Supporting Information
Tripling Magnetite's Thermoelectric Figure of Merit with Rare Earth Doping

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Supplementary Figures

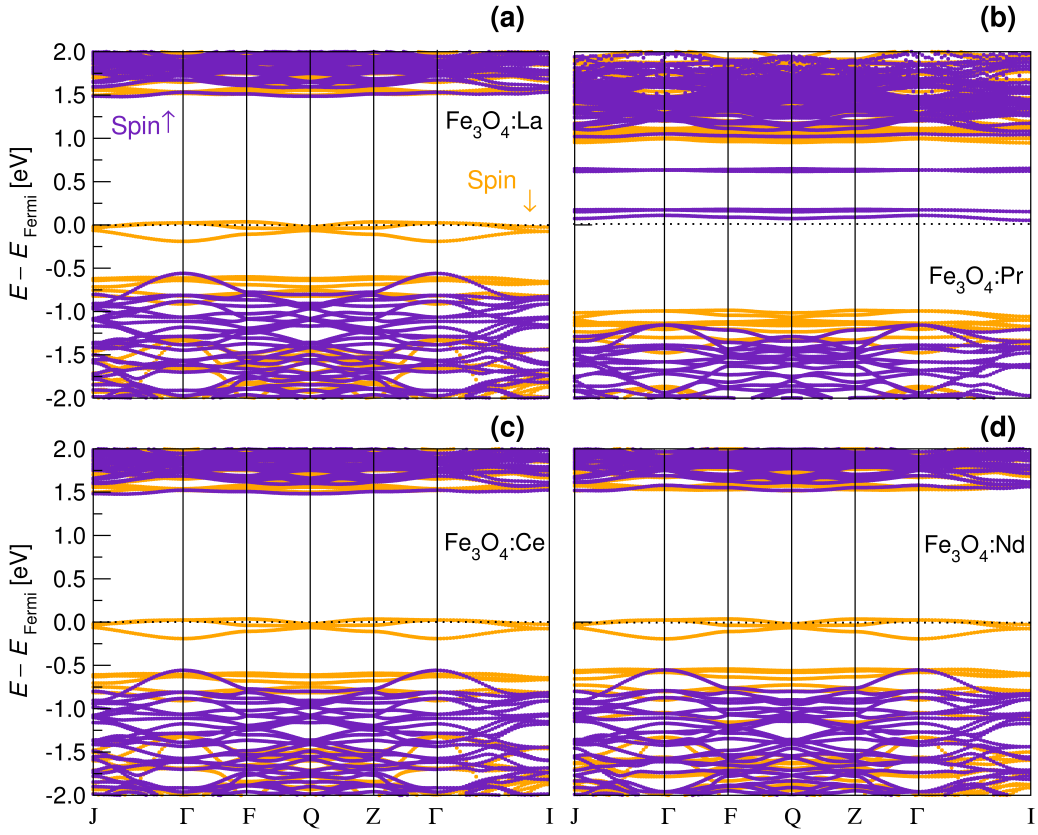

Figure S1: (a) through (d) The band structure of the Fe_3O_4:RE compounds. Purple and orange lines represent spin-up and spin-down bands, respectively.

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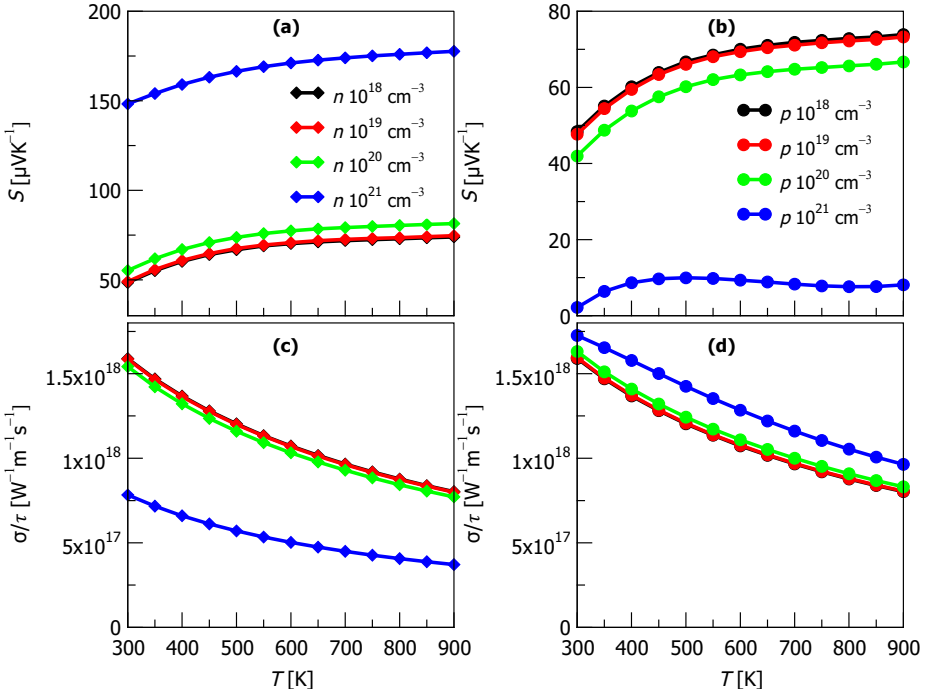

Figure S2: (a) and (b) Predicted S values for Fe_3O_4:La as a function of temperature for different doping concentrations. (c) and (d) The electrical conductivity per relaxation time (σ/τ) for Fe_3O_4:La.

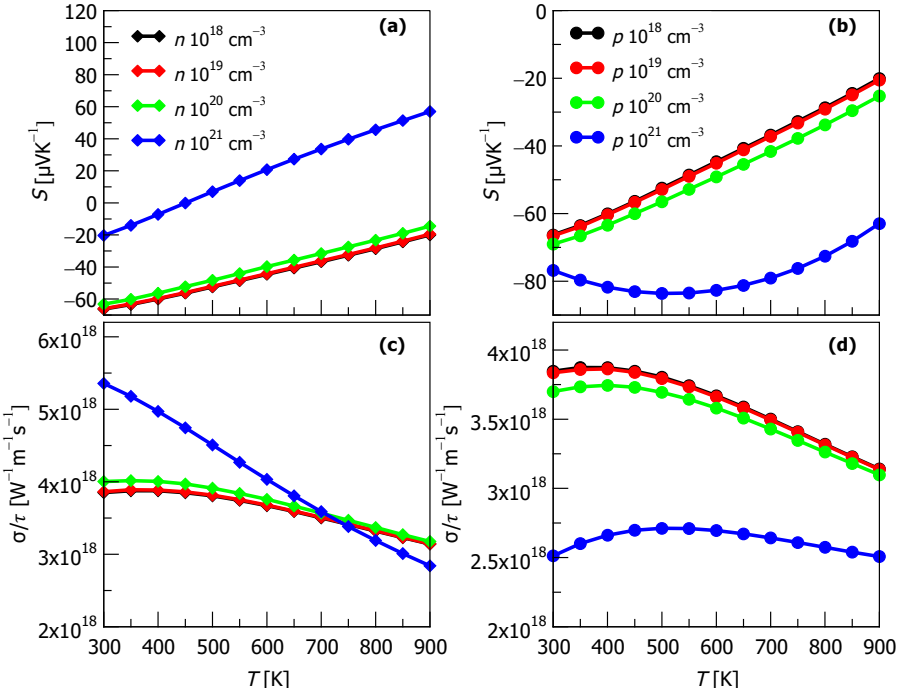

Figure S3: (a) and (b) Predicted S values for Fe_3O_4:Ce as a function of temperature for different doping concentrations. (c) and (d) The electrical conductivity per relaxation time (σ/τ) for Fe_3O_4:Ce.

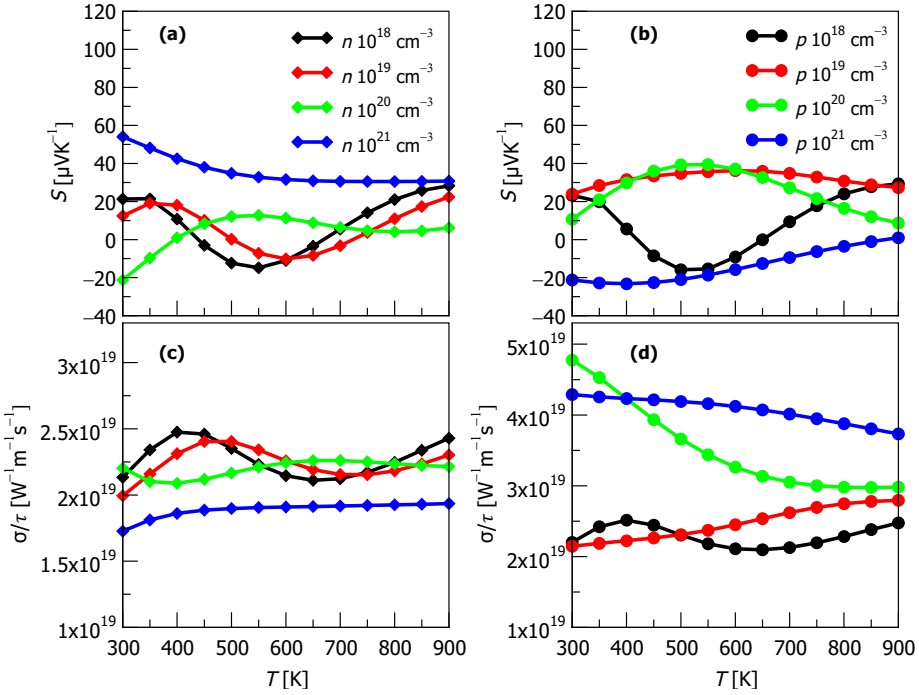

Figure S4: (a) and (b) Predicted S values for Fe_3O_4:Pr as a function of temperature for different doping concentrations. (c) and (d) The electrical conductivity per relaxation time (σ/τ) for Fe_3O_4:Pr.

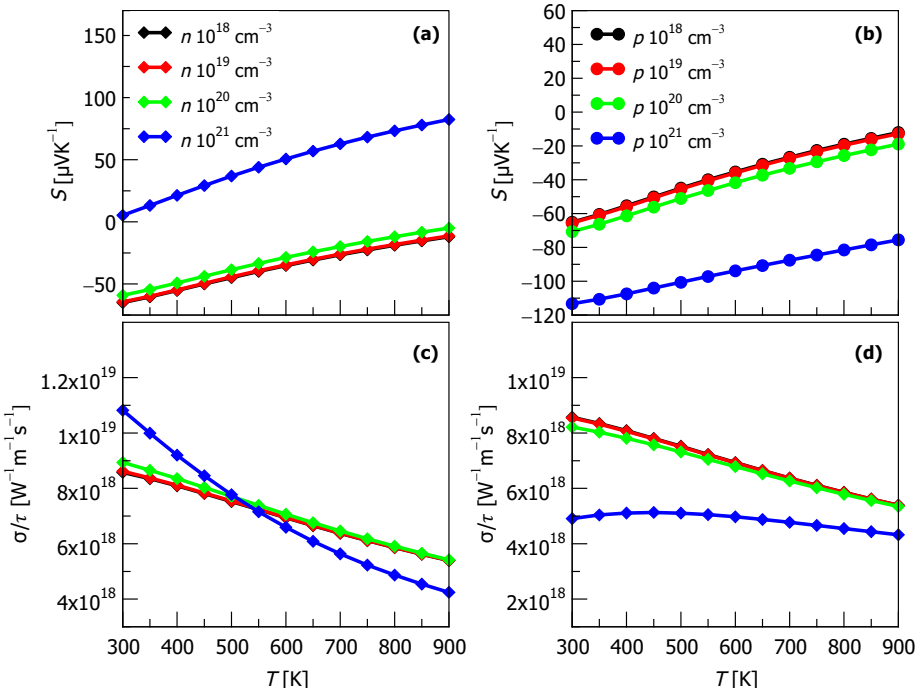

Figure S5: (a) and (b) Predicted S values for Fe_3O_4:Nd as a function of temperature for different doping concentrations. (c) and (d) The electrical conductivity per relaxation time (σ/τ) for Fe_3O_4:Nd.

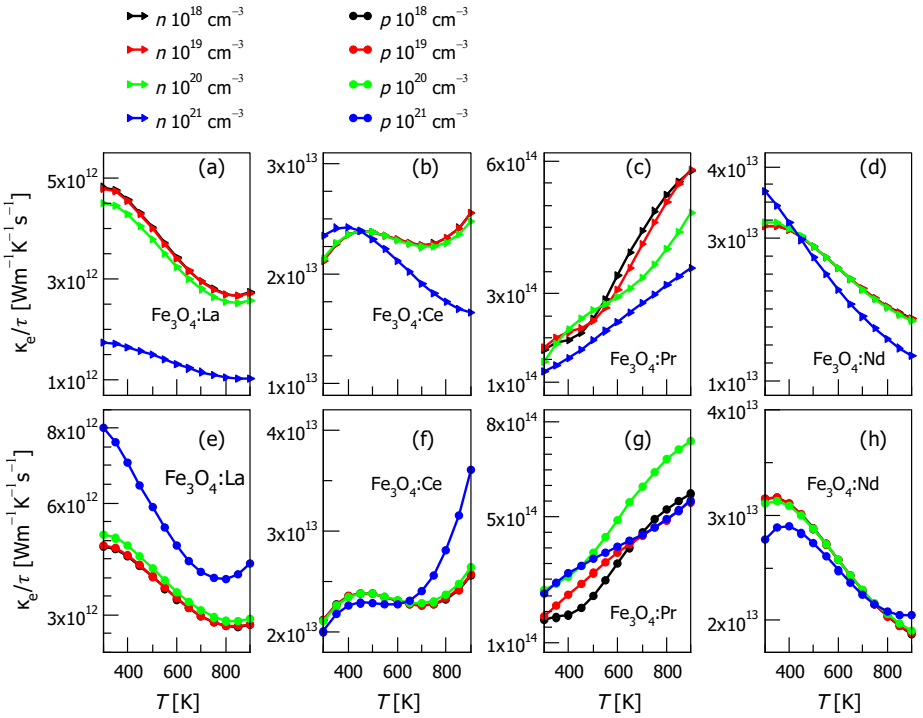

Figure S6: The electronic contribution to the thermal conductivity per relaxation time (κ_e/τ) in Fe_3O_4:RE.

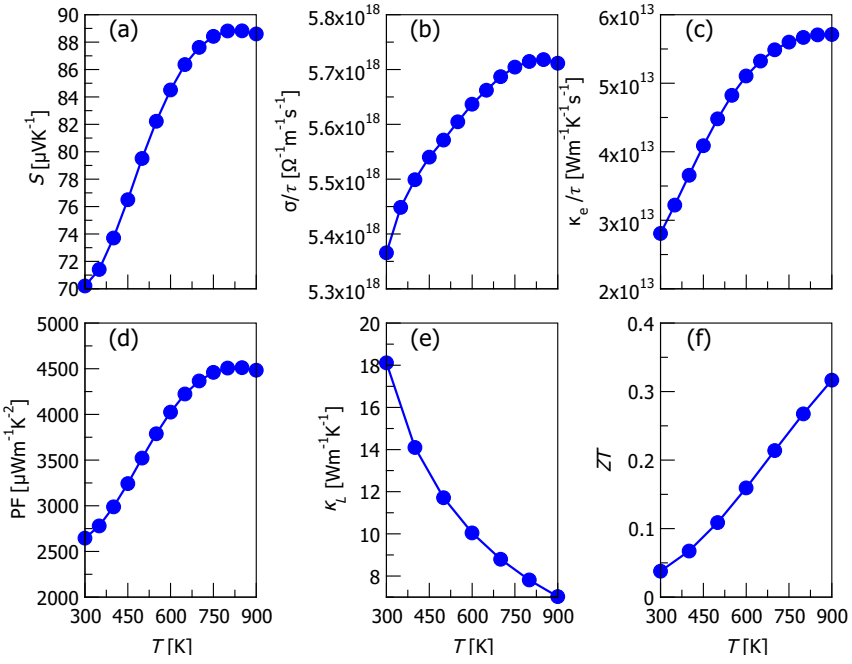

Figure S7: The transport and thermal properties of pristine Fe_3O_4. Here, κ_L was calculated using classical lattice dynamics with Lewis & Catlow force fields that were parameterized by being fit to data derived from experimental measurements of iron oxides [S1]. The obtained κ_L values have the same order of magnitude as the few reported experiments [S2, S3]. However, a perfect match is elusive as experimental samples always had a degree of porosity. It is known that increased porosity can reduce κ_L by an order of magnitude [S4]. The classical lattice dynamics calculations were ~ 100 times more resource-efficient than the ones based on *ab initio* calculations.

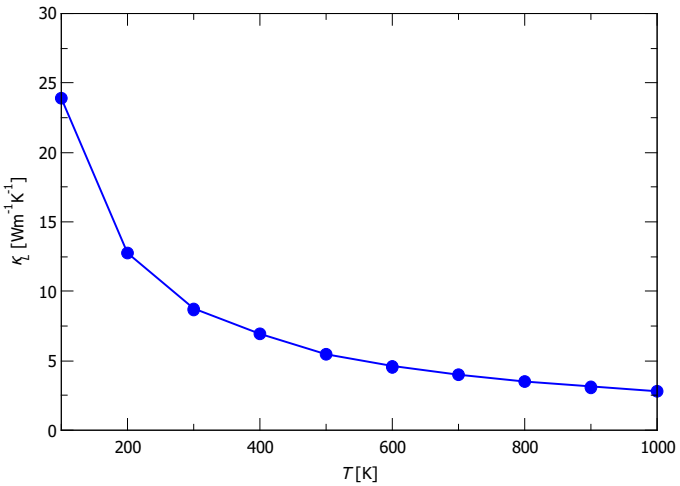

Figure S8: κ_L for Nd-doped Fe_3O_4 calculated using classical lattice dynamics based on Lewis & Catlow force fields. At higher temperatures, there is a reasonable agreement with the machine-learning force field results reported in Fig. 4. At lower temperatures, nonetheless, the results based on Lewis & Catlow force fields are up to ~ 3 times larger than those from machine-learning force fields. The disparity might stem from the fact that in the Lewis & Catlow force fields, the Nd$-$O interaction was parametrized based on neodymium oxide with an equilibrium bond length of 2.365 Å. However, in Nd-doped Fe_3O_4, the Nd$-$O bond length is 2.321 Å. In this case, the technique based on *ab initio* calculations seems more reliable.

Supplementary Table

Table S1: The total DFT energy of the elemental dopants in metal form (E^{DFT}(RE)), the formation energies of the four rare-earth doped magnetite (ΔH) studied, and their corresponding effective mass (m^*). The small variation in m_e^* and m_h^* justifies the constant τ approximation throughout the calculations.

Compound	E^{DFT}(RE) (eV/atom)	ΔH (eV)	m_e^* (m_0)	m_h^* (m_0)
Fe_3O_4:La	−19.1035	4.7820	5.88	−1.18
Fe_3O_4:Ce	−19.0835	4.9327	5.88	−1.18
Fe_3O_4:Pr	−36.2322	4.2288	5.56	−2.95
Fe_3O_4:Nd	−29.1788	4.1644	5.90	−1.26

References

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