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Viewpoint set

Maximum performance of permanent magnet materials

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ABSTRACT

Calculations based on density functional theory conclude that the plausible upper limits of saturation magnetic polarization, magnetic transition temperature, and the magnetocrystalline anisotropy constant of permanent magnet materials could be ~2.7 T, ~2000 K, and ~1000 MJ m^{-3}, respectively. It is suggested that a possible strategy to reach these values for high-performance permanent magnets is to design Fe-based magnets containing a small amount of light rare earth elements, together with some additional elements including V, Cr, Co, Ni, and possibly typical elements as interstitials. The idea of creating high-performance permanent magnets without rare earth elements can be dispensed with.

1. Introduction

Development of permanent magnets with performances in excess of that of the $Nd_2Fe_{14}B$ magnet [1]—currently the strongest permanent magnet—has long been a challenging subject: for example see refs. [2,3]. This has not only been in strong demand for industrial applications but has also attracted much interest from the viewpoint of basic science. Despite intensive efforts, no essentially novel magnet that shows better performance than the $Nd_2Fe_{14}B$ magnet has been developed so far. In this situation, one might have the following question: Is it possible at all to obtain a permanent magnet material that is superior to the currently available maximum performance, or should we follow other directions such as pursuing a magnet less expensive but with reasonable performance?

To answer these questions, it is necessary to have a perspective on the possible (maximum) performance of permanent magnet materials and some guides to approach the final goal.

In terms of the intrinsic properties of permanent magnet materials, the key elements are the saturation magnetic polarization J_S (or magnetization M_S), ferromagnetic transition temperature (Curie temperature) T_C, and magnetocrystalline anisotropy energy (MAE) E_A. In realistic permanent magnets, the lowest order uniaxial magnetic anisotropy constant K_1 can represent MAE. To have large values of these quantities is of primary importance for a permanent magnet materials candidate. The importance of MAE comes from the fact that one of most critical features of permanent magnets, coercivity H_C, is bound by MAE through an empirical relation: $H_C = \alpha H_A$–(demagnetization field), where $H_A = 2K_1/J_S$ [4]. Here, α is a (temperature dependent) phenomenological constant that depends on micro and macroscopic permanent magnet structures, and is said to be 0.1–0.3 except in extreme conditions.

One of the performance measures of permanent magnets is maximum energy product $(BH)_{max}$, whose upper limit is given by $J_S^2/(4\mu_0)$. If we assume 2.45 T for J_S, which is the known largest value of J_S, realized for permendule ($Fe_{0.65}Co_{0.35}$), the upper limit of energy product $(BH)_{max}$ is as high as 1190 kJ m^{-3} [5]. Although there is no evidence that 2.45 T is the largest possible value of J_S, it is reasonable to say that our goal for $(BH)_{max}$ could be ~2000 kJ m^{-3}. Actually this is much higher than the 460 kJ m^{-3} value of $Nd_2Fe_{14}B$. The main obstacle for this goal lies in the smallness of H_C: $H_C > J_S/(2\mu_0)$ is necessary to achieve the maximum $(BH)_{max}$ that is expected from J_S. This indicates that it is crucial to achieve a H_C that matches J_S to improve $(BH)_{max}$.

The magnetic transition temperature T_C is also important from the viewpoint of MAE. This is because in order to guarantee a sufficient value of H_C at operating temperature T_{op}, T_C must be considerably higher than T_{op}: K_1 drops much faster than J_S with increasing temperature. However, considering that T_C for iron is 1044 K and that of cobalt is 1360 K, meeting the condition $T_C > 1.5\ T_{op}$ (to be on the safe side), where T_{op} is typically 300–500 K, might not be unrealistic for many Fe- and Co-based magnetic materials.

From the above consideration, we think that it would be useful to approximate the upper limits of J_S, T_C, and K_1, and make a prospect about the performance of permanent magnets. In the following, we will discuss each of these quantities in a semi-quantitative manner using the data obtained through first-principles calculation.

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In Section 2, the upper limits of J_S, T_C, and K_1 are estimated on the basis of first-principles calculations. Using the thus obtained information, the approach for these upper limits is discussed in Section 3. Section 4 concludes with remarks.

2. Upper limits of intrinsic magnetic properties

For ionic systems, the maximum value of magnetic moment per atom M of 3d ions is $\sim 5.9\ \mu_B$, which is made possible by d^5 (Mn^{2+} and Fe^{3+}) configurations (quenching of orbital angular momentum assumed), and that of 4f ions is $\sim 10.6\ \mu_B$, made possible by f^9 (Dy^{3+}) and f^{10} (Ho^{3+}) configurations. However, these values are achieved only for a system in which magnetic ions are diluted by anions, except for the cases of Dy and Ho elements. Since T_C of Dy and Ho is ~40 K, their large magnetic moments can be meaningful only for limited purposes. All other lanthanides also carry large magnetic moments, but T_C is too low for practical use. Therefore, in the following discussion of J_S and T_C, we will restrict ourselves to the cases where 3d magnetic ions are the main origin of magnetic polarization. On the other hand, 4f ions are of main concern for MAE, though the contribution of 3d–5d transition metal ions cannot be neglected, in particular at high temperature. For this reason, we will discuss MAE in a somewhat different way than we do for J_S and T_C.

The discussions are based on all-electron first-principles electronic structure calculations performed within the framework of the local density approximation (LDA/GGA) of density functional theory (DFT). We used a KKR-CPA package (AkaiKKR) [6], and for the calculation of T_C, Liechtenstein's method [7] was applied.

2.1. Saturation magnetic polarization J_S

Fig. 1 shows the overall behavior of magnetic moment $M(a, Z)$ per atom of 3d elements plotted against the lattice constant a and atomic number Z. The crystal structure is fixed to bcc. Here, the fractional atomic number of a fictitious atom is used: $Z = 25$, 26, and 27 corresponds to Mn, Fe, and Co atoms, respectively. The number of total electrons per atom is equal to Z. For large lattice constant ($a \sim 3.2$ A), M is a decreasing function of Z. For a lattice constant of bcc Fe (2.867 A at room temperature), M shows a usual Slater-Pauling type behavior [8], namely, taking a maximum value of 2.35 μ_B/atom at $Z = 26.2$, decreasing to both sides. The peak position shifts toward larger Z with a decreasing lattice constant. In the meantime, the peak height decreases rapidly. These behaviors are well understood as results of (i) shift of the position where the transition from strong to weak ferromagnetism takes place, and (ii) the shift of the position where the instability of ferromagnetism against volume collapse takes place, both occurring when atomic number decreases.

The behavior of saturation magnetic polarization J_S shown in Fig. 2 does not obey that of the magnetic moment. The most prominent feature is that unlike the magnetic moment, it has a dome-like structure appearing around $a = 2.65$ A and $z = 26.4$, where J_S takes the maximum value of 2.66 T. We do not go into details about the origin of this dome. It is merely pointed out that this is related to the fact that in the bcc structure, the interatomic distance between nearest neighbor pairs becomes small, forming a considerable bonding antibonding splitting and the pseudo gap in between. Unfortunately, the lattice constant $a = 2.65$ A is 7% too small compared with the equilibrium lattice constant of bcc Fe. It should be noticed that, contrary to the behavior of the magnetic moment, the magnetic polarization increases with decreasing a up to the point where the magnetic state starts to rapidly collapse.

Magnetic polarization takes on a large value at one of the corner points in the Z-a plane, $Z = 25$ and $a = 3.2$ A, but this is not real. In this region, the antiferromagnetic state is more stable than the ferromagnetic state, as will be shown in the next subsection. The truth is that in a broad region of the Z-a plane—the region to the left of the dashed line in Fig. 2—the ferromagnetic state is unstable. If we combine this fact with the information given by Fig. 2, we may conclude that a large J_S is expected only in the vicinity of the dome-like structure seen in Fig. 2, and the upper limit of J_S would not exceed ~2.7 T.

All the above observations are based on the calculation performed for the bcc structure. However, similar calculations for the fcc structure confirm that the conclusions do not change drastically: it is

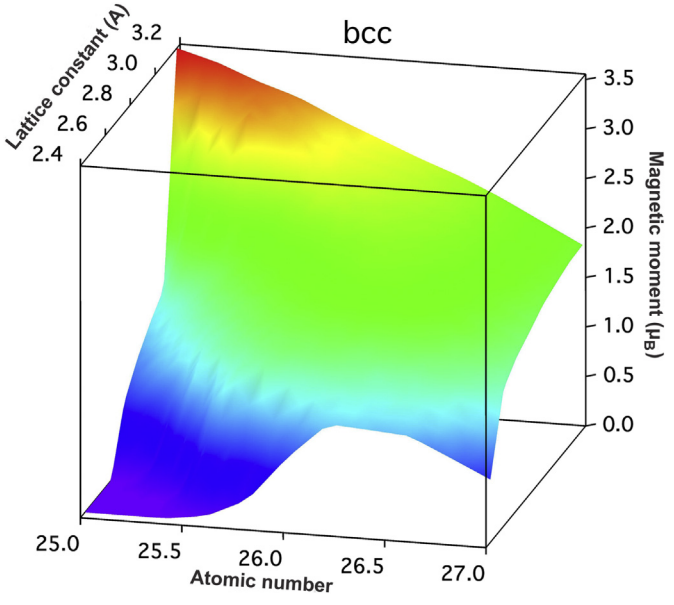

Fig. 1. Magnetic moment per atom M of 3d elements is plotted against the lattice constant and atomic number of a fictitious atom. Here bcc structure is assumed, and the electron number corresponds to the atomic number of fictitious atomic.

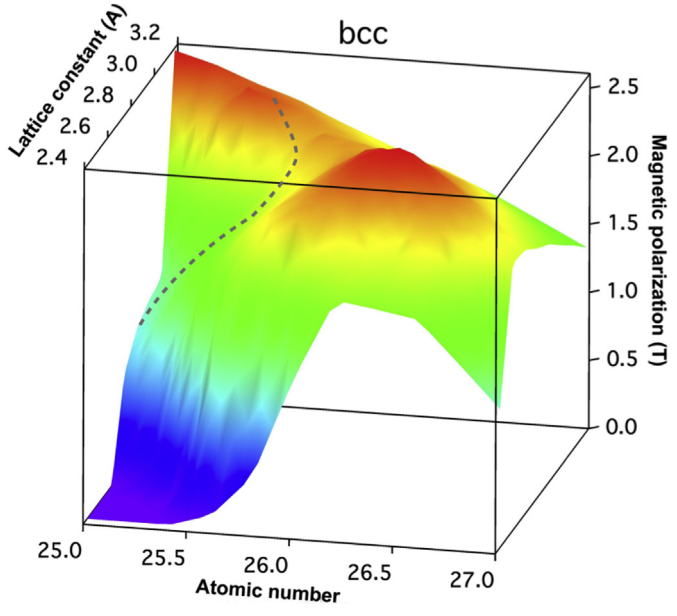

Fig. 2. Saturation magnetic polarization J_S of the system plotted against the lattice constant and atomic number of a fictitious atom. The ferromagnetic state is unstable in the region to the left of the dashed line. Here the bcc structure is assumed, and the electron number corresponds to the atomic number of fictitious atoms.

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