1278
doi:10.1017/S1431927611007264
Microsc. Microanal. 17 (Suppl 2), 2011

Direct Imaging of Hydrogen Atoms in a Crystal by Annular Bright-Field STEM

E. Abe,* R. Ishikawa,* E. Okunishi,** H. Sawada,** Y. Kondo,** and F. Hosokawa**

*Department of Materials Science & Engineering, University of Tokyo, Tokyo 113-8656, Japan
**Electron Optics Division, JEOL Ltd., Tokyo 196-8558, Japan

Enhancing the imaging power of microscopy to identify all chemical types of atom, from low- to high-atomic-number elements, would significantly contribute for a direct determination of material structures. Scanning transmission electron microscopes (STEM) successfully provided images of heavy atom positions by annular dark-field method, but detection of light atoms was difficult owing to their weak scattering power. For detection of lighter atoms with extremely weak scattering, annular-bright-field (ABF) phase-contrast imaging may be preferred, since it requires the atoms only to alter a phase of wave. In fact, a significant sensitivity of ABF-STEM was demonstrated by imaging oxygen[1, 2] and nitrogen[1] atoms in crystalline solids, and further lithium atoms in a LiV_2O_4 compound[3]. In the present work, using ABF-STEM we demonstrate the direct imaging of the lightest hydrogen atoms in a crystalline solid[4].

The optimum conditions of ABF-STEM imaging can be derived based on phase-contrast transfer function (PCTF) of the hollow-cone illumination (HCI) TEM. Reciprocity supports the equivalence between the techniques (Fig. 1) and practically holds within a weak-phase approximation. The HCI is able to improve significantly the resolution as well as signal-to-noise ratio of a phase-contrast by eliminating the effect of wavelength fluctuations of the incident beam, i.e., a chromatic aberration (C_c) that causes the focus instability. It was also shown that, compared with the axial illumination, the achievable resolution of HCI can be almost doubled with the optimized conditions derived by PCTF analysis. The fundamentals of HCI-related TEM/STEM imaging were established as the Rose-Cowley method by their pioneering works. Practically, ABF-STEM may be advantageous rather than dynamic HCI-TEM, which requires continuous and precise beam-locking illuminations.

Optimized HCI condition (11mrad $\leq \theta_c \leq$ 22mrad, θ_c : cone-angle) derived from the C_s-corrected microscope parameters (JEM-ARM200F, operated at 200kV) confirms that the first-zero of PCTF can be extended into 22.5nm^{-1}, which corresponds to a spatial resolution of about 44.4 pm[4]. This is readily realized as the ABF-STEM condition with 11mrad $\leq \theta_d \leq$ 22mrad (θ_d : detector-angle), leading to a successful imaging of the hydrogen atom columns in a crystalline compound YH_2 (Fig. 2). Similar observation has been also demonstrated for the VH_2 hydride[5].

References

[1] E. Okunishi et al., *Microsc. Microanal.* 15 (2009) 164.
[2] S.D. Findlay et al., *Appl. Phys. Lett.* 95 (2009) 191913.
[3] Y. Oshima et al., *J. Electron Microsc.* 59 (2010) 457.
[4] R. Ishikawa et al., *Nature Materials* doi:10.1038/nmat2957 (2011).
[5] S.D. Findlay et al., *Appl. Phys. Express* 3 (2010) 116603.
[6] This work is partly supported by a Grant-in-Aid for Scientific Research on Priority Areas "Atomic Scale Modification'' from the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

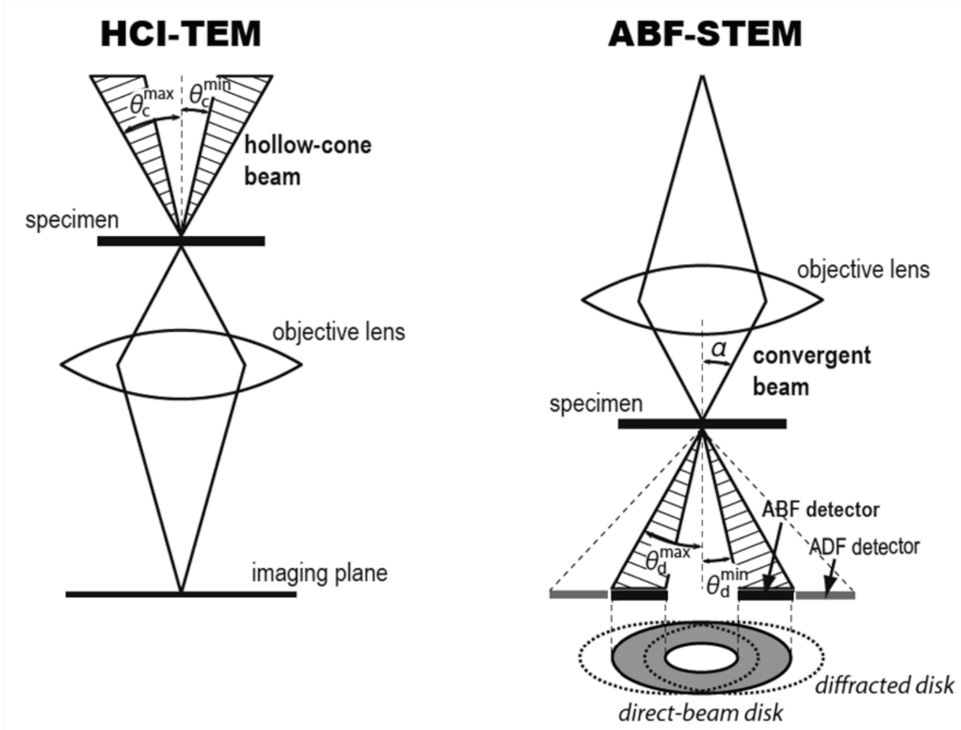

FIG. 1. Schematic ray diagrams of HCI-TEM and ABF-STEM according to reciprocity.

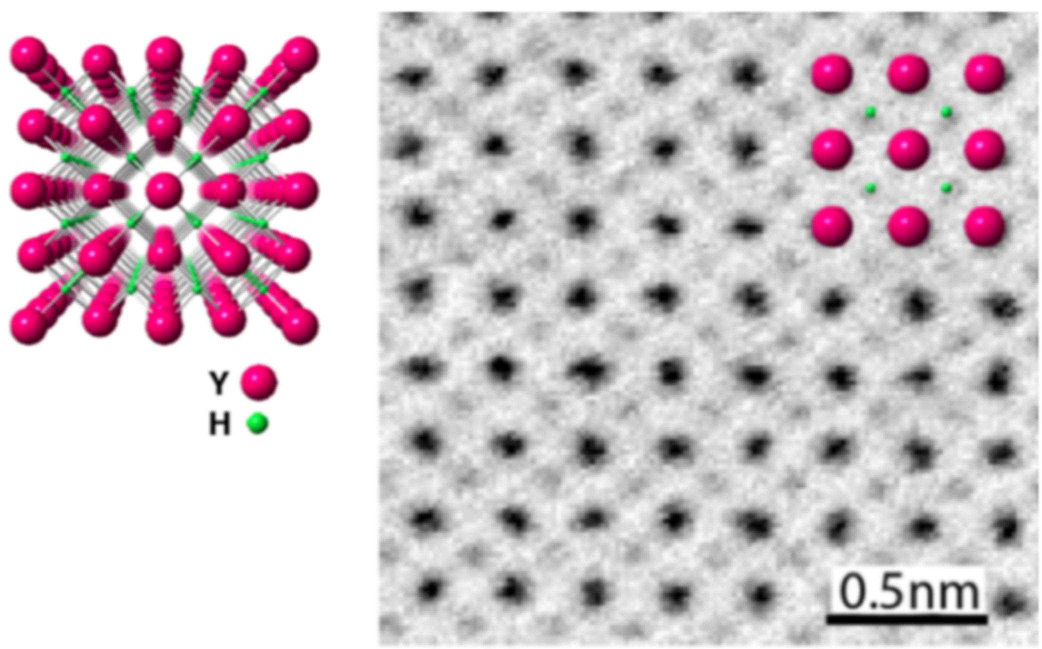

FIG. 2. ABF -STEM image of the crystalline compound YH_2, viewed from the [010] direction. YH_2 unit-cell projection is inserted in the image.