Effects of spin fluctuation on the magnetic anisotropy constant of itinerant electron magnets

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In the disordered local moment picture, we calculated the magnetization \( M \) and magnetic anisotropy energy (MAE) of FePt, CoPt, and MnAl ordered alloys and a body-centered tetragonal FeCo (bct-FeCo) disordered alloy, assuming spatially fluctuated spin configurations at finite temperatures. All alloys exhibit the relation \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) with the exponent \( n \approx 2 \). This is consistent with the two-ion anisotropy model, in contrast to the usual single-ion anisotropy model exhibiting \( n = 3 \). Because these systems have different mechanisms of MAE, we suggest that this relation is a general rule for itinerant electron systems. © 2016 The Japan Society of Applied Physics

Recent remarkable developments in information systems require higher-density magnetic storage devices. However, research and development in this field has faced a serious problem regarding information instability. According to numerous studies, memory bits must satisfy the thermal stability condition given by \( K_1 V / k_B T \geq 60 \), where \( K_1 \) is the uniaxial (or first-order) anisotropy constant, and \( V \) and \( T \) represent the volume of a bit and the temperature, respectively. Thus, the realization of high-density memory by decreasing \( V \) is accompanied by the instability of this condition.

Despite this serious issue in the finite-temperature nature of the magnetic anisotropy energy (MAE), few studies were conducted for 3d transition-metal systems before the precise studies of Okamoto et al.\(^1\) and Thiele et al.\(^2\). In 2002, these studies experimentally demonstrated that the uniaxial MAE of epitaxial FePt films exhibits the relation \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) in the low-temperature region. They additionally showed that the Callen–Callen law\(^3\) \( K_1(T)/K_1(0) = (M(T)/M(0))^n \) relation is missing. Shortly thereafter, Skomski et al.\(^4\) theoretically showed \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) dependence in L10-type CoPt with a classical mean-field two-sublattice model.

Similarly, Myaso\(^5\) et al.\(^5\) demonstrated that the \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) relation could be reproduced from the effective two-ion anisotropy term or from anisotropic exchange interactions between Fe spins mediated by induced Pt spins. They considered the following model Hamiltonian:

\[
H = - \sum_{i,j} J_{i,j} S_i \cdot S_j - \sum_i D_i^{(1)} (S_i^2 - 3/2) - \sum_i D_i^{(2)} m_i^2,
\]

where \( S_i \) and \( m_i \) denote the Fe and Pt spins at the \( i \)-th and \( \nu \)-th sites in each sublattice, respectively. The first term represents the exchange interaction between Fe spins, and the second and third terms are the single-ion anisotropy terms of Fe and Pt spins, respectively. The key feature in this model is that the Pt spin moment \( m_i \) is induced through the exchange field \( \sum_{\nu} J_{\nu,i} S_{\nu} \) from the surrounding Fe spins. Thus, the third term can be rewritten in the form \(-\sum_i D_i^{(2)} S_i^2\) by using the relation \( m_i = -\chi_i \sum_{\nu} J_{\nu,i} S_{\nu} \). Here, \( \chi_i \) implies the spin susceptibility of Pt, and \( D_i^{(2)} \) is defined by \( D_i^{(2)} = \sum_j D_i^{(1)} \chi_j J_{\nu,i} / \chi_i \). This term corresponds to the two-ion anisotropy energy consisting of two different Fe spins. In this sense, the resultant form of the Hamiltonian in Eq. (1) becomes a generalized anisotropic exchange Hamiltonian, which is usually known as the XXZ spin model. On the basis of this scheme, the \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) relation could be successfully realized for the FePt system using the Langevin dynamics simulation.\(^6\) In 2004, Staunton et al.\(^7\) attempted a first-principles calculation for the MAE of FePt at a finite temperature. They also demonstrated the relation \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) and proposed that the anisotropic exchange interaction is responsible for the \( K_1(T)/K_1(0) = (M(T)/M(0))^2 \) relation.

As mentioned above, most previous studies on the relationship between MAE and \( M \) concentrated on the two-sublattice systems: one sublattice has a strong ferromagnetic sublattice and the other sublattice has a large spin–orbit interaction (SOI). In the present work, we study the MAE vs \( M \) relationship for a tetragonally distorted \((c/a = 1.2)\) FeCo disordered alloy,\(^8\) which was theoretically proposed by Burkert et al.\(^9\) to have a large MAE. This alloy is not a two-sublattice system, and thus is completely different from the FePt system. For reference, we also calculated the MAEs of FePt, CoPt, and MnAl L10-type ordered alloys. The reason for employing MnAl is that Al does not have a strong SOI, unlike Pt. Furthermore, as pointed out by Kota and Sakuma,\(^10\) the primary factors realizing a large MAE for these systems are different from each other. In FePt (CoPt), the large MAE originates from the strong SOI in Pt atoms, which hybridize with Fe (Co) 3d orbitals. In the distorted FeCo alloy, on the other hand, 3d bands with \( x^2 - y^2 \) and \( xy \) characters couple through SOI just at the Fermi level, and in the case of MnAl, the matrix elements of SOI between 3d bands have large intensities, which are the main factors providing a large MAE. In this sense, MnAl and distorted FeCo alloys are systems adequate for investigating the general feature of the relationship between MAE and \( M \) in the itinerant systems. In what follows, we explain the calculation model and results.

A standard method for performing practical calculations of the finite temperature magnetism of itinerant electron systems is to adopt the coherent potential approximation (CPA) in terms of the thermal fluctuated spins as scattering potentials for electrons, as performed by Staunton et al.\(^7\). This requires considerable computer resources and time consumption. However, by concentrating only on the relationship between \( M(T) \) and \( K_1(T) \), this expensive approach can be avoided. First, we assume a certain spin configuration \( \{ e_i \} \) (\( e_i \) is the unit vector at the \( i \)-th site) in real space so that the average direction points to \( n \) (unit vector). By arranging artificially the configuration \( \{ e_i \} \) to vary the summation \( (1/N) \sum_{i=1}^{N} e_i \)
from 1 to 0, we can realize the states between \( T = 0 \) and \( T = T_C \) (Curie temperature). Next, under this configuration \( \{ \mathbf{n}; \{ e_i \} \} \) as a molecular field distribution, we calculate the electronic total energy and magnetization defined by \( E(\mathbf{n}; \{ e_i \}) \) and \( M_{\{ e_i \}} \) (\( n \) component of the magnetization), respectively. From the relationship between \( (E(\mathbf{n} = a; \{ e_i \}) - E(\mathbf{n} = c; \{ e_i \})) / V \) and \( M_{\{ e_i \}} \), we obtain \( K_1(T) \) vs \( M(T) \), where \( a \) and \( c \) imply the directions of the \( a \)– and \( c \)-axes, respectively. To further reduce computational resources, we assume that the direction \( e_i \) is restricted to being parallel or antiparallel to \( n \), which means that the spin configuration is always collinear in the line parallel to \( n \). This may give strongly affect the quantitativity in each of \( M(T) \) and \( K_1(T) \). However, we believe that this assumption does not seriously affect the relationship between \( K_1 \) and \( M \), if the temperature dependence of MAE is governed only by the magnetization \( M(T) \). As long as \( M(T) \) and \( K_1(T) \) are treated on the same footing, it is expected that even the collinear model could give a proper relationship between \( K_1(T) \) and \( M(T) \). Note, however, that this is no more than a speculation and is still to be verified somehow. In this work, to see the adequacy of this approach, we first calculated the case for FePt for comparison with the result obtained by Staunton et al.\(^{11} \) For the calculations, we adopt CPA for the random arrangement of spins \( \{ e_i \} \). The fluctuating spin models we considered here are given as \( (\text{FePt})_{1-x}(\text{Fe}1)_x \) for FePt (see Fig. 1) and \( (\text{FePt})_{1-x}(\text{Co}1)_x(\text{Co}1)_{1-x} \) for the FeCo alloy with \( 0 < X < 0.5 \). Here, Fe\( \uparrow \) (\( \downarrow \)) implies the Fe atom whose moment points in the \( \mathbf{n} \) (\( -\mathbf{n} \)) direction. Therefore, \( X = 0 \) implies a complete ferromagnetic state at \( T = 0 \) and \( X = 0.5 \) implies a nonmagnetic state at \( T = T_C \). For each \( X \) and \( n \), we calculate both \( E(\mathbf{n}; X) \) and \( M_{\{ e_i \}}(X) \) by CPA.

For the electronic structure calculations, we employed the tight-binding linear muffin-tin orbital (TB-LMTO) method\(^{11} \) including the SOI under the local density functional approximation. For the disordered FeCo alloy, we applied CPA not only for the spin configuration but also for the arrangement of Fe and Co atoms.

In Fig. 2, we show the calculated total magnetic moment \( M \) per unit cell of each alloy as a function of \( X \). In every alloy, \( M \) decreases linearly with increasing \( X \). This suggests that the magnetic moments in these alloys have similar properties to the local moment model described by the Heisenberg Hamiltonian. In fact, we confirmed that the amplitude of the local moment in these alloys remains constant in the whole \( X \) range. Figure 3 shows the calculated values of \( K_1 \) as a function of \( X \). All \( K_1 \) values monotonically decrease with an increase in \( X \) or a decrease in \( M \). This behavior is understood qualitatively as follows. Generally, in magnetic systems, the magnetic easy direction and energy anisotropy originate from the connection of the orbital moments with the crystal axis. Note that the orbital moments are induced by the spin moments through the SOI, and the average orbital moment determines the anisotropy energy rather than the local orbital moment. Therefore, when the spin moments fluctuate spatially, the average orbital moment decreases, resulting in a decrease in anisotropy energy. As shown by Solovyev et al.,\(^{10} \) \( K_1 \) is expressed by the spatial correlation function of orbital moments, which implies that \( K_1 \) is determined by the expectation value of the orbital moment corresponding to the average orbital moment.

In Fig. 3, we note that the \( K_1 \) values remain finite at \( X = 0.5 \) corresponding to the Curie temperature. This behavior is due to the assumption that restricts the spin moments to be aligned collinearly, from which there remains an energy difference between the cases for \( n = a \) and \( e \) even at \( X = 0.5 \). This result is incorrect because, at \( T = T_C \), the spin configuration should be completely random, yet the spin configuration is the same in these two cases. Thus, we consider that the MAE shows a certain constant value in addition to the \( (M(T)/M(0))^4 \) term.

By comparing Figs. 2 and 3, \( K_1 \) decreases more rapidly than \( M \) with increasing temperature, if \( X \) is regarded as temperature. This implies that the exponent \( k \) of \( (M(T)/M(0))^4 \) is...
function of MnAl, and disordered FeCo (larger than unity. Actually, the curves of
in Fig. 4 clearly exhibit a k value larger than unity. To determine the exponent, we generate log–log plots as shown in Fig. 5. The exponents clearly remain around the value of 2. We should emphasize that the exponent for FePt is 2.2, which is almost the same as that given by experiments and theories. Apart from the constant value of $K_1 (X = 0.5)$, the relationship between $K_1$ and $M$ can be well reproduced by the present approach based on several assumptions. In addition, the cases for the MnAl alloy and bct-FeCo disordered alloy also exhibit an exponent of approximately 2. As mentioned previously, these systems are different from the model proposed by Mryasov et al. introducing the two-ion anisotropy term in the Heisenberg Hamiltonian. Furthermore, the primary factors providing the large MAE of these systems are also different from each other, which may imply that in the metallic system the temperature dependence of MAE is not related to the occurrence mechanism of MAE but is mainly governed by a variation in exchange field due to the temperature change. These findings lead us to believe that the relation $K_1(T)/K_1(0) = (M(T)/M(0))^n (n \approx 2)$ is a general relationship in itinerant electron systems. Even though the amplitude of local moments remains constant in the whole $X$ range (temperature) as shown in Fig. 2, the MAE vs $M$ is apparently different from the case for localized spin systems where $n$ is around 3. As mentioned previously, this difference originates from the different mechanisms of MAE: the $K_1$ of a metallic system is determined through the spatial correlation function of orbital moments, while that of a localized spin system is expressed by the single-ion anisotropy energy.

In summary, we calculated the magnetizations and MAEs of FePt, CoPt, and MnAl ordered alloys and the bct-FeCo disordered alloy using a first-principles approach combined with CPA. Here, we assumed spatially fluctuated spin configurations as a thermal effect at finite temperatures. All alloys exhibit the relation $K_1(T)/K_1(0) = (M(T)/M(0))^n$ with the exponent $n \approx 2$. This is consistent with the two-ion anisotropy model, in contrast to the case for the single-ion anisotropy model exhibiting $n = 3$. From the fact that these systems have different mechanisms of MAE, we suggest that the relation $K_1(T)/K_1(0) = (M(T)/M(0))^n (n \approx 2)$ is widely realized in itinerant electron systems.

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