Chemical pressure effect on T_c in BiS_2-based $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$

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Abstract

We have investigated the crystal structure and superconducting properties of the BiS_2-based layered superconductor $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$.
Bulk superconductivity was observed for $x \geq 0.4$, and the transition temperature was enhanced with increasing Nd concentration. The highest transition temperature was 4.8 K for $x = 1.0$. With increasing Nd concentration, the length of the *a* axis decreased, while the length of the *c* axis did not show a remarkable change. The chemical pressure along the *a* axis upon Nd substitution seemed to be linked with the inducement of bulk superconductivity. We found that the chemical pressure effect did not completely correspond to the external pressure effect.

Keywords: BiS2-based superconductor; chemical pressure effect

1. Introduction

Layered materials have been actively studied in the field of superconductivity because superconductors with unconventional paring mechanisms and/or high transition temperature (T_c) had been discovered in layered crystal structures. Recently, we have reported superconductivity in several layered materials possessing a BiS_2-type superconducting layer [1,2]. The crustal structure composed of an alternate stacking of the BiS_2 superconducting layers and blocking layers is quite similar to those of the Cu-oxide and Fe-based superconductors. So far, three types of BiS_2-based materials, $Bi_4O_4S_3$, $REOBiS_2$ family (RE = La, Ce, Pr, Nd, Yb) and $SrFBiS_2$, have found to become superconducting upon electron doping into the Bi-6*p* orbitals within the BiS_2 layers [1-9]. Electrical resistivity measurements under high pressure revealed that the T_c of BiS_2-based family was sensitive to application of pressure and can be significantly enhanced [10-12] as observed in the Fe-based family [13].

In the $CeOBiS_2$, bulk superconductivity has not appeared under ambient pressure, but appeared under high pressure with T_c of 8 K. While in the $NdOBiS_2$, the bulk superconductivity has appeared under ambient pressure with T_c of ~5 K. These facts indicate that the superconducting properties of BiS_2-based family depend on both the applied pressure and the structure of the blocking layer. In this article, we have investigated the crystal structure and superconducting properties of $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ synthesized to clarify chemical pressure effect on T_c of the $REOBiS_2$ family.

2. Experimental

The polycrystalline samples of $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ (x = 0.0 - 1.0) were prepared by a solid-state reaction method. Bi grains, Ce_2S_3 powder, Nd_2S_3 powder, BiF_3 powder, Bi_2O_3 powder and Bi_2S_3 powder were used as the starting materials. The Bi_2S_3 powder was synthesized by a direct reaction of Bi grains and S grains at 500 ºC in an evacuated quartz tube. Other chemicals were purchased from Kojundo-Kagaku laboratory. The mixture of the starting materials with compositions of $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ (x = 0.0 - 1.0) was well-mixed, pelletized and sealed into an evacuated quartz tube. The $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ pellets were heated at 700 ºC for 10h. The obtained products were ground, sealed into an evacuated quartz tube and heated again with the same heating conditions to homogenize the samples. The obtained samples were characterized by X-ray diffraction using the θ–2θ method. Changes in the lattice volume were discussed with the peak shifts. The temperature dependence of magnetization was measured by a superconducting quantum interface device (SQUID) magnetometer with an applied field of 5 Oe after both zero-field cooling (ZFC) and field cooling (FC).

3. Results and discussion

Figure 1 (a) shows the crystal structure of $REOBiS_2$ (RE = Ce, Nd). The X-ray diffraction pattern for the $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ samples is displayed in Fig. 1(b). Almost all of the obtained peaks were explained using the tetragonal *P*4/*nmm* space group. The profiles exhibit quite similar tendency upon Nd substitution except for the slight differences in peak shifts corresponding to the lattice contraction. The estimated peak positions of the (200) and (004) peaks are plotted in Figure 2. The shift of the (200) peak to a higher angle corresponds to the shrinkage of the *a* axis, while that of (004) peak corresponds to the shrinkage of the *c* axis. Therefore, in this system, the *a* axis decreases with increasing Nd concentration, while *c* axis does not show a remarkable dependence on Nd concentration. Figure 3 (a) shows the temperature dependence of magnetic susceptibility for $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$. Figure 3(b) shows the Nd concentration dependence of the transition temperature (T_c) for $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ estimated from the magnetization measurements. Superconductivity is observed for $x \geq 0.4$. The transition temperature (T_c) is defined as a temperature at which the magnetic susceptibility begins to decrease. The T_c increases with increasing Nd concentration and reach 4.8 K for x = 1.0, $NdO_{0.5}F_{0.5}BiS_2$.

The chemical pressure effect induces the lattice shrinkage along the *a* axis. Bulk superconductivity is observed for $x \geq 0.4$ where the *a* axis is significantly decreased. The contraction of the *a* axis seems to be linked with the appearance of superconductivity in the $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ system. However, the obtained T_c is clearly lower than that observed in the external pressure studies on T_c of $CeO_{1-x}F_xBiS_2$. The pressure dependence of T_c in $CeO_{0.5}F_{0.5}BiS_2$ shows a transition-like behavior and exceeds 6 K under high pressure above 2.61 GPa [11]. In fact, the chemical pressure effect on T_c in $CeO_{0.5}F_{0.5}BiS_2$ does not completely correspond to the external pressure effect. Therefore, further increase of T_c at ambient pressure in the $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ system should require optimization of some crystal structure parameters other than the simple contraction of the *a* axis.

4. Conclusion

In this study, we have systematically synthesized the $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$ polycrystalline samples, and investigated the crystal structure and superconducting properties. On the basis of systematic investigation on the lattice contraction and the susceptibility, we found that chemical pressure effect induced lattice shrinkage along the *a* axis. Bulk superconductivity was observed for $x \geq 0.4$, and the transition temperature was enhanced with increasing Nd concentration. In this system, the chemical pressure along the *a* axis upon Nd substitution seemed to be linked with the inducement of bulk superconductivity. However, the chemical pressure effect on T_c in $CeO_{0.5}F_{0.5}BiS_2$ does not completely correspond to the external pressure effect. To clarify the detailed correlation between superconductivity and crystal structure, studies using single crystals are required.

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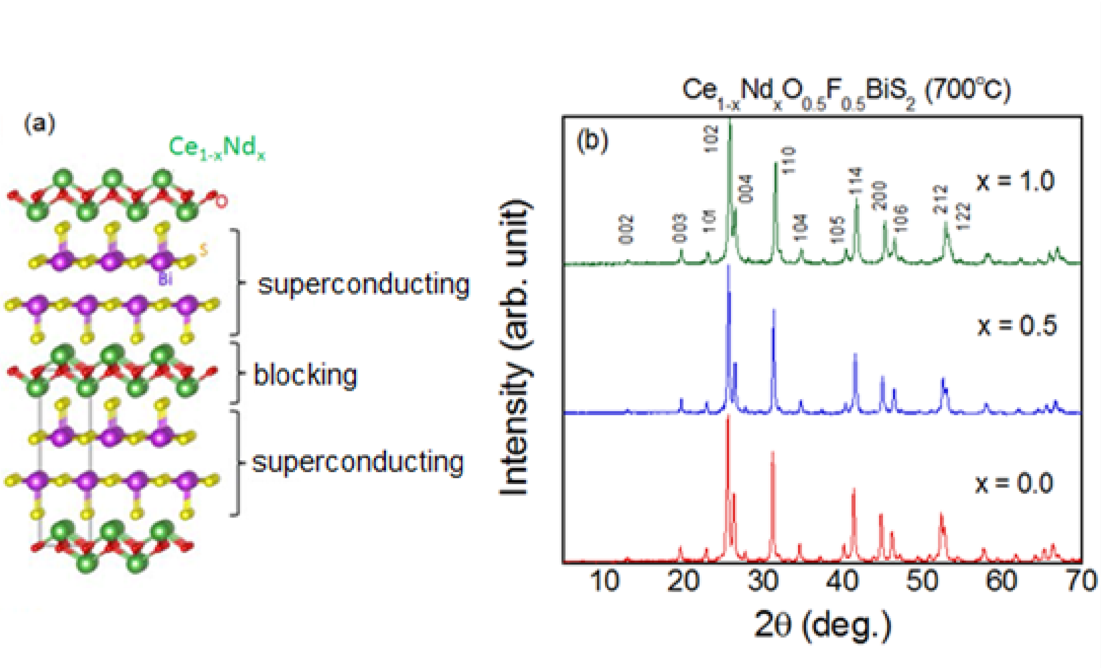

Figure 1. (a) Crystal structure of $REOBiS_2$ (RE = Ce, Nd). (b) X-ray diffraction pattern for $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$. The numbers indicate Miller indices.

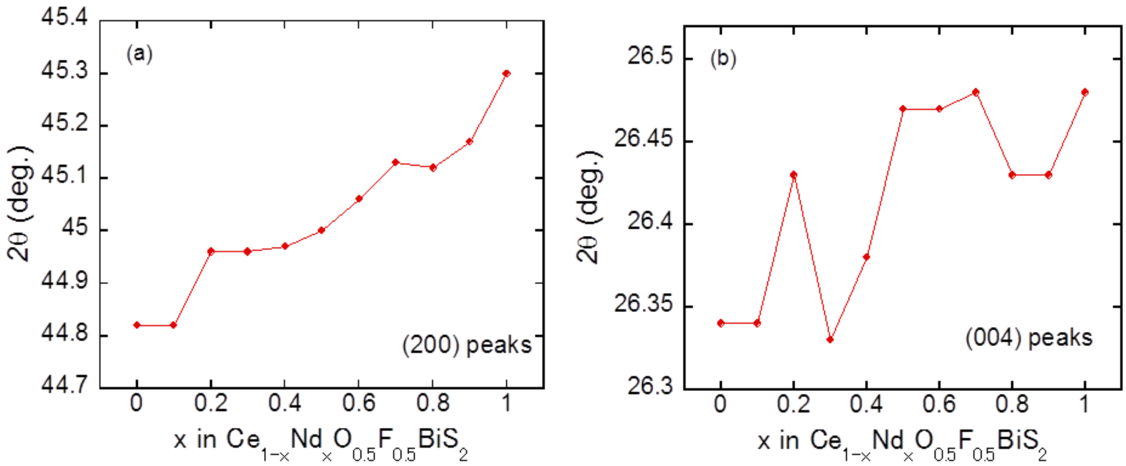

Figure 2. Nd concentration dependence of the peak positions of the (a) (200) and (b) (004) peaks.

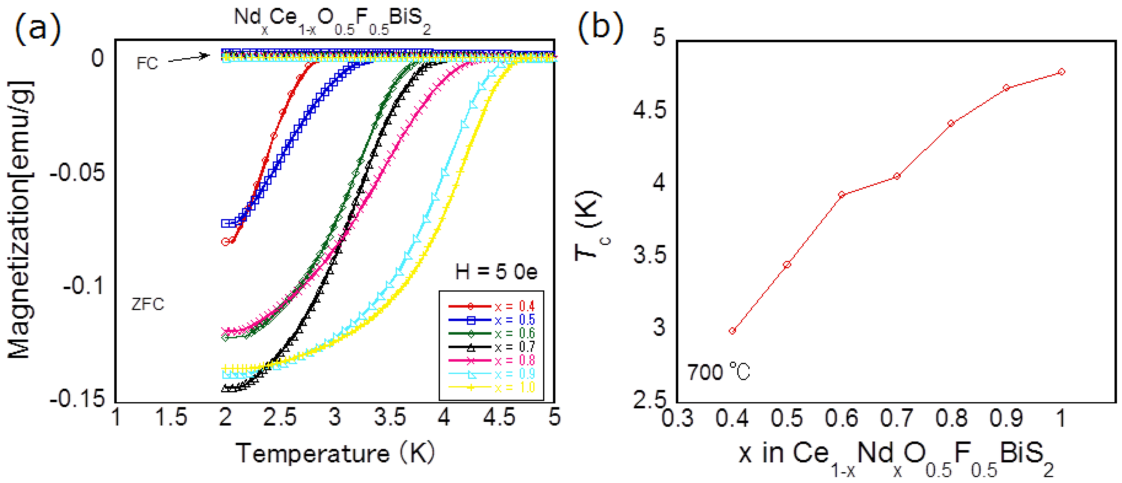

Figure 3. (a) Temperature dependence of magnetic susceptibility for $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$. (b) Nd concentration dependence of the transition temperature (T_c) for $Ce_{1-x}Nd_xO_{0.5}F_{0.5}BiS_2$.