Electron Holography Observation of Vortex Lattices in a Superconductor

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Magnetic lines of force penetrating a superconducting Nb thin film have been directly observed by electron holography. The tilted specimen is examined in the "transmission" mode that allows the 2D magnetic flux distribution to be discerned in an interference micrograph. The phase distribution of the electron wave transmitted through the specimen has been quantitatively measured. It was found that a 2D array of tiny regions, where the phase distribution rapidly changed, coincided spatially with the spots observed by Lorentz microscopy, and, furthermore, these regions were identified to be quantized vortices each having a flux of $h/2e$ by comparing with theoretical calculations.

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Several methods have been developed to observe the presence of quantized magnetic flux lines (henceforth vortices) in superconductors, most notably Bitter magnetic decoration [1,2], scanning tunneling [3], and scanning electron [4] microscopies. Also, scanning Hall probes have been used to resolve individual vortices [5], where the surface field perturbation is related to the flux distribution. These techniques provide a static image of the vortices. A magneto-optical technique has been developed that time resolves the flux distribution; however, individual vortices were not observed [6]. Electron holography [7] has previously been used to investigate the dynamic behavior of magnetic fields close to the surface of a superconductor [8–10], but the 2D vortex lattice could not be observed.

Recently, we succeeded in observing for the first time vortex lattices in a thin specimen by means of Lorentz electron microscopy to study the dynamic behavior of vortices [11]. In this technique, the electron phase shift produced by the vortices in a tilted specimen [12] is manifested in a defocused image.

However, a disadvantage of the Lorentz mode, as well as of the other standard phase contrast methods in electron microscopy [13], is that it is very difficult to extract quantitative information from the experimental data. For instance, the Lorentz micrograph indicates both the location and the polarity of the vortices, but not the degree of flux quantization, which plays an important role in superconductivity. In contrast, electron holography measures the amplitude and phase information on the entire object and therefore it can be used to measure the flux quantitatively. Moreover, holography has a higher spatial resolution than the Lorentz mode.

This paper reports the first results on the direct observation of projected magnetic lines of force of the vortices by electron holography and the theoretical interpretations of the resultant phase distributions.

The experiment was conducted in a 300 keV holography electron microscope developed to provide a highly coherent and bright source of electrons [14]. The microscope is equipped with a specially constructed cold stage to allow magnetic fields to be applied while the specimen, tilted at 45° with respect to both the electron beam and the magnetic field, can be maintained from 30 K down to 4.5 K (Fig. 1). A rotatable electron biprism [15] is used to form holograms.

The thin (70 ± 20 nm) foil specimens used for transmission observation were prepared by chemically polishing 2×2 mm^2 wide by 30 μm thick Nb [$T_c = 9.2$ K, resistance ratio $R(300\text{ K})/R(10\text{ K}) \approx 20$] sections that had been annealed to $\sim 2000\,^\circ$C in a vacuum of 10^{-6} Pa. The annealing resulted in a grain size of 200 to 300 μm with a [110] texture.

Electron holography consists of two steps [7]. First, a hologram containing the amplitude and phase of an elec-

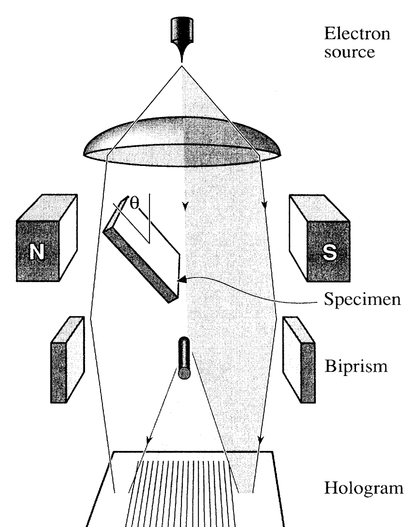


FIG. 1. Schematic of the experiment. The specimen is tilted by θ to the electron beam, and the magnetic field is applied horizontally. Electrons passing through the specimen (object wave) are interfered with the reference wave via the biprism forming a hologram.

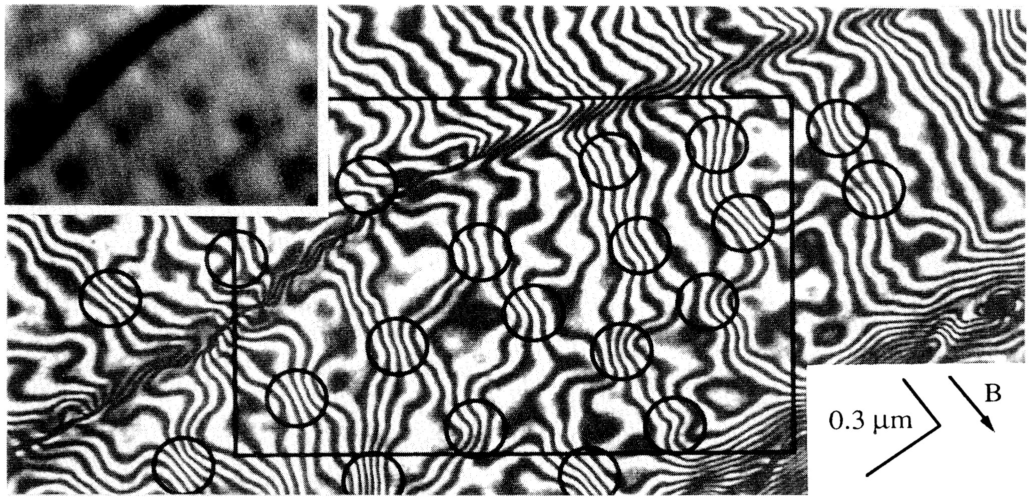


FIG. 2. Interference micrograph of a vortex lattice (phase amplified 16×). Projected magnetic lines of force are directly observed as contour fringes. They are concentrated locally at the circled regions, becoming narrowly spaced. These regions spatially coincide with the spots observed in the Lorentz micrograph (inset), and are identified to be quantized vortices. A bend contour runs diagonally through the Nb foil.

tron beam transmitted through the specimen is recorded on film. The image can then be reconstructed numerically or optically. During reconstruction, a planar comparison wave is interfered with the reconstructed image, via a Mach-Zehnder interferometer, to produce contour fringes of constant phase. By choosing the appropriate conditions of tilting the comparison wave, an electron phase map of the vortex lattice is created. The phase shifts produced by the vortex lattices are rather small and therefore the phase distribution needs to be amplified [16].

The observations were conducted with the objective lens switched off, using the intermediate lens for imaging, such that the holograms had an overall electron optical magnification of $\sim 2000\times$, with a carrier fringe spacing referred to the specimen of 30 nm. Exposure times of about 20 sec were used.

The experimental procedure was as follows. The Nb thin film was cooled to 4.5 K in a magnetic field of 100 G. The vortex lattice was first observed by Lorentz microscopy [11], each vortex being observed as adjacent spots of light and dark contrast. Then, electron holograms were formed. Images were subsequently reconstructed from the holograms as phase amplified interference micrographs. The contours indicate the magnetic lines of force projected along the electron beam direction. We should note that while the reference wave used to form the hologram has been influenced by the applied magnetic field, the contours indicate magnetic lines of force produced only by the vortices.

The resulting interference micrograph, Fig. 2, is 16× phase amplified so that the phase difference between two contours is $\Delta\varphi = \pi/8$. It can be seen from the micrograph that the magnetic lines of force flow in the direction of the applied magnetic field as a whole, but become locally dense at the circled regions. These regions were found to spatially coincide with the spots in the Lorentz micrograph (Fig. 2 inset). The magnetic flux flowing through each vortex appears to be equivalent to a phase difference of $\pi/2$.

Actually, in the case of the vortex shown in Fig. 3, the phase difference was measured to be 0.55π. This reconstruction was obtained by carefully choosing the tilt of the comparison wave in such a way as to obtain a contour map with the overall phase as flat as possible over the whole field of view. This process was necessary for pre-

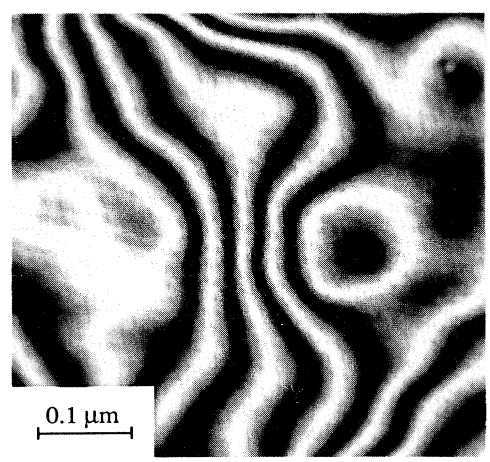


FIG. 3. Interference micrograph of a single vortex (phase amplified 16×). The vortex produces a phase difference (measured from left to right) of 0.55π, consistent with the simulation.

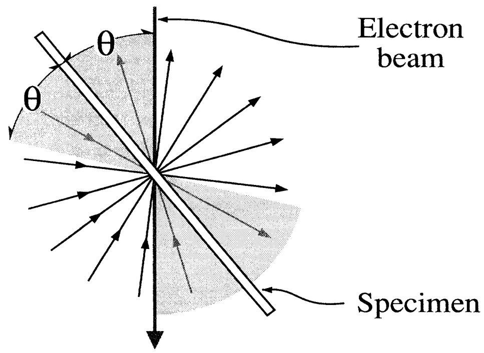


FIG. 4. The effect of specimen tilt on the electron phase difference. Though the vortex contains a single quantum of flux, $h/2e$, portions of the flux (shaded) do not contribute to the measured phase difference.

cise phase measurement in order to correct for any nonuniformity in the specimen thickness and other factors.

The measured phase difference seems to correspond to a magnetic flux one-half the value naively expected from the Aharonov-Bohm effect [17] for a singly quantized flux, that is, $h/2e$. However, the flux flowing through the vortex should be exactly $h/2e$, and not $0.55h/2e$. This fact was proven by theoretical calculations taking into account the effect of tilting the specimen as described below by a simple geometrical argument.

Consider a single vortex in a superconducting specimen (without applied field). When the specimen is tilted, not all of the flux penetrating a vortex contributes to the electron optical phase difference (see Fig. 4). That is, the magnetic flux enclosed by two electron paths passing on either side of the vortex becomes less than $h/2e$ and, accordingly, the electron phase difference is less than π. In the limit of vanishing vortex core radius (L) and specimen thickness (t), the phase difference is given by [12]

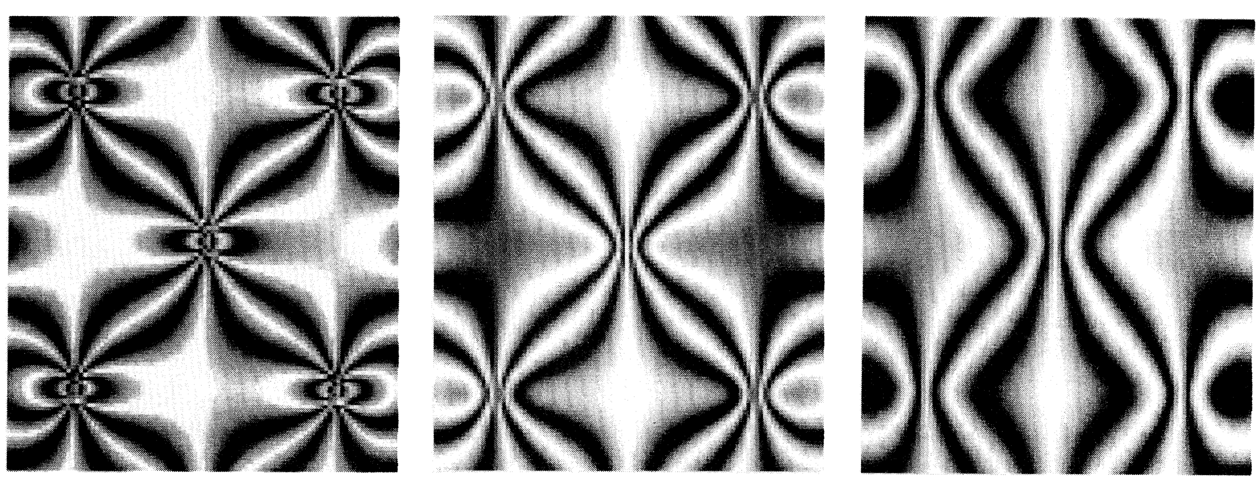
$$\Delta\varphi = 2(\pi - 2\theta)\frac{\Phi}{h/e}, \tag{1}$$

where θ is the tilt angle. Consequently, in the present study ($\theta = 45°$), $\Delta\varphi$ is expected to be $\pi/2$. In previous holography experiments [8–10], the specimen surface was parallel to the electron beam ($\theta = 0°$) and the expected $\Delta\varphi$ of π is recovered.

In the case of an actual specimen, the thickness and the core size are of the same order (e.g., $t \approx 2L$). In this situation, the presence of a finite *length* of vortex core increases the amount of flux enclosed by the electron paths and the resulting $\Delta\varphi$ becomes greater than $\pi/2$. Taking this effect into consideration, vortex lattice images were simulated on the basis of the flux tube lattice model [18]. This model assumes a finite t and a vanishing L, and we have extended the model to the case of a finite L.

The calculated results shown in Figs. 5(a)–5(c) can be directly compared with the experimental data (Figs. 2 and 3). It can be seen that the interference images are sensitive to core sizes, shown for 0 (flux tube case), 30, and 60 nm. The total phase differences across the vortices are π, 0.58π, and 0.57π, respectively. These latter two for finite core sizes are in good agreement with the experimental values.

The interference micrographs are also sensitive to the vortex inner core structure. Indeed, the measured size of the cores is ~100 nm, although it should be pointed out that the resolution of the reconstructed image is of the same order. However, the salient point is that the ability to directly compare the experimental and theoretical data allows some of the troubles associated with the observation of long range fields [19] to be overcome; namely, (i) the arbitrariness of the reference wave in the reconstruc-



(a) (b) (c)

FIG. 5. Calculated interference micrographs (phase amplified 16×): (a) $L = 0$ nm, (b) $L = 30$ nm, and (c) $L = 60$ nm. The vortex spacing is 0.5 μm (five vortices are shown) and the foil thickness is 50 nm.

tion step can be compensated for, and (ii) the influence of the tail of the vortex field on the reference wave in the microscope can be quantitatively evaluated.

In conclusion, we have directly observed for the first time projected magnetic lines of force of the 2D vortex lattices in a superconductor by electron holography, and the obtained results have been interpreted by comparing them with calculations based on a theoretical model accounting for the finite core size of the vortices. The calculated phase difference across a singly quantized vortex agrees with the measured value of 0.55π.

These holographic experiments (whose realization is at the frontier of current instrument performance), in conjunction with the more standard Lorentz microscopy methods, are paving the way for a deeper understanding of the vortex structure and of its interaction with the structural properties of the superconducting specimen, thanks to the high spatial and temporal resolution of transmission electron microscopy techniques.

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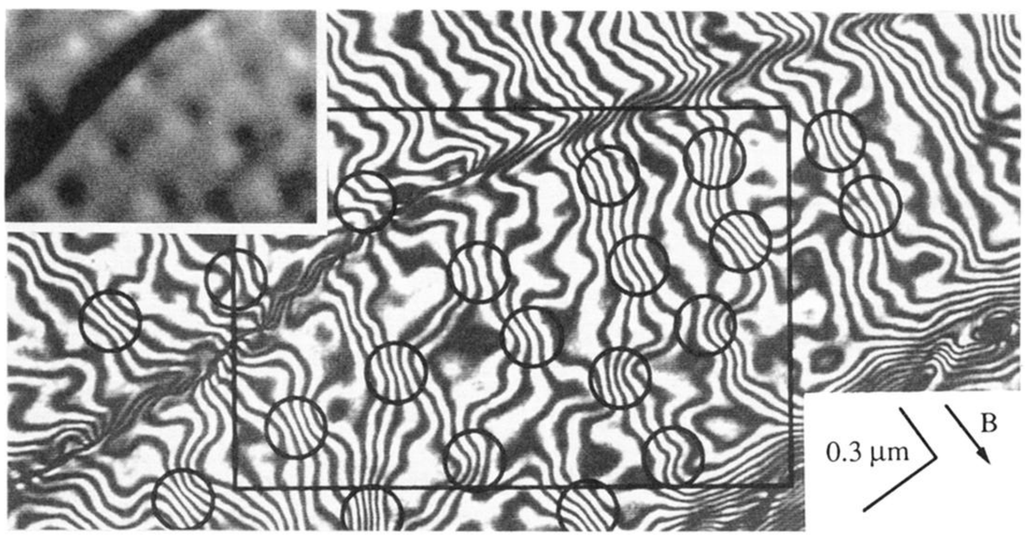

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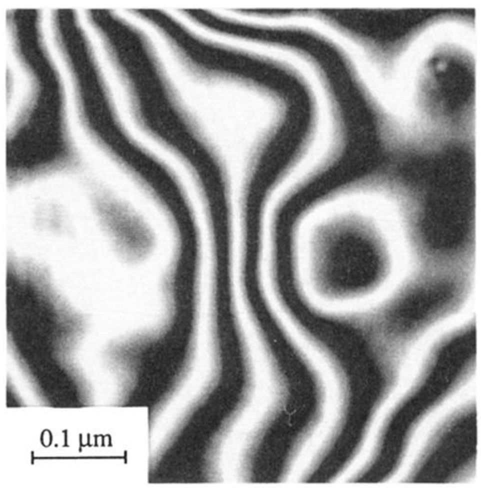

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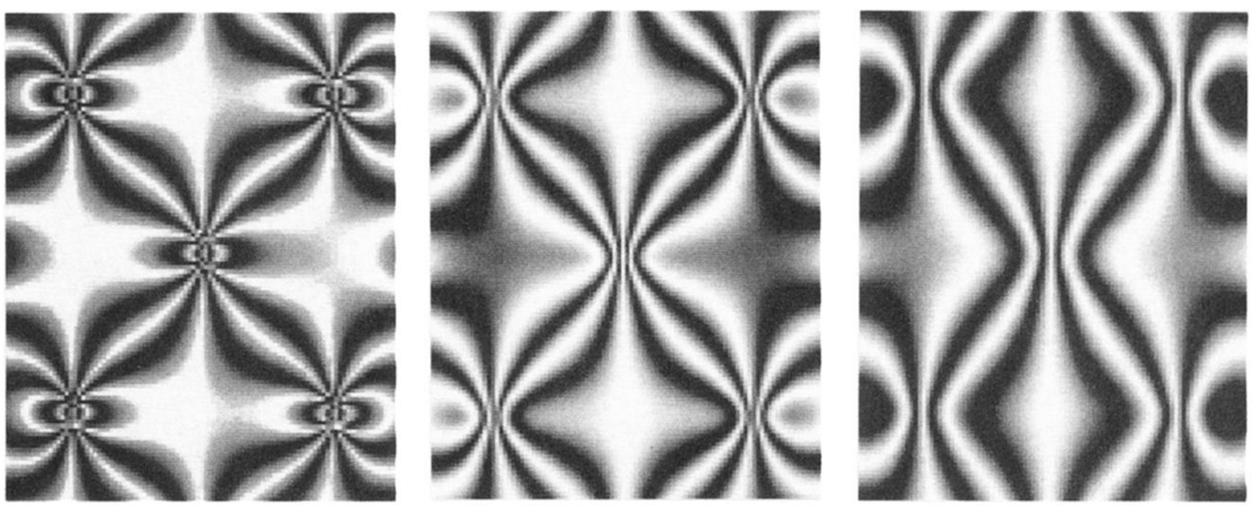

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