Performance and Capabilities of JEM-3000F to Advanced Materials Characterization at Brookhaven National Laboratory

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In this report we describe some of the instrumentation and performance of our JEM-3000F field-emission TEM to the study of technologically important superconductor and magnetic materials. To illustrate the capability of our JEM-3000F, we highlight the more advanced techniques being performed in our lab. We focus on the performance of our microscope to carry out off-axis electron holography at medium- to high-resolution, as well as at low-magnification appropriate to magnetic studies. We present abbreviated results from coherent electron diffraction experiments using magnetic materials recently performed at our laboratory, which complement the higher-magnification holography studies. We further present results of magnetic field calibration of the sample area of our JEM-3000F as a function of objective lens excitation, allowing quantitative in-situ magnetization and demagnetization experiments to be performed. We present a sampling of results from field-calibrated magnetic imaging studies that we have carried out with our microscope that, along with the high- and low-magnification holography and coherent electron diffraction experiments, indicate a highly coherent electron source for our microscope. We also illustrate some of the spectroscopic capabilities of our TEM instrumentation.

1. Introduction

The combination of advanced analysis techniques and cutting-edge instrumentation in transmission electron microscopy (TEM) allows for powerful capabilities to the study of technologically exciting materials. The JEM-3000F field-emission TEM at Brookhaven National Laboratory is just such a microscope equipped with, for example, a Scanning Transmission Electron Microscopy (STEM) attachment, Energy-Dispersive X-ray (EDX) detector, Annular Dark-field (ADF) detector, integrated Gatan Imaging Filter (GIF) for spectroscopy and energy-filtered imaging (giving the option for automatic magnification compensation of the GIF), and electron biprism for holographic studies. The microscope is equipped with two multi-scan CCD cameras mounted before and after the GIF, a Fuji Imaging Plate system replacing the conventional photographic plate system, and two TV-rate CCD cameras mounted, as well, above and below the GIF for in-situ recording and observation. A number of sample holders are also available for our microscope, including heating (to ~1400 K) and cooling (to ~15 K) holders, offering a rather wide range of experimental capabilities exploiting our unique combination of instrumentation. Equipped with a high-resolution pole-piece, our microscope has demonstrated 0.165 nm spatial resolution and energy resolution of 0.7 eV.

In this report we describe results from a sampling of some of the capabilities of our JEM-3000F which highlight the more advanced techniques being performed in our lab. Due to limited space we refer the reader to the literature [1] for the high-resolution performance of our JEM-3000F. We focus here on the recent addition of our ability to perform off-axis electron holography studies where, as the number of research labs equipped for electron holography increases, it is valuable to have an assessment for a particular machine and holography setup to record holograms. An abbreviated discussion of the experimental parameters affecting the quality of holograms is included in Section 2, from which a general approach to characterizing and optimizing the microscope for a given application is presented. It is clear from the discussion that flexibility provided by free-lens control is critical to recording useful holograms for the JEM-3000F. We present an electron holography study of (001) twist boundaries in the \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \) (Bi-2212) high-temperature superconductor in Section 3.1, and demonstrate the ability of this microscope to perform electron holography at medium- to high-resolution. In Section 3.2 we present results from the same Bi-2212 system using newly developed interferometric shadow-imaging-diffraction methods. In Sections 4.1 and 4.2 we present our calibration of the magnetic field at the sample area as a function of objective lens excitation using Hall probe measurements, as well as low-magnification holography and in-situ Foucault imaging studies of \( \text{Nd}_2\text{Fe}_14\text{B} \) permanent magnets. We round out this report in Section 5 by providing an example of the spectroscopy imaging capabilities for our machine.

2. Characterization for Off-Axis Electron Holography

In conventional transmission electron microscopy (TEM), the phase of the image wave is lost on recording since only the image intensity (amplitude-squared) is detected. Off-axis electron holography, however, allows...
retrieval of both the phase and amplitude of the image wave [2]. This is the primary advantage of off-axis electron holography since the phase carries information about the electric and magnetic fields of the sample. Off-axis electron holography is an interferometric technique requiring a coherent electron source, such as provided by a field-emission electron gun (FEG), and an electrostatic biprism situated below the specimen. The sample is positioned off the optic axis of the microscope so that incidental plane-wave illumination passes partially through vacuum (reference wave) and partially through the sample (object wave), as shown schematically in Fig. 1a. The biprism is oriented such that the reference and object waves pass on either sides of the wire, being brought together by a bias applied to the biprism. The resulting interference pattern (hologram) is recorded on a CCD camera, and the complex image wave (amplitude and phase) is mathematically reconstructed from the hologram intensity through knowledge of the interfering reference wave, i.e., plane-wave vacuum reference. Figure 1b shows a schematic of the highly astigmatic illumination used to record off-axis electron holograms. The illumination is generally stretched as much as possible perpendicular to the biprism wire (and, therefore interference fringes) in order to maximize the size of the coherence patch over the width, D, of the hologram.

In practice, the reconstruction process is more involved than implied above and depends critically upon recording holograms with a strong contrast of interference fringes. Additionally, specific experimental objectives impose criteria on the quality and characteristics of the recorded hologram. Experimental parameters directly affecting the fringe contrast include electron wavelength, gun brightness, virtual source size, illumination convergence angle, interference overlap distance, and size of illumination perpendicular to the interference fringes. On the other hand, depending upon experimental objectives, requirements on the interference fringe spacing, overlap distance, and magnification are imposed. Further consideration involving the overall intensity and exposure time required to record the hologram as related to the detection efficiency of the CCD camera is also needed. As a consequence, careful experimental design and detailed knowledge of microscope and CCD imaging characteristics affecting the recordable quality of holograms is needed to efficiently obtain useful data. Complicating matters in this regard is that for a given holography setup, i.e., geometry between microscopic, biprism and CCD camera, the large number of experimental parameters are not necessarily independent from each other. It is evident that both practical considerations and some handle on an optimization approach is required to record useful data.

To begin, the detection quantum efficiency (DQE) of the CCD camera used to record holograms was determined according to the methods described in references [3, 4]. The DQE is a quantitative measure of the detection capabilities of the CCD camera, and depends upon spatial frequency as well as electron dose and exposure time. For our camera the DQE was found to be fairly uniform over the range of spatial frequencies up to the Nyquist limit for typical exposure times (<10 sec) and electron dose (~500 to 10000) the average counts/pix) used in conventional TEM applications. Values of DQE -0.75-0.8 were obtained for low-to-mid spatial frequencies, whereas the DQE dropped to around 0.65-0.7 at frequencies near the Nyquist limit. (A value of DQE=1 indicates ideal detection characteristics.)

For holography applications, however, one is ultimately concerned with maximizing the beam coherence, which necessitates concern over the CCD detection capabilities at low dose and/or long exposure time. Under these conditions the rate at which counts (due to signal) are being generated at the CCD camera becomes important. (i.e., the average beam intensity incident on the camera is such that only a small number of counts per second are being generated, one expects the signal to be lost within thermal and electrical noise generated by the camera, regardless of exposure time.) Figure 2 illustrates the effect of low intensity (average counts per pixel per second) on the DQE for our camera. As seen in the figure, a severe degradation in the camera's detection capability exists for incident intensities <30 counts/sec. This provides a practical guideline in attempting to optimize microscope parameters for electron holography such that an average hologram intensity recorded at greater than about 50 counts/sec (for our camera) is necessary in order that the CCD camera not limit the accuracy of the reconstructed phase and amplitude. Within this limit then, one may examine the effect of various other experimental parameters on the precision of phase and amplitude determination.

The minimum variations in the reconstructed phase and amplitude determination, in general, is inversely proportional to the product DQE (μ/q)N, where μ and q are the interference fringe contrast and spatial frequency, respectively, and Nt is the total electron dose of the hologram [5]. If one restricts consideration to holograms recorded at sufficient intensity as discussed above, the factor DQE(μ/q)N is constant and optimization of parameters amounts to maximizing μN. In terms of experimental parameters, the product μN, can be written as [6, 7]

$$\bar{N} = \frac{A}{\mu q} \exp \left( -\frac{r}{2} \right)$$

where A is the area of the hologram, μ the gun brightness, R, the source size at the sample, d, the illumination convergence semi-angle, θ the exposure time, d, the electron wavelength, and e the electron charge. The overlap distance D is the distance between points in the object plane brought to overlap in the image plane by the biprism, while d, and d, are the size of the illumination perpendicular and parallel to the interference fringes, respectively. (See, also, Fig. 1b.)

Equation 1 illustrates the complex interplay between the wide variety of experimental parameters affecting the quality of recorded holograms, but is not very practical in guiding the choice of those parameters. It is instructive, therefore, to define dimensionless quantities,

$$\frac{d_1}{d_2} \frac{d}{d_1} \frac{d}{d_2} \frac{\bar{N}}{D}$$

where i, is the probe current at the specimen, which relate to particular aspects of the holography experiment. The quantity i, is the coherence parameter of the illumination and is given by the ratio of the coherence width to source size [7]. Values of i, range from 0.1 for essentially incoherent illumination to i,=10 for effectively total coherence. The quantity i, depends on the illumination (as it affects d, but specifically as a ratio D/d, upon the geometry of the holography setup including the intermediate and projector lens excitations. The geometric parameter i, (having, typically, values on the order 10^3) also carries information about the biprism bias as it affects D, therefore changing spatial filtering and magnification implied by the intermediate and projector lens settings. i, carries the information required in consideration of specific experimental objectives such as desired resolution. The parameter i, depends upon the brightness characteristics of the gun, as well as the illumination system, and may be thought of as being described by factors affecting the average hologram intensity, i.e., as an imaging (intensity)
parameter. Typical values for $\varnothing$ are within an order of magnitude or two of $10^6$ for a FEG.

Expressed in terms of these parameters describing various aspects of the holography experiment, the quantity to be maximized in Eqn. [1], $\varnothing \propto \ln n$, is proportional to the reconstructed phase and amplitude signal-to-noise ratio, and given by

$$\Delta \varnothing \propto \exp(-2D^2/P^2). \quad [2]$$

Here, we have written $\varnothing := \frac{D}{f}f$ being the fraction of the interference area ($\sim fD$) recorded, and approximated the gun brightness as $\sim 9$. The factor $D$ is related to the physical size of the CCD detector and microscope magnification, but is somewhat incidental to this analysis and could be absorbed into $\Delta$ without loss of generality. It should be cautioned that the parameters $\Delta$, $\varnothing$ and $\varnothing$ are not independent from each other in terms of experimental variables so that a sanguine maximization of Eqn. [2] is not easily interpretable or even practical. The merit of Eqn. [2] is that the shapes of imaging ($\varnothing$), coherence ($\varnothing$) and geometry ($\varnothing$) in holography experiments are rather simply for their affect on measurement precision, and that the range of practically obtainable values for the parameters are easily known or calculable. This allows an assessment of the importance of each aspect by evaluating how $\varnothing$ varies with each parameter independently.

In particular, eliminating factors common to each partial derivative, and taking the orders of magnitude as discussed above ($\varnothing \sim 10^{-1}$ and $\varnothing \sim 10^{-10}$), one finds

$$\varnothing \sim 10^{-2} \quad \text{(imaging)}$$
$$\varnothing \sim 40(D/d) \times 10^{-10} \quad \text{(coherence)} \quad [3]$$
$$\varnothing \sim (1 - 4D/d^2) \times 10^{-10} \quad \text{(geometry)}$$

for the relative effect of each experimental aspect on the measurement precision. It is clear from Eqn. [3] that the optimization approach to holography must involve, in order, the holography geometry as imbedded in $\varnothing \equiv D/d$ followed by the microscope coherence. Optimization relative to the imaging intensity ought to be relegated to final considerations assuming a sufficient minimum intensity is met. We stress here that Eqn. [3] gives the relative changes of the phase and amplitude measurement precision with respect to the various parameters, hence provides a guide as to which aspect affects most dramatically the measurement precision. The parameters are not truly independent, however, so Eqns. [2 & 3] must be used with caution and practical optimization must proceed in a wider sense. Nevertheless, Eqns. [2 & 3] make a few additional important points to consider.

Firstly, free-lens control of the microscope is critical to obtaining the highest quality holograms for a given application. In particular, control of the imaging and projecting lenses over their affect on the geometric parameter $\varnothing$ is crucial. Evermore so since embodied within $\varnothing$ are the experimental requirements of resolution and field-of-view. Furthermore, control and flexibility over the illumination system is necessary to obtain the most coherent and parallel illumination possible with sufficient beam intensity. Another point made from Eqns. [2 & 3] is that an explicit prescription to optimize the experimental parameters for recording good quality holograms is not very feasible, and that the hologram quality will generally depend upon the application. This makes an overall characterization and assessment of a particular microscope to perform electron holography somewhat abstract or vague, and commands specific demonstration of the microscope’s capabilities for a given application. Bearing in mind this last point, we present results of two electron holography studies with our JEM-3000F TEM having rather different demands on resolution and field-of-view.

3.3 High-Magnification Electron Holography

Our JEM-3000F is equipped for electron holography with a retractable biprism assembly consisting of a platinum wire $\leq 0.6$ nm in diameter. The biprism wire is rotatable $450^\circ$ and located approximately in the selected area aperture plane of the microscope. The multi-scan CCD camera located after the Gatan Image Filter (GIF) attached to the microscope was used to record holograms. (The second multi-scan CCD camera located before the GIF provides considerable flexibility in recording data, but was not used in the present work.) Figure 3 illustrates the performance of our holography setup in terms of holographic fringe contrast, fringe spacing and hologram width for one of the standard holography modes (300 kV accelerating voltage, 100 kX magnification) available with our machine, i.e., no free-lens adjustments made. Measurements were made from holograms recorded $\sim 70$ nm from the smallest spot size and largest condenser aperture generally giving the best available fringe contrast. Adjusting the emission characteristics of the gun (first and second anode voltages) improved the fringe contrast slightly, but not dramatically.

Figure 3a shows the holographic fringe spacing (which controls the resolution of the reconstructed image wave) as a function of voltage applied to the biprism. For example, 2 Å fringes (giving $\sim 6$ Å resolution [13]) requires about 75 V bias applied to the biprism. Figure 3b, then, shows the hologram width and fringe contrast as a function of fringe spacing (consequent to biprism voltage) for the standard 100 kX holography mode. The maximum fringe contrast width (open circles in Fig. 3b) defines the maximum field for holographic reconstruction, but that the actual field-of-view may be limited by the size of the CCD array. More critically for this standard mode, however, is that the fringe contrast (closed circles in Fig. 3b) for higher resolution application is comparatively low: a minimum contrast around 20% being preferred for reliable phase and amplitude reconstruction. For example, a reconstructed image wave with 3 Å resolution requires $\sim 1$ Å fringes, which gives only $\sim 10$ % fringe contrast with this mode. Considerably better quality holograms, however, can be obtained by adjusting the intermediate (I1, I2, and I3) and projector (P) lenses on the imaging system of the JEM-3000F through use of these..
the grain-boundary (upper plot in Fig. 4), one may directly obtain the associated charge distribution (lower plot in Fig. 4), to which structure one may relate physical properties of the interface such as electrical transport properties.

**Figure 5a** shows an electron hologram obtained from a (001) twist boundary in Bi-2212 where the grains have been tilted to a systematic row orientation to assure the interface is parallel to the electron beam. The fringe contrast in Fig. 5a is about 25% (as measured from the vacuum reference image, not shown) at the 70V biprism voltage used, and corresponds to a fringe spacing of 2.1 Å as measured after image calibration by the known lattice spacing of Bi-2212. Under these conditions, the field-of-view on the CCD camera is about 22.1 nm, allowing amplitude and phase profiles extending a few Bi-2212 c-axis unit-cell dimensions (3.1 nm) from the twist boundary to be easily obtained. **Figure 5b and 5c** show, respectively, the numeric reconstruction of the image amplitude and phase from the hologram shown in Fig. 5a. The grain boundary potential, shown in **Fig. 5d**, was directly obtained from the phase image by averaging parallel to the interface and assuming a uniform slope to the sample thickness over the region. As seen in the potential profile, Fig. 5d, the interface potential is on the order of less than a few nanometers in width (where the c-axis unit cell dimension is 3.1 nm), and has a magnitude about 0.8 V. This result is consistent with

![Fig. 3. Summary of JEM-3000F performance to record holograms using standard holography mode at 100kX magnification.](image)

![Fig. 4. Schematic grain-boundary potential, \( V \), and corresponding charge density, \( \rho \), illustrating how recovery of phase information may provide insight to physical properties of grain-bounds as the potential is directly related to reconstructed phase in electron holography. Here, \( \varepsilon_0 \) is the permittivity of the material.](image)

![Fig. 5. (a) Electron hologram of pure (001) twist boundary in Bi-2212 where 2Å holographic interference fringes run nearly perpendicular to the grain-boundary (GB). (b) Reconstructed image amplitude and (c) phase from the hologram shown in (a). (d) Potential obtained from profile of phase image along line AB in (c). Note that the periodic drops in the potential profile correspond to the BiO double-layer positions, which are also clearly seen in (a-c).](image)

![Fig. 6. (a) Experimental shadow-image diffraction (PARODI) pattern and corresponding intensity profile (above) from c-axis systematic row oriented Bi-2212 wedge. (b) Enlarged 0014 experimental disk rotated ~20° counter-clockwise to compare with (c) calculated 0014 disk. (d) Wedge thickness determined from EELS. (e) Measured c-axis charge density over Bi-2212 unit-cell obtained from analysis of similar patterns shown in (a). Measured charge transfer (solid line) with reference to formal valence is shown at the bottom of (e) and compared with first-principle calculations (dashed line).](image)
analysis of holograms obtained from similar pure-phase Bi-2212 (001) twist boundaries where values ranging from about 0.5 to 2 \text{\textdeg} have been found. The dependence of measured grain-boundary potential with twist angle is under investigation, as well as a study of twist boundaries containing local ‘impurity phases’ of Bi-2223 and Bi-2201 where significant charge transfer is observed. These preliminary holography results compliment results obtained from similar boundaries using interferometric shadow-imaging diffraction methods, examples of which we present next.

3.2 Coherent Electron Diffraction
Quantitative convergent-beam electron diffraction (CBED) studies can reveal important information about bonding and ionicity for small unit-cell crystals [14-17]. Conventional CBED techniques, however, are not suitable to study of crystals with a large unit-cell such as high-temperature superconductors, partly because the wide range of incident beam directions prevents the recall of diffraction disks. We have developed a new technique by introducing an additional dimension, thickness, into the diffraction pattern and focusing on the diffraction intensity variation not only as a function of excitation error, but also as a function of crystal thickness [18]. This technique is compared to corresponding CBED methods (which focus the probe on the sample) in that we focus the electron probe above or below the sample. In doing so, we form shadow-images (dark-field images) of a large illuminated area within the diffraction-disks that contain not only orientation information (as with conventional CBED), but thickness, profiles, or Pd differential fringes, as well as for the many simultaneously recorded reflections. Since this method obtains dark-field images within each diffraction disk for many reflections in a single exposure (parallel recording), we term our method parallel recording of dark-field images, or PARODI.

Our PARODI method offers some distinct advantages over other methods. Firstly, dozens of reflections (for large unit-cell crystals) are recorded simultaneously, which ensures that the incident-beam illumination and exposure are exactly the same for all reflections. Secondly, the dark-field images are recorded in the back-focal plane of the objective lens where the objective lens aberration and transfer function can be neglected for quantitative analysis. Thirdly, for a wedge sample the discrete data points within each reflection (corresponding to different sample thickness) are independent which results in a high ratio of experimental observations versus the number of fitting parameters in model calculations giving correspondingly higher levels of confidence. Fourthly, interference fringes due to planar defects (e.g., interfaces) show strong contrast even when the defect is viewed edge-on. And, finally, a wide range of reflections can be simultaneously analyzed in quantitative measurement of charge distribution and lattice displacement since electrons scattered at small angles are sensitive to charge arrangement, whereas electrons scattered at high-angle are sensitive to crystal distortion.

Figure 6a is an experimental PARODI pattern, with a line scan of the reflection intensity shown above it, from a wedge Bi-2212 crystal in systematic (001) orientation using the Fuji Imaging Plate system with our JEM-3000F. The top-left portion of the shadow-images within each disk of Fig. 6a corresponds to vacuum, as easily seen in the 000 disk, and the sample thickness increases from left to right in each shadow-image. Figure 6b shows an enlarged view of the 0014 reflection from Fig. 6a (after ~20° anti-clockwise rotation), and is compared with the intensity shown in Fig. 6c obtained from dynamical Bloch-wave calculations. The intensity oscillation in Fig. 6c originates from the thickness increase of the sample, as shown in Fig. 6d obtained from electron energy-loss spectroscopy. By quantitatively refining model calculations against the experimental patterns obtained by our PARODI method, we can determine the structure factors for low-order reflections very accurately, which can then be used to determine the charge distribution within the crystal. For example, the movement of 0.05 electron holes per unit-cell of YBa2Cu3O6 superconductor between the Bi2212 and CuO planes corresponds to rearranging 1 out of 5000 electrons in the crystal, changes the 001 structure factor by 0.1nm for electron scattering. Correspondingly, by our PARODI method, we can determine this structure factor with an accuracy of 0.09 nm [19]. Figure 6e shows the measured charge density in Bi-2212 overlaid on one unit-cell dimension, where the lattice planes are labeled above the charge density peaks. Below the charge density plot in Fig. 6e are difference profiles (using a formal valence model as reference) showing the charge transfer, \( \Delta \), along the Bi-2212 c-axis unit-cell direction based on our measurements (solid line) and on first principles calculations. These experimental results indicate excess valence electrons associated with the BiO double layers, which is consistent with HRTEM observation [1, 20], and suggests a possible covalent nature to the BiO-BiO bond since such a high charge transfer is observed at the interstitial region of the double-layer. The charge transfer indicates that the charge density and valence distributions in the CuO and SrO planes are nearly equal while Ca is almost completely ionic with negligible charge transfer.

An exciting extension of our PARODI method is to the study of faulted crystals to accurately measure interfacial lattice displacement with a coherent source. Since the method is based on quantitative analysis of shadow images produced by coherent electron diffraction, it is an interferometric technique where the spatial resolution of the measurement is not limited by the wavelength of the fast electrons. Accuracy down to 1pm has been demonstrated in measuring displacements associated with stacking faults and grain boundaries in Bi-2212 [21]. Due to the highly coherent electron source, strong intensity oscillations parallel to the fault are present in the PARODI patterns when a planar fault exists within the shadow-image field-of-view. As an illustrative example, experimental and simulated shadow-images are shown in Fig. 7 (upper row and lower row of disks, respectively) for a stacking fault consisting of an additional (Fig. 7a) or missing (Fig. 7b) (Ca+CuO) bilayer in Bi-2212. After quantitative model refinement against experimental data and careful error analysis, we arrive at displacement vectors \( R=0.320 \pm 0.002 \text{ mm} \) and \( R=0.319 \pm 0.001 \text{ mm} \) for the insertion or deletion, respectively, of a (Ca+CuO) bilayer in Bi-2212. These results are, to our knowledge, about a ten-fold improvement over any existing technique in determining local lattice displacements. Rigid body translations associated with (001) twist boundaries in Bi-2212 were also studied, where, for example, the lattice displacement across a near 13 boundary was determined to be \( R=0.026 \pm 0.004 \text{nm} \). The combination of quantitative electron holography to provide approximate models of potential or charge distribution for complicated interfacial structures, and the powerful refinement capabilities of the PARODI method described here, offer exciting new possibilities to unparalleled studies of grain-boundary interfaces in the high-temperature superconductors.

4. Magnetic Imaging of NdFeB Permanent Magnets
NdFeB magnets are currently the most powerful permanent magnets having found wide application from generators to motors and computer devices. The technological importance of NdFeB magnets is growing rapidly as their magnet properties are improved through chemistry and processing of this compound where its magnetic properties are known to be very sensitive to such microstructures as grain size, grain alignment and the presence of secondary phases [22, 23]. Nevertheless, the energy products thus far achieved for this highly anisotropic material are significantly lower than theoretical upper limits, and our current understanding of the dependence of magnetic structure on microstructure in hard magnets is still quite limited. Knowledge of how magnetic structure is related to microstructure within NdFeB permanent magnets may help address major issues of magnetism in magnetic materials such as an understanding of the role of domain wall depinning and reversed domain nucleation as factors that limit the coercivity of these hard magnets [24]. Light-field electron holography combined with in-situ field-calibrated magnetization and demagnetization Lorentz or Foucault microscopy provides a quantitative approach to examining these important issues.

4.1 Low-Magnification Electron Holography
The standard low-magnification holography mode (objective lens very weakly excited) for our JEM-3000F suffers somewhat poorer performance compared to its standard medium- or high-magnification modes, or compared to similar modes for commercial microscopes from other manufacturers. With low-magnification, holographic fringe contrasts 10 % or lower for bipism voltages around 60 V, which gives 19.1 nm interference fringes and 4.2 \textmu \text{m} hologram width. For this magnification, the field-of-view on our CCD cameras is 8.5 \textmu \text{m} and 0.6 \textmu \text{m}, respectively, for the cameras mounted above and below the GIF. The discussion of Section 1, hence, holds special pertinence here. Efforts to maximize the fringe contrast according to principles discussed above have, so far, met with only partial success. For example, we obtain 8-9 % fringe contrast at 60 V bipism bias in the standard low-magnification mode.

But, by increasing the first intermediate lens from 0.01 V to 2.72 V through free-lens control of the microscope, leaving all other lenses untouched, the fringe contrast is dramatically increased to 58%. This gain, however, is tempered by the fact that the objective mini-lens (OM) cannot be increased sufficiently to focus the sample, and appears to be the primary factor limiting the overall performance of our machine for low-magnification holography applications. Nevertheless, the performance of our microscope can be improved beyond that of the standard-mode lens settings by going over to free-lens control where, though a labor-intensive practice, sufficiently strong contrast can be obtained for particular applications as shown below.

Figure 8a is an electron hologram recorded from a sintered Nd$_2$Fe$_{14}$B permanent magnet, and Figs 8d and 8e are, respectively, the reconstructed amplitude and cosine of the phase images. The hologram shown in Fig. 8a was recorded with lens potentials: C1=3.42, OL=0.35, OM=6.25, I1=0.01, I2=4.60, I3=1.09, and P=3.20; and 60.2 V biprism bias. The fringe contrast (in vacuum) was 16% under these conditions giving 10.8 nm fringes over a 1.46 µm field-of-view on the CCD camera where the hologram covered the entire CCD array. Domain boundaries (marked by arrows in Fig. 8c) appear as discontinuities in the gradient of the phase image. Figure 8d shows a vector map of the calculated phase-gradient, which, neglecting contributions from sample thickness and electrostatic potentials in the sample, represents the projected magnetic induction. (Even accounting for contributions due to sample thickness and electrostatic potentials, one must be aware that the component of magnetic field parallel to the beam does not contribute to the final induction map so that interpretation must be performed with caution.) While being grossly oversimplified, Fig. 8d shows qualitatively the 180°-domain configuration (as thick arrows) present in the sample, and illustrates the potential for our microscope to perform quantitative magnetic studies. The quantitative potentiality of electron holographic methods, combined with more conventional, yet mainly qualitative, magnetic imaging techniques of Lorentz and/or Foucault imaging, discussed next, represents a powerful combination of capabilities of our JEM-3000F to the study of magnetic materials.

4.2 In-Situ Magnetic Imaging
To understand the relation between structure and properties of magnetic materials, it is crucial to be able to magnetize or demagnetize the sample in the microscope so that dynamic behavior of magnetic domain structure, such as orientation dependence and interaction with defects, can be studied in-situ. For magnetic imaging, samples are normally examined with the main objective lens (OL) of the microscope either switched off or slightly exited. The latter case is used for in-situ magnetizing experiments. Exact knowledge of the magnetic field excited in the specimen area of the electron microscope as a function of the OL potential allows one to take qualitative imaging of magnetic domain behavior obtained in the Lorentz and/or Foucault modes to a new level. For example, quantitative description of the evolution of the local domain structure versus the applied field becomes possible, as well as the ability to evaluate processes of magnetization and demagnetization. This may, for example, allow differentiation of the underlying mechanisms between reversed-domain nucleation or domain pinning that determines the coercivity of magnetic materials. To this end, the magnetic field in the sample area versus objective lens potential for our JEM-3000F was carefully calibrated by direct Hall probe measurements using a BELL610 gaussmeter. As plotted in Fig. 9, the magnetic field at the sample area can be varied from 0.02 to 3 Tesla. Table 1, then, shows the magnetic fields present at the sample area corresponding to various standard operating modes of our microscope, including the “Holo Low Mag” mode used for electron holography and conventional “Low Mag” mode used for Lorentz and Foucault imaging.

As an example of the utility of the approach described above, we elucidate the evolution of domain nucleation near grain boundaries of sintered Nd$_2$Fe$_{14}$B magnets, as recorded in the Foucault mode [26]. In the Foucault imaging mode, an aperture is placed slightly off-center from the transmitted beam in the back-focal plane of the imaging lens, i.e., objective mini-

![Fig. 7. Comparison of experimental and calculated 008, 0010 and 0012 shadow-image disks for a stacking fault in Bi-2212 consisting of an (a) extra and (b) missing (Ca+CuO) bilayer. The measured lattice expansion corresponding to insertion is R=0.320±0.002 nm, while for the depletion is R=0.319±0.001 nm.](image)

![Fig. 8. (a) Electron hologram of Nd$_2$Fe$_{14}$B permanent magnet. (b) Reconstructed image amplitude. (c) Cosine of reconstructed phase. Arrows indicate presence of domain boundaries in material. (d) Calculated approximation to projected induction vector map. Thick arrows indicate trend of induction lines suggesting 180°-domain configuration.](image)

![Fig. 9. Magnetic field of objective lens for JEM-3000F as a function of lens potential (excitation) calibrated by direct Hall probe measurements using BELL610 gaussmeter.](image)

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<th>300kV Accelerating Voltage</th>
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<tr>
<td>8000X-1500kX</td>
<td>4.32</td>
<td>27.654</td>
</tr>
</tbody>
</table>

Table 1. Magnetic field at specimen area for JEM-3000F field-emission TEM.
lens of the JEM-3000F. Oppositely oriented domains (in projection), then, appear correspondingly bright or dark in the image depending, respectively, upon whether the magnetic (Lorentz) scattering is into or out of the aperture. Figure 10 shows the domain configuration present in an 84° grain-boundary (GB) region of a sintered NdFeB magnet for two different magnetic fields (3.3 kOe and 2.1 kOe corresponding to Fig. 10a and 10b, respectively) applied normal to the image-plane via the calibrated OL excitation. Due to its crystallographic orientation at 3.3 kOe, the left grain (G1) is near magnetization saturation (single-domain configuration) with a small nucleus of reverse-magnetization present, as shown in Fig. 10a. Upon further reduction of the applied magnetic field to 2.1 kOe, the reverse-magnetization nucleus abruptly expands to encompass the grain with equal width of reversal domains, as shown in Fig. 10b, suggesting grain G1 has reached a demagnetized state. In contrast, the right grain (G2) remains nearly saturated. The magnetization within grain boundaries is further elucidated in Fig. 11 where the applied magnetic field (via the OL excitation) has been decreased to zero and, in fact, reversed. (The ability to reverse the OL lens current (hence, magnetic field) is a non-standard feature installed with our JEM-3000F.) Reversal of domain nucleation near grain boundaries is clearly shown in Fig. 11b wherein in-situ imaging reveals that at zero magnetic field the grain boundaries are relatively parallel to each other while at low magnetic field the grain boundaries are relatively parallel to each other while at low magnetic field. This experiment also demonstrates the ability to control the magnetization of our JEM-3000F. Figure 12b is the corresponding boron map of the same area. Figure 12b shows an increase in boron signal in regions corresponding to positions of the TMV. Extending the above calculations and analysis, we estimate the number of boron atoms per unit area in Fig. 12b to be approximately 10^10 boron atoms per pixel (with a pixel size of 1nm²) in the vicinity of the tobacco mosaic viruses.

6. Conclusions

We have illustrated some of the more advanced techniques to materials characterization that our JEM-3000F field-emission TEM is capable of. Using coherent diffraction techniques newly developed in our laboratory (PARODI), we measure lattice displacements across grain-boundaries in high-temperature superconductors with picometer accuracy, as well as potential variations and charge transfer on sub-nanometer scales. We have taken in-situ magnetization measurements using conventional Lorentz and Foucault imaging modes to new quantitative levels by careful calibration of the magnetic field applied to the sample area as a function of objective lens excitation. We have analyzed the detection limits of our spectroscopic capabilities and illustrated the sensitivity of energy-filtered imaging to detection of light elements. We have also demonstrated the ability to carry out both medium- to high-resolution electron holography studies of superconductor grain-boundaries and low-magnification holography studies of permanent magnets with our JEM-3000F.

Since all of the techniques described above require a highly coherent electron source and good high-tension stability, we may conclude that our JEM-3000F exhibits such qualities. Furthermore, the performance of our machine does not appear hindered by the multitude of various attached detectors and complex integration of experimental capabilities. For holography applications, the ability to freely control all the lens settings was found to be crucial in recording high quality holograms as the programmed settings give poor interference fringe contrast in the holograms. The strength of the objective mini-lens seems to be the limiting factor in low-magnification magnetic applications, while the strength of the condenser stigmators appears to hinder the ultimate capability of our microscope for higher-magnification applications. Nevertheless, combined with the advanced techniques being performed in our laboratory, the demonstration to perform electron holography studies adds another dimen-
sion to the already uniquely powerful capabilities of our TEM instrumentation to the study of advanced materials.

7. Acknowledgements

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References


Fig. 10. Foucault images of domain configuration at 84° grain-boundary (GB) in Nd₃Fe₁₄B sample recorded at (a) 3.3kOe and (b) 2.1kOe fields applied normal to image plane via calibrated objective lens excitation. A small reverse-magnetization domain (circled) is present in grain G1 of (a) which expands to encompass the grain interior as the applied field is further lowered, as shown in (b).

Fig. 11. (a) Successive frames (1-6) of reverse-domain nucleation process near grain-boundary (GB) as recorded at 24 frames/sec in Foucault imaging mode during decrease and reversal of applied magnetic field normal to image plane. (b) Enlarged from circled regions of frames 4-6 showing domain splitting.

Fig. 12. (a) Zero-loss energy-filtered image of tobacco mosaic virus (TMV) and (b) corresponding energy-filtered boron map showing the concentration of boron associated with the virus.

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