

Rapid heat-assisted polarization reversal in a ferroelectric thin film

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ABSTRACT

We demonstrate that switching of ferroelectric thin films sandwiched between metallic electrodes can be controlled by laser-assisted heating, reminiscent of heat-assisted magnetic recording. We employ electrical switching cycles that quantify the electrically switchable remanent polarization P_r and show that 300 ns voltage pulses alone change the polarization by less than $\Delta P < P_r$. Transient heating of the metallic top electrode by synchronized ns laser pulses induces a reversal $\Delta P^L > P_r$ of the average polarization. The transient average temperature modeled by the heat equation can rationalize the polarization change observed for different relative timings Δt of the laser pulse if it arrives before the electrical pulse.

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Recent advances in the field of heat-assisted magnetic recording (HAMR)¹ have underscored the pivotal role of thermal energy in overcoming energy barriers in ferromagnetic systems. This energy can be provided in tiny amounts via nanophotonics.² HAMR has spurred significant progress in data storage technologies—with mass production on the market since 2024—while providing deeper insights into the fundamental physics of ferroelectric materials, i.e., systems that can be switched between two states. The spontaneous polarization in ferroelectric materials can equally well represent bits, and the double-well potential landscapes exhibit striking parallels to ferromagnets, both in their functional characteristics and in the underlying mechanisms governing their behavior,³ including ultrafast photoferroic recording.⁴ In particular, ferroelectric thin films have garnered widespread attention for their enhanced polarization stability, scalability, and tunability at the nanoscale.^{5–7} They are ideal candidates for applications in non-volatile memory devices, sensors, and energy-efficient electronic components.⁸ Sufficiently large electric fields can overcome the energy barriers separating the stable polarization states. However, achieving efficient and rapid polarization switching remains challenging.^{9,10} Light control of rapid ferroelectric switching was achieved in various ways. Photoconductive switches rapidly provide voltage.¹¹ Direct photoexcitation reduces the barrier by changing the electronic

structure,^{12,13} screening the depolarization field,¹⁴ or via interface effects.¹⁵ THz light fields couple directly or indirectly to the ferroelectric soft mode^{16,17} and enable switching. UV light and femtosecond lasers have been successfully applied for permanent laser poling of ferroelectrics.^{18–21} However, infrared pulses that heat the ferroelectric via the adjacent metal electrode address different physics without inter-band excitation of the ferroelectric.

Indirect laser heating brings the film close to its Curie temperature, effectively lowering the activation energy for polarization switching, as initially demonstrated by Merz in seminal studies on BaTiO₃,^{22,23} and more recently confirmed in ferroelectric superlattices.²⁴

In this study, short infrared laser pulses that do not photoexcite the ferroelectric deposit thermal energy in the metal electrode, which transfers heat to the nanoscale ferroelectric thin film, enabling rapid control of the system's free energy landscape by light pulses. We explore heat-assisted polarization reversal (HAPR) by integrating pulsed laser excitation into a ferroelectric testing environment and show that the simultaneous excitation by thermal energy and external fields exploits the fundamental nonlinear physics associated with double-well potentials, as the result is not a linear superposition of the individual excitations. We find experimental conditions where the laser pulse alone only produces a very weak pyroelectric response, and

the electric field alone is insufficient to switch the average polarization. Only when the laser heats the film during the application of the electric field, the polarization is reversed. If the laser pulse arrives before the electrical field pulse, the polarization response to both excitations depends approximately on their relative timing as predicted by one-dimensional heat conduction. Since the photon energy is below the bandgap, the laser is exclusively absorbed in the metal electrode. Thus, we conclude that the energy barrier separating the two polarization states is transiently reduced by the laser-heating transferred from the electrode, facilitating polarization switching under an external electric field. This laser-induced thermal activation represents a powerful tool for manipulating the polarization dynamics of ferroelectric films and opens new doors for practical applications in energy-efficient electronics and beyond.

The devices consist of a 20 nm thick epitaxial SrRuO₃ layer (bottom electrode) grown on SrTiO₃ (100) by pulsed laser deposition, a 60 nm Pb(Zr_{0.2}Ti_{0.8})O₃ thin film and 20 nm thick Pt top electrodes with a diameter of $d = 340 \mu\text{m}$. The experimental design builds on previous studies, in which we investigated the response of similar ferroelectric thin film devices, for which we correlated the structural response—measured by x-ray diffraction²⁵—with the electrical response for sub-coercive fields²⁶ and high frequencies.²⁷ In the present study, we added a Q-switched Ti:Sapphire laser system that emits 10 ns laser pulses at a wavelength of 780 nm. We quantify the fluence $F = 100 \text{ mJ/cm}^2$ on the sample by measuring the laser spot diameter

of $d = 340 \mu\text{m}$ via a CCD camera at the sample position. Thus, the heating is rather homogeneous across the top electrode.

Figure 1(a) illustrates the concept of HAPR by showing the free energy F of the system as a function of polarization P modeled as a Taylor expansion.²⁸ Without applied voltage (upper panel) a clear energy barrier separates the two polarization states, even upon considerable heating, as long as $T < T_c$. A strong applied electric field E tilts the potential energy landscape (lower panel), which prefers one ferroelectric polarization over the other. Now, the same temperature rise can essentially remove the barrier. Because of the energy barrier, the polarization switching is driven by domain nucleation and growth, according to the Avrami model.²⁹ Ultimately, suppression of the barrier for a few ps and concomitant ultrafast switching should be possible in nanoscale volumes by optimized thermal management.³⁰

Figure 1(d) shows a standard hysteresis loop measured via a triangular voltage ramp at 1 kHz. The remanent polarization $\pm P_r$ is given by the two values of the polarization for zero applied voltage. Figure 1(c) shows the applied voltage sequences that mimic a classical ferroelectric testing cycle. The black voltage sequence is used to cross-check the hysteresis loop from panel (d) via a positive-up-negative-down (PUND) sequence. As usual, the polarization change (black line in panel f) is found as the integral of the measured current. The gray line in panels b and c indicate the much shorter first negative voltage pulse, which is used in the laser-assisted measurements. Also, the fourth voltage pulse (positive) is shortened to make the excitation

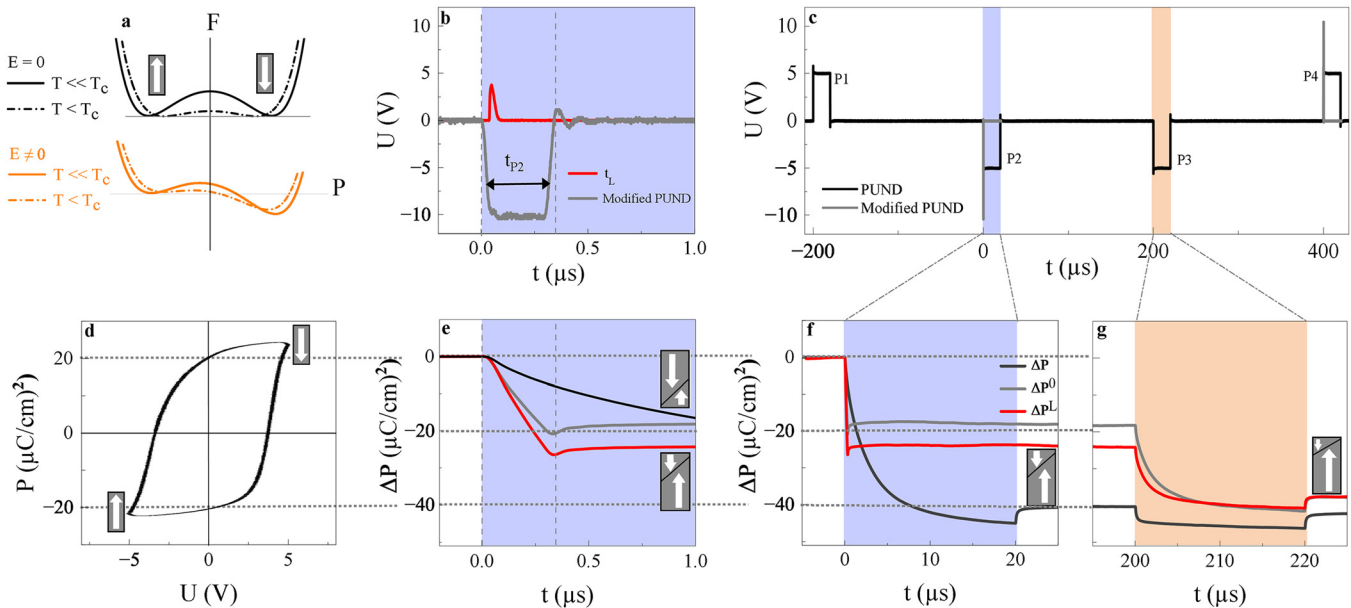


FIG. 1. (a) Model of the free energy F of a ferroelectric system as a function of polarization P , showing the effect of temperature and electric field. (b) Measured timing of the 300 ns short voltage pulse (gray) and synchronized laser pulse (red). Note the blue shade references to panels (c), (e), and (f). (c) Voltage pulse sequence for standard PUND (black) with 20 μs pulses with 5 V amplitude and modified sequence with short voltage pulses (gray) with 10 V amplitude used in the laser-assisted experiments. (d) Hysteresis loop measured for a triangular waveform at 1 kHz. The arrows indicate polarization up when negative voltage is applied to the top electrode. (e) Polarization change ΔP for 300 ns electrical pulses without (gray) and with (red) laser pulse plotted together with the polarization P (black) derived from the PUND sequence shown in panel (c). Note the dashed lines indicating the timing in panels (b) and (e) and the dotted lines connecting the remanent polarization P_r measured in panel (d) with the time-resolved polarization measurements (ΔP), which have the same amplitude. (f) Same as (e) on a longer timescale. (g) Cross-check of the remaining switchable polarization after the short voltage pulse with (red) and without (gray) laser with a saturating pulse. Indeed, less polarization can be switched by this long saturating pulse if the laser had assisted the switching by the previous short voltage pulse. Note that the new domain state is stable on the measured timescale.

symmetric, to enhance the lifecycle of the device. The pulse sequence loops at 1 kHz synchronized to the 10 ns laser pulse at 780 nm wavelength. Each short electrical field pulse is followed by a saturating long voltage pulse of the same polarity and preceded by a saturating long pulse of opposite polarity. There are three relevant timescales: the electric field pulse duration, the field-free time, and the time between the laser pulse and the field pulse. The zoom in Fig. 1(b) details the timing of the laser pulse t_L (red) and the voltage pulse duration t_{p2} (gray). Throughout the manuscript, we refer to voltages and not to electric fields, because this is the measured quantity, and the internal field may strongly depend on the dielectric constant of interface layers.

Figure 1(e) shows the most relevant result: The gray line indicates the polarization change ΔP^0 , which takes place during the 300 ns negative voltage pulse without laser excitation. The quantitative comparison to the hysteresis loop in Fig. 1(d) shows that $|\Delta P^0| < P_r$, i.e., the average polarization is not reversed. The red line, in contrast, indicates that the simultaneous application of the laser pulse increases the polarization change $|\Delta P^L| > P_r$, indicating an average polarization reversal of the device. The polarization change is faster under laser-heating, and therefore, the total number unit cells switching during the 300 ns electric field pulse is larger. The same result is shown again in Fig. 1(f) for an extended timescale that allows for a comparison with the result from the standard PUND sequence (black line) that corresponds to the color-coded voltage in panel (c). For technical reasons, the data acquisition takes about 1 min for each trace shown. This corresponds to 60 000 laser pulses and the same number of switching cycles. On the one hand, this demonstrates the robustness of the measurement; on the other hand, it is true that in some devices, significant leakage occurs, which increases with measurement time. In order to cross-check that the increased ΔP^L in the laser-assisted measurement is indeed due to increased polarization reversal and not due to increased leakage, we additionally quantified the remaining polarization reversal that occurs during the second negative pulse, which has a long duration of 40 μ s and saturates the polarization. Figure 1(g) shows that the saturating voltage pulse induces less polarization switching when the laser pulse had been applied during the preceding electrical pulse (red), as partial switching already occurred during the short negative voltage pulse.

Figure 2 reports how the timing of the laser pulse with respect to the voltage pulses influences the polarization change. Panel (a) shows the short voltage pulse (gray) together with the laser pulse timings (red) for four selected time delays. Note the break in the time axis. The applied voltage alone induces a current (I^0) composed of a positive charging contribution and a positive ferroelectric switching current, as shown in Panel (b). This ferroelectric contribution additionally increases immediately upon laser excitation, such that the currents (I^L) depend on the laser arrival time t_L . Upon switching off the voltage, there is a negative current contribution due to the discharging associated with the dielectric contribution of the capacitor. When the laser arrives at $\Delta t = -60$ μ s, i.e., long before the 300 ns voltage pulse, the response shows a tiny pyroelectric current, in addition to the current induced by the applied voltage pulse only (I^0). Panel (c) shows the integrated current signal, i.e., the polarization change without (ΔP^0) and with (ΔP^L) laser excitation for each timing. For $\Delta t = -60$ μ s, net zero polarization change due to the purely pyroelectric response is confirmed (see inset). The small difference in polarization change ΔP^0 and ΔP^L at $\Delta t = 60$ μ s can be attributed to fatigue of this particular

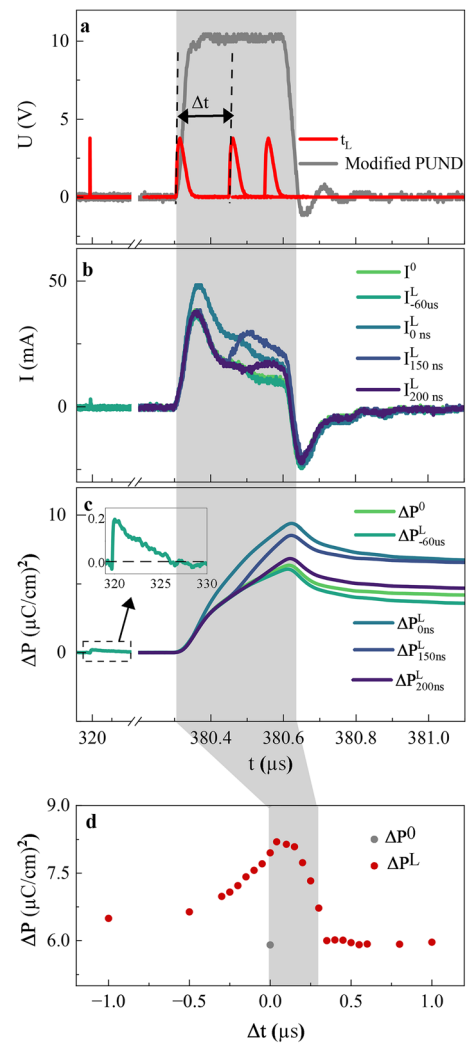


FIG. 2. (a) Applied voltage pulse (gray) and four different laser timings (red) relative to the onset of the voltage pulse. (b) Current response without laser pulse excitation (I^0) and with laser excitation (I^L) for the four selected laser pulse timings t_L . (c) Polarization without laser pulse excitation (ΔP^0) and with laser excitation (ΔP^L) for various t_L . Note the gray shade indicating the on-time of the electric field pulse. The time window 320–325 μ s in panels (b) and (c) highlights the pyroelectric current and polarization, respectively. The inset in panel (c) shows the pyroelectric response due to laser excitation when no voltage is applied. (d) Polarization change (ΔP) as a function of laser timing (Δt) relative to the onset of the voltage pulse.

device under the current illumination conditions. Although each device shows slightly different fatigue, the presented results are robust. Panel (d) further illustrates the polarization change due to laser excitation ΔP^L measured for several Δt between the laser and the voltage pulse. ΔP^L peaks at $\Delta t = 40$ ns when the laser pulse coincides with the maximum of the voltage pulse. This suggests that optimal switching occurs when the thermal activation and the applied voltage act simultaneously. When the laser pulse arrives at $\Delta t > 100$ ns, ΔP^L rapidly falls to the constant ΔP^0 (black dot), which is measured without laser excitation.

When the laser pulse interacts with the electrode at times before the onset of the voltage pulse, i.e., at $\Delta t < 0$, the polarization change ΔP^L gradually decreases because the ferroelectric has the highest temperature shortly after the laser pulse has been absorbed and cools down as the thermal energy dissipates toward the substrate. In order to check this hypothesis, we implemented a 1D heat transport model in the udkm1Dsim modeling package,³¹ assuming literature parameters of the thermophysical constants (see Table I). We start the modeling with the Pt film at the temperature given by the absorbed photon energy, and the rest of the sample at room temperature. Figure 3(a) shows the calculated temperature change when we assume a rather poor heat transport across the PZT/SRO interface. The decay of the temperature with time delay $|\Delta t|$ matches very well with the data points reproduced from Fig. 2(d) for negative $\Delta t < 0$. In order to achieve this excellent fit, we assumed a 2 nm SRO layer with a much poorer heat conductivity of $\kappa = 0.0018$ W/(m K) to model interface resistance. The agreement is taken as an indication that the timescale of heat transport is relevant to the heat-assisted switching.

In Fig. 3(b), we present an additional set of data where laser excitation approximately doubles the relative amount of polarization change. This is shown by the black filled circles in the figure for different voltage pulse durations t_{p2} for a relative delay of $\Delta t = 0$ of the laser pulse with respect to the onset of the applied voltage pulse with $U = 5$ V. The laser-assisted polarization change ΔP^L amounts to about 240 % of the switching achieved without laser for $t_{p2} = 0.5$ ns. For an even shorter fixed voltage pulse duration of $t_{p2} = 300$ ns, we varied the time delay Δt (squares) and found the maximum increase in switching efficiency for the smallest $\Delta t = 0$ ns, i.e., when the laser heats the film immediately when the electric field pulse is applied. For voltages ranging from 6 to 8 V, we found increases of switching efficiency up to 275 %. When the voltage pulse duration is short, the applied electric field has limited time to induce switching. In this regime, the transient laser heating significantly lowers the energy barrier, strongly enhancing the polarization reversal. For longer voltage pulses, some of the laser-induced heat dissipates before the switching process completes for a given duration, reducing polarization reversal.

We are convinced that much better contrast ratios can be achieved with optimized devices and optimized voltage and laser pulse sequences. In particular, a smaller area and improved electrical contact, as well as thinner electrodes and ferroelectric films, should drastically decrease the heat load and concomitant fatigue. The voltage pulse duration is ideally as short as the timescale over which heating is significant. In smaller structures, this can easily go down to 100 ps, and then similarly short voltage pulses can be used, which significantly reduce the polarization switching without a laser pulse. Different material compositions and strain conditions may help to improve the domain wall propagation velocity. The faster the process and the smaller the ferroelectric volume that needs to be heated, the less energy will be

TABLE I. Relevant thermophysical properties of constituent materials.

Material	λ_{pen} (nm)	κ_{th} (W/(m K))	C_p (J/(kg K))
Pt (20 nm)	7.8 (Ref. 1)	5 (Ref. 2)	132
PZT (50 nm)	∞	5.72 (Ref. 3)	300 (Ref. 3)
SRO (20 nm)	∞	1 (0.0018) (Ref. 3)	481 (Ref. 3)
STO substrate	∞	10 (Ref. 3)	697 (Ref. 3)

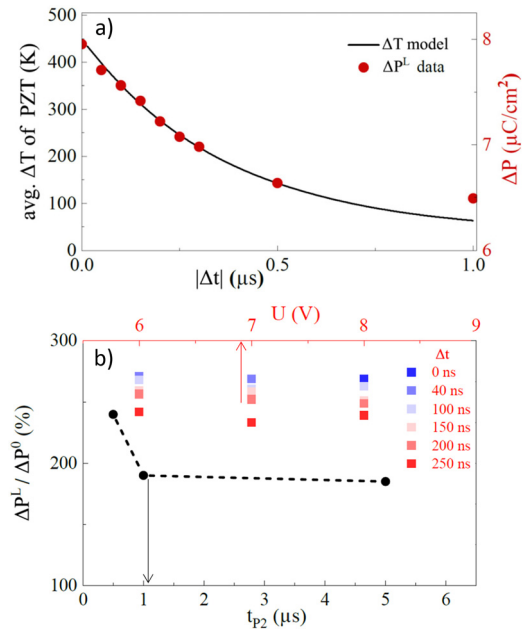


FIG. 3. (a) The average temperature change ΔT in the PZT film is extracted from modeling the heat transport through the heterostructure. It approximately reproduces the dependence of the laser-induced polarization change ΔP^L on the time delay Δt between the laser pulse and the electrical pulse. (b) Dots: relative polarization enhancement as a function of voltage pulse duration (t_{p2}) with a pulse delay of $\Delta t = 0$ and $U = 5$ V. Squares: the same for a constant voltage pulse duration $t_{p2} = 300$ ns and as a function of applied voltage U for various Δt . Data in this figure were recorded on a different device from Figs. 2 and 3.

consumed for the laser-assisted switching. As an order of magnitude estimate for the small energy of the light pulse required to raise the temperature by $\Delta T = 100$ K for an optimized barrier, we take a 10 nm thick ferroelectric film with a 200 nm electrode diameter. The pulse energy is only $W = C_V V \Delta T = 0.1$ pJ, using a specific heat of about 2.6×10^6 J/(m³ K).

In conclusion, our experimental results show that ferroelectric polarization switching is enhanced when a laser pulse deposits thermal energy into the metallic electrode, which is subsequently transferred to the adjacent ferroelectric thin film, thereby lowering the energy barrier. The enhancement is robust across various applied voltages and time delays Δt between the laser and voltage pulses. Under favorable conditions, the combination of heat and electric field can lead to switching of the average polarization direction. This laser-assisted polarization switching is indeed a heat-assisted polarization reversal (HAPR), since the light only interacts with the metal electrode, which increases the temperature of the ferroelectric. Optimization of the device material and thickness, together with proper lateral size scaling and heat management, should improve the switching contrast dramatically. These advancements make HAPR a promising approach for the development of energy-efficient electronics, non-volatile memory devices, and next-generation data storage technologies.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Rekikua Alemayehu: Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Visualization (lead); Writing – original draft (lead). **Steffen Zeuschner:** Formal analysis (supporting); Software (lead); Validation (supporting); Writing – review & editing (supporting). **Alexander von Reppert:** Resources (supporting); Supervision (supporting); Writing – review & editing (equal). **Matthias Rössle:** Methodology (equal); Resources (equal); Supervision (equal); Writing – review & editing (equal). **Marin Alexe:** Funding acquisition (equal); Methodology (equal); Project administration (equal); Resources (equal); Writing – review & editing (equal). **Matias Bargheer:** Conceptualization (lead); Funding acquisition (lead); Methodology (equal); Resources (lead); Supervision (lead); Writing – original draft (lead); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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