Influence of structural disorder on the temperature dependence of the transport and magnetic properties of β -phase $\text{Co}_x \text{Al}_{1-x}$ alloy films

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The influence of the order-disorder structural transition on the magnetic and transport properties of Co-rich β -phase Co_xAl_{1-x} (x=0.54 and 0.62) alloy films has been investigated. The disordered state in these alloy films was obtained by vapor-quenching deposition onto glass substrates cooled in liquid nitrogen. The transport properties of the ordered and disordered Co_xAl_{1-x} alloy films have been measured between 2 and 300 K with and without a magnetic field of 0.5 T. The influence of the order-disorder structural transition on the temperature dependence of the resistance was discussed in the context of magnetic (magnetization, magnetic susceptibility, and ferromagnetic resonance) measurements, and analyzed in terms of the partial localization of the electronic states and variable-range hopping conductivity. [S0163-1829(99)03101-X]

I. INTRODUCTION

The electronic structures, physical properties, and thermal stability of 3d transition metal (TM) monoaluminides and gallides in connection with peculiarities of their crystalline structures have been focuses of many investigations during recent years. The transport properties of 3d TM monoaluminides and gallides were also widely investigated.^{1–12} Most of these investigations were concerned with the study of the concentration and temperature dependence of the resistivity of the bulk samples. Many of the results have the following common features: (i) the existence of a prominent resistivity minimum at low temperatures and (ii) a strong concentration dependence of the resistivity and the temperature coefficients of resistivity near the equiatomic composition (Refs. 5, 6, and 9 for Co-Al alloys, Refs. 8 and 12 for Fe-Al alloys, Ref. 10 for Ni-Al alloys, and Ref. 1 for Co-Ga alloys). There are different points of view on the nature of these phenomena, which are based on the peculiarities of either the magnetic properties or the electronic structures of these compounds.

 β -phase 3*d* TM-Al(Ga) alloys have a CsCl-type (*B*2-type) crystal structure in a wide concentration range of 3*d* TM.¹³ In perfectly ordered stoichiometric 3*d* TM-Al(Ga) compounds 3*d* TM and Al(Ga) atoms form interpenetrating primitive cubic lattices, where each 3*d* TM atom has eight Al(Ga) atoms as the nearest neighbor and vice versa. There is a general consensus that these compounds are not magnetic even at very low temperatures.

On the other hand, a slight atomic disorder or a minute compositional deviation from the equiatomic to the 3d TM rich one leads to the occurrence of local magnetic moments. It is widely believed that these moments are induced from so-called antistructure 3d TM atoms (3d TM-ASA)—3d TM atoms at Al sites. Indeed, the appearance of one 3d

TM-ASA results in the formation of a nine-atom 3d TM cluster which can carry a magnetic moment. Taking into account the similarity between the physical properties of 3d TM aluminides and gallides, and the results of Wachtel, Linse, and Gerold¹⁴ and Amamou and Gautier¹⁵ for the Co-Ga system, it can be supposed that the ferromagnetic behavior of the 3d TM-Al(Ga) alloy arises essentially from interactions between at least a couple of 3d TM-ASA's. Therefore, the 3d TM-ASA's or their clusters can be considered as scattering centers for free electrons.

According to Sellmyer, Caskey, and Franz the resistivity minimum (T_{\min}) at low temperatures of the nearly equiatomic Co-Al bulk alloys is related to the magnetic nature and could be explained by the following. (i) Kondo effect: s-d scattering of the conduction electrons from noninteracting (extreme dilution limit) localized magnetic moments, (ii) interacting moments: s-d scattering of the conduction electrons from localized magnetic moments which are coupled with themselves, for example, by an indirect exchange interaction, and (iii) spin fluctuations: a hypothetical scattering of the conduction electrons from excitations which are associated with the incipient ferromagnetism of the whole crystal at low temperatures. Zayer, Neumann, and Ziebeck⁸ also thought that a T_{\min} at about 50 K of Fe_xAl_{1-x} alloys with x = 0.54 - 0.73 is caused by a ferromagnetic to spin-glass magnetic phase transition, since an appearance of randomly oriented magnetic moments below the freezing temperature $(T_f:$ a temperature below which magnetic moments are frozen with random orientations and without long-range order) in the spin-glass state can be considered as additional scattering centers for free electrons. An observed prominent cusp at 30 K in the temperature dependence of the ac susceptibility allows the identification of a T_f of 30 K. The closeness of T_{\min} and T_f was suggested as evidence for the magnetic ori-

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gin of the resistivity minimum.8

The temperature dependence of scattering from magnetic inhomogeneities reflects that of magnetic transformation. The magnetic phase diagrams^{1,16} as well as results on the temperature dependence of the magnetic properties of 3dTM-Al(Ga) alloys^{17,18} show that various magnetic phase transitions such as "paramagnetic→spin glass," "paramagnetic-ferromagnetic-cluster spin glass," and "paramagnetic→ferromagnetic→reentrant ferromagnetic" are possible as the temperature is lowered. At a very low concentration (about 5%) of the 3d TM-ASA T_f is directly proportional to the concentration of the magnetic impurities and below T_f the material becomes a canonical spin glass. In a more concentrated region a mictomagnetism or cluster spin glass is found. Beyond this concentration point of the 3dTM-ASA an inhomogeneous reentrant region with ferromagnetic or antiferromagnetic long-range order appears. Therefore, each of these processes at low temperatures induces additional scattering centers for free electrons and could be considered as the origin of an exponential growth of the resistivity at low temperatures.

On the other hand, the electronic structures of 3d TM monoaluminides (gallides) have been studied extensively by both theoretical and experimental methods.^{1,19–36} The general features of all the calculated dispersion curves for 3d TM monoaluminides (gallides) are reflected in the density-of-states (DOS) curves. All the DOS curves have two intense maxima mostly due to *d* electrons, which are separated by a narrow and deep minimum (pseudogap). The Fermi level position depends on the 3d TM and is located on the steep slope of one of the peaks. Generally the dispersion curves are very flat near the Fermi level. This implies a high effective mass which is a possible reason for a high resistivity of these alloys. It can be expected that a change in alloy composition leads to a change in the Fermi level position and hence in the transport properties of the alloy.

In contrast to the aforementioned magnetic origin of the resistivity minimum, Whittle *et al.*¹ suggested that an exponential decrease in resistivity with temperature in the low-temperature range is of nonmagnetic origin. That is, this behavior is a direct consequence of the band structure of the alloy.¹ This term can be quite well explained with hopping or tunneling conduction between localized states, while the overall temperature coefficient of resistivity (sometimes negative) is modelled by incorporating the Debye-Waller factor into Mattheisen's rule.

The resistivity of thin film samples of 3d TM aluminides exhibits some peculiarities which depend on the film thickness. For example, the temperature dependence of the resistivity for ultrathin (about 5 nm) Co-Al single-crystalline films grown by molecular beam epitaxy between AlAs and GaAs layers shows a T_{min} at about 5 K,⁹ which is lower than that of the bulk Co-Al samples (about 45 K).⁶ The resistivity minimum in β -phase Co-Al alloys was explained by the Kondo effect in Ref. 9, where it was also emphasized that transport measurements of the ultrathin Co-Al films revealed a significant reduction of Kondo temperature, compared with that of bulk Co-Al, due to a difference in the DOS and Fermi surface between three-dimensional bulk and quasi-twodimensional ultrathin film. The thickness dependence of the resistivity minimum for β -phase Ni-Al alloy films buried between GaAs and AlAs is opposite to that of the Co-Al alloy films—the thinner the Ni-Al film the higher T_{min} , for which an increased interface roughness is responsible.¹⁰

A difference in the compositional dependence of the resistivity minimum between bulk and thin film 3d TM-Al alloys was also observed. For example, a T_{min} at 50 K for Fe_xAl_{1-x} alloy films is nearly independent of the Al content for x between 0.54 and 0.73.⁸ In contrast, a decrease in x from 0.885 to 0.731 for bulk Fe_xAl_{1-x} alloys leads to a shift in T_{min} from 15 up to 22 K. Thus, it is clear that there is no common point of view yet on the nature of the resistivity minimum at low temperatures of 3d TM aluminides and gallides.

It is well known that the electronic structure and magnetic properties of 3d TM aluminides and gallides strongly depend on their crystalline structure. It can be expected that the introduction of a structural disorder into 3d TM aluminides changes the magnetic state and/or induces additional localized electronic states. Taking into account the very steep energy dependence of the DOS near the Fermi level position, it is also expected that the concentration dependence of the transport properties is significant. To the best of our knowledge, the influence of the structural disorder on the resistivity of 3d TM monoaluminides has not been investigated yet. This work is devoted to a study of the structural and compositional dependence of the transport and magnetic properties of β -phase Co-Al alloy films.

II. EXPERIMENTAL PROCEDURE

Co-rich β -phase Co_xAl_{1-x} alloys of two compositions were prepared by melting Co and Al pieces of 99.99% purity in an arc furnace with a water cooled Cu hearth. In order to obtain volume homogeneity the ingot was remelted twice and then annealed at 1000 K for 6 h.

 $Co_x Al_{1-x}$ alloy films with a total thickness of 100–150 nm were prepared by flash evaporation of the crushed alloy powders of 80–100 μ m diameter onto glass and singlecrystalline Si substrates in a high vacuum of 5×10^{-5} Pa. The deposition rate was about 2 nm/sec. Such a high deposition rate allows the reduction of the volume contamination and oxidation. An equilibrium ordered β -phase in these films was reached by the deposition onto heated substrates at 720 K. In order to obtain a disordered state in the film, vaporquenching deposition was employed, where the gas is condensed onto substrates cooled by liquid nitrogen. The substrate temperature during the deposition was maintained at ~150 K.

X-ray fluorescence analysis on the prepared samples of both compositions revealed that x is equal to 0.54 and 0.62, respectively. Structural analysis of the films was performed by transmission electron microscopy (TEM) on films of 60–100 nm thickness, prepared under similar conditions on fresh chips of single-crystalline NaCl.

The resistance measurements were carried out with the physical properties measurement system of the Quantum Design company by using the four probe technique in a temperature range of 2-300 K with and without an external magnetic field of 0.5 T, directed in the film plane. The magnetic properties of the films were investigated by measuring the ac magnetic susceptibility at 200 Hz in the temperature



FIG. 1. Transmission electron diffraction patterns of the $Co_{0.54}Al_{0.46}$ alloy film deposited at (a) 720 K and (b) 150 K, respectively.

region of 4–100 K. The field dependence of the magnetization for the disordered and ordered $\text{Co}_x \text{Al}_{1-x}$ alloy films was measured using a vibrating sample magnetometer (VSM) at room temperature (RT) in a magnetic field up to 1.8 T for in-plane geometry. The ferromagnetic resonance (FMR) of the films was also studied at 9.3 GHz and RT in a parallel configuration.

III. RESULTS AND DISCUSSION

According to the results of the TEM study the film deposition onto glass substrates at 720 K leads to the formation of the ordered alloy structure with a mean grain size of about 30 nm. Only B2 structures are detected in the TEM patterns: a series of additional superstructure rings, which are attributed to the reflections from the (100), (111), and (210) atomic planes, are clearly seen (see Fig. 1). This result shows that a stable phase of the superlattice is formed under our deposition conditions, even though the degree of long-range order (η) cannot be estimated quantitatively. We note that we obtained this result for an alloy composition which slightly exceeds the threshold for the existence of the β phase, predicted by the equilibrium phase diagram for the bulk Co-Al system. A decrease in the substrate temperature to 150 K leads to the formation of considerably disordered ($\eta \sim 0$) polycrystalline alloy films with a mean grain size of 10–15 nm, which is identified with diffraction rings of the bcc lattice only.

The in-plane magnetization loops M(H)'s normalized with respect to the film volume, of ordered and disordered Co_xAl_{1-x} (x=0.54 and 0.62) alloy films exhibit ferromagnetic behavior, and nearly the same field dependence: an abrupt growth of the magnetic moment in a rather narrow region of magnetic field up to saturation and then a slight growth of the moment with field (see Fig. 2). It can be supposed that this growth is caused by paraprocess or concerned with an induced magnetic moment by the external magnetic field. The transition of the Co_xAl_{1-x} alloy films from the ordered to disordered states causes an increase in magnetization from 259–424 emu/cm³ and 446–550 emu/cm³ for x= 0.54 and 0.62, respectively.

A similar situation is observed in the FMR spectra of the ordered and disordered Co_xAl_{1-x} alloy films (see Fig. 3). The FMR spectra for the ordered alloy films exhibit broad

absorption lines at a resonance field of 296 and 225 mT for x=0.54 and 0.62, respectively. The transition to the disordered state leads to a shift of the resonance magnetic field to a lower field of 221 and 145 mT for x=0.54 and 0.62, respectively. A simple calculation for the parallel configuration of FMR reveals that $4 \pi M_{\text{eff}}$'s are equal to 122 mT (x=0.54) and 250 mT (x=0.62) for the ordered states, and to 248 mT (x=0.54) and 560 mT (x=0.62) for the disordered states. It is seen that the obtained results correlate qualitatively with M(H) data by VSM. As far as the magnetic moments in the Co-rich β -phase $\text{Co}_x \text{Al}_{1-x}$ alloys result from the Co-ASA (Co clusters), such a behavior of M(H) indicates that the structural disordering forms more Co-ASA than in the ordered state.

The temperature dependence of the resistance of the ordered and disordered Co_xAl_{1-x} (x=0.54 and 0.62) alloy films normalized with respect to the resistance at RT is presented in Figs. 4 and 5, respectively. The following regularities for both the investigated Co_xAl_{1-x} alloy films are observed: (i) all the Co_xAl_{1-x} alloy films exhibit a prominent resistance minimum in the low-temperature region, (ii) the temperature dependence of the normalized resistance for both alloy films in the ordered state shows a convex curvature in a region of 50–300 K, while for the disordered state a concave curvature is typical, and (iii) the structural disorder



FIG. 2. Magnetization loops of $Co_x Al_{1-x}$ films with x = 0.54 (1, 2) and 0.62 (3, 4) for the ordered (1, 3) and disordered (2, 4) states.



FIG. 3. FMR spectra of the Co_xAl_{1-x} film with x = 0.54 (a) and 0.62 (b) for the ordered (dashed-dotted lines) and disordered (solid lines) states.

shifts the resistance minimum to a higher temperature.

We emphasize that there is a certain compositional dependence of the observed phenomena. An increase of the Co content in the $\text{Co}_x \text{Al}_{1-x}$ alloy films results in a shift of the resistance minimum to lower temperatures. Namely, T_{\min} of the ordered and disordered $\text{Co}_{0.54}\text{Al}_{0.46}$ alloy films is equal to



FIG. 4. Variation of the normalized resistance with temperature in zero external field for the ordered (a) and disordered (b) $Co_{0.54}Al_{0.46}$ alloy films. Insets show the temperature derivatives of the resistance.



FIG. 5. Variation of the normalized resistance with temperature in zero [(a),(b)] and 0.5 T (c) external magnetic fields for the ordered (a) and disordered [(b),(c)] Co_{0.62}Al_{0.38} alloy films. Insets show the temperature derivatives of the resistance.

13.70 and 56.03 K, while for $Co_{62}Al_{38}$ these values are equal to 9.36 and 26.82 K, respectively.

The various scattering mechanisms which contribute to the total resistivity can be expressed as (Matthiessen's rule)

$$\rho(d,T) = \rho_{bd} + \rho_{if}(d) + \rho_{ep}(T) + \rho_{ee}(T) + \rho_{em}(T),$$

where ρ_{bd} is due to bulk defect scattering, $\rho_{if}(d)$ to interface roughness scattering, $\rho_{ep}(T)$ to electron-phonon scattering, $\rho_{ee}(T)$ to electron-electron interactions, and $\rho_{em}(T)$ to the spin-dependent scattering on magnetic impurities. ρ_{bd} depends on the structural state of the alloy film, but is independent of temperature. Considering that the electron mean free path in such a film is much shorter than the film thickness (*d*) and does not increase significantly with decreasing temperature in a highly disordered material, we believe that the $\rho_{if}(d)$ contribution to the total resistivity is negligible in our case.

Let us analyze the temperature-dependent contributions to the total resistivity $\rho_{ee}(T)$, $\rho_{ep}(T)$, and $\rho_{em}(T)$. Although $\rho_{ee}(T)$ is proportional to T^2 , its contribution is very weak compared to the other temperature-dependent terms, and hence it can be safely excluded from consideration.³⁷

According to Hopfield's electron-phonon scattering rate,³⁸ the temperature dependence of ρ_{ep} must be linear if $T > \Theta_D/10$ where Θ_D is the Debye temperature; that is,

$$\rho_{ep}(T) = \frac{2 \pi m_b k_B T}{e^2 \hbar N},$$

where m_b is the weighted average of the effective mass over all bands, N is the total number of charge carriers, and k_R , e, and \hbar are the Boltzmann constant, electron charge, and Planck's constant divided by 2π , respectively. However, the experimental R(T) dependencies show a very significant deviation from the linearity for the ordered as well as disordered $Co_x Al_{1-x}$ alloy films. These deviations are more clearly seen in the $\partial R / \partial T$ functions (see insets of Figs. 4 and 5). The deviation from linearity at high temperatures is common in many metals. Mott and Jones³⁹ and Dugdale⁴⁰ explained this deviation by $s \rightarrow d$ transitions. For TM the d electrons are significantly localized, and the current is mostly carried by the *s* electrons. However, the resistance is determined mainly by the scattering processes in which electrons jump from the s band to the d band.³⁹ The scattering rate is not constant over the equienergy surfaces in the range of $k_B T$ centered at the Fermi level and the resistivity is proportional to $(1 - AT^2)$ where A depends on $dN_d(E)/dE$, $N_d(E)$ being the DOS of the *d* band.

For $T < \Theta_D/20$ the scattering of charge carriers by lattice vibrations is proportional to T^5 . It is clear that this kind of scattering can not be responsible for the resistivity growth with decreasing temperature. Incorporating the Debye-Waller factor in ρ_{ep} can explain the negative temperature coefficient of the resistivity, but does not elucidate the exponential growth of *R* with decreasing temperature in the lowtemperature region.¹

Let us consider the $\rho_{em}(T)$ term. The Kondo effect cannot be accepted as a reason for the resistivity minimum of the investigated β -phase Co_xAl_{1-x} alloy films, because they are ferromagnetically ordered at RT. According to Sellmyer, Caskey, and Franz⁶ in nearly equiatomic Co-Al bulk alloys whose Co concentrations are more than 0.6% above the stoichiometric point, the ferromagnetic interactions between the Co-ASA's become apparent so that the resistance minimum disappears. This process is also accompanied by a change in curvature of the R(T) dependence from concave to convex type. In a view of the ferromagnetism below RT of the bulk ordered $Co_x Al_{1-x}$ alloy investigated by Sellmyer, Caskey, and Franz,⁶ such a behavior of the R(T) dependence may be explained in terms of spin-disorder scattering as discussed by Coles.⁴¹ Although there is a correspondence in the Cocontent dependence of T_{\min} between Ref. 6 and the present work (the higher the Co concentration in the nearly equiatomic $Co_x Al_{1-x}$ alloys, the lower the temperature of the resistance minimum), the nature of these phenomena, to our mind, seems different.

Figure 6 presents the temperature dependence of the imaginary part (χ'') of the ac magnetic susceptibility for the disordered Co_{0.62}Al_{0.38} alloy films. It is seen that there is a prominent cusp near 10 K. The resistance minimum for the disordered Co_{0.62}Al_{0.38} alloy films is located close to this



FIG. 6. Variation with temperature of the imaginary part of the ac magnetic susceptibility for the disordered $Co_{0.62}Al_{0.38}$ alloy film. The ac frequency was equal to 200 Hz and its amplitude was 10 Oe. The dc field was 0 Oe.

temperature. This result suggests the magnetic origin of this phenomenon. The aforementioned compositional dependence of the resistance minimum is also consistent with the magnetic point of view for the origin, if the investigated Co_xAl_{1-x} alloys fall into an interval for "ferromagnetic—cluster spin glass" transition in the magnetic phase diagram.

Therefore, if the nature of the low-temperature upturn and the resistance minimum of the Co_xAl_{1-x} alloy film is magnetic in the origin, it can be expected that near T_{min} the "ferromagnetic—cluster spin glass" transition is observed. If T_{min} is coincident with T_f , this temperature corresponds to a boundary between stable and unstable regions in the T-Hdiagram (the de Almeida-Thouless line).⁴² According to Ref. 42 the high field asymptote of this line can be described as

$$\ln T = a - bH^2.$$

For the Ising model of a system of two-component spins with elementary moments $\mu = \mu_B$, $a = C \ln(J/k_B)$ and $b = \mu_B^2/(2J^2)$, where *C* is a constant, and $J = k_B T_{H_0}$ is an exchange integral of the intercluster interaction. T_{H_0} is a value for *T* at H=0. According to this equation, the higher the magnetic field, the lower the temperature of the magnetic phase transition.

The influence of the magnetic field on the resistance of the disordered $\text{Co}_{0.62}\text{Al}_{0.38}$ alloy films is illustrated in Fig. 5(c). It is seen unexpectedly that T_{\min} is not lowered in the presence of the external magnetic field. T_{\min} at H=0 is about 25 K, while T_{\min} at H=0.5 T is near 29 K. It should also be noted that the curvatures of the R(T) dependence for the $\text{Co}_{0.62}\text{Al}_{0.38}$ alloy films with and without external magnetic field are nearly the same.

This makes us consider another possible origin of the resistance minimum. $Co_x Al_{1-x}$ alloys are highly resistive ones. This property arises from the peculiarities of the electron energy structure. It was mentioned above that the Fermi level for the CoAl compound is located on the left side of a narrow intense peak mainly due to the Co sites near the deep minimum (pseudogap) of the DOS curve, which separates bonding and antibonding peaks of *d* character. Mott and Davis⁴³ have stated that if $N(E_F)$ is less than about one third of the free electron value, conduction at low temperature is thermally activated hopping or tunneling, and shows an exponential dependence with temperature. At very low temperature the tunneling to the nearest neighbors becomes severely reduced and the variable-range hopping, in which the tunneling to the states with lower electronic energy, but spatially separated, is more plausible, becomes the main transport mechanism. For this kind of conductivity ($\sigma = 1/\rho$) the temperature-dependent contribution to Matthiessen's rule can be expressed as follows:⁴³

$$\sigma(T) = \sigma_0 \left(\frac{T_0}{T}\right)^p \exp\left[-\left(\frac{T_0}{T}\right)^{1/4}\right], \qquad (1)$$

where σ_0 is a constant, *p* is the exponent of the preexponential term, and T_0 can be found from the slope of the $\ln(R/T^p)$ vs $T^{-1/4}$ curve. Both σ_0 and T_0 depend on the localization length of the electrons involved in conduction at low temperature. Various theories show that *p* could be 0.5,⁴⁴ 0.25,⁴⁵ and 0.35.⁴⁶

Since the normalized resistance curves show the minima, we can determine the numerical value of p from the relationship between T_{\min} and T_0 using an iterative method. By differentiating the resistivity [the inverse of Eq. (1)] with respect to temperature we can obtain

$$T_{\min} = \frac{T_0}{(4p)^4}$$

For the first iteration we assume p=0 and find slope T_0 in $\ln R$ vs $T^{-1/4}$ plots using the linear regression method. Using T_0 and T_{\min} we can obtain p. This is used for the next iteration and determines the slope in the $\ln(R/T^p)$ vs $T^{-1/4}$ plot. The newly found T_0 and T_{\min} give a new value of p and, if the difference between the new and old p's satisfies the certain criterion $(|p^{\text{new}} - p^{\text{old}}| < 1 \times 10^{-6})$, the iteration stops, otherwise, p^{new} is used for the next iteration to determine the slope in the $\ln(R/T^p)$ vs $T^{-1/4}$ plot and iteration continues. Since the resistance change below T_{\min} is relatively small for our samples, the slope T_0 is small too. We found that the numerical values of p's are in a range of 0.004–0.009, which are so small that the inclusion of the pre-exponential term in the fitting procedure does not significantly alter the results. Therefore, we can ignore the temperature dependence of the pre-exponential term in the fitting below T_{\min} and Figs. 7 and 8 show the ln R vs $T^{-1/4}$ plotting for the investigated ordered and disordered $Co_x Al_{1-x}$ alloy films at low temperatures. However, we do not think that the physical origin of the resistance minimum is due to the pre-exponential term of Eq. (1). Rather, we argue that the resistance minimum occurs because there is a competition between electron-phonon interaction and the variable-range hopping in the lowtemperature region.

This kind of conductivity at low temperature is due to the hopping between localized states near the Fermi level. Recently, Lamba, Rastogi, and Kumar⁴⁷ applied the theory of Anderson localization to the cluster compound and successfully explained the localization of the states near the Fermi level and the occurrence of ferromagnetism due to localization. The authors gave three conditions for the localization of the electrons near the Fermi surface: (i) the band character at



FIG. 7. $\ln R$ vs $T^{-1/4}$ for the (a) ordered and (b) disordered $\operatorname{Co}_{0.54}\operatorname{Al}_{0.46}$ alloy films. The low-temperature parts are approximated by straight lines which are plotted by the least square fit.

the Fermi level is dominantly d type, (ii) the bands at the Fermi level are very narrow, and (iii) they disperse only along one family of crystallographic directions.⁴⁷ Indeed the preliminary band structure calculations⁴⁸ using a tightbinding linearized-muffin-tin-orbital method revealed the fact that the band structure of the ordered equiatomic CoAl alloy satisfies the above three conditions (see Fig. 9). Furthermore, the Fermi level is located at the rising edge of the deep minimum of the DOS curve, which separates the bonding and antibonding states (see Fig. 9) and gives a negative temperature coefficient.⁴⁰ The band character of the DOS at



FIG. 8. $\ln R$ vs $T^{-1/4}$ dependencies for the (a) ordered and [(b),(c)] disordered $Co_{0.62}Al_{0.38}$ alloy films in (b) zero and (c) non-zero (H=0.5 T) magnetic field. The low-temperature parts are approximated by the straight lines which are plotted by the least square fit.



FIG. 9. Total density of states of CoAl (dashed line) and Co_9Al_7 (solid line). The Fermi level is located at 0 energy.

the Fermi level is mainly a t_{2g} -like one,⁴⁸ which is the most likely to be localized among the d characters. Therefore, the states near the Fermi level in Co_rAl_{1-r} alloy films can be easily localized even in the case of slight disorder. To confirm this fact we also calculated the electronic structures of nonequiatomic Co_5Al_3 ($Co_{0.5625}Al_{0.4375}$) and Co_9Al_7 $(Co_{0.625}Al_{0.375})$ alloys using a supercell method similar to that of Ref. 49. The calculation was spin polarized, and the result shows that the alloy is ferromagnetic with a magnetic moment of 1.609 μ_B for the Co-ASA, which is much greater than that of the neighboring eight Co atoms $(0.0226\mu_B \text{ per}$ atom). The detailed calculation results will be published in a separate paper.⁴⁸ The calculated DOS curve shows a narrow and sharp peak very close to the Fermi level, and the band character of the peak is mainly a d type of minority spin, from both the Co-ASA and its neighboring eight Co atoms (see Fig. 9). This is another signature for the localization of states near the Fermi level due to structural disorder. Therefore, we argue that the upturn of the resistance at low temperatures is caused by variable-range hopping, in which the hopping mechanism dominates the charge transport.

Mott and Davis⁴³ postulate that if some states at a given energy are localized, then for that energy they must all be localized. Localized and nonlocalized states cannot coexist at the same energy for a given configuration.⁵⁰ Therefore, one can have a finite and continuous DOS in which all states are localized.⁴³ It is natural to suppose that the structural disorder in the investigated alloys leads to a broadening of the energy interval with localized electronic states, in other words, to a higher $T_{\rm min}$. It is then expected that in the ordered alloy, variable-range hopping dominates at lower temperatures than the disordered one. The experimentally observed behavior of the temperature dependence of the resistance for the Co_xAl_{1-x} alloy films agrees with the expectation, and can serve as evidence for the hopping conductivity mechanism.

IV. SUMMARY

(1) The disordered state in $Co_x Al_{1-x}$ (x=0.54 and 0.62) alloy films was obtained by vapor-quenching deposition onto glass substrates cooled by liquid nitrogen. (2) It was shown experimentally that the loss of translational invariance in the disordered state leads to significant changes in the magnetic and transport properties of the $Co_x Al_{1-x}$ alloy films. (3) The observed temperature and structural dependencies of the resistance minimum and the exponential $T^{-1/4}$ dependence of the resistance at low temperature in the investigated $Co_x Al_{1-x}$ alloy films can be explained in terms of the partial localization of the electronic states and variable-range hopping conductivity. (4) The significant deviation from the linearity of the R(T) dependence for the ordered and disordered $Co_x Al_{1-x}$ alloy films can be explained by $s \rightarrow d$ scattering.

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