Alloys made by equimolar mixtures of more than five elements have recently been developed and several unusual properties were discovered, including (a) higher electrical resistivity ($10^{-6} - 10^{-7} \, \Omega \cdot m$), (b) higher hardness [Vickers hardness (HV) = 700], (c) higher wear resistance (1.2–1.6 m/mm²), (d) higher thermal stability, and (e) superior corrosion resistance, compared with ordinary alloys. This multiple element system has been termed as high entropy alloys (HEAs) because its configurational entropy ($\Delta S_{\text{conf}} = 2.2R - 2.7R$) is greater than that of ordinary alloys (1.1R). According to previous studies, addition of boron to HEAs occasionally produces microprecipitates of borides and the material's hardness thus increases from HV = 232 to HV = 736. In this work, we find that the permeability of Fe–Co–Ni–Al–B–Si alloy firstly decreases exponentially; it then increases significantly at gigahertz. Data reveal that the magnetic resonance at gigahertz involves the hyperfine structure arising from microprecipitates of magnetic borides.

Fe–Co–Ni–Al–B–Si, Fe–Co–Ni–Al–B–Si–Cr, and Fe–Co–Ni–Al–B–Si–Cr–B were studied, in comparison with boron-free Fe–Co–Ni–Al–B–Si and ferromagnetic Fe–Co–Ni. Samples were made by a standard arc-melting technique and elemental ratio in each HEA is equimolar (e.g., 16.6 at. % for Fe–Co–Ni–Al–B–Si). As-made products were cut into films ($7 \times 3 \times 0.5 \, \text{mm}^3$) and sliced samples were measured by a constant magnetic field equipped with an ac signal detector (Permeance meter, PMF-3000, 0–3 GHz), x-ray diffraction (XRD), electron probe x-ray microanalysis (EPMA), superconducting quantum interference device (SQUID) magnetometer, and electron paramagnetic resonance (EPR).

Figure 1(a) displays the permeability of HEAs in comparison with Fe–Co–Ni (yellow) and all samples show an exponential decrease between 0 and 1.4 GHz. Surprisingly, a significant increase is present in Fe–Co–Ni–Al–B–Si (red) above 1.4 GHz (arrow) while others continuously level off. This outcome has been verified by repeated experiments. Permeability seen here for Fe–Co–Ni–Al–B–Si resembles tradeoff between domain wall motion resonance at low frequency and gyromagnetic coupling at high frequency, previously discussed by Dionne. The onset of angular resonance frequency for transverse permeability is at $\omega_c = \gamma H_K$ and is limited at $\omega_c = \gamma (H_K + 4\pi M_s)$, where $\gamma$ is anisotropy constant, $2.78 \times 10^6 \, \text{Hz/Oe}$, $H_K$ is coercivity, and $M_s$ is saturation magnetization. We find that hysteresis loops of Fe–Co–Ni–Al–Si–Cr, Fe–Co–Ni–Al–Si–Cr–B, Fe–Co–Ni–Al–B, and Fe–Co–Ni are similar and their coercivity is around 140–160 Oe with saturation magnetization at 1000–1200 Oe (an example for Fe–Co–Ni–Al–Si–Cr–B is shown in the supplemental material). In contrast, the Fe–Co–Ni–Al–B–Si alloy shows smaller coercivity (75 Oe) and higher saturation magnetization [2500 Oe, Fig. 1(b)], which is fairly unusual compared with other HEAs studied here. Firstly, although most HEAs with Fe (or Co and Ni) behave as hard magnetic materials, saturation magnetization above 2000 Oe is not previously observed. Secondly, if one substitutes $H_K = 75$ Oe and $M_s = 2000$ Oe [Fig 1(b)] into the equation, the $\omega_c$ range appears to be $10^6 – 10^{10}$ Hz, namely, onset of angular resonance frequency for transverse permeability is lower than in Fig. 1(a) (arrow) by one order of magnitude.
for thin permalloy films coupled with antiferromagnetic layers; however, we do not anticipate a similar effect for the current study because our samples are ferromagnetic in nature. Here we firstly focus on the origin of higher saturation magnetization seen in Fe–Co–Ni–Al–B–Si. According to the equation \( \Delta E / h = g \beta B / h \) (Hz), where \( \Delta E \) is energy difference between spins, \( g \) is Landé splitting factor, \( \beta \) is Bohr magnetron \( (=9.273 \times 10^{-24} \text{ J T}^{-1}) \), and \( B \) is magnetic field, if one inserts \( B=1000 \text{ Oe} \) and \( g=2.0023 \) (free electron) into the equation, the \( \Delta E \) appears to be very small \( (0.85 \times 10^{-22} \text{ eV}) \), indicating that spins have aligned before \( B =1000 \text{ Oe} \). This outcome is obviously inconsistent with saturation magnetization at 2500 Oe [Fig 1(b)]. Evidences provided by XRD, EPMA measurements, and calculation seem to suggest that higher saturation magnetization seen in Fe–Co–Ni–Al–B–Si probably originates from magnetic domains which broaden the distribution of spin directions. The Fe–Co–Ni–Al–B–Si alloy is a bcc structure with lattice constant \( a \) of 0.29 nm, slightly greater than \( \alpha\)-Fe by \(-0.03\)–0.05 nm (Fig. 2). Additional reflections at 301, 321, and 411 correspond to tetragonal Fe3B, and the EPMA also confirms the presence of Fe–B enriched domains \((-10 \, \mu \text{m})\) embedded in bcc matrix (Fig. 3). XRD and EPMA did not detect enriched Fe–B domains in Fe–Co–Ni–Al–Si–Cr–B and Fe–Co–Ni–Al–B–Si matrix. Tetragonal domains embedded in bcc matrix imply large anisotropy energy for Fe–Co–Ni–Al–B–Si sample; for example, the Bloch wall energy of tetragonal Fe3B is \( 10–20 \, \text{ergs/cm}^2 \) and its easy axis of magnetic symmetry lies along the \( c \) axis. The Wall energy of the bcc matrix is expected to be much lower than that of borides because the easy axis of magnetization for bcc structure usually lies along cubic edges and the corresponding wall energy is around \( 1–2 \, \text{ergs/cm}^2 \), e.g., the \( \alpha\)-Fe along the 100 direction is 1 \( \text{erg/cm}^2 \). Here we estimate the anisotropy energy density of the bcc structure using the expression \( U_k = K_1 \left[ (\alpha_1 \alpha_2)^2 + (\alpha_2 \alpha_3)^2 + (\alpha_3 \alpha_1)^2 \right] \), where \( \alpha_i \) is cosine angle of magnetization in arbitrary direction with respect to the cubic edge and \( K_1 \) is \( 4.2 \times 10^5 \, \text{ergs/cm}^3 \) at room temperature. If one sets \( \alpha=45^\circ \) as upper limit the \( U_k \) appears to be \( 3.1 \times 10^6 \, \text{ergs/cm}^2 \), which resembles \( \alpha\)-Fe. In other words, the wall energy of the bcc matrix would approximate the \( \alpha\)-Fe and higher saturation magnetization in Fe–Co–Ni–Al–B–Si alloy is truly attributed to large anisotropy energy.

We now discuss the permeability increase at gigahertz from Fe–Co–Ni–Al–B–Si. Due to quenching of spin–orbit paramagnetic by electrostatic within the crystal lattice, the metals and alloys are usually weakly temperature dependent of spin susceptibility. HEAs are less atomic ordered and Coulomb field established within the crystal lattice is weaker; therefore resurrection of spin–orbit paramagnetic is expected. Our SQUID profiles truly show a clear temperature dependence of spin susceptibility (\( \chi_m \)) for Fe–Co–Ni–Al–B, Fe–Co–Ni–Al–Si–Cr, and Fe–Co–Ni–Al–B–Si–Cr (Fig. 4); the \( \chi_m \) of Fe–Co–Ni–Al–B–Si alloy (red) is weakly temperature dependent, similar to ternary (orange). At low temperature, the \( \chi_m \) of HEAs is the sum of spin paramagnetic and spin–orbit coupling; the latter decreases as temperature rises (Fig. 4); the former dominates the magnetic property of HEAs at room temperature, which is consistent with EPR data. In our study, EPR profiles of Fe–Co–Ni–Al–B, Fe–Co–Ni–Al–Si–Cr, and Fe–Co–Ni–Al–Si–Cr are similar (an example for Fe–Co–Ni–Al–B–Si–Cr is shown in the supplementary material) and are briefed as follows. Firstly, EPR...
is such that 3 previously observed in transition metal ions and borides. Hyperfine interaction occurs when an unpaired electron is very close to nuclei so spin instantly couples with circulating current must arise from paired 4$s$ electrons of Fe; hence $I=1$, consistent with a triplet state. Hyperfine coupling can resonate at the gigahertz range via rapid transition between paired spins ($\uparrow\uparrow$) and parallel spins ($\downarrow\downarrow$) under applied magnetic field, so-called electron nuclear double resonance process. This is supported by calculation as follows. According to Fig. 5, the triplet appears around 300–1000 Oe and $g$-value lies between 3 and 8 so adsorption frequency can be evaluated by substituting $B=300–1000$ Oe and $g=3–8$ into the equation $\Delta E/h=g\beta B/h$, which gives adsorption frequency around 1.2–5 GHz, in good agreement with Fig. 1(a) (arrow).

Selection of electromagnetic adsorption materials is largely determined by natural resonance frequency and materials with large anisotropic energy can promote resonance frequency to the gigahertz range, e.g., $\alpha$-Fe/Fe$_3$B/Y$_2$O$_3$. This work discovers that Fe–Co–Ni–Al–B–Si system contains microprecipitates of magnetic boride and its onset of magnetic resonance frequency appears at gigahertz, consistent with report.

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FIG. 5. EPR profiles at different temperatures for Fe–Co–Ni–Al–B–Si.

band intensity significantly decreases with increasing temperature, indicating a reduction of spin concentration consistent with SQUID, and secondly, the band broadens towards the lower $g$-value region with increasing temperature, attributed to phase transition from spin-orbit interaction to spin-spin coupling as temperature increases. For Fe–Co–Ni–Al–B–Si alloy a triplet is present between $g=3.450$ and 6.822 at 4 K along with a broad adsorption of free electrons centered at $g=2.003$ (Fig. 5). Resonance line separation of the triplet is $\sim200$ Oe, which is similar to a hyperfine splitting previously observed in transition metal ions and borides. Hyperfine interaction occurs when an unpaired electron is very close to nuclei so spin instantly couples with circulating current around the nucleus, leading to spin splitting into $2I+1$ ($I$: nuclear spin) and variation of local electric quadrupole interaction. For Fe–Co–Ni–Al–B–Si alloy the hyperfine interaction probably arises from boride enriched regions because (a) other HEAs (i.e., boride-domain-free) do not show a triplet feature and (b) the electronic configuration of Fe$_3$B is such that 3$d$ electrons with $T_{2g}$ symmetry hybridize with $p$ orbit of B atoms to form nonmagnetic bonding and the remaining $d$ electrons with $E_s$ symmetry become available for hyperfine coupling with nuclei. This means that the circulating current must arise from paired 4$s$ electrons of Fe; hence $I=1$, consistent with a triplet state. Hyperfine coupling can resonate at the gigahertz range via rapid transition between paired spins ($\uparrow\uparrow$) and parallel spins ($\downarrow\downarrow$) under applied magnetic field, so-called electron nuclear double resonance process. This is supported by calculation as follows. According to Fig. 5, the triplet appears around 300–1000 Oe and $g$-value lies between 3 and 8 so adsorption frequency can be evaluated by substituting $B=300–1000$ Oe and $g=3–8$ into the equation $\Delta E/h=g\beta B/h$, which gives adsorption frequency around 1.2–5 GHz, in good agreement with Fig. 1(a) (arrow).

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