

Magnetic Field Observation of a Single Flux Quantum by Electron-Holographic Interferometry

Tsuyoshi Matsuda, Shuji Hasegawa, Masukazu Igarashi, Toshio Kobayashi, Masayoshi Naito,
Hiroshi Kajiyama, Junji Endo, Nobuyuki Osakabe, and Akira Tonomura
Advanced Research Laboratory, Hitachi, Ltd., Kokubunji, Tokyo 185, Japan

Ryozo Aoki^(a)

Department of Physics, Faculty of Science, Kyusyu University, Hakozaki, Higashi-ku, Fukuoka 812, Japan

(Received 1 March 1989)

The magnetic lines of force of a single flux quantum (fluxon) penetrating a superconducting film (Pb) were observed directly and individually by the electron holography technique using the Aharonov-Bohm effect. The phase contours of the electron wave not only confirm the quantized flux value $h/2e$ but also reveal, by phase amplification, internal structure of a single fluxon. With the film thickness $\lesssim 0.5 \mu\text{m}$, each fluxon, after penetrating the film, fans out or makes a U shape returning to another point on the film surface. With thicker films, fluxons form a bundle with a flux amounting to several times $h/2e$.

PACS numbers: 74.60.Ge, 61.16.Di, 74.70.Be

Many of the fruitful studies of superconductivity dealt with magnetic effects, such as the Meissner effect, the magnetic flux quantization,¹ and the lattice formation of fluxons in type-II superconductors.² One is naturally led to attempt observing the structure as well as the dynamical behavior of a single fluxon.

The first observation of fluxons was achieved by Essman and Träuble;³ they developed a high-resolution Bitter technique to observe a replica of the distribution of fine cobalt particles deposited on a type-II superconductor surface with an electron microscope, verifying Abrikosov's prediction² that the fluxons would form a triangular lattice.

The fluxons were observed also by electron interferometry utilizing the Aharonov-Bohm effect,⁴ in which two electron waves get a relative phase shift of π when their paths enclose a magnetic flux of $h/2e$, which is equal to the fluxon value. Lischke⁵ and Wahl⁶ detected the leakage of the fluxons trapped in a superconducting tube. Boersch *et al.*⁷ took a step forward to observe thermally activated jumps of pinned fluxons from one pinning center to another. Indeed, it is the merit of this

technique that it enables one to see the magnetic field pattern directly without recourse to its still replica, such as in the Bitter technique,³ thereby providing a new way to trace the dynamical behavior of fluxons. So far, however, the fluxon has been detected merely as a line of dislocations of parallel interference fringes by half of their spacing, the line not being sharp enough to permit a clean determination of the motion, not to mention the internal structure, of the fluxon. Another type of observation of a single fluxon has recently been achieved with scanning tunneling microscopy;⁸ this technique probes the electronic structure surrounding the fluxon at the superconductor surface, while the Aharonov-Bohm effect in the above experiments and ours senses the magnetic field structure.

The present Letter is the first report of our electron-holographic studies of the fluxons. We have succeeded in observing the magnetic field structure of a single fluxon penetrating a superconducting thin film. Recalling that the electron-holographic interferometry⁹ with n -times phase amplification (see below) produces one spacing displacement of fringes for a pair of electron



FIG. 1. Interference micrographs of magnetic fluxes penetrating superconducting Pb films (phase amplification, $\times 2$). Film thickness (a) $0.2 \mu\text{m}$ and (b) $1.0 \mu\text{m}$.

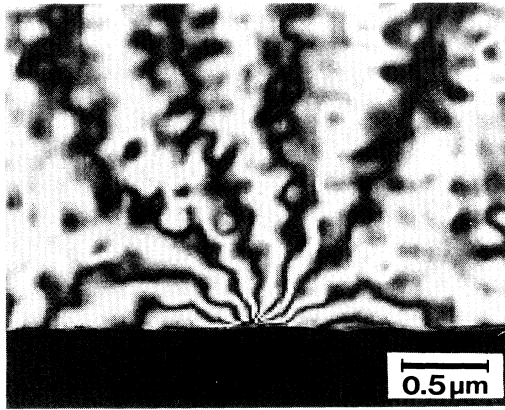


FIG. 2. 16-times phase-amplified interference micrograph of a single fluxon (film thickness $=0.2 \mu\text{m}$ and sample temperature $=4.5 \text{ K}$).

paths enclosing a magnetic flux of h/ne , we see in Fig. 1 ($n=2$) (a) isolated single fluxons that have penetrated a Pb film as thin as $0.2 \mu\text{m}$, and (b) a bundle of fluxons in the case of a Pb film of thickness $1 \mu\text{m}$. A closer look can be taken of a single fluxon by increasing the magnitude n of the phase amplification (Fig. 2, $n=16$). Further discussions will be given below after a brief description of our experimental procedure.

Our superconducting films were fabricated by evaporating Pb on one side of a tungsten wire (diameter of $30 \mu\text{m}$) at room temperature, whose surface was made clean and smooth in advance by flash heating to 2000 K with an electric current. A sample is shown in Fig. 3. The films were made up with grains of single crystals, so that special attention was paid to preparing films almost free from surface roughness, pinholes, and cracks on grain boundaries. The characteristics of the prepared samples were critical temperature $T_c=7.2 \text{ K}$, and residual resistance ratio $\rho_{300 \text{ K}}/\rho_{7.5 \text{ K}}=50-80$.

We note that, although Pb is a type-I superconductor, an applied magnetic field produces penetrating fluxons such that they are well separated from each other when the thickness of the Pb film is less than $0.5 \mu\text{m}$ (Ref. 10) as is the case for Fig. 1(a).

Our experiment consisted of two steps: electron-hologram formation and optical image reconstruction. The setup for the first step is shown schematically in Fig. 4. The electron microscope differs from a conventional one in four respects. First, a 150-kV field-emission gun is used so that the electron beam may be highly coherent and well collimated (illumination angle $=5 \times 10^{-8} \text{ rad}$). Second, it is equipped with a newly developed low-temperature stage, which can keep a sample at low temperatures down to 2 K . Third, it has a controllable electromagnet to apply a magnetic field of $0-100 \text{ G}$ on the sample in a horizontal direction. And fourth, an electron biprism¹¹ is installed to form an interference pattern between object and reference beams.

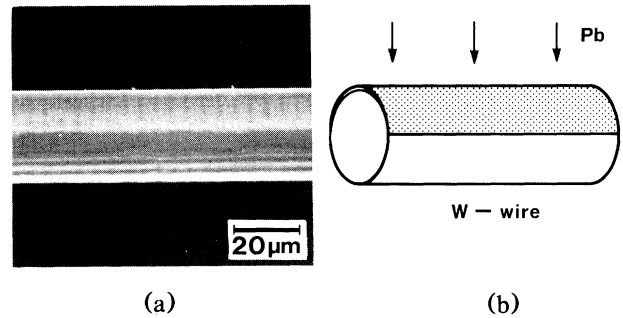


FIG. 3. Superconducting sample. (a) Scanning electron micrograph; (b) sketch.

In this experiment, we apply a weak magnetic field of $0.2-1.0 \text{ G}$ perpendicularly to the sample, and then cool the sample down to 4.5 K on the low-temperature stage. One-half of the collimated electron beam illuminates the sample for the observation of the magnetic fields penetrating the sample, and the other half acts as the reference beam. They are led to form an interference pattern on the image plane by the electron biprism. The image is formed through the intermediate lens and not through the usual objective lens, since the latter has to be turned off so that its magnetic field will not affect the sample. The image is enlarged $1000-2000$ times by electron lenses and is recorded on film to make a hologram, of which the spacing and the total number of interference fringes are $75 \mu\text{m}$ and 200 , respectively.

Optical reconstruction from the hologram using a He-Ne laser makes interference micrographs. The process is

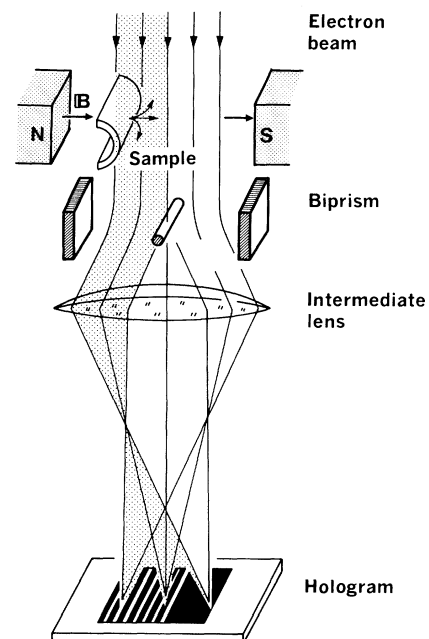


FIG. 4. Electron-optical system for hologram formation.

rather simple; a collimated laser beam illuminates the electron hologram to produce two diffracted beams, one carrying a reconstructed image and the other carrying its conjugate. A Mach-Zehnder-type interferometer makes these two images overlap to form a twice phase-amplified interference micrograph, taking advantage of the fact that two image amplitudes are complex conjugate to each other. This micrograph, prepared in the form of an interferogram, can be used as a twice phase-amplified hologram to repeat the above process to attain the higher phase amplification. Experimental details of the process are described in Ref. 12. An amplified interference micrograph can also be obtained using a digital image analysis technique.¹³

Let us now discuss the interference micrographs thus obtained for the magnetic fields penetrating the superconducting film. Figure 1 shows the twice phase-amplified contour fringes, which can be directly interpreted as projected magnetic lines of force, each representing a flux of $h/2e$.¹⁴ We note that, although a uniform external field is applied to the sample, only the magnetic fields generated by the current induced in the superconductor are observed here, because the uniform field affects equally the object electron beam passing by the sample and the reference beam passing far away. The magnetic lines of force are quite different in the two micrographs, Figs. 1(a) and 1(b), where film thicknesses are 0.2 and 1.0 μm , respectively. In the right half of Fig. 1(a), a magnetic line of force penetrates the film in an extremely localized region, and then fans out into free space. Its flux is $h/2e$ and therefore it is identified as a single fluxon. This identification is confirmed by further experiments as will be described below.

In addition to such an isolated fluxon, we observed an antiparallel pair of fluxons connected by a U-shaped line of force, as shown in the left half of Fig. 1(a). The antiparallel pair of fluxons may have been created when the film was cooled through the Kosterlitz-Thouless regime¹⁵ just below T_c , the presence of which is expected from the two-dimensional character of the thin film. The fluxon oriented against the applied magnetic field may survive to be observed as long as the field is not too strong and the pair is pinned by some mechanism so that the two would not meet to annihilate each other.

We emphasize that the antiparallel pair of fluxons has never been observed by any method, say, Bitter's, so far available, since none of them can tell the polarity of the magnetic field.

In the micrograph 1(b), magnetic flux penetrates the film in a bundle of several fluxons. This is a case of a thicker film, of thickness $\sim 1 \mu\text{m}$; it is known that the intermediate state occurs in a film thicker than 0.5 μm , causing the film to split into normal and superconducting domains.¹⁰

Internal structure of a fluxon line can be observed in highly phase-amplified interference micrographs. An ex-

ample is shown in Fig. 2; the amplification ratio n is 16, and consequently each line of force represents a flux of $h/16e$. We note that the number of lines here is 8, and hence the total flux amounts to $h/2e$ in agreement with the fluxon value. The diameter (half-width) of the fluxon at the superconductor surface is determined¹⁶ from this micrograph to be approximately 1500 $\text{\AA}$, which value is not inconsistent with the penetration depth $\sim 500 \text{\AA}$ (Ref. 17) of Pb. In order to extrapolate the fluxon profile into the superconductor, theoretical calculations are now in progress. Direct observations of the fluxons inside are also being planned with a higher-energy electron beam that can traverse the superconducting film.

In order to make sure that the magnetic fluxes we observed in the above experiments are due to supercurrents, we confirmed by the same electron-holographic interferometry (i) that the fluxes remain frozen even after the applied magnetic field is removed, and (ii) that the trapped fluxes disappear completely when the sample temperature is raised above T_c .

Thus, we have developed a method and observed the detailed structure of the magnetic field of a single fluxon. Our expectations are that this method will enable us to investigate various kinds of previously inaccessible fundamental features of superconductors. For example, it should help in determining the mechanism of anisotropic superconductivity in high- T_c materials, in clarifying the flux pinning mechanism limiting critical currents, and in searching for a possible flux quantum different from $h/2e$.^{18,19}

The authors would like to express special appreciation to Professor Chen Ning Yang of the State University of New York, Dr. Murray Peshkin of Argonne National Laboratory, Professor Hiroshi Ezawa of Gakushuin University, and Dr. Eizabro Yamada of Hitachi Central Research Laboratory for their valuable and stimulating discussions. Gratitude is extended to Dr. Ushio Kawabe, Dr. Yoshinobu Tarutani, and Dr. Masaaki Futamoto of Hitachi Central Research Laboratory for their helpful discussions and advice. Yukio Honda of Hitachi Central Research Laboratory and Hisashi Nagano, Takao Matsumoto, and Seiichi Kondo of Hitachi Advanced Research Laboratory also deserve thanks for their assistance during the experiments.

(a)Present address: Department of Electrical Engineering, Faculty of Engineering, Osaka University, Yamadaoka, Suita 565, Japan.

¹B. S. Deaver and W. M. Fairbank, Phys. Rev. Lett. 7, 43 (1961); R. Doll and M. N  bauer, Phys. Rev. Lett. 7, 51 (1961); N. Byers and C. N. Yang, Phys. Rev. Lett. 7, 46 (1961).

²A. A. Abrikosov, Zh. Eksp. Teor. Fiz. 32, 1442 (1957) [Sov. Phys. JETP 5, 1174 (1957)].

- ³U. Essman and H. Träuble, Phys. Lett. **24A**, 526 (1967).
See also P. L. Gammel *et al.*, Phys. Rev. Lett. **59**, 2592 (1987).
- ⁴Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959).
See also A. Tonomura *et al.*, Phys. Rev. Lett. **56**, 792 (1986).
- ⁵B. Lischke, Phys. Rev. Lett. **22**, 1366 (1969).
- ⁶H. Wahl, Optik (Stuttgart) **28**, 417 (1968).
- ⁷H. Boersch *et al.*, Phys. Status Solidi (b) **61**, 215 (1974).
- ⁸H. F. Hess *et al.*, Phys. Rev. Lett. **62**, 214 (1989).
- ⁹A. Tonomura, Rev. Mod. Phys. **59**, 639 (1987).
- ¹⁰M. Tinkham, Phys. Rev. **129**, 2413 (1963); R. N. Goren and M. Tinkham, J. Low Temp. Phys. **5**, 465 (1971); G. J. Dolan, J. Low Temp. Phys. **15**, 111 (1974).
- ¹¹G. Möllenstedt and H. Dücker, Naturwissenschaften **42**, 41 (1954).
- ¹²A. Tonomura *et al.*, Phys. Rev. Lett. **54**, 60 (1985).
- ¹³S. Hasegawa *et al.*, J. Appl. Phys. **65**, 2000 (1989).
- ¹⁴A. Tonomura *et al.*, Phys. Rev. Lett. **44**, 1430 (1980).
- ¹⁵J. M. Kosterlitz and D. Thouless, J. Phys. C **6**, 1181 (1973).
- ¹⁶The fluxon diameter could not be estimated for all of the observed fluxon patterns, since some of the fluxons penetrated the superconductor sample at regions which were not at the extreme edge of the sample shadow. Their magnetic fields just at the superconductor surface were hidden in the shadow and consequently could not be measured.
- ¹⁷For example, H. R. Kerchner and D. M. Ginsberg, Phys. Rev. B **10**, 1916 (1974).
- ¹⁸V. B. Geshkenbein and A. L. Larkin, Phys. Rev. B **36**, 235 (1987).
- ¹⁹P. W. Anderson *et al.*, Phys. Rev. Lett. **58**, 2790 (1987).

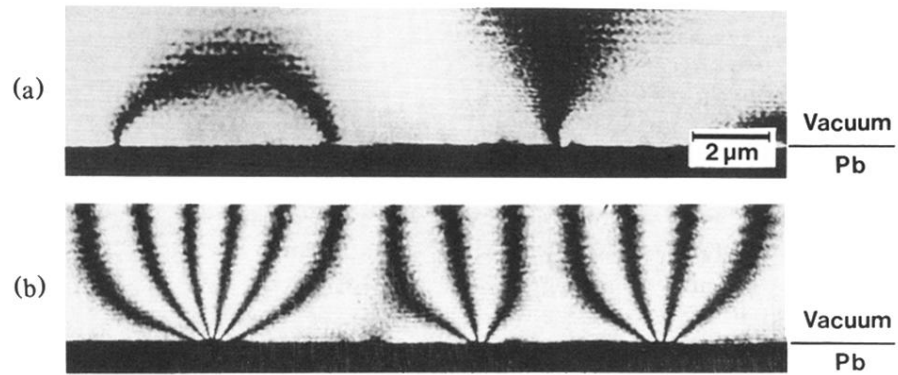


FIG. 1. Interference micrographs of magnetic fluxes penetrating superconducting Pb films (phase amplification, $\times 2$). Film thickness (a) $0.2 \mu\text{m}$ and (b) $1.0 \mu\text{m}$.

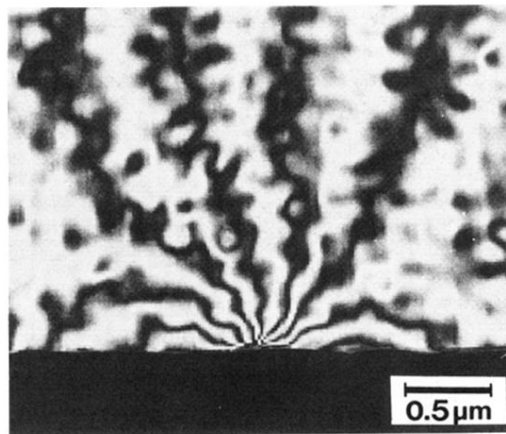


FIG. 2. 16-times phase-amplified interference micrograph of a single fluxon (film thickness = $0.2\text{ }\mu\text{m}$ and sample temperature = 4.5 K).

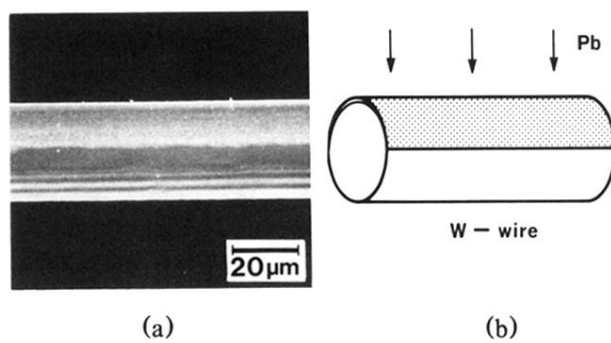


FIG. 3. Superconducting sample. (a) Scanning electron micrograph; (b) sketch.