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The Challenge of Observing Physical Phenomena via Electron Microscopy

V-shaped double-slit electron-interference experiments bring scientists one step closer to understanding mysterious wave/particle duality

RIKEN Senior Scientist Ken Harada, in collaboration with Osaka Metropolitan University, Meijo University, and Hitachi, has conducted experiments probing wave/particle duality. The results of these experiments, conducted from 2018 to 2019, bring us one step closer to understanding the mysterious wave/particle duality of quantum mechanics—and promise to help clarify the relationship between interference and electron propagation trajectories. We asked Dr. Harada to explain how he uses electron holography to see exotic physical phenomena.

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Ken Harada, PhD

In 1991, Ken Harada completed his PhD in applied physics at the Osaka University Graduate School of Engineering and joined Hitachi Advanced Research Laboratory, where he worked on observing magnetic flux quanta in superconductors. From 2001 to 2007 he was assigned to **RIKEN's Frontier Research** System, where he assembled the research team that later evolved into his current research team, focusing on the development of biprismbased optical systems for two-stage electron beams. In 2015 he joined RIKEN, where his research interests have included double-slit experiments, electron vortex beams, and schlieren optical systems.



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Experimental Results Offer Clues for Which-way Experiments

According to quantum mechanics, electrons have both wave-like and particle-like properties, but the laws of nature seem to prohibit these two types of properties from being measured simultaneously. Perhaps for this reason, the results of double-slit experiments have never moved beyond "exhibiting the mystery of wave/particle duality." The notion that individual electrons-which are only detected as particles-could simultaneously pass through two slits seems far removed from everyday human intuition. Indeed, if electrons are particles it should be possible to trace the trajectories of their motion, and this reasoning has motivated many researchers to conduct experiments using a broad array of instruments and techniques. For double-slit experiments with particles, one strategy is to detect interference fringes after determining which slit the particle passes through; experiments based on this approach are known collectively as which-way experiments.

With the goal of clarifying the relationship between electron trajectories and interference phenomena, Harada and his collaborators began experiments in 2017 using a holography electron microscope, which offers access to the highestcoherence electron beams available in the world today.

"I'm targeting the very observation of physical phenomena themselves," Harada explains, "and to that end I've thought endlessly about questions like how to exploit highly coherent electron waves, how to assemble rational optical systems, and what sorts of physical phenomena we should optimize our apparatus to observe."

In 2018, the researchers used a 1.2 MV field emission transmission electron microscope (FE-TEM) to conduct an asymmetric double-slit interference experiment, with biprisms controlling the width of each slit. To determine which slit an electron passed through, they used pre-Fraunhofer conditions corresponding to reductions in the travel distance from the double slit to the image plane; however, when interference fringes form, it becomes impossible to distinguish which slit the electron passed through. This is because, when electrons behave like waves, they are diffracted after passing through the slits even if the propagation distance is short, creating interference between pairs of electron waves. This illustrates the difficulty of controlling interference when electron waves propagate naturally, and Harada says it was their desire to solve this problem that led the team to devise an optical system operating under infocus conditions.

The experiments mentioned in the title proceeded as follows.

In the FE-TEM system, two electron biprisms were placed beneath the objective lens to control and superpose waves; this gave the researchers control over interference fringes consisting of dot images. Then they used a V-shaped double slit to observe and record interference phenomenon under pre-interference conditions, interference conditions, and post-interference conditions—via a direct-detection camera system.

"We set up an electron-optics system in which our V-shaped double slit allowed us to observe all three interference conditions (pre-interference, interference, and post-interference) simultaneously in a single field of view," Harada explains. "The upper biprism was positioned on the image plane of the double slit used to deflect the propagation of waves placed at the objective plane. The lower biprism was positioned between the objective lens and the image plane to deflect waves in such a way as to ensure that the two waves overlapped to produce a slit-shaped pattern on the image screen. Thus we designed an electron-wave doubleslit experiment in which the double-slit positions produced an image precisely at the detector surface under conditions corresponding to zero propagation distance in the optical sense."

As a result, the team concluded that interference fringes could be observed only when information on the trajectories of individual electrons could not be obtained.

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Ken Harada at the controls of a Hitachi HF-3300S TEM system. In the darkened microscopy laboratory, Harada inspects captured images with an intimidating intensity of focus.



An Idea for Observing Magnetic Flux Quanta Redefines the Term Performance

As it turned out, it was another double-slit experiment that set Harada on the road to his current research using electron microscopes. This experiment, conducted by the late Akira Tonomura—a Hitachi fellow—clearly demonstrated wave/particle duality and showcased the mysterious nature of quantum mechanics. In 1987, while a master's course student at Osaka University, Harada saw a movie of Tonomura's experiment at a physics conference in Nagoya—and remembers to this day the shock it delivered.

"To save money, I bought a discount railway ticket and traveled to Nagoya on a rickety local train that departed Osaka at 4 AM," Harada recalls. "The moment I joined Hitachi Advanced Research Laboratory in 1991, I found Dr. Tonomura and told him I wanted to perform that experiment," Harada recounts with a smile, "and he said 'Whoops! I already threw away that camera.""

Thereafter, Harada began his career as a researcher working under Tonomura, with an initial assignment to observe magnetic flux quanta in superconductors.

"At that time, there were instances in which superconducting materials been observed, but we

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were the first in the world to take on the challenge of observing the actual phenomena," says Harada. "When I started my experiments I wasn't sure if I would be able to see anything or not, but Tonomura said 'Well, if the conclusion is that the phenomenon is unobservable, then that's the conclusion—but don't ask me to bail you out when someone else succeeds in observing it!' Happily, the experiment was successful, and in 1992 we observed magnetic flux quanta in the conventional superconducting metal niobium. After that we turned our attention to Bi-based high-temperature superconductors, and in 1993 we successfully carried out a dynamical observation of magnetic flux quanta in a hightemperature superconductor."

In high-temperature superconductors, superconductivity occurs at the temperature of liquid nitrogen, which is much easier to work with than the ultra-low temperatures required for conventional superconductors. However, one problem is that electrical current flows can produce Joule heating, which may destroy the superconductivity. It was considered that Joule heating is caused by the motion of magnetic flux quanta, and the successful observation of the dynamics of such quanta was a major breakthrough that sent shockwaves throughout the research field.

Harada recounts this achievement with humility: "Of course this was not just my work—it was built on a foundation created by many previous experiments. I just happened to receive the final handoff of the baton—and had the good fortune to get involved at precisely the right time." Harada's breakthrough clearly represented dramatic progress, but further conceptual advances were needed to achieve the goal of using electron microscopes—originally developed to observe materials—for the purpose of observing phenomena.

"Of course, if you want to look at smaller materials, increasing the magnification is the



The 1 MV holography electron microscope. Akira Tonomura scoured Japan to find a site with the optimal geology and environment for this instrument, and eventually settled on its current location, Hatoyama. The instrument was built on a foundation of extremely hard ground rock.



1MV ホログラフィー電子顕微鏡

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obvious thing to do," Harada notes. "What was truly revolutionary here was that we did the opposite: we worked at low magnification—less than 100×—and worked to improve the parallelism of the world's highest-coherence electron beam. Dr. Tonomura's realization that direct observation of physical phenomena wouldn't require high magnification redefined the meaning of the term performance."

Torrential Cascades of Ideas Reveal a Spirit of Intellectual Adventure

Harada's research style is to conceive of a technique for observing some subject, develop an experimental apparatus, and use it to observe a physical phenomenon—the phenomenon itself—that nobody had seen before. Keiko Shimada, a member of RIKEN technical staff, says that Harada "is passionate about seeing things—when he gets an idea he can't rest until he's tried it out." Harada admits as much: "Sometimes it occurs to me that if I just do things a little differently, a goal that was previously out of reach may suddenly become achievable—and when that happens I get very excited and start making all sorts of requests—and since I can't help but want to attempt it, I end up asking them to do various things for me.

In response, "He has so many ideas that it's very

important for me to write them down so I don't forget them," she notes with a smile.

Today's world tends to emphasize and reward rapid results, but too much focus on the short term will eventually deplete our store of fundamental knowledge. What drives Ken Harada as a researcher seems to be a powerful spirit of adventure—a desire to see and learn about new things without known precedents, even if it's unclear what benefits may result.

The technology that connects Harada to Hitachi is the holography electron microscope (HEM). In this specialized type of electron microscope, designed to take maximal advantage of the wave nature of electrons, a base transmission electron microscope



Harada confides that he considers all work in the field of electron-wave interference to be something that he should be doing. Even with regard to double-slit experiments, his passion for research knows no bounds: "You can't be satisfied with simply achieving a demonstration."

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(TEM) system is equipped with a field emission electron gun emitting a high-brightness, highcoherence beam of electrons, together with electron biprisms to deflect this beam. By exploiting the phase change that occurs when electron waves pass through electromagnetic fields, HEMs can measure electromagnetic fields and related phenomena with high sensitivity and atomic-scale resolution.

Tonomura developed the world's first practical system in the 1970s and went on to develop higherperformance versions that notched up a number of key accomplishments in physics, including Harada's observation of magnetic flux quanta in superconductors.



Center for Exploratory Research is surrounded by an expanse of untrammeled nature, with wild weasels and raccoon dogs making frequent appearances.

The history of electron microscopy dates back to the early 20th century, with the first instrument developed in Germany in 1931. In Japan, where information from the outside world was hard to come by during World War 2, a national development effort led to Hitachi's first prototype in 1941 and, in 1942, to the market launch of Japan's first commercial electron microscope, the Hitachi HU-2. "The fact that Japanese researchers had to think through everything, starting from scratch, turned out to be very useful for the later development of the technology," says Harada. "I agree with the late Noboru Masuko, an Emeritus Professor at the University of Tokyo, who said 'With technology, you don't necessarily need to be its parent, but if you don't have memories of it from the cradle then it won't bear fruit."

Using Electron-wave Interference to Demonstrate Violation of Bell's Inequality

In the V-shaped double-slit experiment mentioned above, the operating positions of the two electron biprisms were rigorously defined by electron optics, a point that Harada says was a subject of vigorous discussions. Harada hoped not only to repeat an experiment he had witnessed over 30 years earlier, but to create his own—more advanced—version, a purpose for which Hitachi technology provided no small amount of assistance.

"In the absence of electron-wave interference, we had the information needed to determine whether the electron passed through the left or the

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right slit," Harada explains. "Our understanding had progressed at least that far. Of course, demonstrating this still required surmounting some rather tall hurdles, but my view is this: You may not be as smart as Einstein or Bohr, but as long as you have an electron microscope and a camera then you're able to do experiments. And if you're able to do experiments, then declining that challenge is really not an option."

Thanks to recent technological advances, doubleslit phenomena—long the province of thought experiments devised by ingenious theorists and accessible only by sophisticated, groundbreaking experimental techniques—are slowly shedding their exotic mystique and acquiring a more realistic real-world flavor. Ken Harada, for one, gives every indication of having become even more motivated by these developments.

"The experimental demonstration of quantum entanglement, which won the Nobel Prize for Physics in 2022, was based on experiments using photons," he notes, then continues with a note of resolve: "This may sound like a pipe dream, but I'd love to try using electrons to demonstrate violations of Bell's inequality."





Editor's Postscript

Double-slit experiments and wave/particle duality, which so captivated a young Ken Harada traveling to Nagoya on discount rail tickets, have been a subject of continuous experimental inquiry for more than half a century—and it's easy to see why: exploring and interpreting phenomena that so thoroughly confound our everyday intuition is no simple task. And yet, in facing such deep riddles, Harada's motto—"If you' re able to do experiments, then declining that challenge is really not an option"seems to me a perfect crystallization of the researcher's mission and mindset. Which slit did the particle pass through? Reporting this story leaves me eagerly awaiting the day when this question finally finds an answer.

> (Reported and written by Toshinari Yamaguchi; photography by Yuki Akiyama. Interviewed on January 13, 2023)