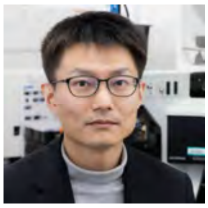
Ultrashort Electron Beam Science RIKEN-Hakubi Research Team（2024）
RIKEN Hakubi team leader: Yuya Morimoto (Ph.D.)



(0) Research fields

CPR Subcommittee**:** Chemistry

Keywords: ultrashort electron beam, pulsed laser, molecular science, attosecond science, electron microscopy

(1) Long-term goal of laboratory and research background

Electron beams are used, for example, in electron microscopy and electron-beam lithography, where high spatial resolution is required. By using state-of-the-art laser and electron-beam technologies, we aim at controlling the temporal structure of an electron beam with ultimate attosecond resolution (attosecond = one quintillionth of a second) and applying the controlled electron beams for imaging and controlling chemical reactions. We explore the atomic-scale dynamics of electrons in a material, which is the initial step of most photochemical reactions.

(2) Current research activities (FY2024) and plan

(A) Design and installation of an electron-beam energy analyzer

The generation and detection of the attosecond electron beam, which will be used in the aforementioned experiment, requires measuring the energy of fast electron beams with a resolution of a few eV. Furthermore, such an energy analyzer is well-suited for electron energy loss spectroscopy and can be used for ultrafast spectroscopy when combined with our pulsed electron-beam technology. We designed and constructed a static magnetic field 90-degree deflection type energy analyzer.

The analyzer consists of two parallel permalloy plates, each wound with copper wire, through which the electron beam travels. The beam path is maintained under ultrahigh vacuum. We optimized the analyzer's shape and evaluated its performance by calculating the three-dimensional magnetic field distribution in the beam path using magnetic-field calculation software. We then simulated the electron flight path using the calculated magnetic field. Specifically, we obtained the optimal design for a dual-focusing type spectrometer where beam focusing occurs in both directions: along the energy dispersion plane and perpendicular to it. The energy dispersion was estimated to be 8 μm/eV for 30 keV electrons, with an energy resolution of 0.53 eV (full width at half maximum). These estimated values demonstrate that the analyzer we designed possesses sufficient performance to achieve the objectives.

Next, the energy analyzer was fabricated and installed. Permalloy was magnetically annealed after machining to maximize its performance. The coils were also wound in-house. An octupole lens was fabricated to project the energy-dispersed electrons onto the detector. Subsequently, performance evaluation was conducted in combination with a home-built pulsed electron beam machine, achieving a resolution of 20 eV (FWHM).

Future plan, 1) The resolution is limited by the stability of the DC power supply used to generate the static magnetic field. Therefore, by introducing a highly stable power supply and implementing noise reduction measures, a resolution of less than 1 eV as designed should be achieved. 2) The generation of attosecond electron beams will be confirmed by modulating the pulsed electron beam with light and measuring its energy spectrum. Subsequently, attosecond pump-probe experiments will be conducted.

(B) S-matrix theory for the scattering of attosecond electron beams

Advances in electron microscopy made it possible to generate electron beams with spot sizes below the angstrom scale and temporal widths on the order of hundreds of attoseconds. However, in most existing electron scattering theories, electrons are assumed to be plane waves. We derived the S-matrix theory that treats the electron beam as a three-dimensional wave packet and allows non-perturbative treatment of the interaction between electrons and target atoms. We then calculated the scattering probability of an attosecond electron beam by an isolated atom in vacuum.

Calculated angular distributions of attosecond electron beams scattered by hydrogen and argon atoms revealed strong azimuthal asymmetry when the target atom was displaced from the beam center. The azimuthal asymmetry exhibits both the one-fold symmetry, where the sign reverses at 180 degrees, and the two-fold asymmetry, where the sign changes every 90 degrees. We found that the two-fold asymmetry dominates for hydrogen atoms, while the one-fold asymmetry is dominant for argon atoms. Detailed analysis revealed that the two-fold asymmetry originates from the spatial coherence of the electron beam, meaning that the azimuthal asymmetry is attributed to the beam's quantum nature rather than the target atom. Conversely, the one-fold asymmetry arises from phase changes in the electrons during scattering by the target atom. Argon atoms have a larger atomic number and cause a greater phase shift, resulting in the one-fold asymmetry being stronger than the two-fold asymmetry.

Furthermore, we derived from S-matrix theory an optical theorem generalized for the three-dimensional wave-packet that provides the conservation of beam flux. Numerical calculations confirmed that the theorem derived indeed shows flux conservation.

Future plan, 1) The calculation was performed on atomic targets in the ground state. Future work will extend the scope to molecules and solid crystals. Additionally, the research will expand to systems in non-equilibrium states where the target's electronic and vibrational states evolve in time, providing theoretical predictions for attosecond electron diffraction and attosecond microscopy. 2) In the calculation, we assumed the simplest Gaussian-shaped wave packet as the attosecond electron beam. Using the latest experimental techniques, wave packets having various temporal, spatial, and momentum distributions can be generated. Therefore, calculations will be performed for electron beams with different wave packet shapes to confirm whether any changes occur in the collisional process between attosecond electrons and matter.

(3) Members

(RIKEN Hakubi team leader)
Yuya Morimoto

(Special postdoctoral researcher)
Yuichi Tachibana
Marie Ouillé

(Technical staff)
Yui Yamashita

(4) Representative research achievements

1. Y. Morimoto and P. Baum, "Field-induced rocking curve effects in attosecond electron diffraction", Phys. Rev. Lett. **132**, 216902 (2024).
2. Y. Morimoto and L. B. Madsen, "Scattering of ultrashort electron wave packets: optical theorem, differential phase contrast and angular asymmetries", New J. Phys. **26** 053012 (2024).
3. L. Brückner, T. Chlouba, Y. Morimoto, N. Schönenberger, T. Shibuya, T. Siefke, U. Zeitner and Peter Hommelhoff, "Mid-infrared dielectric laser acceleration in a silicon dual pillar structure", Opt. Exp. **32**(16), 28348-28355(2024).
4. Y. Tachibana and Y. Morimoto, "Design principles of a magnetic-field-type electron energy analyzer", Sho-totsu **21** (2) R003（2024).
5. Y. Morimoto, "Ultrafast Transmission Electron Microscopy with an optically modulated Beam", Kenbikyo **59**(2), 57-61 (2024).

Laboratory Homepage

https://epulse.riken.jp
https://www.riken.jp/research/labs/hakubi/m_ultrashort_electron_beam_sci/index.html