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Magnetic structures and their stability in Mn_3Rh ordered and disordered alloys

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The electronic and magnetic structure of Mn_3Rh, including noncollinear structures, in the ordered and disordered states have been investigated by the tight-binding linear-muffin-tin orbital method. A magnetic long-range order of the triangular ($T1$) structure in the ordered state is reflected in a dip around the Fermi level in the density of states. This characteristic feature can be retained in the multiple-Q spin-density-wave (SDW) structures in the disordered alloy, implying that a stable antiferromagnetic order also remains in the disordered alloy. The most stable structure in the disordered alloy is suggested to be the $3Q$-SDW structure. From the effective exchange constant, the Néel temperature in the disordered alloy is estimated to be about 680 K, close to the experimental value of about 700 K. Compared to the Néel temperature of pure γ-Mn metal, such a high value is pertinent to the change in the electronic structure caused by the addition of Rh.

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I. INTRODUCTION

Most Mn-based alloys exhibit antiferromagnetism due to their occupation of $3d$ orbitals close to half-filling. Among them, a number of face-centered-cubic (fcc) alloys such as FeMn and Mn-based γ-phase alloys are considered to have complex (noncollinear) spin structures in which the relative angle between the neighboring spins deviates from 0° or 180°. However, a detailed discussion on their magnetic structures has been lacking because of difficulties in both of experimental and theoretical techniques, and the scatter of data due to different experimental methods.

On the other hand, antiferromagnetic Mn-based alloy systems have recently become important as an exchange biasing film in the giant magnetoresistance (GMR) and tunnel magnetoresistance (TMR) devices. Especially, in contrast to a MnPt $L1_0$-type alloy,[1] Mn-based systems such as Mn-Rh (Refs. 2 and 3) and Mn-Ir (Refs. 4–8) γ-phase-disordered alloys require no post-heat-treatments in industrial processes, and hence these γ-phase alloys are promising candidates for the devices mentioned above. As is well known, the exchange-biasing characteristics and the blocking temperature are closely interrelated with the spin structures and the magnitude of the Néel temperature.[9–12] Therefore, the microscopic foundations such as the magnetic structures of these systems are strongly desired to develop excellent properties for GMR and TMR.

Accordingly, from both fundamental and practical viewpoints, investigations of the spin structures and the Néel temperature of γ-phase antiferromagnetic Mn-based alloy systems are very important. Experimentally, Krén and co-workers[13,14] and Kouvel and Kasper[15] suggested the magnetic structure of a Mn_3Rh $L1_2$-type ordered alloy to be $T1$, the so-called "triangular" structure, and its Néel temperature to be 855 K,[14,16] being relatively high among Mn-based alloys. Theoretically, on the other hand, Kübler *et al.*[17] performed first-principles band calculations for the magnetic structures including noncollinear structures of a Mn_3Rh $L1_2$-type alloy, and pointed out that the triangular structure is most stable. In a recent study, it was reported that Mn_3Rh in the disordered state also has a high Néel temperature of about 700 K,[18] which is still higher than that of pure γ-Mn metal. As for γ-phase-disordered alloys, however, the magnetic structure has not yet been investigated both experimentally and theoretically.

In the present work, the magnetic and electronic structures of Mn_3Rh ordered and disordered alloys are calculated by a tight-binding (TB) linear-muffin-tin orbital (LMTO) method based on the local-spin-density (LSD) functional approximation. Noncollinear magnetic structures are treated through the local rotation of the spin quantization axis on each site. In both the disordered alloy and paramagnetic state, the coherent potential approximation (CPA) is adopted for each sublattice within the local frame of the spin axis. The effective exchange constant,[19,20] acting on each Mn moment, is also calculated to examine the magnetic stability and to give a rough estimation of the Néel temperature. Before going into the results, let us first give a brief description on the methodology in Sec. II. The details were presented in Ref. 20.

II. METHODOLOGY

The Green function (GF) in the TB-LMTO is given by the equation

$$\overline{G}(\omega)=\Delta^{-1/2}g^{\gamma}(\omega)\Delta^{-1/2}, \tag{1}$$

with

$$g^{\gamma}(\omega)=[p^{\gamma}(\omega)-\overline{S^{\gamma}}]^{-1}, \tag{2}$$

where $g^{\gamma}(\omega)$ is the so-called auxiliary GF constituted of the potential function $p^{\gamma}(\omega)$ and the screened structure constants $\overline{S^{\gamma}}$ given by $p^{\gamma}(\omega)=[(\omega-C)/\Delta]\delta_{L',L}\delta_{\sigma',\sigma}$ and $\overline{S^{\gamma}}=\overline{S}(1-\gamma\overline{S})^{-1}$, respectively. Here we define $L=(i,l,m)$ (i denotes the site, and l and m are orbital indices) and σ as the spin state. The quantities C, Δ, and γ are the potential parameters determined within each atomic sphere by using the LSD. The local-spin quantization axis pointing to (θ_i, ϕ_i), that is, the direction of magnetic moment on the ith site, is introduced in $\overline{S}$ through $\overline{S}_{L'\sigma',L\sigma} = [U(\theta_{i'}, \phi_{i'}) S_{L',L} U^{+}(\theta_i, \phi_i)]_{\sigma',\sigma}$ with the spin rotation matrix U. Thus the site-diagonal part (i,i component) of the GF in Eq. (1) is diagonal with respect to the spin space, because the GF in Eq. (1) is defined in the local frame of the spin axis. The GF $G(\omega)$ measured in the global spin axis can be obtained by the transformation $G_{ii}(\omega) = U^{+}(\theta_i, \phi_i) \overline{G}_{ii}(\omega) U(\theta_i, \phi_i)$, with which the density matrices in the global spin space is

$$\rho_i^{\sigma',\sigma} = \frac{i}{2\pi} \int^{E_F} d\omega [G_{ii}^{\sigma',\sigma}(\omega + i\delta) - G_{ii}^{\sigma',\sigma}(\omega - i\delta)], \tag{3}$$

where E_F is the Fermi energy. The magnetic moment on each site can be given by $\mathbf{m}_i = (2\,\mathrm{Re}\,\rho_i^{\uparrow\downarrow}, 2\,\mathrm{Im}\,\rho_i^{\uparrow\downarrow}, \rho_i^{\uparrow\uparrow} - \rho_i^{\downarrow\downarrow})$.

Applying the CPA for the disordered alloys, the most localized (β) representation[21] is used for the potential function and the structure constants through the transformation as $p^{\beta} = p^{\gamma}[1 - (\beta - \gamma) p^{\gamma}]^{-1}$ and $\overline{S^{\beta}} = \overline{S^{\gamma}}[1 - (\beta - \gamma)\overline{S^{\gamma}}]^{-1} = \overline{S}(1 - \beta \overline{S})^{-1}$ with $\beta_{l=0} = 0.3485$, $\beta_{l=1} = 0.0530$, and $\beta_{l=2} = 0.0107$. Using these quantities, the CPA condition for the system constituted of plural sublattices is

$$\langle T_i \rangle = \sum_a c_{ia} [p_{ia}^{\beta}(\omega) - \tilde{p}_i(\omega)] \times \{1 + [p_{ia}^{\beta}(\omega) - \tilde{p}_i(\omega)] \Phi_i(\omega)\}^{-1} = 0, \tag{4}$$

with

$$\Phi_i(\omega) = \frac{1}{N} \sum_k [(\tilde{p}(\omega) - \overline{S_k^{\beta}})^{-1}]_{ii},$$

where $\tilde{p}_i(\omega)$ is the coherent potential function, and c_{ia} denotes the density (probability) of an a atom occupying the sub-lattice i. Accomplishing the self-consistent calculation, the auxiliary GF in the β representation is given by

$$g_{ia}^{\beta}(\omega) = \Phi_i(\omega) \{1 + [p_{ia}^{\beta}(\omega) - \tilde{p}_i(\omega)] \Phi_i(\omega)\}^{-1} \tag{5}$$

for the a atom at the ith sublattice. One can obtain the GF in Eq. (1) in the local frame of the spin axis by transforming from the β representation to the γ representation by means of the following identity equation:

$$g^{\gamma} = (p^{\gamma})^{-1} - (p^{\gamma})^{-1} p^{\beta} (p^{\gamma})^{-1} + (p^{\gamma})^{-1} p^{\beta} g^{\beta} p^{\beta} (p^{\gamma})^{-1}. \tag{6}$$

Once the GF $g^{\gamma}(\omega)$ is obtained, the effective exchange constant[19,20] which can be regarded as the amplitude of the exchange field acting on an atom denoted by 0 by surrounding moments, $J_0 = \Sigma_{i \neq 0} J_{io} \boldsymbol{e}_i \cdot \boldsymbol{e}_0$, is expressed by

$$J_0 = -\frac{1}{4\pi} \mathrm{Im} \int^{E_F} d\omega \mathrm{Tr}_{\mathrm{Im}} \{\Omega_0(\omega)(g^{\gamma\uparrow\uparrow}_{00}(\omega) - g^{\gamma\downarrow\downarrow}_{00}(\omega)) + \Omega_0(\omega) g^{\gamma\uparrow\uparrow}_{00}(\omega) \Omega_0 g^{\gamma\downarrow\downarrow}_{00}(\omega)\}, \tag{7}$$

with $\Omega_i(\omega) \equiv (p_i^{\uparrow}(\omega) - p_i^{\downarrow}(\omega))$. This equation also works for the noncollinear case both in ordered and disordered alloys, as pointed out in Ref. 20. According to a generalized molecular field theory from Ref. 22, the magnetic transition temperatures such as the Curie temperature T_C and the Néel temperature T_N can be estimated from $2J_0/3k_B$.

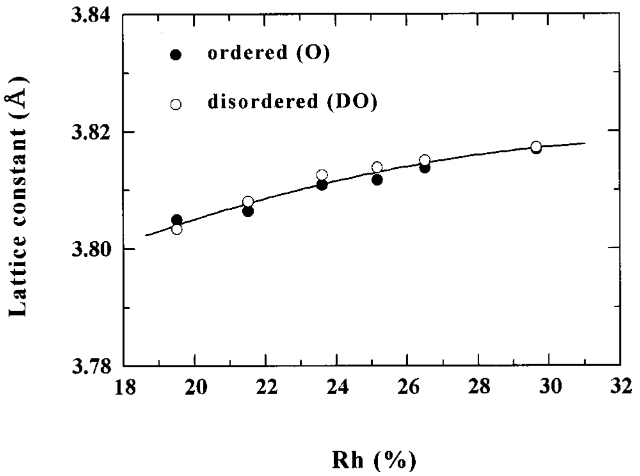

FIG. 1. Concentration dependence of the room-temperature lattice constant a of Mn_3Rh $L1_2$-type ordered (O) and γ-phase disordered (DO) alloys.

III. RESULTS AND DISCUSSION

The concentration dependence of the room-temperature lattice constant a for Mn-Rh $L1_2$-type ordered and γ-phase disordered alloys is given in Fig. 1,[16,18] together with additional data. The experimental procedures were given in Refs. 16 and 18. It should be noted that no remarkable difference between the ordered and disordered states is observed. The room-temperature lattice constant a and the Néel temperature T_N of Mn_3Rh $L1_2$-type ordered (O) and γ-phase disordered (DO) alloys are listed in Table I. To obtain T_N of the disordered alloy definitely is difficult because the ordering is liable to occur on the heating process during measurements.[18]

The magnetic structure shown in Fig. 2 was suggested by Kouvel and Kasper,[15] and called the triangular ($T1$) structure. Later on, Kübler *et al.* showed that this structure is most stable among possible magnetic structures.[17] Here we examine the stability by evaluating the effective exchange constants acting on a Mn moment, together with a comparison to the paramagnetic state. Figure 3 shows the local density of states (DOS) of a Mn_3Rh $L1_2$-type ordered (O) alloy with a triangular ($T1$) magnetic structure. The lattice constant at the ground state ($T = 0$ K) is settled as 3.81 Å, deduced from the observed value obtained at room temperature in Fig. 1 and

TABLE I. The room temperature lattice constant a and the Néel temperature T_N of Mn_3Rh $L1_2$-type ordered (O) and γ-phase-disordered (DO) alloys.

Mn_3Rh	Lattice constant, a (Å)	Néel temperature, T_N (K)
Ordered (O)	3.81_2	855
Disordered (DO)	3.81_4	~700

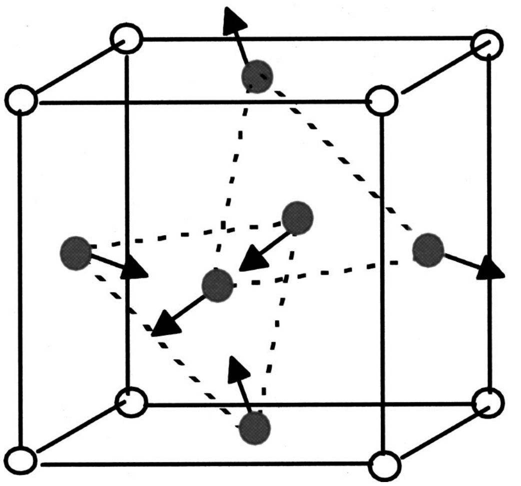

FIG. 2. The magnetic structure of $T1$ for a $\mathrm{Mn_3Rh}$ $L1_2$-type ordered (O) alloy. The open and shaded circles denote Rh atoms and Mn atoms, respectively.

Table I. The result was drawn from the imaginary part of the GF in Eq. (1), not from the usual technique with secular equations, in order to make a comparison with the later results by the CPA for the disordered alloy. Although it was confirmed that this procedure reproduces well the DOS and the moments obtained by the usual secular equation, the present Mn moment of $2.78\mu_B$ is smaller than $3.10\mu_B$ obtained by the ASW method by Kübler *et al.*[17] In addition, the experimental value of about $3.6\mu_B$ (Refs. 13 and 14) is much larger than the values obtained by both the calculations mentioned above. It is not clear at present what brings about such a difference. It should be noted that the Rh moment is zero in both the calculations.

Shown in Fig. 4 is the local DOS in the disordered local moment (DLM) state as a paramagnetic (PM) state of the $L1_2$-type ordered (O) alloy. The DLM state can be calculated by the CPA for the system where the Mn moments pointing upward [Mn(↑)] and those pointing downward [Mn(↓)] are distributed randomly in the Mn sublattices in Fig. 2. The Mn moment is reduced to $2.57\mu_B$, smaller by as much as less than 10% of the $T1$ structure, implying that the present alloy seems to have a rather local moment character. The behavior of the local DOS below -0.2 Ry is retained in both the $T1$ and the PM states, whereas the dip around E_F found in the $T1$ state is swept out in the PM state. This situation is similar to the case of MnPt $L1_0$-type alloy in which the pseudogap realized in the antiferromagnetic (AF) state completely vanishes in the PM state,[20] which means that the gap is closely connected with the AF long-range order. The energy in this PM (DLM) state is higher than that in the $T1$ state as much as about 9 mRy/atom, giving the same order of the Néel temperature.

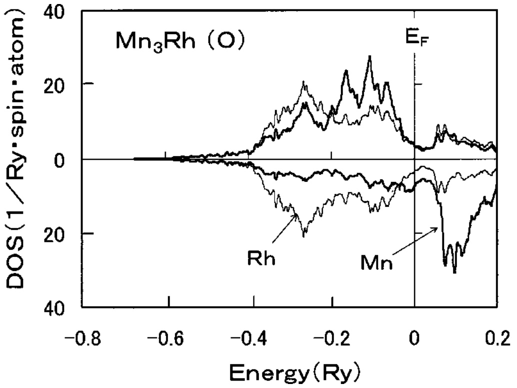

FIG. 3. Density of states of a $\mathrm{Mn_3Rh}$ $L1_2$-type ordered (O) alloy with a $T1$ structure.

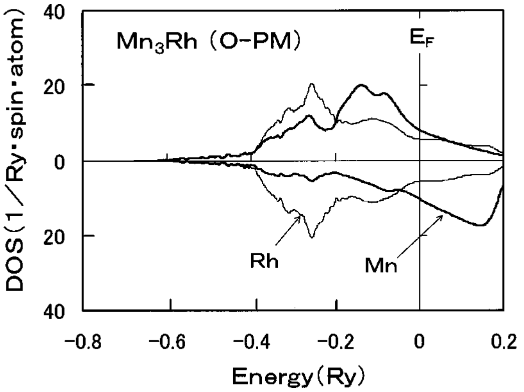

FIG. 4. Density of states of a $\mathrm{Mn_3Rh}$ $L1_2$-type ordered (O) alloy in the paramagnetic state.

To examine the magnetic stability in the $T1$ state, the effective exchange constant J_0 is calculated by using Eq. (7). Figure 5 shows the value of J_0 of the Mn moment as a function of the Fermi level E_F which gives an upper limit of the integral range in Eq. (7) in the rigid-band scheme. The actual E_F is located at the origin of the abscissa. It is found that J_0 has a maximum value of about 160 meV at the actual $E_F(=0)$. This value is close to the energy difference between the $T1$ and PM states, and $2J_0/3k_B$ gives the Néel temperature of 1250 K. The higher value of the Néel temperature than the experimental value by 20–30 % would be adequate within the molecular field approximation for the three-dimensional system.

The magnetic structure in the γ-phase disordered (DO)

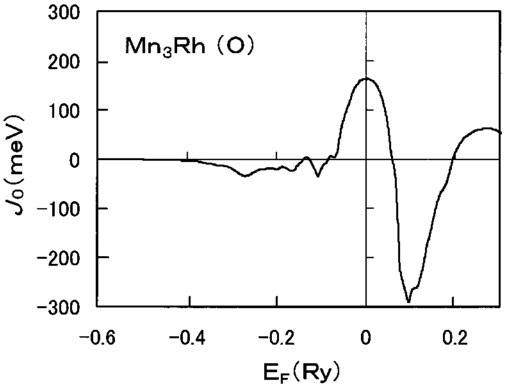

FIG. 5. Effective exchange constant J_0 of a $\mathrm{Mn_3Rh}$ $L1_2$-type ordered (O) alloy with a $T1$ structure as a function of the Fermi level E_F (band filling). The actual E_F is located at the origin of the abscissa.

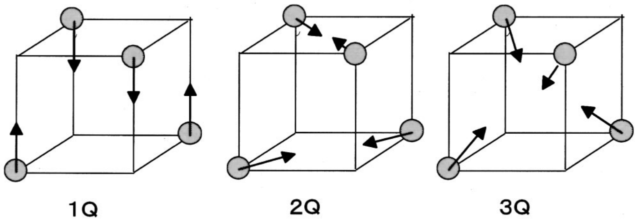

FIG. 6. The $1Q$, $2Q$, and $3Q$ spin structures in the fcc lattice.

alloy considered here is restricted in the $1Q$, $2Q$, and $3Q$ spin structures shown in Fig. 6 as the multiple-Q spin-density wave (MQSDW) structures in the fcc lattice.[23–25] The local DOS of the $1Q$, $2Q$, and $3Q$ spin structures of Mn_3Rh in the disordered state are presented in Fig. 7. The lattice constants of Mn_3Rh in both the O and DO states are practically the same, as seen from Table I. Therefore, the lattice parameter of the disordered alloy is used the same value of the ordered alloy. Contrary to the case of the MnPt $L1_0$-type alloy,[20] the local DOS of Mn and Rh sites maintains well the characteristic features of DOS of the $T1$ structure in the ordered alloy. In particular, it should be remarked that the DOS of the $3Q$ structure is closest to the $T1$ structure, and exhibits a relatively large dip around E_F, which is comparable to that of the $L1_2$-type ordered alloy. This leads us to infer that any of the MQSDW structures in the DO alloy have a lower energy, compared with that in the PM (DLM) state in the O alloy. Actually, as shown in Table II, the MQSDW states in the DO alloy are more stable than the PM state in the O alloy. This is in contrast to the MnPt $L1_0$-type alloy, where the MQSDW structures in the disordered alloy have a higher energy than in the PM state of the ordered alloy.[20] From Table II, the $3Q$ structure is found to have the lowest energy in the disordered state. In the table, the energy of the PM state in the DO state is also shown by using the CPA for the random alloy of the composition $Mn(\uparrow)_{0.375}Mn(\downarrow)_{0.375}Rh(\uparrow)_{0.125}Rh(\downarrow)_{0.125}$. The energy in the PM state is higher by about 7 mRv/atom than that in the MQSDW states, which means that the MQSDW states in the DO alloy are stable, comparable with the $T1$ state in the O alloy. A neutron diffraction study of Mn-Ir γ-phase-disordered alloys was made by Yamaoka, Mekata and Takaki and the possibility of a $1Q$ structure was pointed out,[26] which is different from the present calculations. We have carried out the band calculations for Mn_3Ir γ-phase-disordered alloy, and arrived at the same conclusion as that of Mn_3Rh γ-phase-disordered alloy. Further details of these results will be reported elsewhere.

Shown in Fig. 8 is the effective exchange constants J_0 for each MQSDW structure as a function of E_F, the band filling in the rigid band scheme. All of them exhibit the similar behavior as that of the $T1$ structure in Fig. 5, though the magnitude of the ordinate is slightly reduced. Especially for the $1Q$ structure, the actual value of J_0 at $E_F = 0$ becomes small, with a slight shift of the peak position from $E_F = 0$. The order of magnitude of J_0 in these magnetic structures is consistent with the relative energy shown in Table II, and hence the results suggest that the $3Q$ structure is most stable in the γ-phase DO alloy. The Néel temperature T_N estimated from J_0 is about 680 K, comparable with the experimental value of about 700 K given in Table I. The calculated Mn moments of the $1Q$, $2Q$, and $3Q$ structures in the DO alloy are slightly smaller than the value in the O alloy. On the other hand, a small Rh moment is induced in the disordered state, whereas no moment is confirmed in the ordered state. We have examined the following additional calculations. The value of J_0 in the disordered state with a smaller lattice constant of $a = 3.75$ Å, being the same as that of pure γ-Mn metal, does not change within the numerical accuracy and is still larger than the value of $J_0 = 64$ meV for pure γ-Mn metal. Conversely, J_0 of pure γ-Mn metal with the same lattice constant $a = 3.81$ Å of Mn_3Rh is confirmed to be almost the same as that of pure γ-Mn metal with $a = 3.75$ Å.

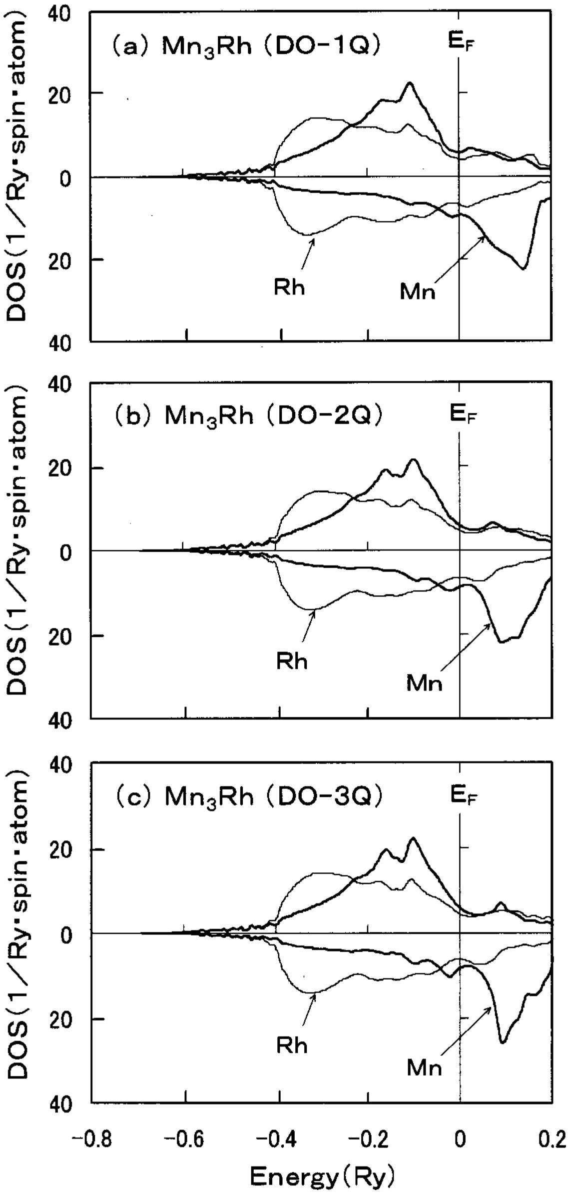

FIG. 7. Density of states of Mn_3Rh γ-phase disordered (DO) alloys for each spin structure. (a) $1Q$ structure. (b) $2Q$ structure. (c) $3Q$ structure.

TABLE II. The spin structure, calculated total energy (ΔE in mRy/atom), magnetic moment (M in μ_B), effective exchange constant (J_0 in meV), and Néel temperature ($T_N^{\rm cal}$ in K) estimated from $2J_0/3k_B$ for $\mathrm{Mn_3Rh}$ $L1_2$-type ordered (O) and γ-phase-disordered (DO) alloys.

Phase	Spin	ΔE	$M_{\rm Mn}$	$M_{\rm Rh}$	J_0	$T_N^{\rm cal}$
Ordered	$T1$	0	2.78	0.00	163	1250
(O)	PM	9.1	2.57	0.00		
	$1Q$	8.6	2.57	0.04	39	300
Disordered	$2Q$	7.6	2.62	0.07	80	620
(DO)	$3Q$	7.3	2.66	0.08	88	680
	PM	15.1	2.48	0.00		

These results strongly suggest that the enhancement of the Néel temperature T_N is not attributed to the lattice expansion but to the change in the electronic structure caused by the addition of Rh to pure γ-Mn metal.

IV. CONCLUSION

Electronic and magnetic structures, including noncollinear structures, of $\mathrm{Mn_3Rh}$ $L1_2$-type ordered and γ-phase-disordered alloys have been investigated by the tight-binding linear-muffin-tin-orbital method combined with the CPA. The magnetic stability in both the states is also examined with the effective exchange constant J_0 acting on the Mn moments. In the ordered state, that is, in an $L1_2$-type alloy, the magnetic moment of the so-called $T1$ (triangular) structure is about $2.78\mu_B$ for a Mn atom. The moment is slightly reduced to $2.57\mu_B$ in the paramagnetic state, which implies that the system seems to be characterized by a rather local moment picture. The long-range order of the $T1$ structure in the ordered state is reflected in a dip structure around the Fermi level in the DOS which vanishes in the paramagnetic state. The electronic total energy of the $T1$ structure is fairly low, about 9 mRy/atom lower than that in the paramagnetic state. The calculated J_0 of about 160 meV also indicates a high stability of the $T1$ structure. The Néel temperature T_N estimated from J_0 within the molecular field approximation for the spin system is about 1250 K, about 1.5 times higher than the experimental value of 855 K. The characteristic feature observed in the DOS in the ordered state can be also retained in the MQSDW structures in the disordered state, which means that an antiferromagnetic long-range order remains in the γ-phase disordered alloy. The most stable structure is suggested to be the $3Q$ structure in the disordered state. The Néel temperature T_N, estimated from J_0, is about 680 K, close to the experimental value of about 700 K. Such a high value in comparison with that of pure γ-Mn metal is not related to the lattice expansion but to the change in the electronic structure caused by the addition of Rh.

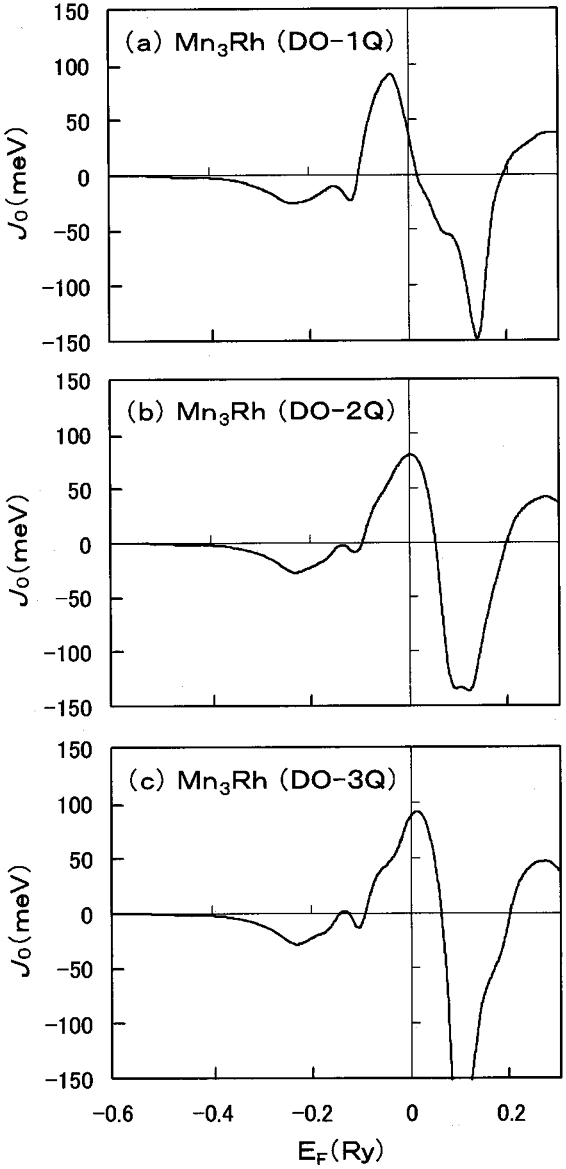

FIG. 8. Effective exchange constant J_0 of $\mathrm{Mn_3Rh}$ γ-phase disordered (DO) alloys as a function of the Fermi level E_F (band filling) for each magnetic structure. (a) $1Q$ structure. (b) $2Q$ structure. (c) $3Q$ structure.

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