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Structural and chemical analysis on doped ceramics by transmission electron microscopy

In complex oxides, metallic and oxygen atoms occupy several crystallographically non-equivalent sites, on which the electronic states of the impurities are strongly sensitive. By employing wave-front reconstruction techniques on high-resolution TEM images taken under Cs-corrected conditions and Z-contrasts in atomically resolved STEM, we can directly clarify the favorable sites for dopant atoms.

Understanding the optical properties of materials requires a detailed investigation of their microstructure, especially regarding the exact location of the required dopants (e.g. rare earth elements such as Ce^{3+}, Nd^{3+} or Yb^{3+}). For example, it was recently shown by careful Transmission Electron Microscopy (TEM) observations that Ce strongly segregates at grain-boundaries within YAG (Yttrium Aluminium Garnet $Y_3Al_5O_{12}$) [1].

The oxide materials to be investigated in the current proposal includes YAG (yttrium-aluminum garnet) doped with RE (rare-earth), such as cerium or ytterbium, prepared either in a single or polycrystalline form in INSA-Lyon. We use FEI Titan 80-300 (S)TEM installed in Tohoku University as a major characterization tool (Fig.1), and the images taken will be analyzed quantitatively with a help of several simulation techniques, including the one developed in INSA-Lyon.

We have performed both High Resolution (HR) and HAADF imaging on pure YAG and Yb-doped YAG polycrystals, using a TITAN FEI electron microscope, operating at 300 kV. On the one hand, this instrument is equipped with a C_s aberration corrector on the objective lens, allowing a resolution down to 0.1 nm to be obtained in the HREM mode[2]. On the other hand, it is also equipped with an annular detector collecting electrons in a 70-210 mRad angular range. Nanoprobe chemical analysis was also simultaneously performed using an EDAX EDX (Energy-Dispersive X-ray) analyser mounted on the microscope.

Fig. 1 Cs corrector at the objective lens, which makes the resolution of phase contrast image better than 0.1 nm.

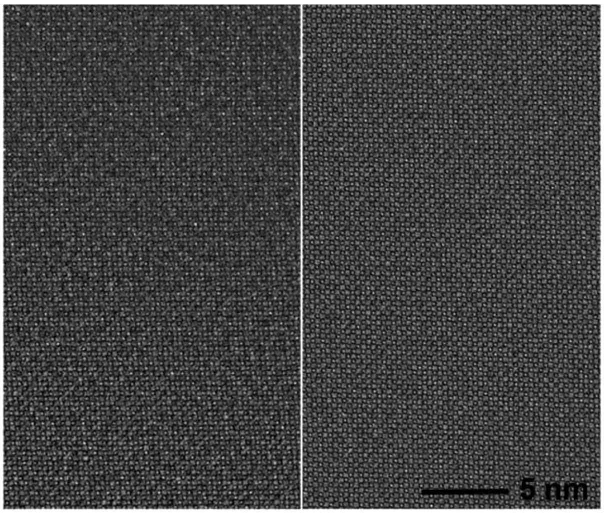

Fig. 2 HAADF-STEM images of two regions of similar thicknesses (as attested by low-loss EELS spectra) from a 1.4 at. % Yb-doped sample (left) and a pure YAG sample (right).

Whereas Cs-corrected HREM failed to reveal the distribution of Yb-containing atomic columns, the STEM-HAADF imaging mode appeared to be more efficient owing to its sensitivity to Z. Figure 1 shows a comparison of a 1.4 at.%Yb-doped and a pure YAG samples when observed along the [001] azimuth. A high density of brighter columns is observed for the doped material, which can be consistently analysed in terms of statistics as imaging Yb-containing atomic columns. Dedicated HAADF-STEM image simulations confirm this finding. This work shows that no segregation, neither clustering of Yb ions occurs in polycrystalline YAG.

To summarize, we have demonstrated that dopant distribution in oxide materials can be performed with aberration corrected scanning transmission electron microscopy.

References

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Key Words

Optical ceramics, Yb-doped YAG, atomic STEM-HAADF

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