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$Fe_{1-x}Rh_x$ layers are grown with varying rhodium fraction x on (001)-oriented MgO substrates by molecular-beam epitaxy. Film structural, morphological, magnetic, and transport properties are investigated. At room temperature, layers are ferromagnetic (FM) for $x < 0.48$ and antiferromagnetic (AF) for $x > 0.48$. Separating the two magnetically ordered phases at $x = 0.48$ is an abrupt change in the $Fe_{1-x}Rh_x$ lattice parameter of $\Delta a = 0.0028$ nm ($\Delta a/a = -0.9\%$). For AF layers, the FM state is recovered by heating across a first-order phase transition. The transition leads to a large resistivity modulation, $\Delta\rho/\rho = 80\%$, over a narrow temperature range, $\Delta T = 3$ K, in stoichiometric $Fe_{0.50}Rh_{0.50}$/MgO(001). For samples with compositions deviating from $x = 0.50$, fluctuations broaden ΔT and defect scattering reduces $\Delta\rho/\rho$. *Published by AIP Publishing.*
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FeRh ($Pm\bar{3}m$, B2, CsCl structure) is a fundamental component in memory cells,[1,2] magnetocaloric refrigerators,[3,4] and logic devices.[5,6] Its diverse functionality stems from an entropy-driven first-order transition[7] between ferromagnetic (FM) and antiferromagnetic (AF) states which persists when deposited as films—a prerequisite for integration in device heterostructures. Accompanying the intrinsic magnetic transition is a large resistivity modulation which rivals giant magnetoresistance effects observed in magnetic multilayers.[8,9] The rhodium fraction x is suspected to strongly affect $Fe_{1-x}Rh_x$ transport characteristics, but its role has not yet been systematically investigated in epitaxial films. Instead, work has been focused on understanding size effects,[10,11] annealing treatments,[12–15] and transition mechanics.[16–20] The few compositional studies on $Fe_{1-x}Rh_x$ films omit transport properties entirely, emphasizing magnetic attributes,[21] or are based on inhomogeneous polycrystalline layers containing secondary phases.[22] Here, we systematically examine the structural, morphological, magnetic, and transport properties as a function of rhodium fraction x of phase-pure epitaxial $Fe_{1-x}Rh_x$ films with the CsCl structure deposited on (001)-oriented MgO substrates.

$Fe_{1-x}Rh_x$/MgO(001) films are grown via molecular-beam epitaxy to a thickness of ~35 nm in a Veeco GEN10 system (base pressure: 1×10^{-8} Torr $= 1.3 \times 10^{-6}$ Pa) by simultaneously supplying iron (99.995% pure) and rhodium (99.95% pure) from independent effusion cells. Rhodium fractions x are controlled by adjusting iron and rhodium cell temperatures within 50 °C of 1150 and 1600 °C, respectively, while maintaining a total atomic flux of $\sim 4 \times 10^{13}$ atoms/cm^2 s, corresponding to a growth rate of ~0.3 nm/min. x values determined[24] from Rutherford backscattering spectra agree with x-ray reflectivity (XRR) deposition rate calibrations based on pure iron and rhodium layers (linear correlation coefficient $r = 0.997$), demonstrating that atomic incorporation probabilities are unaltered by chemistry. From the calibrated atomic fluxes, deposition times are set to produce layers with a thickness of ~35 nm. A substrate temperature $T_s = 420$ °C (estimated from a thermocouple in indirect contact with the growth surface and concealed from incident molecular fluxes) is employed for film growth and subsequent 30-min-long *in situ* anneals. High homologous growth temperatures ($T_s/T_m = 0.37$ for FeRh with melting temperature $T_m \approx 1600$ °C) are necessary[25] to order bcc $Fe_{1-x}Rh_x$ alloys into the B2 CsCl-structure intermetallic with iron and rhodium residing on distinct positions of the two-atom basis.

X-ray diffraction (XRD) θ–2θ scans, collected using Cu $K_{\alpha 1}$ radiation (wavelength $\lambda = 0.154056$ nm), establish a phase diagram consisting of four regions: single-phase bcc-Fe(001) ($x \lesssim 0.20$), single-phase B2 $Fe_{1-x}Rh_x$ ($0.20 \lesssim x \lesssim 0.60$), two-phase mixtures of (001)-textured B2 $Fe_{1-x}Rh_x$ and fcc-Rh ($0.60 \lesssim x \lesssim 0.80$), and single-phase fcc-Rh(001) ($x \gtrsim 0.80$). The phase boundaries of our epitaxial films grown on MgO(001) are in close agreement with reports for bulk samples:[23,26] the rhodium-deficient limit, for which the bcc solid solution orders into the CsCl structure, agrees exactly, while the rhodium-rich limit extends 0.08 rhodium fractions above the bulk boundary ($x = 0.52$) due to epitaxial stabilization.[27–31]

A representative XRD θ–2θ scan is presented in Fig. 1(a) for stoichiometric $Fe_{0.50}Rh_{0.50}$/MgO(001). Five peaks are observed over the 2θ range 10–110°: the three reflections at $2\theta = 29.94$, 62.18, and 101.6° are indexed as $Fe_{0.50}Rh_{0.50}$ 00l; the two peaks at 42.92 and 94.05° are identified as MgO 00$2l$. Sharp mixed-integer film reflections (no systematic absences) indicate CsCl-type ordering. The lack of additional reflections together with pole figure and grazing-incidence scans (not shown) establish that films with $0.20 \lesssim x \lesssim 0.60$ are phase-pure untwinned epitaxial layers oriented with a 45° in-plane

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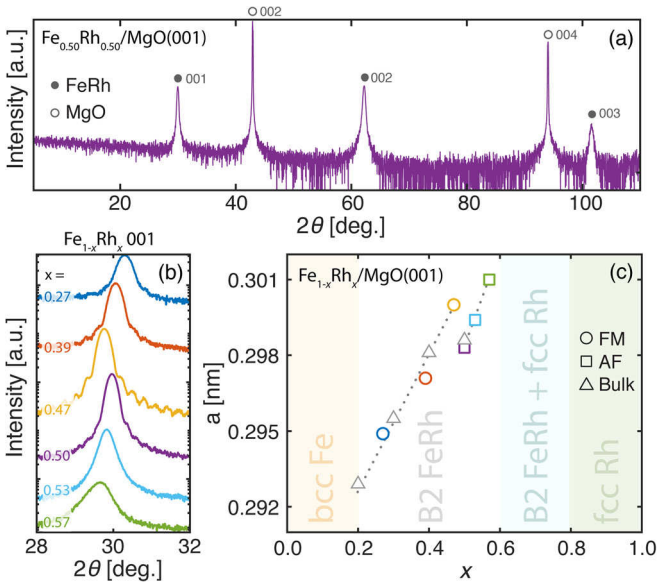

FIG. 1. (a) XRD θ-2θ scan of an ~35-nm-thick stoichiometric $Fe_{0.50}Rh_{0.50}$ film with the B2 CsCl-structure grown on MgO(001) at 420 °C by molecular-beam epitaxy. (b) θ-2θ scans showing the $Fe_{1-x}Rh_x$ 001 peak for rhodium fractions $0.20 \lesssim x \lesssim 0.60$. (c) Film out-of-plane lattice parameters (circles and squares) as a function of composition together with bulk lattice parameters[23] (triangles) for reference. Circles indicate ferromagnetic ordering and squares indicate antiferromagnetic ordering at room temperature.

rotation with respect to their MgO substrates: $(001)_{Fe_{1-x}Rh_x} \parallel (001)_{MgO}$ and $[110]_{Fe_{1-x}Rh_x} \parallel [100]_{MgO}$.

Diffracted intensities near the $Fe_{1-x}Rh_x$ 001 reflection are plotted as a function of x in Fig. 1(b). As x increases across the single-phase field, $Fe_{1-x}Rh_x$ peaks shift—with one exception—to lower 2θ angles. Figure 1(c) shows out-of-plane lattice parameter a values obtained[32] from θ–2θ peak positions. a increases approximately linearly from 0.2950 ($x = 0.27$) to 0.3000 nm ($x = 0.47$), contracts sharply to 0.2983 nm ($x = 0.50$), and then continues increasing to 0.3010 ($x = 0.57$). Film lattice parameters values $a(x)$ are in excellent agreement with reports for bulk polycrystals [also shown in Fig. 1(c)].[23] Regression analyses yield a slope of 0.04 ± 0.01 nm per rhodium fraction, in close agreement with 0.06 expected based on the larger metallic radius[33] of rhodium (134 pm) versus iron (126 pm), suggesting that rhodium substitutes for iron across the $Fe_{1-x}Rh_x$ single-phase field. The lattice parameter discontinuity of $\Delta a = 0.0028$ nm ($\Delta a/a = -0.9\%$) at $x = 0.48$ occurs as $Fe_{1-x}Rh_x$ undergoes a first-order transition[7] from a FM ($x < 0.48$) to an AF ($x > 0.48$) state.[34] The contracted AF cell corresponds to the new equilibrium geometry[35] after spins ferromagnetically aligned on iron (3.2 μ_B) and rhodium (0.9 μ_B) leave rhodium (0.0 μ_B) magnetically inactive and reorganize antiferromagnetically along {001} on iron (3.3 μ_B).[26,36,37]

The structural quality of the films is assessed from ω-rocking curves of $Fe_{1-x}Rh_x$ 001 reflections and atomic force microscopy (AFM) elevation maps. Rocking curve scans and corresponding peak full-width-at-half-maxima (FWHM) are plotted in Figs. 2(a) and 2(b). Reflections are broad at $x = 0.27$ and 0.57 due to crystalline mosaicity, but sharpen as x approaches 0.50. FWHM values decrease from 0.65° ($x = 0.27$) and 1.28° ($x = 0.57$) to 0.23° ($x = 0.47$) and 0.32° ($x = 0.50$) indicating increasing crystalline perfection. MgO 002 rocking curves, measured for reference, are found to consist of split peaks with individual peak FWHM values of ~0.005° (18 arcsec) and an ensemble FWHM of ~0.06° (216 arcsec); the splitting results from the formation of subgrains (separated by small-angle grain boundaries) and are commonly observed in commercial MgO substrates.[38]

Figure 2(c) depicts representative AFM height images. Root-mean-square surface roughness values ρ_{rms} determined independently from AFM and XRR (not shown) are plotted as a function of x in Fig. 2(d). At $x = 0.27$, the surface morphology ($\rho_{rms} = 3.0$ nm) is comprised of 150-nm-wide mesas separated by 1.5-nm-deep trenches preferentially aligned along $Fe_{1-x}Rh_x$ ⟨100⟩. Such features are the hallmark of unfavorable substrate wetting and three-dimensional island growth.[39] For $x = 0.47$, the mesas fuse leaving a smooth surface with sub-monolayer height fluctuations ($\rho_{rms} = 0.1$ nm). Further increasing x to 0.57 is accompanied by the appearance of mounds faceted along $Fe_{1-x}Rh_x$ ⟨100⟩ due to the combination of high surface energies and high diffusivities.[40,41] Thus, the smoothest films with the highest structural perfection are obtained near $x = 0.50$.

Figure 3(a) shows the in-plane room-temperature magnetization M of $Fe_{1-x}Rh_x$/MgO(001) films measured as a function of applied magnetic field H using a vibrating sample magnetometer. Films with $x \leq 0.48$ display hysteretic behavior characteristic of FM ordering with saturation magnetizations of ~4 μ_B/f.u., consistent with prior reports.[36] Coercive fields H_c, defined as the value of H where M changes maximally, decrease with increasing x from 235 ($x = 0.27$) to 129 ($x = 0.39$) and 59 Oe ($x = 0.47$). Fitting $H_c(x)$ with a

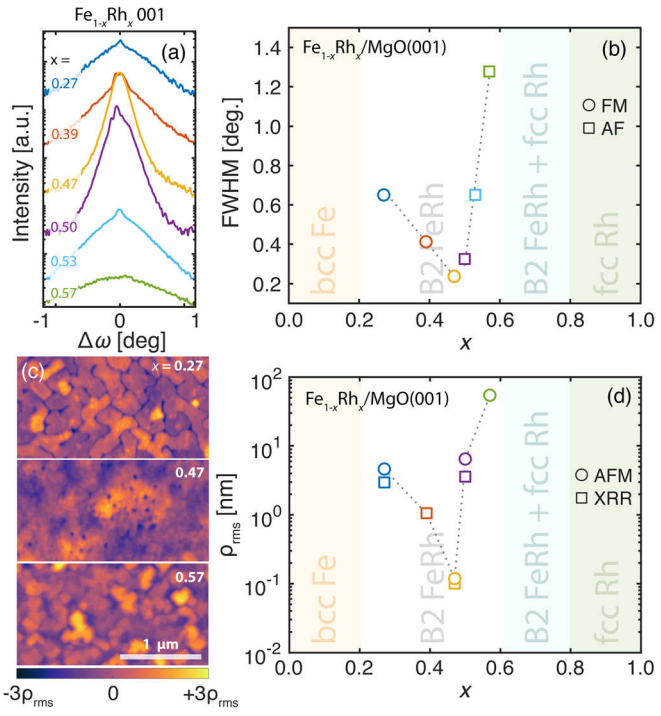

FIG. 2. (a) XRD ω-rocking curve scans of $Fe_{1-x}Rh_x$ 001 reflections and (b) corresponding FWHM values as a function of rhodium fraction x. (c) Representative AFM height images of $Fe_{1-x}Rh_x$/MgO(001) layers as a function of composition x across the B2 single-phase field. MgO[100] and $Fe_{1-x}Rh_x$[110] are aligned with the horizontal image axis. (d) Root-mean-square surface roughness values determined as a function of x independently using XRR and AFM.

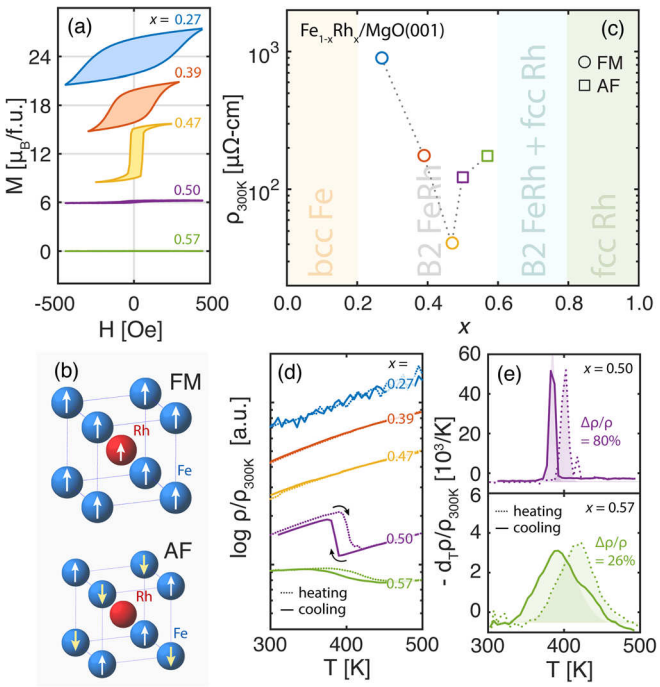

FIG. 3. (a) Magnetization M of $Fe_{1-x}Rh_x$/MgO(001) films versus applied magnetic field H as a function of rhodium fraction x. Curves are offset by 6 μ_B/f.u. for clarity. (b) The crystal structure and spin configurations of ferromagnetic (FM: $x < 0.48$) and antiferromagnetic (AF: $x > 0.48$) $Fe_{1-x}Rh_x$. (c) Room-temperature $Fe_{1-x}Rh_x$ resistivities $\rho_{300\,K}(x)$ for $x = 0.20$ through 0.80, spanning the B2 single-phase field. (d) Temperature-dependent resistivities $\rho(T)$ as a function of x; curves are vertically offset for clarity. (e) Negative temperature-derivative of $\rho(T)$ for samples exhibiting AF-FM transitions ($0.48 < x \lesssim 0.60$).

mean-field behavior, $H_c \propto \sqrt{x - x_c}$, yields a critical rhodium fraction of $x_c = 0.48$ below which $Fe_{1-x}Rh_x$ is FM. For x above x_c, $Fe_{1-x}Rh_x$ films are macroscopically demagnetized at room temperature, but recover their magnetization when heated above ~400 K. Since symmetries are necessarily restored by heating across any phase transition,[42] the loss of magnetization in films with $x > 0.48$ implies AF ordering, for which heating leads to the recovery of additional symmetry operations and the emergence of a FM state. These conclusions are in agreement with Mössbauer spectroscopy[26] and neutron scattering[36] results. The crystal structure and spin configurations of ferromagnetic and antiferromagnetic $Fe_{1-x}Rh_x$ are illustrated in Fig. 3(b).

Room-temperature resistivities $\rho_{300\,K}(x)$ of ~35-nm-thick $Fe_{1-x}Rh_x$/MgO(001) films are shown in Fig. 3(c). As x is varied across the single-phase field, $\rho_{300\,K}$ decreases from 898.3 $\mu\Omega$ cm ($x = 0.27$) to 40.9 $\mu\Omega$ cm ($x = 0.47$), rises rapidly to 122.3 $\mu\Omega$ cm ($x = 0.50$), and continues increasing slowly to 174.9 $\mu\Omega$ cm ($x = 0.57$). The resistivity obtained here for stoichiometric $Fe_{0.50}Rh_{0.50}$, which represents the lowest value reported in the literature,[43] reflects the structural perfection and chemical purity of the layer. The large $\rho_{300K}(x)$ values near the $Fe_{1-x}Rh_x$ phase field boundaries stem predominately from increased structural disorder.

Temperature-dependent $Fe_{1-x}Rh_x$ resistivities $\rho(T)$ between 300 and 500 K are plotted in Fig. 3(d). For rhodium-deficient films ($0.27 \leq x \leq 0.47$), $\rho(T)$ increase linearly with T demonstrating metallic phonon-limited conduction. The superposition of resistivity curves measured during heating and cooling reflect the stability of these layers in air. At $x = 0.50$, a drop in resistivity is observed near $T_c \approx 392$ K, associated with a transition[7] between AF ($T < T_c$) and FM ($T > T_c$) states. The negative temperature-derivative of $\rho(T)/\rho_{300\,K}$, plotted in Fig. 3(e), shows that the transition is sharp, hysteretic, and symmetric—attributes consistent with first-order transitions—and occurs at 385 ± 3 and 401 ± 3 K during heating and cooling, respectively. The pronounced modulation in resistivity observed, $\Delta\rho/\rho \equiv (\rho_{AF} - \rho_{FM})/\rho_{FM} = 80\%$, represents the highest thermally induced value reported[6,10,22,43–45] and is consistent with the 85 ± 6% theoretical maximum realizable for well-ordered films;[22] the narrow transition widths, $\Delta T = 3$ K, are the smallest observed to date.[6,10,11,22,43–45] For bulk stoichiometric samples, a comparable resistivity change was observed at room temperature by driving the AF-FM transition with pulsed magnetic fields exceeding 15 T; thermally induced resistivity changes were not investigated, but a T_c of 405 K, in close agreement with our measured values, was deduced from temperature-dependent heat capacity measurements.[46]

Rhodium-rich films with $x = 0.57$ also exhibit a similar transition. In this case, the resistivity changes by only 26% (versus 80% for $x = 0.50$) as AF regions slowly transform into FM domains at 418 ± 32 K and back at 393 ± 40 K [Figs. 3(d) and 3(e)]. The smaller $\Delta\rho/\rho$ values for $x = 0.57$ results from defect scattering, which simultaneously raises ρ_{AF} and ρ_{FM}. The broader transition stems from fluctuations, as expected for a film characterized by chemical disorder, crystalline mosaicity, and high surface roughness.

In summary, ~35-nm-thick epitaxial $Fe_{1-x}Rh_x$/MgO(001) films are grown at 420 °C by molecular-beam epitaxy and systematically investigated as a function of rhodium fraction x. Within the CsCl-structure $Fe_{1-x}Rh_x$ single-phase field ($0.20 \lesssim x \lesssim 0.60$), rhodium replaces iron producing a linearly increasing lattice parameter due to its larger metallic radius (134 versus 126 pm).[33] B2 CsCl-type ordering is evidenced by pronounced x-ray diffraction from mixed-integer film reflections. A lattice parameter discontinuity of $\Delta a = 0.0028$ nm ($\Delta a/a = -0.9\%$) is observed at $x_c = 0.48$, below (above) which films are FM (AF). The perfection and surface smoothness of the layers are optimized near $x = 0.50$. Room-temperature resistivities $\rho_{300\,K}(x)$ exhibit a minimum of 40.9 $\mu\Omega$·cm at $x = 0.47$. For AF layers ($x \geq 0.48$), FM ordering can be recovered by heating across the first-order phase transition. Temperature-dependent resistivity measurements demonstrate sharp, hysteretic, and symmetric transitions at 385 ± 3 K and 401 ± 3 K during heating and cooling of stoichiometric $Fe_{0.50}Rh_{0.50}$/MgO(001) films. The large resistivity modulation achieved, $\Delta\rho/\rho = 80\%$, represents the largest thermally induced value observed to date for $Fe_{1-x}Rh_x$ films. In rhodium-rich layers, the transition is broadened by fluctuations and the percent resistivity change is reduced due to defect scattering.

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