Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

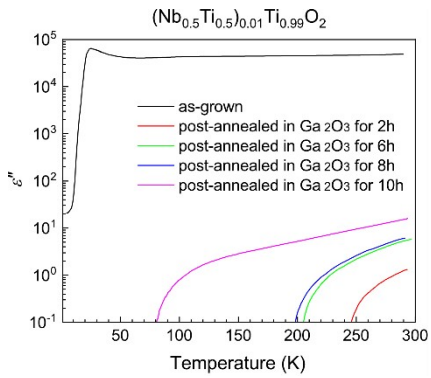

Fig. S1: Temperature dependence of ϵ'' for the as-grown and post-annealed single crystals of NTTO-001, measured along the [001] direction. The post-annealing time in the Ga_2O_3 powder is systematically varied from 2 h to 10 h, as indicated. Note that the steep drop observed in the post-annealed sample is an artifact due to a value of ϵ'' in the low-temperature region that is too small to measure accurately.

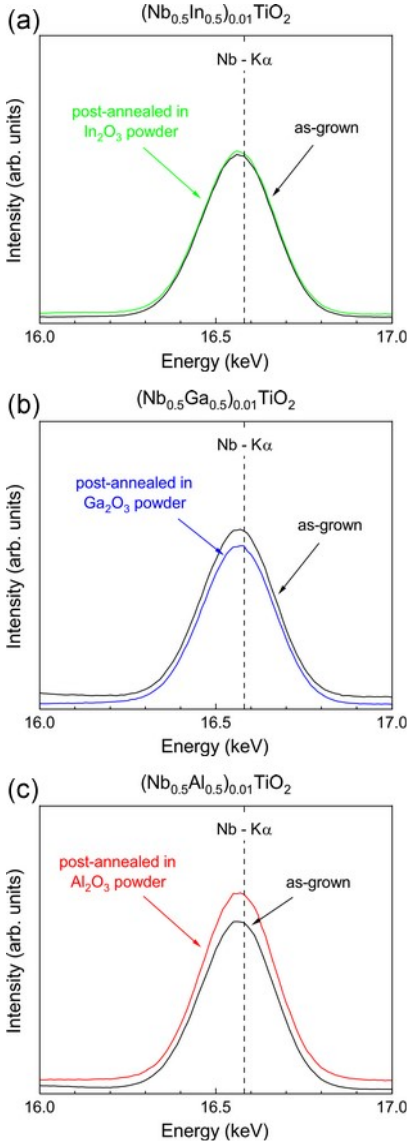

Fig. S2: X-ray fluorescence spectra for (a) (NI)TO-001, (b) (NG)TO-001, and (c) (NA)TO-001 as-grown and post-annealed single crystals, where the position of the Nb-Kα line is indicated by the dashed line in each panel. Note that the intensity of each spectrum is normalized with the most intense peak of Ti-Kα, around 4.5 eV.

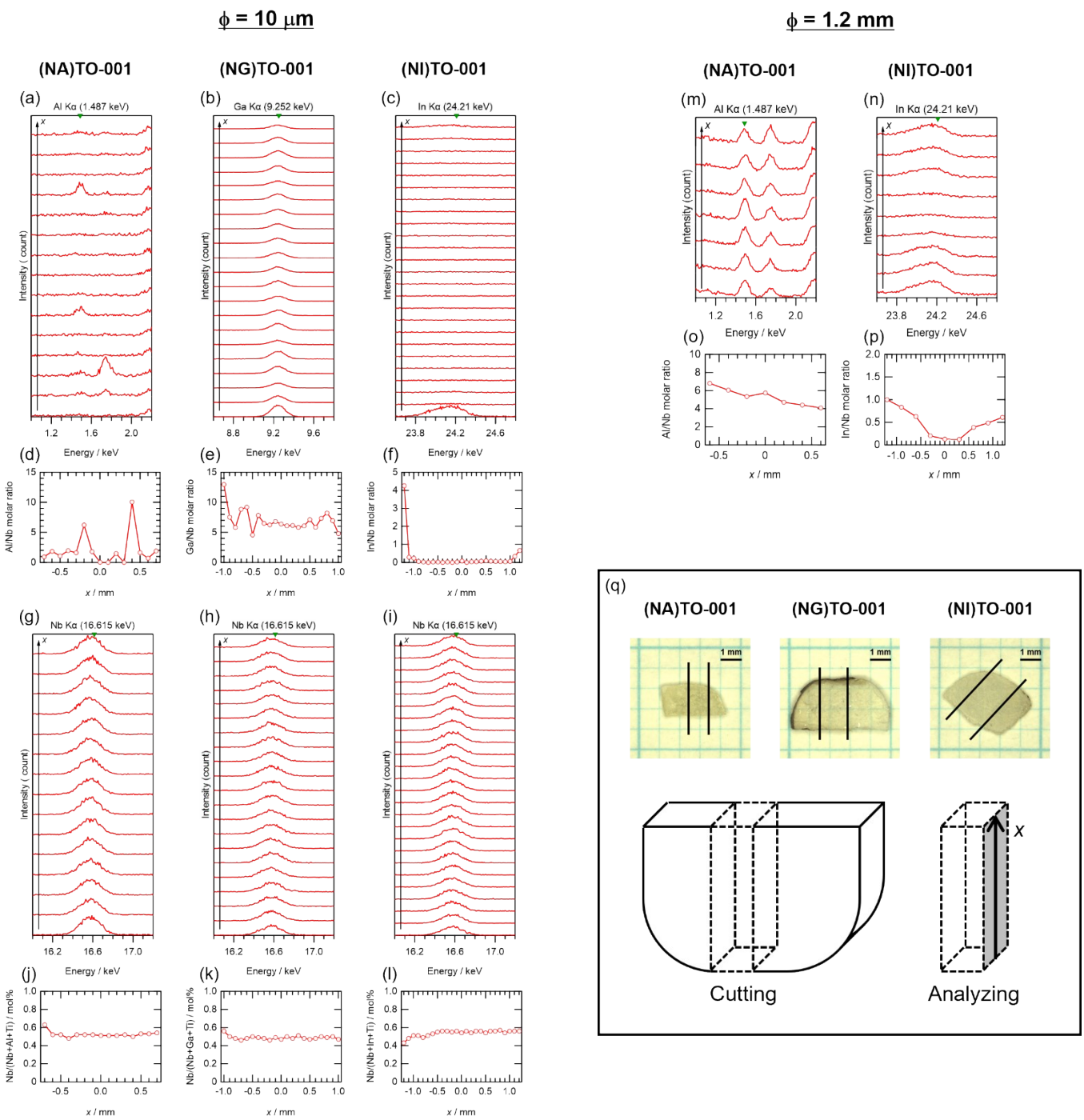

Fig. S3: X-ray fluorescence (XRF) spectra measured along the depth direction of the post-annealed single crystals of (a,g) (NA)TO-001, (b,h) (NG)TO-001, and (c,f) (NI)TO-001, where panels (a-c) and (g-f) present the ka lines of the trivalent dopants and Nb, respectively. The diameter of focus used in these XRF measurements was 10 mm. Panels (d), (f), and (f) respectively denote the Al/Nb, Ga/Nb, and In/Nb molar ratios, which were estimated from the XRF spectra. Panels (j), (k), and (l) present the Nb/(Nb+Al+Ti), Nb/(Nb+Ga+Ti), and Nb/(Nb+In+Ti) molar ratios, respectively. As shown in panels (a-l), In and Al show an inhomogeneous distribution, where In condenses around the surfaces while Al is distributed in a patchy fashion. In marked contrast to In and Al, Ga and Nb are found to spread all over the sample homogeneously. Panels (m)-(p) denote the XRF spectra and the molar ratios of Al/Nb and Ga/Nb, which were obtained with a focus diameter of 1.2 mm. The sample preparation method and scanning direction for the XRF measurements are schematically illustrated in panel (q).

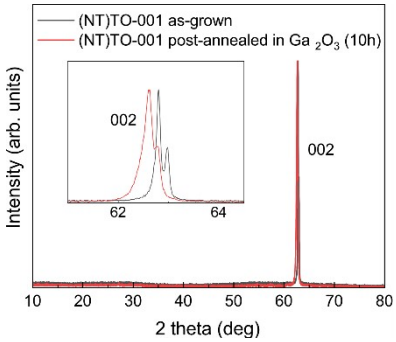

Fig. S4: X-ray diffraction patterns of the as-grown and post-annealed (NT)TO-001 single crystals, where the post-annealing was performed in the Ga_2O_3 powder for 10 hours at 1400 ℃.