ADDENDUM

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A strong ferroelectric ferromagnet created by means of spin–lattice coupling

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This Letter determined that $EuTiO_3$, when appropriately strained, becomes a strong ferroelectric ferromagnet, in agreement with prediction. Strong ferroelectrics are proper ferroelectrics, having polarization as their order parameter, with high paraelectric-to-ferroelectric transition temperatures (T_c). Such ferroelectrics are manifested by a high T_c and a high peak at T_c in the dielectric constant versus temperature behaviour, signifying that ferroelectricity is driven by the soft mode, which is indicative of proper ferroelectricity. Our measurements of strained $EuTiO_3$ demonstrate both of these characteristics (shown in Fig. 3 of our Letter), and led us to conclude that strained $EuTiO_3$ is a strong ferroelectric. In contrast, all well-established prior single-phase ferroelectric ferromagnets are improper or pseudoproper ferroelectrics (that is, with weak ferroelectricity resulting in minuscule P_s). We did not present P_s values in our Letter. Second harmonic generation measurements do not provide quantitative values of P_s and attempts to determine P_s via pyroelectric measurements (Yan, L., Li, J. F. & Viehland, D., personal communication)[1] resulted in unphysically high values, presumably owing to electrical leakage. Nonetheless, the magnitude of the P_s of our strained $EuTiO_3$ films can be estimated as follows. In their classic work, Abrahams, Kurtz, and Jamieson[1] established a correlation between P_s and T_c for displacive ferroelectrics. By studying numerous displacive ferroelectrics they found[1]

$$T_c = (2.00 \pm 0.09) \times 10^4 (\Delta z)^2 \qquad \text{(equation (1) of ref. 1)}$$

and

$$P_s = (258 \pm 9)\Delta z \qquad \text{(equation (5) of ref. 5)}$$

where T_c is the paraelectric-to-ferroelectric transition temperature in K, Δz is the atomic displacement of the 'homopolar' metal atom in Å, and P_s is the spontaneous polarization of the ferroelectric in $\mu C\,cm^{-2}$. Combining these equations to eliminate Δz allows P_s to be estimated from T_c in displacive ferroelectrics. The huge anomaly of the soft optical phonon near T_c that we observe (Supplementary Fig. 1 of our Letter) shows that strained $EuTiO_3$ is a displacive ferroelectric, making the aforementioned correlation applicable. Plugging in our measured value of T_c (Fig. 3 in our Letter) yields $P_s = 29 \pm 2\ \mu C\,cm^{-2}$ for our strained $EuTiO_3$ films from this established correlation. This rough estimate is consistent with our first-principles theoretical predictions—$P_s = 21\ \mu C\,cm^{-2}$ for $EuTiO_3$ under +1.1% biaxial tension, corresponding to the strain of our commensurate $EuTiO_3$ films grown on (110) $DyScO_3$. Thus, the data in our Letter shows that appropriately strained $EuTiO_3$ is a strong ferroelectric ferromagnet.

1. Abrahams, S. C., Kurtz, S. K. & Jamieson, P. B. Atomic displacement relationship to Curie temperature and spontaneous polarization in displacive ferroelectrics. *Phys. Rev.* **172,** 551–553 (1968).