron pulse.
- Attosecond molecular dynamics using the re-collision approach.
- Visualization of a complex wavefunction using an attosecond laser pulse.

"Sub-laser-cycle electron pulses for probing molecular dynamics"
H. Niikura, F. Legare, R. Hasbani, M. Ivanov, A. D. Bandrauk, D. M. Villeneuve and P. B. Corkum, **Nature 417**, 917-922(2002).

"Probing molecular dynamics with attosecond resolution using correlated wave packet pairs",
H. Niikura, F. Legare, R. Hasbani, M. Ivanov, D. M. Villeneuve and P. B. Corkum,
Nature 421, 826-829 (2003).

"Tomographic Imaging of Molecular Orbitals",
J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pepin, J. C. Kieffer, P. B. Corkum and D. M. Villeneuve,
Nature 432, 867-871 (2004).

"Mapping attosecond electron wave packet motion"
H. Niikura, D. M. Villeneuve and P. B. Corkum, **Phys. Rev. Lett. 94**, 083003 (2005).

"Coherent imaging of an attosecond electron wave packet",
D.M.Villeneuve, P. Hockett, M. J JVrakking and H. Niikura ,
Science 356, 1150 (2017)

"High-resolution attosecond imaging of an atomic electron wave function in momentum space", T. Nakajima, T.Shinoda, D. M. Villeneuve and H. Niikura,
Phys.Rev. A106, 063513(2022).

"From *Femto* to *Atto* Clock"