agreement with the experimental value of 175 kV. Measurements at lower power were consistent with $P^{2/3}$ scaling, suggesting the usefulness of Eq. (4) as a scaling relation.

We conclude that the observed heating of the target's support structure at large distances is dominantly the Ohmic heating caused by the return electron current. The description of the hot-electron–driven return current given above also implies that the electrons on the high-energy tail of the Maxwellian electron distribution are emitted, thereby modifying the distribution function in the plasma. These reported measurements should be useful in modifying computer simulations that presently lack net return-current flow. The magnitude of the net current flow is large enough to affect magnetic field distributions that have been reported in laser-heated targets.

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Extended Fine Structure on the Carbon Core-Ionization Edge Obtained from Nanometer-Sized Areas with Electron-Energy-Loss Spectroscopy

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Extended fine structure, equivalent to extended x-ray–absorption fine structure, has been observed on carbon K-ionization edges obtained with electron energy-loss spectroscopy utilizing 0.75- to 5-nm radius probe sizes, and sampling as few as 10^4 atoms in times as short as 4 min. Radial distribution functions show the expected behavior for graphite and an amorphous carbon sample. A second "amorphous" sample, however, shows structure which suggests the existence of local tetrahedral coordination.

There has been interest recently in the use of the inelastic scattering of fast electrons (electron-energy-loss spectroscopy or EELS) as a probe of the inner core excitations of materials to determine electronic band structure[1,2] and area-averaged structural information from extended electron-energy-loss fine structure (EXELFS)[3-6] equivalent to extended x-ray–absorption fine structure (EXAFS) in the photon absorption experiment. In addition, it has been re-

ported that EXAFS is obtainable from as few as 10^{13} atoms on a surface by use of Auger transitions excited by synchrotron radiation.[7] We report in this Letter the presence of identifiable EXELFS from as few as 10^4 atoms with 0.75- to 5-nm probe radii. These data were obtained in times as short as 4 min with a field-emission, electron-gun–equipped, scanning transmission electron microscope (STEM).

The cross section for inelastic scattering of electrons from a specimen is written in the dipole approximation as

$$\partial^2\sigma/\partial\Omega\partial\omega \propto q^{-4}\sum_f |\langle f|\vec{q}\cdot\vec{r}|i\rangle|^2 \times \delta(\omega-\omega_f-\omega_i), \quad (1)$$

where $\hbar\omega$ (or ΔE) is the energy lost by fast electrons scattered with a change of wave vector $\hbar\vec{q}$ in the process of producing a change of state from $|i\rangle$ to $|f\rangle$.[8] This representation is adequate for $q<1$ Å^{-1} ($r<0.3$ Å for the carbon 1s electron), corresponding to a scattering angle of 7 mrad for electrons of incident energy $E_e=80$ keV. The half width ($\equiv\Delta E/2E_e$)[9] for 300-eV losses is about 1.8 mrad and therefore about 70% of the core signal satisfies this. EXAFS results[10] from a change in $|f\rangle$ when ejected core electrons are backscattered towards the origin by near-neighbor atoms. The EXAFS modulation for a one-component material is approximately given by

$$\chi(k)\approx\frac{A(k)}{k}\sum_j\frac{N_j}{R_j^2}\mathrm{Im}\{\exp(i2kR_j)\exp[i\varphi(k)]\}, \quad (2)$$

where k is the wave vector of the ejected core electron; N_j is the number of atoms in the jth-neighbor shell; R_j is the shell distance; $\varphi(k)$ is the phase shift suffered by the ejected electron during the process of ejection and backscattering; and $A(k)$ is the backscattering amplitude.

The present work was done at the Cavendish Laboratory on a VG Microscopes, Ltd., model HB5 STEM equipped with a high-brightness field-emission electron source.[11] This instrument produces a 0.3- to 5-nm radius probe of 80-keV electrons with a probe convergence half angle of 1.5 to 16 mrad. Therefore cross-section integration is usually done by the incident probe angle rather than by a collector angle. Current into this probe ranges from about 10 nA at 5 nm down to ~0.3 nA at 0.3 nm. The specimens were self-supporting and about 10 nm thick. The number of atoms routinely sampled therefore lies between 10^3 and 10^5 rather than 10^{13} and 10^{19} as is the case in instruments with millimeter-sized beams. This illustrates the point made recently[12] that for situations in which very small structures would benefit from an analysis of EXAFS, an electron scattering experiment with a field-emission source will provide substantially better performance than photon absorption experiments using synchrotron radiation.

Data from 10-nm thick Ticonderoga graphite were obtained with a 0.75-nm radius probe subtending a half angle of 8 mrad (1.2 Å^{-1}). The electron spectrometer was adjusted for 3-eV energy-loss resolution which necessitates a maximum collection angle of 0.6 mrad. Fifty energy-loss points were taken between 270 and 400 eV during a 45-min period. The average count per channel was 2500, producing a statistical accuracy of 2%. Data from the 10-nm amorphous carbon samples were obtained from a 5-nm radius probe subtending a half angle of 16 mrad (2.4 Å^{-1}). The spectrometer collection half angle was 1.0 mrad producing an 8-eV resolution. One hundred points were measured between 250 and 600 eV in a 4-min period. The average number of events per channel was about 300.

The raw-data result for graphite is shown in Fig. 1(a). The K-edge onset occurs at 285 eV for core to π-band transitions,[6] and core to σ-

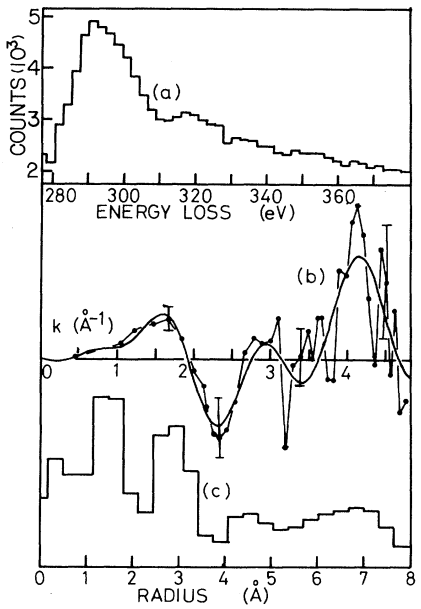

FIG. 1. (a) Raw data for graphite K core excitation. (b) Residual EXELFS intensity weighted by k^2. The overlay includes frequencies only below 3.5 Å in the RDF. Error bars include statistical and multiple-scattering considerations. (c) Radial distribution function.

band transitions occur near 292 eV. A peak near 318 eV is likely a composite of the first EXELFS peak and multiple-scattering intensity involving a core to σ-band transition followed by $\sigma+\pi$ plasmons of about 27 eV.[13] Examination of the plasmon intensity in the region below 50 eV indicates that for this sample, up to 50% of the K core intensity near 318 eV is due to this multiple scattering. Finally, the remaining fluctuations are attributed to EXELFS. A detailed comparison with the low-angle result of Kincaid, Meixner, and Platzman[6] is not possible here, because the present result is an angular integration of the scattered intensity by the incident probe. Equation (1) indicates that EXELFS may vary with angle, and preliminary work by us appears to confirm this view. Therefore we would not expect to be able to compare this result with Ref. 6 without obtaining the differential intensity at various scattering angles with a small angular resolution.

We have extracted the EXELFS modulation from the data by a least-squares fitting and subtraction of an edge structure quadratic in ΔE over regions large enough to avoid eliminating EXELFS modulations corresponding to radii above 0.8 Å. We show Fig. 1(b) the residual intensity multiplied by k^2 to compensate partially for the term $A(k)/k$ in Eq. (2). The magnitude of the Fourier transform [radial distribution function (RDF)] of this intensity, after correction for $\varphi(k)$,[14] is shown in Fig. 1(c). This shows the near-neighbor spacing of 1.4 Å, and a composite of the 2.5- and 2.8-Å spacings because of the poor spatial resolution of about 0.7 Å necessitated by the rather narrow range of k space considered (about 1.5 to 5 $Å^{-1}$). The error bars are largely related to curve a by Poisson statistics. However in the region 2 to 3 $Å^{-1}$ they have been increased to include the residual which is obtained when the above-mentioned multiple-scattering effects are approximately included. The ratio of area of the second peak to the first is $0.9 \pm 20\%$, agreeing with the expected value of 0.9 obtained by calculating $N_j/R_j{}^2$ for the first three neighbor shells in graphite (three neighbors at 1.4 Å, six at 2.5 Å, and three at 2.8 Å). We note that additional intensity due to the multiple scattering mentioned above would appear near 1.1 Å and therefore may be the source of the larger background intensity in this region.

In Fig. 2(a), we show the K-edge intensity (weighted by ΔE^3 to emphasize the EXELFS modulations) for an amorphous carbon sample which

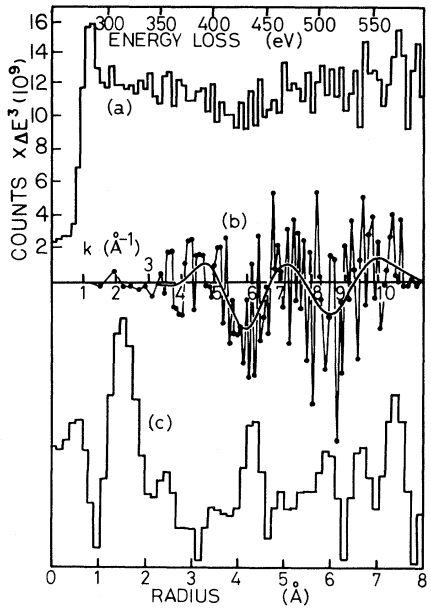

FIG. 2. (a) Raw data for the carbon sample which was evaporated onto mica. The intensities are weighted by ΔE^3, (b) EXELFS residual weighted by k^2. The overlay consists of frequencies below 2 Å. (c) Radial distribution function.

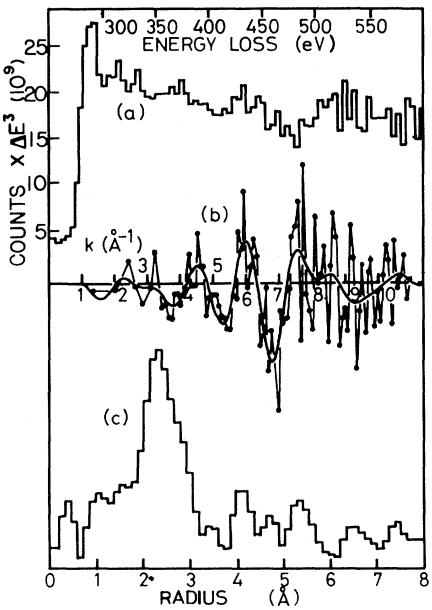

FIG. 3. (a) Raw data for the carbon sample which was evaporated onto KCl. The intensities are weighted by ΔE^3. (b) EXELFS residual weighted by k^2. The overlay consists of frequencies only below 3 Å. (c) Radial distribution function.

was prepared by vacuum evaporation onto mica; 2(b) shows a very weak, but reproducible, modulation in the residual intensity; and finally, 2(c) shows that this modulation corresponds to a 1.5-Å spacing only, indicating that the atomic positions are indeed uncorrelated beyond their near neighbors.

In Fig. 3(a) we show the K-edge intensity for a carbon sample prepared by vacuum evaporation onto KCl. The EXELFS modulations, in 3(b), are more prominent and are clearly of a higher frequency than those in Fig. 2. Figure 3(c), the RDF, shows a strong 2.5-Å peak with a shoulder near 1.4 Å. A possible explanation for this can be found by considering the distribution expected for diamond (four neighbors at 1.4 Å, twelve at 2.5 Å, and twelve at 2.9 Å). Combining the second two shells into one peak, we find two resultant peaks with an area ratio of about 1.7. Figure 3(c) implies a value between 2 and 4, depending on assumptions about background intensity and peak width. The experiment therefore favors a diamondlike, rather than a graphitelike, basis for a structural model of this particular sample. This apparent structural difference between the two amorphous carbon samples is supported by an elastic electron-diffraction experiment performed on the STEM, and by high-resolution examination by electron microscopy. It should be cautioned, however, that the difference in substrate may have been only coincidental, since other evaporation parameters are unknown for these two samples.

These experiments provide clear evidence that EXELFS intensity from very small volumes is potentially useful as a structural tool. Presently, however, two major problems limit the accuracy obtainable. First, for these particular examples, contamination by carbon deposition during the experiment time severely obscures the EXELFS signal. This was the primary limit on the counting time. For core signals reasonably well separated from the carbon edge, this will not be a problem and recording times approaching several hours will be possible. Secondly, the efficiency of the present experimental apparatus is limited. For instance, in the graphite experiment, the incident probe subtended a half angle of 8 mrad. This intensity is roughly isotropic and, combined with the 1.8-mrad characteristic half width of the electron-energy-loss scattering, produces isotropic scattering out to 8 or 9 mrad at the spectrometer entrance aperture. However, the spectrometer collection half angle for 3-eV resolution is only 0.6 mrad. Therefore, only 0.6% of the available intensity was collected. Work by Crewe, Isaacson, and Johnson[15] has shown that this collection angle, at least in one dimension, is expandable to >20 mrad. Work is presently underway, therefore, on new spectrometer schemes and on prespectrometer lens systems to overcome this problem. We fully expect these efforts to produce collection efficiencies close to unity.

In conclusion, we have shown that structural information via an analysis of EXELFS is obtainable from very small volumes (~30 nm^3) and very small numbers of atoms (~10^4). This result confirms predictions[12] of the usefulness of field-emission source, electron-scattering experiments. With proper control of electron trajectories into electron spectrometers to increase their collection efficiency, we expect that routine EXELFS analysis will be possible with data from small volumes, surfaces, and interfaces within thin samples. This should provide considerable new information about problems of current interest such as the segregation into precipitates of light elements in structural alloys, and the presence of impurities at interfaces within semiconductors and at semiconductor-to-metal contacts.

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Experimental Test of the Extended-Scaling Hypothesis in the Spin-Flop System $CsMnBr_3 \cdot 2D_2O$

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This neutron-scattering study deals with the behavior of the order parameters $M_{st}^{\parallel}(H,T)$ and $M_{st}^{\perp}(H,T)$ in $CsMnBr_3 \cdot 2D_2O$. The data provide a first direct test of the extended-scaling theory near the bicritical point of a spin-flop system.

The (H,T) phase diagram for an antiferromagnet with weak orthorhombic spin anisotropy contains a bicritical point,[1] where two distinct types of critical behavior are simultaneously present, namely an ordering of the magnetic moments parallel to the easy axis in the antiferromagnetic (AF) phase, as well as an ordering perpendicular to it in the spin-flop (SF) phase. As the bicritical point is approached along either of the two critical lines separating the AF and SF phases from the paramagnetic (P) phase, an abrupt crossover is expected from Ising-like critical behavior (number of relevant spin components $n=1$) to XY-like bicritical behavior ($n=2$).[2] The crossover can be described by a scaling theory first introduced by Riedel and Wegner[3] and subsequently developed further by a so-called extended-scaling hypothesis.[4] On the basis of this hypothesis, which may be formulated[3,4] as the postulate that the Gibbs free energy and its derivatives are generalized homogeneous functions, the shape of the phase boundaries can also be predicted.

In the last few years, experimental studies on a number of spin-flop systems have been undertaken to verify the extended-scaling hypothesis and its implications. However, the results so far do not extend further than an accurate determination of the shape of the phase boundaries and a comparison of these data with the theoretical predictions.[5-7] Here we shall report the results of an extensive scaling analysis of the variation of the order parameters in the AF and SF phases, i.e., the components of the staggered magnetization parallel ($M_{st}^{\parallel}$) and perpendicular ($M_{st}^{\perp}$) to the easy axis, respectively, near the bicritical point in $CsMnBr_3 \cdot 2D_2O$ (CMB). To our knowledge such a direct experimental verification of the extended-scaling hypothesis for spin-flop systems has not been reported earlier.

CMB is a pseudo-one-dimensional ($d=1$) Heisenberg antiferromagnet.[8,9] The crystal structure is orthorhombic with space group $Pcca$. Below $T_N \simeq 6.30$ K the symmetry of the AF ordering is described by the magnetic space group $Pc'c'a'$.[10]

