

Supplementary Information

Nanoceramics of Metastable ϵ -Fe₂O₃: An Effect of Sintering on The Magnetic Properties and Sub-Terahertz Electron Resonance

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§ 1. Synthesis and X-ray diffraction analysis

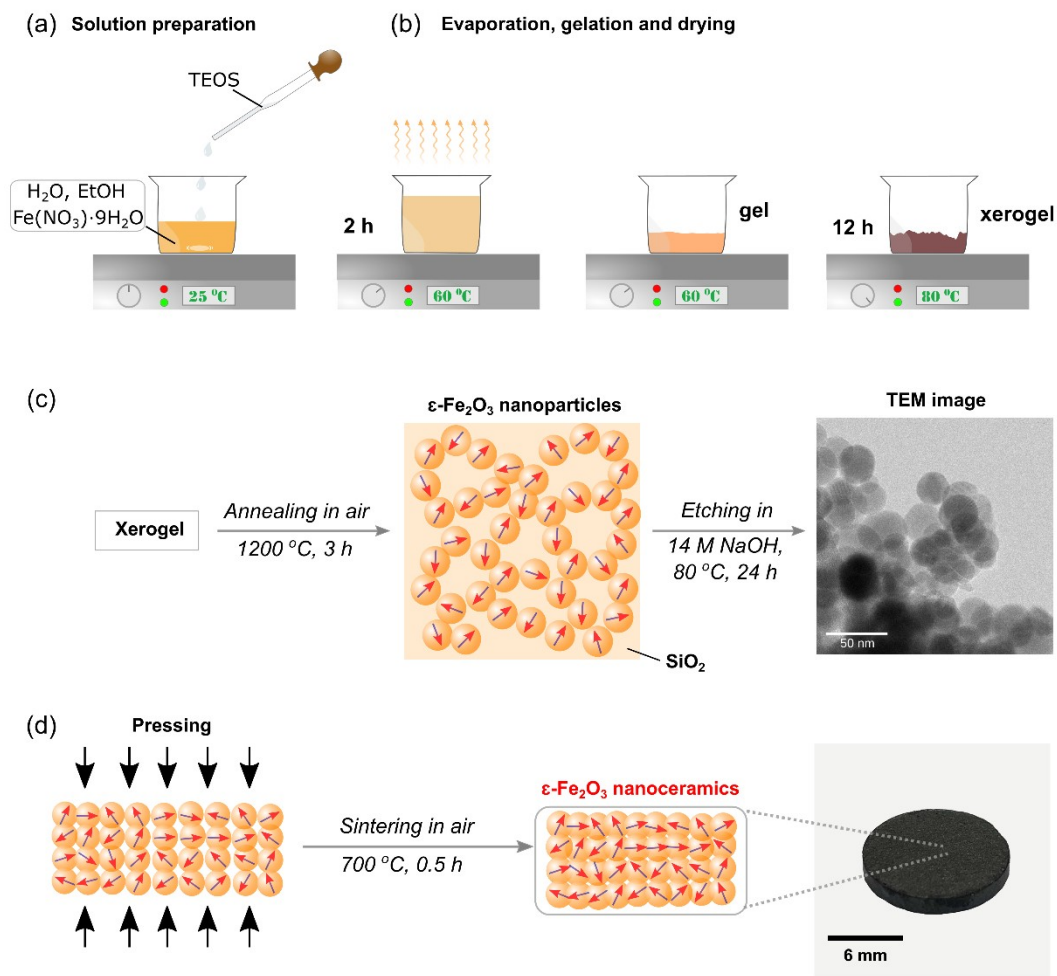


Figure S1. Flowchart describing the production of ϵ -Fe₂O₃ nanoceramics. (a) The preparation of a solution of iron nitrate within a mixture of water, ethanol, and tetraethoxysilane (TEOS) in a mole ratio of 1:6:6, respectively. (b) Stage of obtaining xerogel: evaporation of a part of ethanol and water, gelation of the solution, and drying of the gel. (c) Annealing the xerogel to obtain ϵ -Fe₂O₃/SiO₂ composite, leaching the ϵ -Fe₂O₃ nanoparticles. (d) Forming a pellet of the ϵ -Fe₂O₃ nanoparticles by cold pressing and sintering.

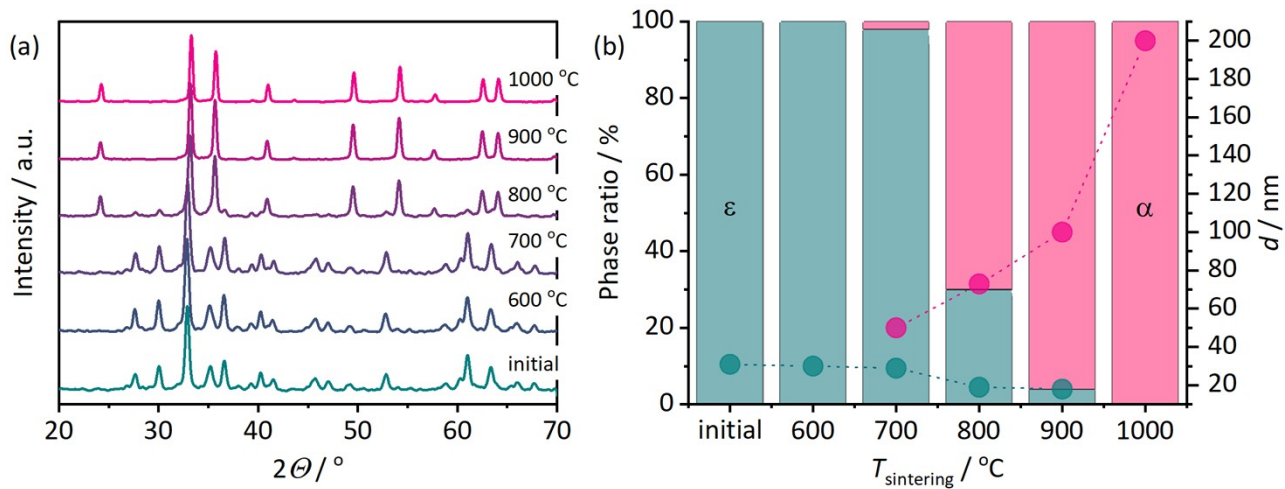


Figure S2. (a) X-ray powder diffraction patterns of the initial ϵ - Fe_2O_3 sample and the pressed samples annealed at 600 – 1000 °C for 30 min. (b) The phase composition in weight % and particle diameter for ϵ and α iron oxide phases calculated by the full-profile analysis.

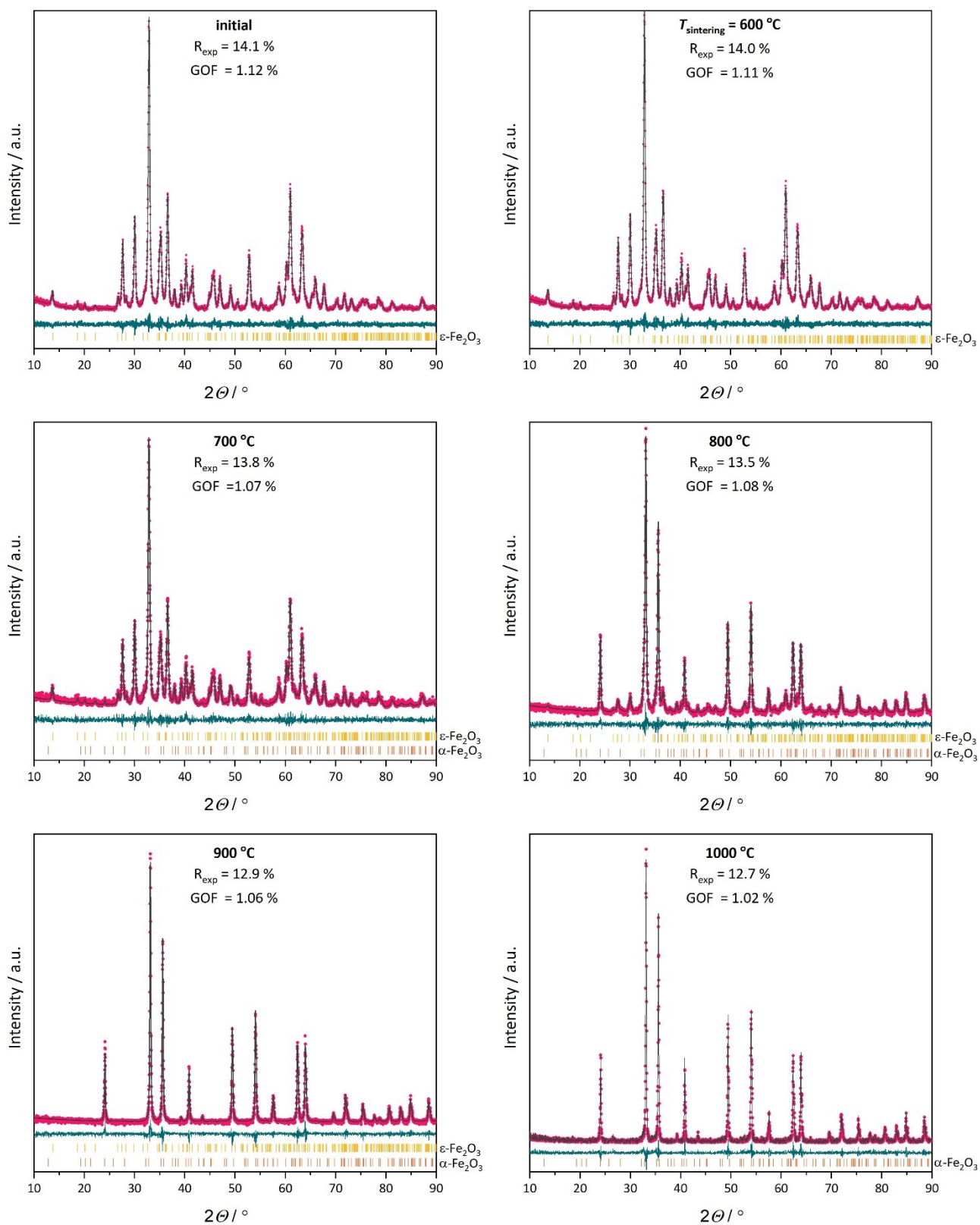


Figure S3. Results of the full-profile analysis of powder XRD patterns and the corresponding R-factors.

§ 2. Study of the microstructure

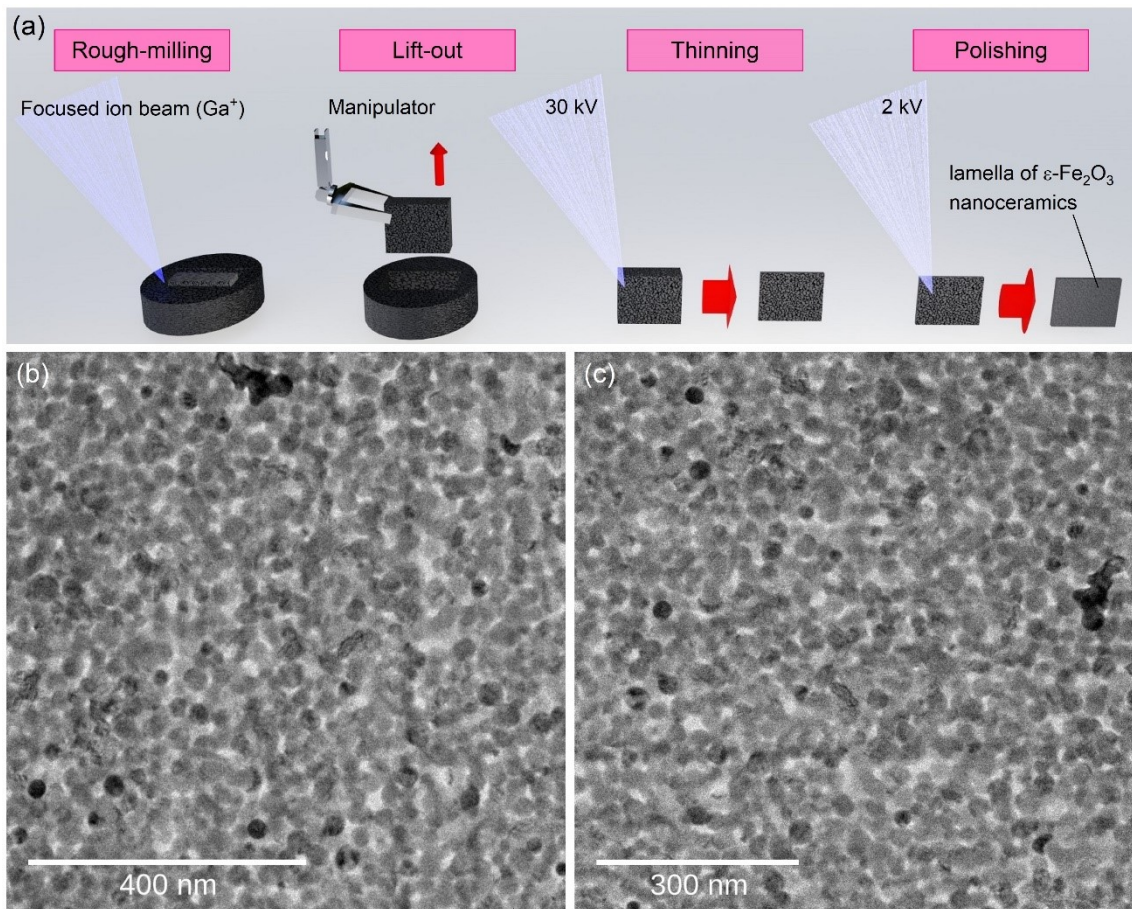


Figure S4. (a) A scheme of a typical process of lamella preparation from a ceramic sample. (b, c) TEM images of the epsilon iron oxide nanoceramics.

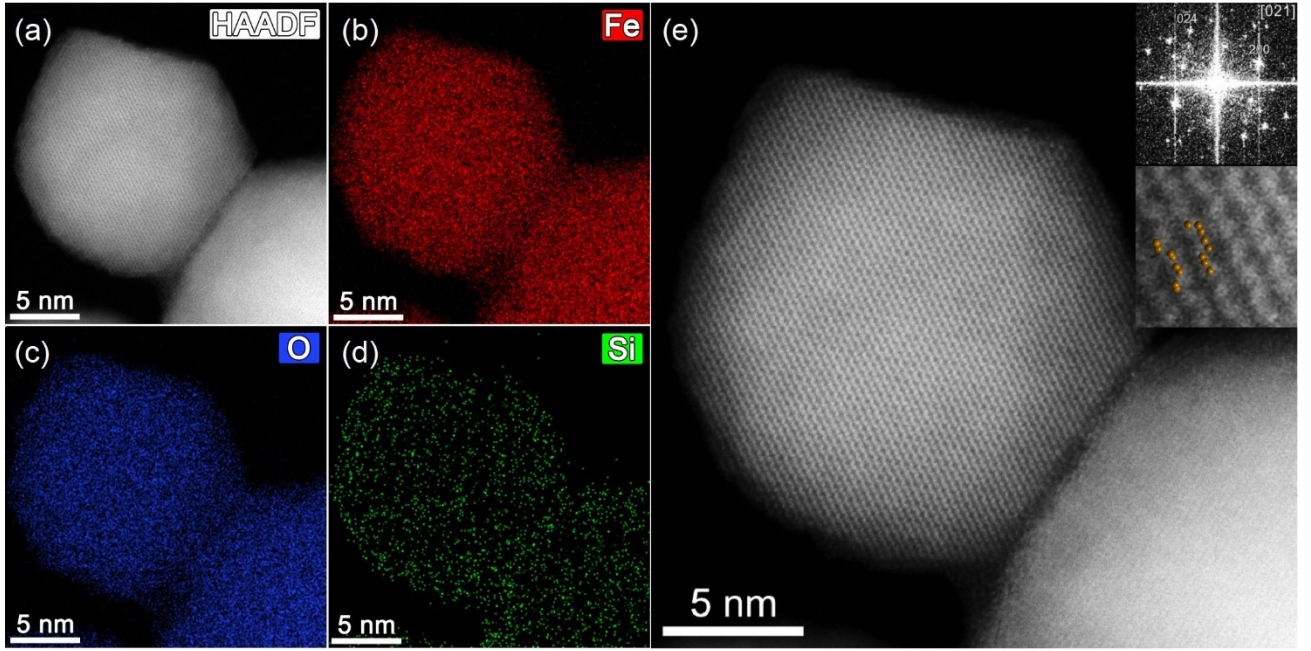


Figure S5. (a-c) HAADF-STEM image of two connected ϵ -Fe₂O₃ crystallites after annealing at 700 °C with corresponding color-coded STEM-EDX compositional map and elemental maps of Fe and Si. (e) High-resolution HAADF-STEM image of ϵ -Fe₂O₃ crystallites after annealing. FFT image as well as magnified HAADF-STEM image with overlaid [021] projection of ϵ -Fe₂O₃ crystal structure (only Fe atoms are present as brown spheres) are given in the insets.

§ 3. Modeling spin current

By definition the spin current can be expressed as ¹:

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

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

To write this expression in a more physical form, we calculate $M \times M$ 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) $H_a \ll H_E$, 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), $\gamma = 0.0028 \text{ GHz Oe}^{-1}$, and $\alpha = 0.001$.

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: $H_a = 8800 \text{ Oe}$, $H_E = 556\,000 \text{ Oe}$, $M_0 = 590 \text{ G}$ at 0 K, and the highest $f_r = 261 \text{ GHz}$ at 4.2 K from ²; and $\gamma = 0.0028 \text{ GHz Oe}^{-1}$, $\alpha = 0.001$. Thus, the $J_z^{\text{dc}} \approx 2.5$ units of $(\hbar/4\pi)g_r^{\uparrow\downarrow}h^2$.

References

- (1) Baltz, V.; Manchon, A.; Tsoi, M.; Moriyama, T.; Ono, T.; Tserkovnyak, Y. Antiferromagnetic Spintronics. *Rev. Mod. Phys.* **2018**, *90* (1), 015005. <https://doi.org/10.1103/RevModPhys.90.015005>.
- (2) Johnson, F. M.; Nethercot, A. H. Antiferromagnetic Resonance in MnF₂. *Phys. Rev.* **1959**, *114* (3), 705–716. <https://doi.org/10.1103/PhysRev.114.705>.