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Optical Writing of Magnetic Properties by Remanent Photostriction

V. Iurchuk,¹ D. Schick,² J. Bran,¹ D. Colson,³ A. Forget,³ D. Halley,¹ A. Koc,^{2,4} M. Reinhardt,^{2,4} C. Kwamen,^{2,4}
 N. A. Morley,⁵ M. Bargheer,^{2,4} M. Viret,³ R. Gumenuik,⁶ G. Schmerber,¹ B. Doudin,¹ and B. Kundys¹

¹*Institut de Physique et Chimie des Matériaux de Strasbourg (IPCMS), UMR 7504 CNRS-UdS 23 rue du Loess, 67034 Cedex 2, Strasbourg, France*

²*Institute for Methods and Instrumentation for Synchrotron Radiation Research, Helmholtz-Zentrum Berlin, Albert-Einstein-Str. 15, 12489 Berlin, Germany*

³*Service de Physique de l'Etat Condense, DSM/IRAMIS/SPEC, CEA Saclay URA CNRS 2464, 91191 Gif-Sur-Yvette Cedex, France*

⁴*Institut für Physik & Astronomie, Universität Potsdam, Karl-Liebknecht-Str. 24-25, 14476 Potsdam/Golm, Germany*

⁵*University of Sheffield, Department of Materials Science and Engineering, Mappin Street, Sheffield S1 3JD, United Kingdom*

⁶*Institut für Experimentelle Physik, TU Bergakademie Freiberg, Leipziger Str. 23, 09596 Freiberg, Germany*

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1 We present an optically induced remanent photostriction in BiFeO₃, resulting from the photovoltaic effect, which is used to modify the ferromagnetism of Ni film in a hybrid BiFeO₃/Ni structure. The 75% change in coercivity in the Ni film is achieved via optical and nonvolatile control. This photoferromagnetic effect can be reversed by static or ac electric depolarization of BiFeO₃. Hence, the strain dependent changes in magnetic properties are written optically, and erased electrically. Light-mediated straintronics is therefore a possible approach for low-power multistate control of magnetic elements relevant for memory and spintronic applications.

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Multiferroic phenomena are often summarized in a Venn diagram showing the intersection of ferromagnetic, ferroelectric, and ferroelastic orders [1], each with its own control field. Numerous electric methods of magnetization control use elastic strain to leverage magnetoelectric (ME) properties in solids [2–34] and in magnetostrictive-electrostrictive or ferroelectric structures [5–8]. The expected technological benefit is the possibility of low-power [9–11] operation down to the nanoscale [12–15]. Indeed, strain-mediated electric control of magnetic performance of tunnel junctions has been reported [16]. Furthermore, by using the ferroelastic effect of remanent strain, multiple nonvolatile states can be written on piezoelectric substrates [17,18]. Here we present the optical analog of this memory imprint approach, based on photostriction in BiFeO₃ (BFO) [19], a well-studied benchmark multiferroic material [20] exhibiting cross-linked ferroic orders. Light brings a new layer of functionality to multiferroics [21–24]. In particular, photoferroelectric [25] effects associated with above-band gap photovoltaic (PV) properties, [26–28] can mediate light-induced changes of the ferroelastic order. While it is increasingly well established that BFO exhibits strain under illumination [29–31], the possibility of remanent strain states suggests a new approach [32]. The optical control of strain is particularly important for BFO, which possesses both high photostrictive efficiency [32] and large optoelastic coupling [33]. Furthermore, the magnetoelastic coupling in BFO has been shown to dominate its ME properties [34] that can provide a bridge for ME coupling between magnetic and electric orders [35]. These effects, together with the strain-tunable magnonic response in BFO thin

films [36] provide an attractive strain-engineering prospective [37]. Photostriction control can also be extended to miniaturized structures using light-polarization-dependent functionality in ferroelectric domain walls in BaTiO₃ [38] offering an optical degree of control in spin-based devices [39,40]. Here we will first show that light can impact the internal electric field of BFO through the PV effect to produce optically induced ferroelastic remanent states, and then demonstrate the use of this ferroelastic deformation to stress a superposed ferromagnetic film, thereby achieving strain-mediated optical control of the magnetic anisotropy.

Illuminating a material which is ferroelectric (FE) and PV results in above-band gap voltage generation that changes the internal electric field in the sample [41]. The former process can be compared to the action of "subcoercive" electric fields insufficient to saturate the polarization, resulting in minor (nonswitching) FE loops [42]. Figure 1(a) illustrates how light excitation can be an alternative to the electric field, and generate a minor remanent polarization state via the PV effect [Fig. 1(b)]. A continuous wave (cw) 404 nm laser with a 3 ns rise time was used as the illumination source through an optical fiber. The sample was illuminated through a thin (20 nm) Au film, used as contact transparent electrode for depolarizing the substrate. Under constant illumination, a steady-state photocurrent results in an increase of polarization saturating after ~70 sec (not shown). The light induced change in electric polarization partly persists in ~5.5% after the light is switched off [Fig. 1(a)]. One can conclude from Fig. 1(a) that different remanent polarization levels can result from different illumination times. The electric polarization of the

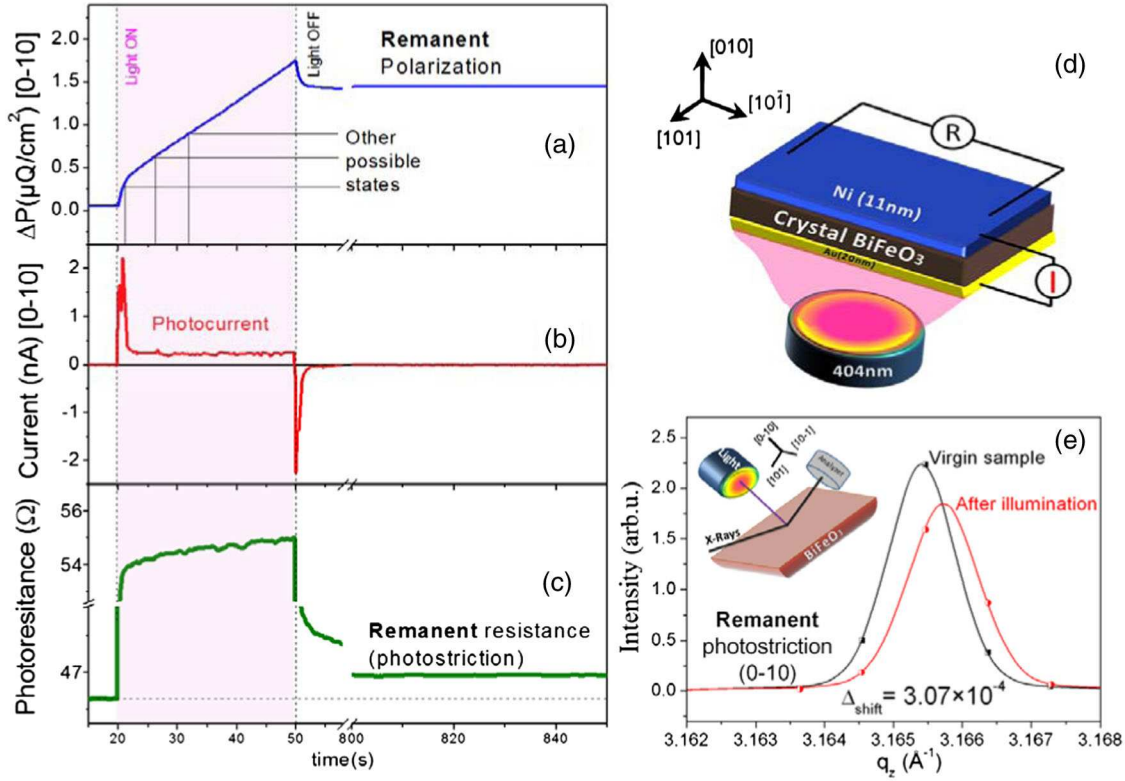


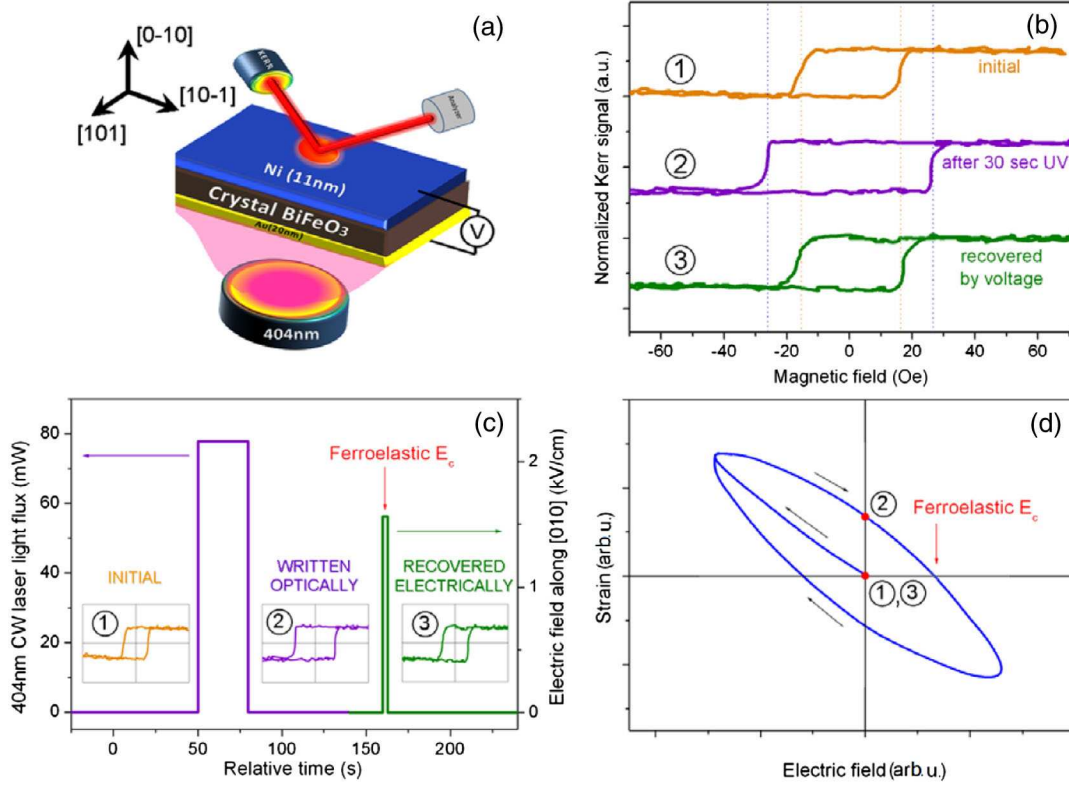
FIG. 1. (a) The remanent polarization state (a) created by 30 sec UV light. (b) Corresponding photocurrent. (c) Remanent photostriction detected by measuring the resistance of the Ni film (d) and by static x-ray diffraction of BFO (e).

84 BFO is the primary order parameter and it results in a
 85 change in strain (which is the secondary order parameter)
 86 that is linearly related to the polarization in the subcoercive
 87 region through the piezoelectric response of the oxide [43].
 88 Figure 1(c) shows the remanent photostriction detected
 89 using a resistive measurement of a Ni thin film adlayer in
 90 the setup illustrated in Fig. 1(d). The overall remanent
 91 strain of the sample in Fig. 1(d) is tensile in the (010) plane
 92 and results in an in-plane expansion of the Ni film. In order
 93 to verify the remanent deformation of the BFO substrate,
 94 we carried out static x-ray diffraction experiments
 95 [Fig. 1(e)] at the XPP/KMC3 beam line in the synchrotron
 96 facility BESSY II (Berlin, Germany) [44]. A similar BFO
 97 crystal with the same orientation (but without adlayer) was
 98 used to determine the lattice spacing along the [010]
 99 direction in the as-grown [45] state and after 3 sec of light
 100 illumination. In this case, a femtosecond pulsed laser was
 101 used yielding a similar integral number of photons to that
 102 used for the switching in Figs. 1(a)–1(c) with the cw laser.

103 The pulsed laser consists of a multistage oscillator and
 104 amplifier system (Impulse, Clark-MXR) and delivers 250 fs
 105 long pulses of 10 μ J pulse energy at a central wavelength of
 106 1030 nm and a repetition rate of 208.3 kHz. They are then
 107 passed through a third harmonic setup at the beam line to
 108 generate the laser pump pulses of 350 nm with a final
 109 average power of 80 mW incident on the sample in a spot
 110 size of $277 \times 176 \mu\text{m}^2$ (FWHM) under an incidence angle

111 of 20° between laser beam and sample surface. The x-ray
 112 photon energy was set to 9 keV with a relative bandwidth of
 113 $\Delta E/E = 10^{-3}$. The x-ray spot size on the sample was
 114 approximately $100 \mu\text{m}^2$ and the experiment was conducted
 115 on a 4-axis goniometer in $\theta/2\theta$ geometry, with the diffracted
 116 photons detected by a DECTRIS Pilatus 100k
 117 hybrid-pixel 2D detector.

118 After illumination, the x-ray scan reveals a remanent
 119 shift of $\Delta q = 3.07 \times 10^{-4} \text{ \AA}^{-1}$, which corresponds to a
 120 relative lattice contraction of 1×10^{-4} along [010] direc-
 121 tion. It is accompanied by a peak broadening in the out- and
 122 in-plane directions, which may be attributed to increase of
 123 intrinsic nanoscale inhomogeneities, possibly related to
 124 ferroelastic domains. No significant sample heating is
 125 expected during the x-ray scan as this would yield lattice
 126 expansion, contrary to our findings. The observed contrac-
 127 tion along the [010] direction leads to an overall lattice
 128 expansion in the (010) plane due to Poisson's ratio and
 129 agrees well with Fig. 1(c) showing tensile remanent photo-
 130 striction. The light is therefore able to induce anisotropic
 131 deformation in BFO that can be used to stress the
 132 magnetostrictive overlayer, as in piezoelectric-magneto-
 133 strictive structures. This possibility is demonstrated by
 134 the experiment in Fig. 2(a), where the 11 nm thick Ni
 135 film was deposited on the flat side of the BFO crystal in an
 136 e -beam evaporator at a rate of 0.1 nm/s for $M(H)$ loop
 137 measurements [Fig. 2(a)]. The remanent photostriction



F2:1 FIG. 2. (a) Schematics of the experiment. (b) Room-temperature ferromagnetic loops of an 11 nm thick Ni film on top of a BFO single
 F2:2 crystal before (1) and after (2) excitation by 404 nm light (fluence 250 J cm^{-2}). The initial $M(H)$ loop (1) can be recovered (3) by an
 F2:3 electric pulse (c) that corresponds to the ferroelastic coercive force E_c as represented by an example sketch (d) [46].

138 largely modifies the magnetic properties of the Ni thin film
 139 [Fig. 2(b)], as revealed by the longitudinal magneto-optic
 140 Kerr effect (MOKE) magnetometry. The shape of the initial
 141 $M(H)$ loop is modified after light exposure, with a change
 142 in coercivity of 75%, which remains stable over a long
 143 period. For this particular sample, we waited 5 days before
 144 electrical recovery tests, but other samples showed that the
 145 effect persisted for more than a month. The scenario
 146 explaining how light can impact magnetic properties is
 147 clearly seen from Fig. 1(a). When the light is turned on, the
 148 concentration of free carriers (electrons and holes) starts to
 149 increase due to the above-band gap PV effect, and the
 150 photocurrent across the BFO crystal stabilizes. This creates
 151 an electric field in the bulk of the crystal that tends to
 152 influence the net polarization [Fig. 1(a)]. Since the magni-
 153 tude of this light-induced electric field is small compared to
 154 the ferroelectric coercive field, there is no polarization
 155 reversal but only slight displacements of the ferroelastic
 156 domains in BFO which contribute to its net deformation.
 157 After the light is turned off, the generation of free carriers
 158 ceases and the ferroelastic domains gradually relax to a new
 159 equilibrium configuration that determines the remanent
 160 photostriction. This optically induced strain is imprinted in
 161 the magnetostrictive Ni adlayer.

162 Successful electrical erasing, namely, recovery of the
 163 initial ferroelastic configuration of BFO, can be achieved in

164 two ways. If the coercive ferroelastic force is known, it can
 165 be done by applying the voltage corresponding to the
 166 ferroelastic coercive force [Fig. 2(d)]. The electric field
 167 amplitude of $5\text{V}/32 \mu\text{m}$ was enough to recover a close to
 168 initial “virgin” $M(H)$ loop in the sample (Fig. 2).
 169 Alternatively, an oscillating damped voltage procedure
 170 analogous to ac demagnetization can be used, as in the
 171 case of electrically written states [17,18]. When the initial
 172 spontaneous ferroelastic state is not characterized, the ac
 173 electrical erasure may be more convenient.

174 The possibility of direct ME coupling at the interface
 175 [47] can be discarded because the optical writing [Fig. 2(a)]
 176 was also demonstrated for samples where a 5 nm Au film is
 177 inserted between the BFO substrate and the Ni film to
 178 screen any electric charges at the interface. The Au film
 179 also excludes the possibility of direct magnetic coupling
 180 between the BFO and the Ni.

181 All MOKE loop measurements were performed at room
 182 temperature after excitation and are therefore free of Joule
 183 heating artifacts. The data shown in Fig. 1(a) obtained
 184 during excitation suggest a negligible heating effect of the
 185 laser light, because the polarization of BFO should
 186 decrease when warming to its ferroelectric Curie temper-
 187 ature of $\sim 1143 \text{ K}$ [48]. A temperature increase of 12.3 K,
 188 detected with a thermal camera during the 30 s illumination
 189 had no influence on the $M(H)$ loops of the Ni film. Even

190 after heating to 325 K (13 K more than detected by the
 191 thermal camera), the $M(H)$ loops remained unchanged. We
 192 can therefore safely infer that the optical modification of
 193 the magnetic properties has a photovoltaic-photostrictive
 194 origin, as confirmed by the electrical erasure test we
 195 performed. Our data indicate that the magnetostriction of
 196 the Ni adlayer explains the modification of its magnetic
 197 properties, originating from the remanent strain state
 198 imprinted by light on the BFO substrate.

199 In conclusion, we have demonstrated that ferroelastic
 200 deformation states can be written optically in BFO, and that
 201 it is possible to erase them electrically. The remanent
 202 photostriction naturally depends on the remanent ferro-
 203 electric state of the sample. The possibility to recover the
 204 initial state of the functional materials is of key importance,
 205 as we observed that the ferroic electric or elastic orders
 206 results in remanent states values that depend on the
 207 sample's history (spontaneous polarization). This observa-
 208 tion requires a special care when performing repetitive
 209 experiments (e.g., pump and probe procedures) with
 210 unsaturated FE samples in order to guarantee proper reset
 211 of the initial polarization. The observed photopolarization
 212 induces a deformation that can be coupled to a ferromag-
 213 netic adlayer, resulting in optically controlled magnetic
 214 anisotropy. This optically induced effect manifests itself in
 215 a 75% change in the ferromagnetic coercivity, exceeding by
 216 55% the well-known electric control in the BaTiO₃/Fe
 217 structures [49] with the nonvolatile and wireless advantage,
 218 thus opening the technologically interesting possibility of
 219 multistate magnetic operation [Fig. 1(a)]. The ultrafast
 220 photostriction in BFO films [50–52] and ceramics [53]
 221 combined with the possibility of ultrafast gating [54],
 222 provides a perspective for light-controlled magnetic switch-
 223 ing devices and magnetoresistive memories on sub-ns time
 224 scales. Furthermore, the fact that photostriction can exist in
 225 a number of different materials [32,55] expands the horizon
 226 of photo-magneto-elastic interactions beyond inorganic
 227 compounds [56].

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