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Measuring the Hole State Anisotropy in MgB_2 by High-Resolution Angular-Resolved Electron Energy- Loss Spectroscopy

R.F. Klie[1], P.D. Nellist[2], Y. Zhu[1], J.W. Davenport[1] and J. Tafto[3]

[1] Brookhaven National Laboratory, Dept. of Materials Science, Upton, NY 11973
[2] Nion Co, Kirkland, WA 98033
[3] University of Oslo, Department of Physics, Blindern, 0316 Oslo, Norway

Here, we present a high-resolution study of the anisotropy of empty p-states near the Fermi-level in MgB_2 by angular-resolved EELS. DFT calculations [1, 2] suggested that the p_{xy} states, in the highly anisotropic compound, have a high density up to 0.8 eV above the Fermi level before the density of states drops to near zero and then starts to rise again about 5 eV above the Fermi level. These incompletely filled p_{xy} states at the Fermi level onset are believed to play an important role in the superconductivity of this two-dimensional electron-hole BCS superconductor.

The EELS results have been acquired using the JEOL3010F STEM/TEM, equipped with a 300 keV field emission gun and a post column GIF. The microscope and spectrometer were setup for “parallel illumination” with a convergence angle of α =0.7mrad and varying collection angles. Figure 1 shows the B K-edge with the incident beam parallel (a) and perpendicular (b) to the c-axis for three different effective collection angles centered at the forward direction. We note that the shoulder at the edge onset decreases with increasing collection angle in fig. 1a), whereas the trend is opposite in fig. 1b) with the incident beam normal to the c-axis. These features can be understood by considering the momentum transfer selection of the spectrometer entrance aperture, and the DFT calculations of the B K-edge[1]. These show that the p_z states have a nearly uniform and rather high density of states at the Fermi level, while the p_{xy} states within this energy range have the narrow high peak confined to an energy window of 0.8 eV at the Fermi level. Larger orientation selectivity can be achieved by displacing the incident beam so that we effectively collect electrons that are scattered at least 0.5mrad. Figure 2a) shows two spectra that fulfill these experimental conditions. Spectrum a) is taken with the incident beam perpendicular to the c-axis, and the center of the entrance aperture is displaced by θ_D=1.2 mrad towards the (001) diffraction sort; the collection angle ranges θ_c=0.7mrad to θ_c=1.7mrad. The shape of the B K-edge exhibits a high intensity pre-peak that remains constant over the range of 5 eV, which is in agreement with the first-principle calculations. Spectrum b) shows the results of the displaced entrance aperture with the c-axis parallel to the incoming electron beam. With the displaced aperture more than 90 % of the total spectral contribution stems from the transitions into the p_{xy} states. We clearly see the pre-edge peak with a high intensity at the edge-onset and a subsequent drop in intensity, as predicted from the first-principle calculations. [2]

Figure 2b) shows an experimental spectrum acquired with the Nion VG HB501, a DSTEM, having a cold-field emission source operated at 100 keV with a nominal energy resolution of 0.4 eV. In addition, this microscope is equipped with a C_s-corrector [3] that enabled us to achieve a convergence angle of 22mrad and an electron-probe size of 1.4 Å. The spectrometer collection angle was about 30mrad. The main difference between this spectrum and previously published spectra from conventional STEM/TEM instruments, is that an additional shoulder in the B K-edge pre-peak can be clearly identified at 189 eV. The DFT results, averaged over all possible directions, and the two orientation-resolved components, all energy broadened by 0.5 eV are also shown. The

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comparison with the DFT simulation clearly shows that a splitting of the pre-peak is expected, due to the individual contributions from the p_{xy} and the p_z hole-states. The intensity at the edge-onset (186 eV) can be now identified as the sum of both the p_{xy} and the p_z states, whereas the second shoulder (189 eV) is caused only by transitions into the p_z states. [4]

We will show that the individual contributions of the Boron K-edge pre-peak can be separated by angular-resolved EELS and that the p_{xy}-peak at the Fermi level can be resolved directly. This will allow us to study the effects of dopants, point defects and grain boundaries on the local superconducting charge carrier concentration directly by EELS on the nanometer scale. [5]

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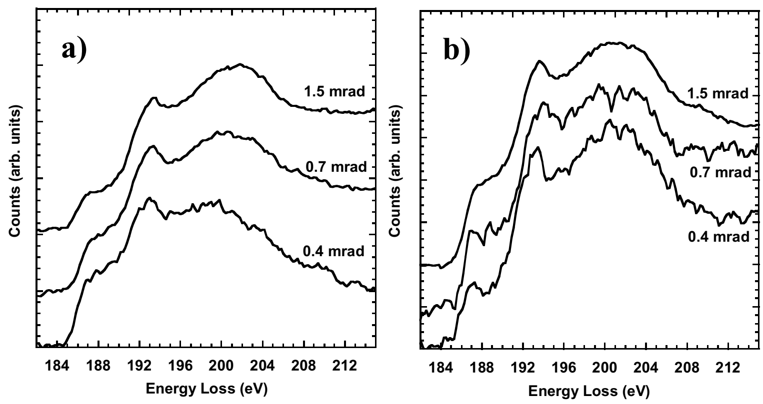

Figure 1: EEL spectra with the incident beam a) along and b) perpendicular to the c-axis with effective collection angles of 0.4, 0.7 and 1.5 mrad.

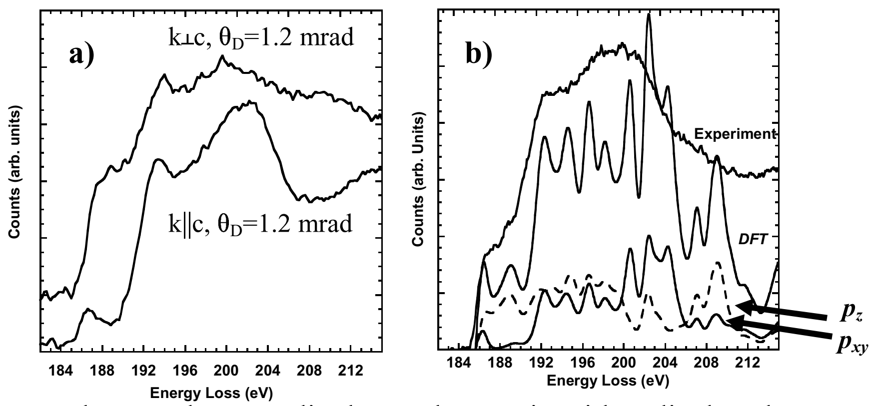

Figure 2: a) Spectrum along and perpendicular to the c-axis with a displaced entrance aperture by 1.2 mrad; b) experimental spectrum and *DFT* simulation averaged over all possible directions and the individual components.

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