First-Principles Calculation of Electronic Structure and Magnetic Properties of Sm2Fe17Nx

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Sm2Fe17N3 with the Th2Zn17 structure has excellent properties as a permanent magnet material [1]. It shows a much larger magnetic anisotropy and a higher Curie temperature than Nd2Fe14B, which is commonly used today, although its saturated magnetization is a little smaller than that of latter. The main obstacles in using Sm2Fe17N3 are that it is not resistant to heat treatment and it is difficult to sinter. Hence, it is used only as a bonded magnet at present. However, Sm2Fe17N3 is interesting from the viewpoint of basic physics because the mechanisms that govern the magnetic properties of the system, especially magnetic anisotropy, do not seem very trivial.

The most prominent feature of Sm2Fe17N3 is that adding N has enhanced the magnetism of the system. Introducing N into Sm2Fe17 increases the saturation magnetization by 12% and the Curie temperature by 93%. The magnetic anisotropy changes from in-plane to along the c-axis, and the magnitude of the anisotropy is markedly increased. The origin of such enhancement of the magnetism of ferromagnetic materials triggered by typical elements has long been discussed theoretically. One of the reasons for the enhancement is the volume expansion effect by the insertion of N. Another reason is the hybridization between the d states of magnetic ions and the p states of typical elements. Concerning Sm2Fe17N3, according to the first-principles electronic structure calculation by Steinbeck et al. [2], the enhancement of the saturation magnetization mainly comes from the volume expansion. Although an enhancement in the magnetization by N insertion was also seen in their calculation where the volume was fixed, the change was rather small compared with that otherwise obtained. Steinbeck et al. investigated the magnetic anisotropy by calculating the crystal field parameters at the Sm site and concluded that, in contrast to the magnetization, the crystal field was not affected by the volume but increased by the insertion of N. Asano and Yamaguchi pointed out that, in RFe12X and R2Fe17X3 (R is a rare-earth element and X is a typical element), the hybridized states between the X-p and R-d states increase the anisotropic electron distribution and crystal field [3].
In this study, we investigate the role of N in the magnetic properties of Sm2Fe17Nx on the basis of first-principles calculation. We treated the Sm-f states as valence states whereas the open-core treatment has often been used for the R-f states. We took the self-interaction correction (SIC) into account and remedied this problem. For the magnetic anisotropy we evaluated it from the total energy of the system. Accordingly, we could compare our results directly with the experimental data. We studied the N content dependence of magnetic properties.

We found that the treatments of the Sm-f states do not markedly affect much the other states. For example, the magnetization as a whole is mainly determined by the electronic structure of Fe. In this respect, the results of the present calculation are consistent with those obtained by the open-core method. However, it is clearly seen that the Sm-f states are affected by the surrounding N-p states that can never be seen by the open-core method. This effect has an impact on the magnetic anisotropy. We also discuss the results for the systems where N is replaced by other typical elements such as B and C.

Figure shows the magnetocrystalline anisotropy constant K1 of Sm2Fe17Nx. The results of K1 are also within the range of experimental values [4–8] (in experiments, K1 does not depend much on the temperature). The anisotropy is very small for Sm2Fe17 and increases linearly as the N content increases. This linear behavior is also consistent with the experimental observation. In contrast to the magnetic moments, the structure dependence of K1 is small. As the source of the anisotropy, the contribution of the band energy is large, and the change in the electronic structure of the Sm-f states due to the N insertion affects the increase in the anisotropy. We also calculated Sm2Fe17Nx systems. When we proceed from X=C to X=N, the magnetic moment markedly increases owing to the change in the bonding nature between X and Fe. A correlation between the magnetic anisotropy and the total magnetic moment is clearly observed.

References