Impurity Position in a Mn-Doped Bi_2Te_3 Topological Insulator Investigated by X-Ray Fluorescence Holography

Mn $K\alpha$ X-ray fluorescence holography (XFH) measurements were carried out on a single crystal of a $Bi_2Te_3Mn_{0.1}$ topological insulator at 100 K to search for impurity sites of Mn atoms. Three-dimensional atomic images were reconstructed using an L_1-regularized linear regression. The possible Mn impurity sites are twofold: an interlayer site with an octahedral symmetry and a substitutional site of Bi or Te. This finding for the impurity sites can be obtained only by using XFH measurements, not by the usual diffraction or XAFS experiments.

Since Laue's discovery of diffraction by crystals [1], this method has been the standard for structural characterizations of materials for a long time. Diffraction can clarify the long-range periodicity of averaged crystal arrays by using the interference phenomena of waves scattered by atoms. One of the serious disadvantages of diffraction for structural characterizations is the difficulty of determining impurity sites in a crystal due to the lack of periodicity for the impurity sites.

X-ray fluorescence holography (XFH) is a newly developed technique for atom-resolved structural characterizations, and enables three-dimensional (3D) atomic images to be drawn around a specific element emitting fluorescent X-rays [2]. Due to the interference between the direct incident X-rays (reference wave) and those scattered by the surrounding atoms (object wave), the fluorescent X-ray intensity slightly modulates with the angle of incident X-rays by some 0.1%, from which 3D images of the neighboring atoms can be obtained by simple Fourier-transform-like approaches without any special models. Thus, this technique can be used to find impurity sites in a crystal.

Bi_2Te_3 has recently attracted much attention as a topological insulator, which is obtained by adding transition metals such as Fe and Mn [3]. Undoped Bi_2Te_3 has a layer structure with a hexagonal form. Most researchers believe that the plausible site for Mn is in an interlayer position, but no experiments can exclude the interstitial sites or substitutional sites with the Bi or Te atoms. To answer this question, we carried out Mn $K\alpha$ XFH experiments on a single crystal of a $Bi_2Te_3Mn_{0.1}$ topological insulator to search for the impurity sites of Mn atoms [4]. The XFH experiments were carried out at BL-6C at 100 K. The details of the experimental technique are given elsewhere [2]. The obtained holographic data were analyzed using a sophisticated algorithm of an L_1-regularized linear regression (SPEA-L1 algorithm [5]) representing a sparse modeling approach.

Figure 1 shows 3D atomic images around an Mn impurity atom. The dashed lines indicate the hexagonal structure on the (001) plane when the Mn impurity atom is substitutionally replaced with a Bi or Te atom. Large images are seen inside the hexagonal positions on the (001) plane at z = 0 nm. Small but prominent atomic images are also observed at z = ±0.13 nm. Note that these images do not form a hexagonal shape, but have a threefold rotational symmetry. It should also be noted that the directions of the triangle images at z = ±0.13 nm differ from each other.

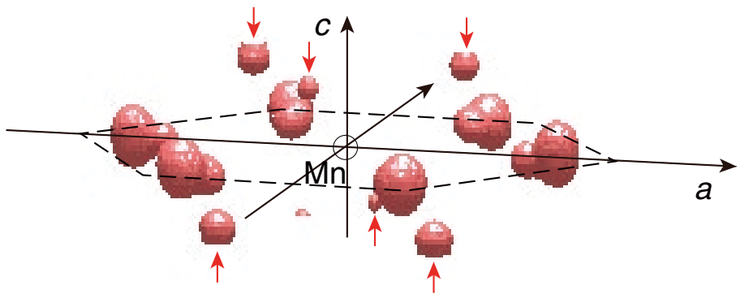

Figure 1: 3D atomic images around an Mn impurity atom. The figure is taken from Ref. [4].

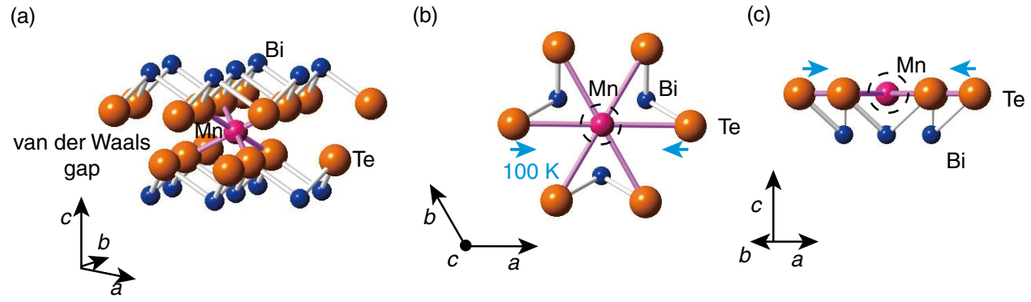

Figure 2: Structural models for the Mn impurity in Bi_2Te_3 crystal. The figure is taken from Ref. [4].

From the obtained atomic images around the Mn impurity atom, two models can be proposed for the Mn impurity sites. The triangle images at z = ±0.13 nm are suitable for the atomic configuration as shown in Fig. 2(a), where the Mn impurity enters an interlayer site with an octahedral arrangement with six Te atoms. The other prominent images with hexagonal shape at z = 0 nm are a substitutional site replaced with a Bi or Te atom as shown in Figs. 2(b) and (c), viewed parallel and perpendicular to the c axis, respectively.

Figures 2(b) and (c) illustrate the position of the Mn atom at the Te site of the layer surface. However, due to the same hexagonal arrangements around the Bi and Te neighboring atoms, the same model is applicable for the Bi site. The neighboring element can be identified with a resonant approach to XFH using the anomalous atomic form factor near an absorption edge of the neighboring element; such work is now in progress.

REFERENCES

[1] W. Friedrich, P. Knipping and M. Laue, *Ann. Phys.* **41**, 971 (1913); M. Laue, *Ann. Phys.* **41**, 989 (1913).
[2] K. Hayashi, N. Happo, S. Hosokawa, W. Hu and T. Matsushita, *J. Phys.: Condens. Matter* **24**, 093201 (2012).
[3] H. -J. Kim, K. -S. Kim, M. D. Kim, S. -J. Lee, J. -W. Han, A. Ohnishi, M. Kitaura, M. Sasaki, A. Kondo and K. Kindo, *Phys. Rev. B* **84**, 125144 (2011).
[4] S. Hosokawa, J. R. Stellhorn, T. Matsushita, N. Happo, K. Kimura, K. Hayashi, Y. Ebisu, T. Ozaki, H. Ikemoto, H. Setoyama, T. Okajima, Y. Yoda, H. Ishii, Y. -F. Liao, M. Kitaura and M. Sasaki, *Phys. Rev. B* **96**, 214207 (2017).
[5] T. Matsushita, *e-J. Surf. Sci. Nanotech.* **14**, 158 (2016).

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