

Spin Hall effect in the high-resistivity high-entropy alloy Al-Cr-Mo-W

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We study thin films of the high-entropy alloy system $\text{Al}_x(\text{Cr-Mo-W})_{1-x}$, grown on Ta seed layers by magnetron cosputtering. Between $x = 0.2$ and $x = 0.6$, a resistivity larger than $100 \mu\Omega \text{ cm}$ is achieved, with a peak of $180 \mu\Omega \text{ cm}$ at $x = 0.5$. Around the stoichiometric composition Al-Cr-Mo-W, the alloy forms a bcc solid solution. The harmonic Hall method was used to characterize the spin Hall angle of the alloy series, where a maximum spin Hall angle of $\theta = -0.12 \pm 0.01$ is observed for $x = 0.25$. The implied spin Hall conductivity is $\sigma_{\text{SH}} \approx -72\,000 \hbar/(2e) \text{ S/m}$. The experimental results show excellent agreement with density-functional-theory calculations, which show similar trends and values. The results demonstrate that high-entropy alloys with a main-group-element component can form a simple crystal structure and show high resistivity. This suggests that a whole new class of materials for spin Hall device engineering is available with simple methods.

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I. INTRODUCTION

The spin Hall effect (SHE) converts a charge current density j into a transverse spin current density j_s , with the spin Hall angle (SHA) $\theta = j_s/j$ [1,2]. In cubic concentrated alloys and pure elements, the SHA is determined by the intrinsic contribution, which is derived from the band structure of the material [3]. It can be written as $\theta_{\text{SH}} = \sigma_{\text{SH}}\rho$, where σ_{SH} is the spin Hall conductivity (SHC) and ρ is the resistivity. Thus, a material with a large SHA requires a large SHC and high resistivity. The two elements with the highest SHC are Pt (positive SHC) and β -W (negative SHC) [4–6]. While Pt crystallizes with the fcc structure and thin films thereof exhibit resistivities below $50 \mu\Omega\text{cm}$, the β phase of W crystallizes with the A15 structure and shows resistivities well above $100 \mu\Omega\text{cm}$ [4,7–10]. Therefore, β -W routinely shows large SHA values in experiments. However, the β phase is metastable and depends critically on the growth conditions and the built-in defects (such as trapped oxygen or nitrogen) [11–14]. The stable bcc phase

α -W has both much smaller SHC and smaller resistivity [6]. However, bcc-W still has the largest SHC among all stable allotropes of the elements, with the exception of Pt. Alloying of W and related elements has been proposed to optimize the Fermi level and maximize the SHA [10,15]. It has been shown that the Fermi-level tuning of the SHC is possible with Ta-Re alloys, reproducing the SHC of W at intermediate stoichiometries [16]. Resistivity tuning by alloying is a straightforward method to enhance the resistivity and, thereby, the spin Hall angle; this has been predicted for multiple Pt alloys and demonstrated experimentally [17–20].

In the present study, we explore the spin Hall effect of a high-resistivity high-entropy alloy (HEA) based on W and the other elements of the same group. A high-entropy alloy forms a simple bcc or fcc crystal structure, which is mainly stabilized by the configurational entropy rather than formation enthalpy [21,22]. Typically, materials with at least four elements are referred to as HEAs. We refer the reader to the Appendix for a more complete description of what characterizes a HEA in detail. Only few reports on the SHE of HEAs have been published. An early report characterizes a Ta-Nb-Hf-Zr-Ti HEA, for which an SHA of $|\theta| \approx 0.033$ is reported [23]. However, in this report the material is amorphous and is thus not a proper high-entropy alloy by definition, where the entropy effect stabilizes a particular simple crystallographic phase (e.g., fcc or bcc).

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Amorphous metals usually have high resistivity and can thus yield a large spin Hall angle [16,24]. A recent study investigates Mn-Nb-Mo-Ta-W alloys, which are found to grow as a single bcc phase. Analysis of the spin Hall magnetoresistance indicates a spin Hall angle of $|\theta| = 0.12$ [25]. Another recent study shows a very large SHA in Ir-Ru-Al-Ga-based HEAs with a partially ordered tetragonal structure grown epitaxially on MgO. SHAs larger than $|\theta| = 0.6$ and huge resistivities of more than $200 \mu\Omega\text{cm}$ are reported for multiple compositions [26]. According to the findings of a recent theoretical high-throughput calculation study, HEAs with a main-group element should exhibit high resistivity, in excess of $100 \mu\Omega\text{cm}$ [27]. To form a bcc HEA with a large SHC and large resistivity, we start from an equiatomic mixture of W, Mo, and Cr. The same-group elements Mo and Cr have lower SHCs, but still the largest within their respective periods among the bcc elements [28]. By screening the main-group elements for suitable values of ΔH_{mix} , Ω , and δ (for the definitions, see the Appendix), we find that Al is a suitable candidate for HEA formation. While the binary Cr-W and Cr-Mo phase diagrams show miscibility gaps because of the atomic size mismatch, the other binary phase diagrams contain numerous line compounds and W-Mo forms a solid solution for all compositions. We obtain $\Delta H_{\text{mix}} \approx -8 \text{ kJ/mol}$, $\Omega \approx 3.32$, and $\delta \approx 4.3\%$. The valence-electron concentration (VEC) is 5.25, so formation of a bcc solid solution, similar to the elemental structures of Cr, Mo, and W, is expected.

II. METHODS

Thin films of the type seed layer (5 nm)/(Al-Cr-Mo-W) (20 nm)/Cr (1 nm) (with seed layer = none, Cr, Mo, W) and Ta (3 nm)/Al_x(Cr-Mo-W)_{1-x} (8 nm)/Co₄₀Fe₄₀B₂₀ (3 nm)/Ta (0.5 nm)/TaO_x (1 nm) were grown by room-temperature dc magnetron (co)sputtering with varying Al concentrations on commercially available Si/SiO₂ (200 nm) wafers. The TaO_x layer was formed by reactive sputter deposition in an Ar-O₂ gas mixture. Typical power levels of the 2-in. sputter sources were between 50 W and 70 W. The individual elemental particle fluxes from Al, Cr, Mo, and W targets were determined with a calibrated film-thickness sensor to obtain the nominal concentrations. The typical reproducibility errors between the film-thickness-sensor results and the calibrations is below 5%. The total growth rate was typically 0.3 nm/s and the Ar working pressure was 2×10^{-3} mbar. The base pressure of the deposition system was always lower than 5×10^{-8} mbar before the deposition runs. We measured the film resistivity using a standard four-point technique as $\rho_{\text{eff}} = kR_{4w}t_{\text{total}}$, a numerically determined correction factor $k \approx 2.7$ for samples of size $10 \times 10 \text{ mm}^2$, and a parallel conductor model to obtain the alloy resistivity.

X-ray reflectivity confirmed the thicknesses and the crystallization was monitored by x-ray diffraction (XRD) with a diffractometer with a Cu anode. Afterward, the samples were laterally patterned using photolithography, ion-beam etching, and lift-off lithography for contact lines. The spin Hall angle and spin Hall conductivity were determined with the harmonic Hall technique with magnetic fields up to 1 T, as described in our previous work [24]. We explicitly take the anomalous Nernst effect into account and correct for it. The parameters are obtained as averages over at least five Hall crosses per composition.

Additionally, ferromagnetic resonance (FMR) was used to determine the effective magnetization M_{eff} and the Gilbert damping parameter α . We used our OpenFMR broadband FMR system [29] for the detection of the FMR.

We computed the electrical resistivity, spin Hall conductivity, and the spin Hall angle of the Al_x(Cr-Mo-W)_{1-x} (ACMW) series using the spin-polarized and relativistic Korringa-Kohn-Rostoker (SPR-KKR) density-functional-theory code with the atomic spheres approximation (ASA). All calculations were performed at lattice constants given by Vegard's law and Debye temperatures for SHA calculations were similarly interpolated between the elemental values. The SHA and resistivity calculations were performed for $T = 300 \text{ K}$ with the alloy-analogy model [30]. Chemical substitution was treated with the coherent potential approximation (CPA) with a primitive bcc unit cell. Convergence of energy integration and Brillouin-zone integration were carefully tested. The Perdew-Burke-Ernzerhof generalized gradient approximation was used for the exchange-correlation functional. The angular-momentum cutoff for the wave-function expansion was set to $\ell_{\text{max}} = 3$. Details of our methods are given elsewhere [16,18,24].

III. RESULTS

In Fig. 1(a), we show the XRD measurements of 20-nm-thick Al-Cr-Mo-W films grown on different seed layers. We observe a textured growth with a (011) preferred orientation, as expected for a bcc material. No other X-ray reflections (except higher orders and substrate) were observed. According to the Scherrer formula, the width of the reflections is compatible with a columnar growth and full crystallization across the thickness of the films. The high-entropy alloy Al-Cr-Mo-W thus appears to grow easily on multiple seed layers with similar quality.

However, as we will show later, Al-Cr-Mo-W has a high resistivity, whereas the seed layers have low resistivity and will thus shunt the current in the electrical characterization of the spin Hall properties. Therefore, we have studied the growth of thinner Al-Cr-Mo-W and its dependence on the Al concentration on Ta (3 nm) as the seed layer. The results are shown in Fig. 1(b). We find the maximum diffraction intensity at the equiatomic composition, i.e., Al 25%. At

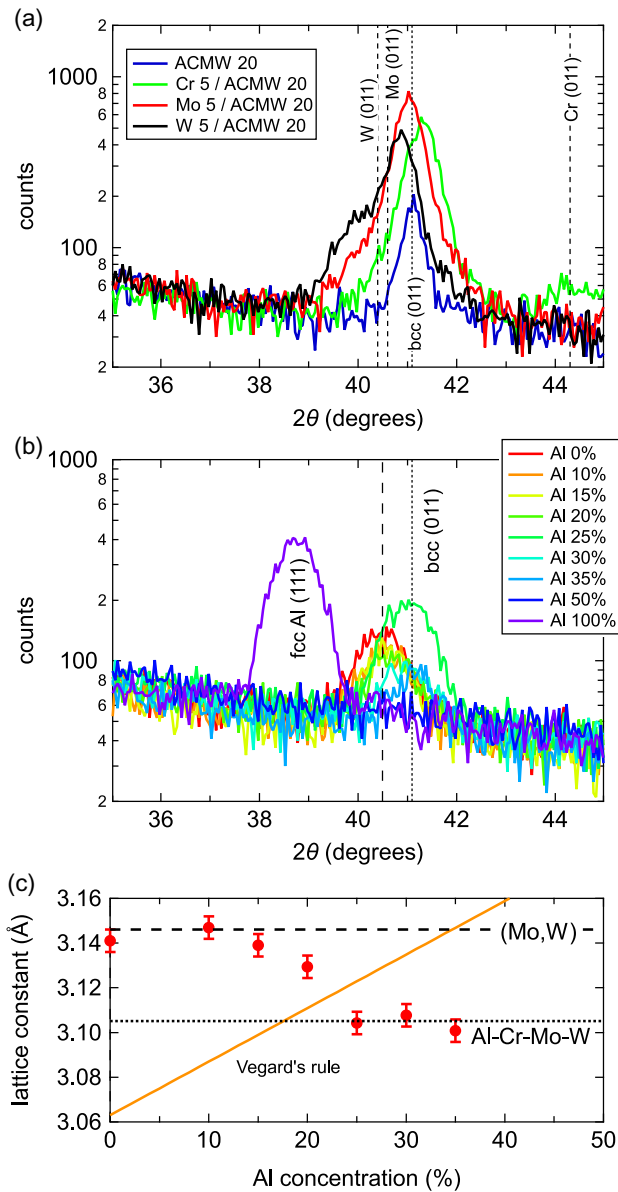


FIG. 1. (a) X-ray-diffraction (XRD) measurements of seed layer (5 nm)/Al-Cr-Mo-W (ACMW) (20 nm)/Cr (1 nm) samples. (b) XRD measurements of Ta (3 nm)/Al_x(Cr-Mo-W)_{1-x} (8 nm)/Co₄₀Fe₄₀B₂₀ (3 nm)/Ta (0.5 nm)/TaO_x (1 nm) samples. (c) Lattice constants obtained from the XRD measurements in (b).

Al 50% and above, no crystallization is observed, with the exception of pure Al, which shows a strong (111) texture. Again, the peak widths are compatible with columnar growth and crystallization across the thickness of the films with $D = K\lambda / (IB(2\theta) \cos \theta) \approx 7.8$ nm at the stoichiometric composition, where $K \approx 1$, $\lambda \approx 1.542$ Å and $IB(2\theta)$ is the integral breadth in radians. The lattice constant at the equiatomic composition is $a = 3.104$ Å [dotted line in Fig. 1(b)], very close to the expected value of $a = 4\bar{R}/\sqrt{3} = 3.12$ Å by simply taking the average atomic radius $\bar{R}$

and $R_{\text{Al}} = 143$ pm, $R_{\text{Cr}} = 125$ pm, $R_{\text{Mo}} = 136$ pm, and $R_{\text{W}} = 137$ pm (obtained from the room-temperature lattice constants of the elements). For the Cr-Mo-W alloy (0% Al) and for a 10% Al content, we find $a \approx 3.144$ Å [dashed line in Fig. 1(b)], which is much larger than the expected value of $a = 3.063$ Å. However, noting the miscibility gap in both the Cr-W and Cr-Mo phase diagrams, a decomposition into Cr + (Mo, W) seems likely here, and we observe only the bcc solid solution of (Mo, W) with an expected lattice constant of $a = 3.156$ Å. A steplike reduction of the lattice constant with increasing Al content is observed, with a transition between Al 20% and Al 25% [see Fig. 1(c)]. This seems unexpected, because the Al atom is larger than the transition-metal atoms; thus an increase of the lattice constants would be expected here according to Vegard's rule. However, we can explain the decrease by the high-entropy effect: the addition of Al helps dissolve the smaller Cr atoms in the solid solution, thereby forming a homogenous alloy. At the stoichiometric composition, its crystalline volume is maximized and we find the largest x-ray peak intensity with nearly the expected lattice constant. Further added Al is not dissolved in the lattice (the lattice constant does not increase from here); instead, we finally get an amorphous alloy at high Al concentration, where the entropy is reduced again. Thus, the homogeneous solid solution only forms in a narrow range of Al concentration, where the entropy is maximized.

All samples (except Al 100%) showed smooth growth according to x-ray reflectivity (XRR) analysis (Fig. 2). Typically, the stack roughness parameters of the Gaussian roughness model are $\sigma \approx 3.2$ Å, where the individual layers are hardly distinguishable. The total thickness of all samples is between 16 nm and 17 nm, in agreement with the sum of the individual nominal thicknesses. The Al 100% film shows a faster decay of the Kiessig fringes, which indicates a larger roughness compared to the other samples. The chosen Ta layer is thus a suitable seed layer to promote crystallinity and smooth growth across the compositional range.

Broadband FMR measurements on unpatterned samples were performed up to 30 GHz with the OpenFMR system. The Kittel analysis of the resonance positions and the Gilbert damping analysis of the resonance widths show substantial reductions with increasing Al content across the compositional series (Fig. 3). The value of the effective magnetization M_{eff} appears correlated with the crystallinity of the samples, with a plateau at low Al concentrations and a drop toward high Al concentration. This may point to either a reduction of the saturation magnetization or an increase in the uniaxial out-of-plane anisotropy due to structural modifications of the films. The reduction of the Gilbert damping parameter α indicates a reduction of the spin-mixing conductance as a result of reduced spin-orbit coupling and less effective spin absorption in the HEA with increasing Al concentration.

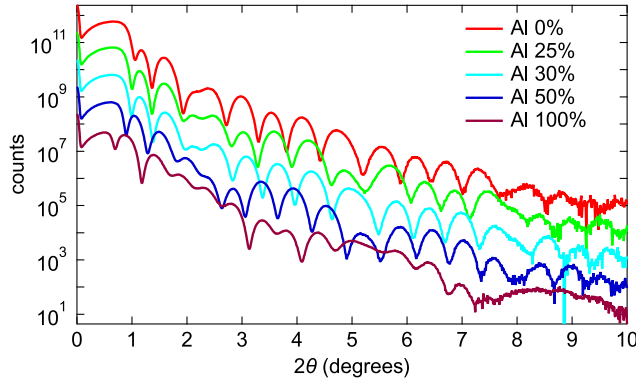


FIG. 2. X-ray reflectivity measurements on selected samples. The measurements are multiplied by 10 with respect to one another for clarity.

Alternatively, this may also indicate a longer spin-diffusion length with increasing Al concentration, which is not saturated at our sample thickness. This would reduce the observable effect of spin pumping, thus reducing the additional effect of this loss channel on the overall damping. A more detailed understanding would require film-thickness series for every composition. For comparison, two reference samples show the following values: Ta (5.5 nm)/Co₄₀Fe₄₀B₂₀ (2.5 nm), $M_{\text{eff}} = 1064$ kA/m and $\alpha = 0.013$; and Pt (5.5 nm)/Co₄₀Fe₄₀B₂₀ (2.5 nm), $M_{\text{eff}} = 900$ kA/m and $\alpha = 0.023$. Thus, our HEA series is, unsurprisingly, more similar to Ta films. The results show that the Co₄₀Fe₄₀B₂₀ layer has well-defined magnetic properties and is suitable as a spin-detector layer for the harmonic Hall measurements.

The harmonic Hall results for the SHA are compared to the *ab-initio*-calculated SHA in Fig. 4(a). The general trend agrees well and shows a peak of the SHA around the equiatomic composition. The SHC can be approximated by normalizing the observed torque in the harmonic Hall method directly to the electric field E applied along the

Hall cross, i.e.,

$$\sigma_{\text{SH}} \approx \frac{2e}{\hbar} \frac{B_{\text{DL}} M_s t_{\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}}}{E}, \quad (1)$$

where B_{DL} is obtained as a fit parameter from the harmonic Hall measurements and $M_s \approx 1050$ kA/m is the saturation magnetization of Co₄₀Fe₄₀B₂₀. This method is easier to apply and more accurate than applying $\theta = \sigma_{\text{SH}} \rho$ and applying a parallel-resistor model. The SHC and the alloy resistivities are shown in Figs. 4(b) and 4(c). Here, we observe an excellent matching of the harmonic Hall result and the *ab-initio*-calculated SHC. Around the equiatomic composition, we find $\sigma_{\text{SH}} \approx -72\,000$ ($\hbar/2e$) S/m, a spin Hall angle of $\theta \approx -0.12 \pm 0.01$, and a resistivity of $\rho \approx 140$ $\mu\Omega\text{cm}$. These values are very similar to those of Ta thin films with the complex β -Ta structure [31]; however, here we obtain these values for a crystalline bcc alloy. The SHA is also larger than that of our Pt reference sample; however, the SHC is much larger in Pt. The SHC of the suspected Cr-(Mo, W) alloy is also close to the theoretical result of (Cr, Mo, W). This can be explained by looking at the calculated SHCs of Cr and (Mo, W), which are $-30\,700$ ($\hbar/2e$) S/m and $-112\,000$ ($\hbar/2e$) S/m, respectively. Then, the composition-weighted average is approximately $-84\,900$ ($\hbar/2e$) S/m, very similar to our experimental finding ($-83\,500$ ($\hbar/2e$) S/m) and slightly smaller than the calculated value of (Cr, Mo, W), $-94\,200$ ($\hbar/2e$) S/m.

We emphasize that our calculations explicitly account for vertex corrections, i.e., extrinsic contributions (skew scattering and side jump). As can be seen in Fig. 4(b), these clearly play only a very small role in determining the total SHC of the alloy; in particular, for intermediate Al concentrations, the results with and without vertex corrections are indistinguishable. Thus, it is a common misconception that the SHE in chemically disordered alloys would be dominated by extrinsic effects. Instead, it is solely given by the intrinsic contribution due to suppression of the vertex corrections in concentrated alloys.

Comparing the measured and *ab-initio*-calculated resistivities, we observe that both follow the same trend: a huge enhancement of the resistivity toward 50% Al concentration and the expected Nordheim-type inverted parabola. Our measurements, surprisingly, show slightly lower resistivity values compared to the calculated values. Usually, one would expect the opposite, due to finite-thickness effects, imperfect crystallization, grain-boundary scattering, and static disorder contributions neglected in the calculation. Here, the slightly lower resistivity could indicate an inhomogeneous solid-solution formation (i.e., a subregular solid solution [21]), where both locally Al-rich and Al-poor fractions would have lower resistivity, respectively, in agreement with our XRD result. For the end points of the composition series, we have measured

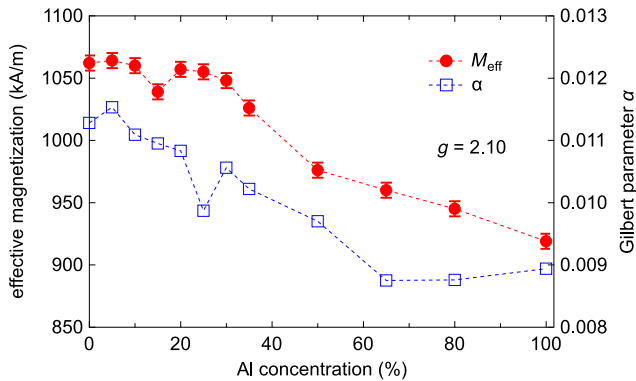


FIG. 3. The effective magnetization and the Gilbert damping parameter α as a function of the Al concentration. The Lande factor was held constant at $g = 2.1$.

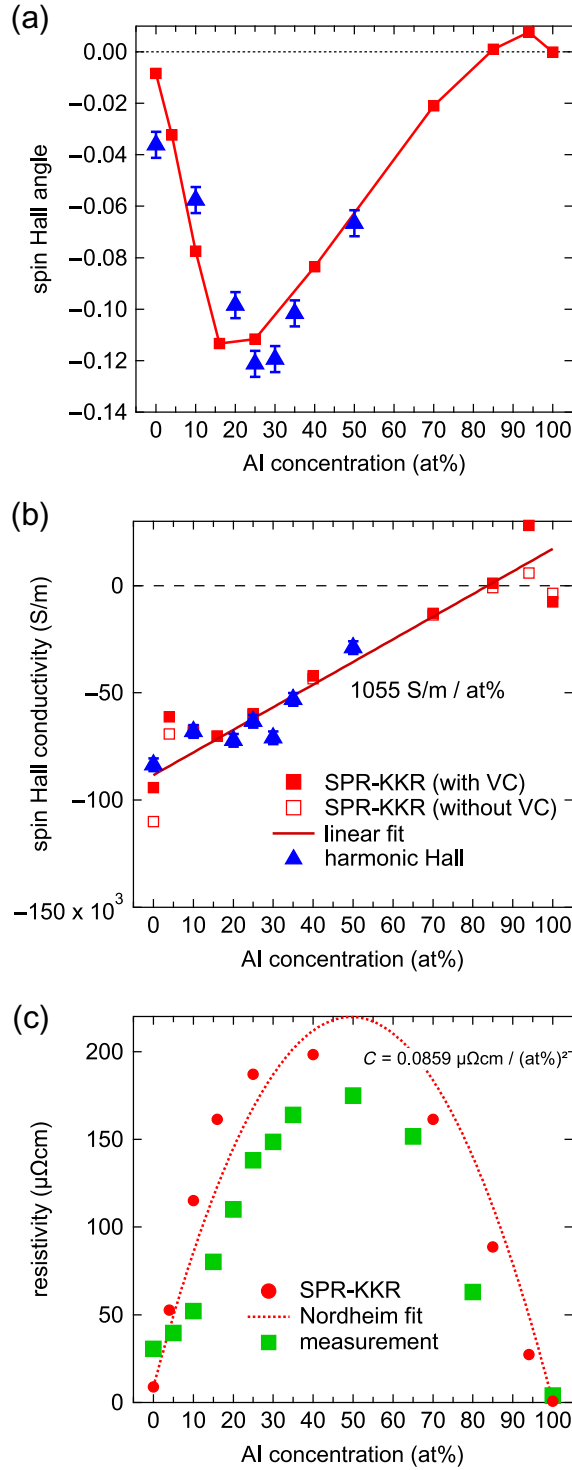


FIG. 4. A comparison of the measured and calculated data: (a) the spin Hall angle; (b) the spin Hall conductivity; and (c) the resistivity. The red squares represent calculations and the blue triangles are harmonic Hall results. In (c), the solid green squares represent four-point measurements. The Nordheim fit represents a parabolic fit with fixed end points to the spin-polarized and relativistic Korringa-Kohn-Rostoker (SPR-KKR) calculation results.

$\rho = 30.5 \mu\Omega\text{cm}$ for Cr-(Mo, W), and $\rho = 4.0 \mu\Omega\text{cm}$ for Al. The calculated value of $\rho = 8.9 \mu\Omega\text{cm}$ for Cr-Mo-W is much smaller than the measured value, but resistivity values around $30 \mu\Omega\text{cm}$ are quite typical for bcc transition-metal thin films with our deposition conditions, and the segregated Cr+(Mo, W) phase is expected to have a higher resistivity than a homogeneous phase due to additional grain-boundary scattering. The Nordheim fit is obtained as $\rho(x) = x\rho_0 + (1-x)\rho_1 - Cx(1-x)$. The magnitude of the parameter C is outstanding: in typical alloys, only very modest enhancements of the resistivity can be observed. A resistivity of about $150 \mu\Omega\text{cm}$ in a crystalline alloy is, thus, quite noteworthy. This can be explained along the lines of the resistivity theory in metals, which states that the impurity contribution to the resistivity can be roughly written as $\rho_{\text{imp}}(x) \propto x(1-x)N(E_F)(\Delta V)^2$. The density of states at the Fermi energy, $N(E_F) \approx 0.85$ states per electronvolt remains mostly constant across the composition series and drops only toward the Al-rich side to $N(E_F) \approx 0.55$ states per electronvolt. The scattering potential ΔV arises as the difference of the local scattering potentials with the surrounding matrix. In transition-metal main-group alloys, this effect is naturally large due to the presence or lack of open d shells, respectively. With maximum disorder, the resistivity is maximized, i.e., around the equiatomic composition. While trivial in binary alloys, in our quaternary alloy this leads to a skew of the calculated resistivity maximum toward lower Al concentration. However, the measurements reveal the maximum at 50% Al concentration. This result has, however, to be taken with care due to the multiple phases present in the system, from low to high Al concentration: Cr + bcc-(Mo, W) $\rightarrow$ bcc-(Al, Cr, Mo, W) $\rightarrow$ amorphous $\rightarrow$ fcc-Al.

IV. CONCLUSIONS

We have demonstrated the polycrystalline textured growth of quaternary high-entropy alloy thin films. A Ta seed layer is a good choice to obtain a homogenous bcc-crystallized phase around the equiatomic composition and to avoid shunting. A high resistivity of $140 \mu\Omega\text{cm}$ was achieved in the thin film of Al-Cr-Mo-W. The spin Hall effect was characterized, where a spin Hall conductivity of $-72\,000 (\hbar/2e) \text{ S/m}$ and a spin Hall angle of -0.12 ± 0.01 were observed around the equiatomic composition. This work demonstrates the existence of chemically disordered high-resistivity high-entropy alloys with a sizeable spin Hall effect and a simple lattice structure. Further studies should investigate the possibility to enhance the phase homogeneity with thermal treatment and the epitaxial deposition of the alloy on single-crystalline substrates. Temperature-dependent investigations will shed light on the electronic transport mechanism in the high-resistivity state. This will allow for further and deeper understanding

of the spin Hall effect in this crystallographically simple high-resistivity alloy.

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The authors have no conflicts of interest to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available upon reasonable request from the corresponding author.

APPENDIX: HIGH-ENTROPY ALLOYS

High-entropy alloys (HEAs) are single-phase multielement alloys stabilized by the configurational entropy [21]. The configurational entropy of mixing for an ideal solid solution is given by

$$\Delta S_{\text{mix}} = -R \sum_{i=1}^N c_i \ln(c_i), \quad (\text{A1})$$

where R is the universal gas constant, N is the number of constituents, and c_i is the concentration of the i th element. For equiatomic alloys, the configurational entropy is $\Delta S_{\text{mix}} = R \ln(N)$. A value of $\Delta S_{\text{mix}} > 1.5R$ (i.e., at least five components) is often associated with a high-entropy alloy, whereas broader definitions also include four-components alloys ($\Delta S_{\text{mix}} = 1.39R$ for a equiatomic four-component alloy), emphasizing their stabilization by entropy (the *high-entropy effect* [21]). The stability of the HEA is assessed by comparing the Gibbs mixing energy $\Delta G_{\text{mix}} = \Delta H_{\text{mix}} - T\Delta S_{\text{mix}}$ with the mixing enthalpy of the components, which is approximated as [32]

$$\Delta H_{\text{mix}} = \sum_{i=1}^N \sum_{j>i}^N 4\Delta H_{ij} c_i c_j, \quad (\text{A2})$$

where ΔH_{ij} is the binary mixing enthalpy of elements i and j . For the entropy effect to be dominant, the mixing enthalpy needs to fulfill $-15 \text{ kJ/mol} < \Delta H_{\text{mix}} < +5 \text{ kJ/mol}$ (to avoid formation of binary phases or decomposition into the elements, respectively). The stability is usually expressed in terms of a thermodynamic parameter

Ω as

$$\Omega = \frac{T_m \Delta S_{\text{mix}}}{\Delta H_{\text{mix}}}, \quad (\text{A3})$$

where T_m is the average melting point of the constituent elements. For the formation of the HEA from the melt, $\Omega > 1.1$ is required. For the stability at lower temperature, additionally the Gibbs mixing energy at that temperature must remain lower than the formation energies of competing phases. Furthermore, the atomic sizes must be similar, quantified by a size mismatch parameter

$$\delta = \sqrt{\sum_{i=1}^N c_i \left(1 - \frac{r_i}{\bar{r}}\right)^2}, \quad (\text{A4})$$

where R_i is the atomic radius of the i th atom and $\bar{r}$ is the concentration-weighted mean atomic radius. $\delta < 6.6\%$ is usually seen as favorable for the formation of a HEA. The VEC per atom is indicative of the crystal structure of the solid solution, where $\text{VEC} < 6.87$ induces the bcc structure, $\text{VEC} > 8$ typically leads to the fcc structure, and intermediate values tend to result in phase mixtures. The required binary mixing enthalpies can be obtained from Ref. [33].

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