Supplementary Information

Hard Ferrite Magnetic Insulators Revealing Giant Coercivity and Sub-Terahertz Natural Ferromagnetic Resonance at 5 – 300 K

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§1. XRD, SEM, terahertz time-domain spectroscopy

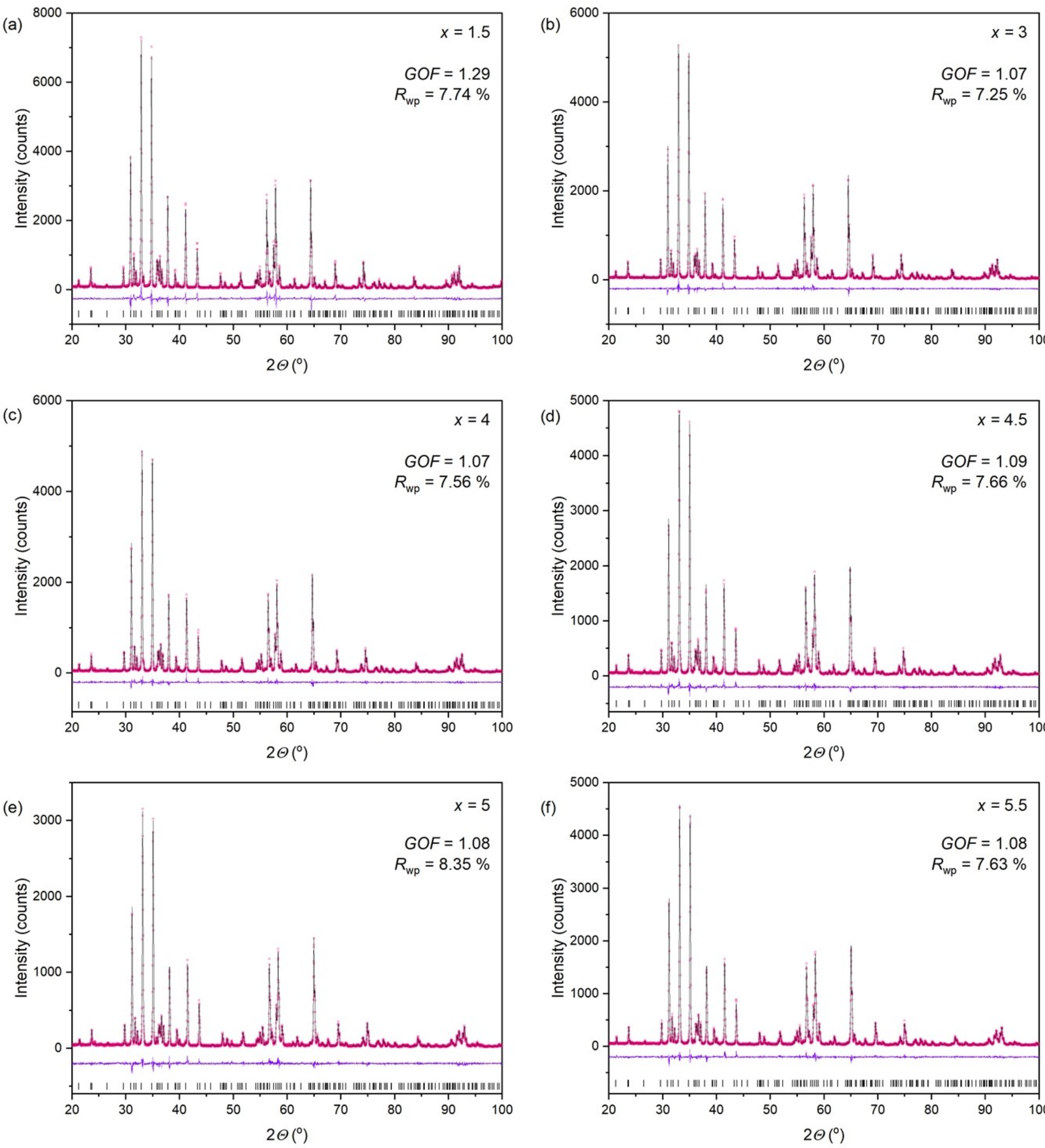

Figure S1. Rietveld refinement of the hexaferrite samples $Sr_{1-x/12}Ca_{x/12}Fe_{12-x}Al_xO_{19}$ (x = 1.5 – 5.5). The experimental (pink circles) and calculated (black line) data, their difference (purple curve), and the bars correspond to the M-type hexaferrite phase.

Table S1. Unit cell parameters (*a*, *c*, *V*), the Curie temperatures (T_C), and T_{max}/T_C ratio (β), where T_{max} corresponds to a maximum of anisotropy field of hexaferrite samples. $Sr_{1-x/12}Ca_{x/12}Fe_{12-x}Al_xO_{19}$.

x	*a*, Å	*c*, Å	*V*, Å^3	T_C, K	β
1.5	5.8499(1)	22.922(3)	679.33(1)	648	0.60
3	5.8099(1)	22.7843(7)	666.05(1)	561	0.62
4	5.7858(1)	22.7016(3)	658.08(3)	510	0.59
4.5	5.7738(6)	22.659(3)	654.21(1)	466	0.54
5	5.7597(7)	22.613(4)	649.69(2)	443	0.45
5.5	5.7461(9)	22.571(5)	645.41(2)	410	0.44

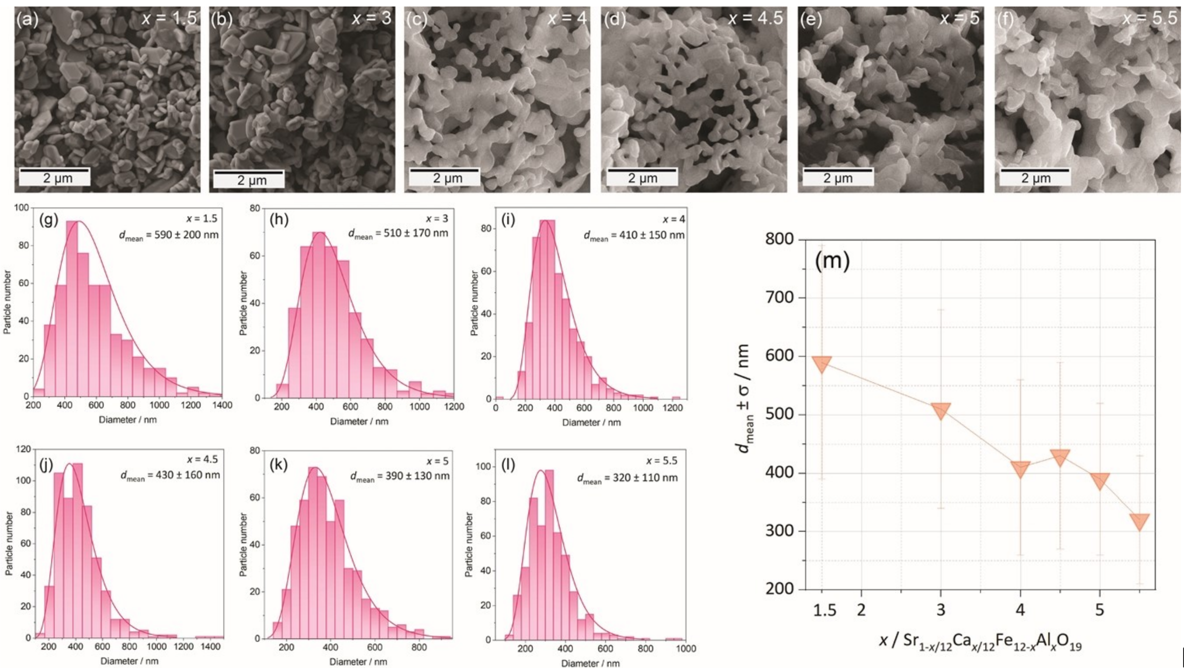

Figure S2. SEM images (a – f) and particle size distribution histograms (g – l) of the single-domain hexaferrite samples $Sr_{1-x/12}Ca_{x/12}Fe_{12-x}Al_xO_{19}$ (*x* = 1.5 – 5.5). (m) Mean diameter *vs* aluminum concentration (*x*).

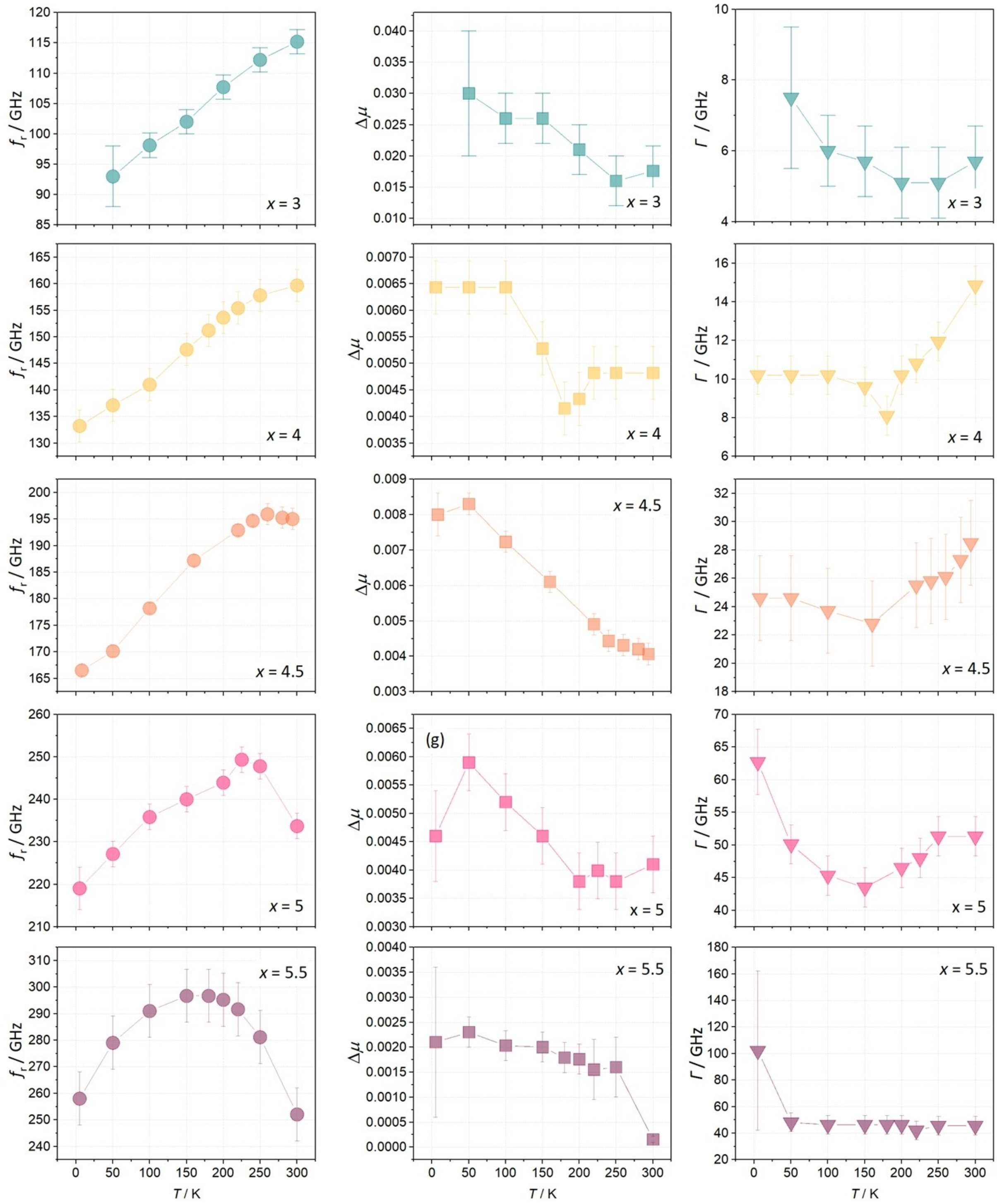

Figure S3. Temperature dependence of the parameters of natural ferromagnetic resonance absorption observed in the single-domain hexaferrite samples $Sr_{1-x/12}Ca_{x/12}Fe_{12-x}Al_xO_{19}$ (x = 3 – 5.5) and processed with Lorentzian expression for complex magnetic permeability $\mu^*(f) = \Delta\mu f_r^2(f_r^2 - f^2 + if\Gamma)^{-1}$: resonance frequency ($f_r$), magnetic contribution ($\Delta\mu$), damping factor (Γ).

§2. Modeling spin current

By definition the spin current can be expressed as [2]:

$$J_s = (\hbar/4\pi) g_r^{\uparrow\downarrow} M \times \dot{M},$$

where $g_r^{\uparrow\downarrow}$ denotes the spin-mixing conductance.

To write this expression in a more physical form, we calculate $M \times \dot{M}$ by parameterizing as:

$$M = \begin{cases} m_i cos(ft) \\ \pm m_i sin(ft) \\ M_{i0} \end{cases}$$

Considering a precession of the magnetic moment around the *z*-axis, it can be concluded that the currents flowing in the plane, which is basal to the axis, will compensate each other when averaging over time. Thus, to fully judge the spin current, it is enough to consider only its *z*-component. By solving the Landau-Lifshitz equations for the corresponding magnetic systems, we obtained the *z*-projection of the dc spin current:

$$J_{s,z}^{dc} = (\hbar/4\pi) g_r^{\uparrow\downarrow} f_r m_\pm^2 = (\hbar/4\pi) g_r^{\uparrow\downarrow} f_r \chi_r''^2 h^2$$

Also, solving Landau-Lifshitz for the simplest case (for two-magnetic-sublattice ferrimagnetic) for the condition the anisotropy field is much lower than the molecular field (exchange field) $H_a << H_E$, we get the expression for the imaginary part of the magnetic susceptibility:

$$\chi_r'' = \frac{\gamma M_{SV}}{\alpha f_r}$$

To calculate the dc spin current J_z^{dc} in units of $(\hbar/4\pi) g_r^{\uparrow\downarrow} h^2$ for the hexaferrite samples, we have taken the measured NFMR frequency, the saturation magnetization at 90 kOe multiplied by the crystallographic density of the compound (obtained from the Rietveld refinement), γ = 0.0028 GHz Oe^{-1}, and α = 0.001.

In the same way, for the antiferromagnetic resonance the following expressions are valid:

$$f_{AFMR\pm} = \gamma[\sqrt{(2H_E + H_a)H_a} \pm H_{app}],$$

where H_{app} is the applied magnetic field.

$$\chi_r'' = \frac{\gamma M_0 H_a}{\alpha (H_E + H_a) f_r}$$

To model J_z^{dc} for the antiferromagnetic resonance in MnF_2, we have taken the following parameters: H_a = 8800 Oe, H_E = 556 000 Oe, M_0 = 590 G at 0 K, and the highest f_r = 261 GHz at 4.2 K from [1]; and γ = 0.0028 GHz Oe^{-1}, α = 0.001. Thus, the $J_z^{dc} \approx 2.5$ units of $(\hbar/4\pi) g_r^{\uparrow\downarrow} h^2$.

References

[1] F. M. Johnson, A. H. Nethercot, *Phys. Rev.* **1959**, *114*, 705.
[2] V. Baltz, A. Manchon, M. Tsoi, T. Moriyama, T. Ono, Y. Tserkovnyak, *Rev. Mod. Phys.* **2018**, *90*, 015005.