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Equivalence of direct and converse magnetolectric coefficients in strain-coupled two-phase systems

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We demonstrate an equivalence between direct and converse magnetolectric effects in two-phase systems comprising piezoelectric and magnetostrictive materials. This was achieved by recasting the Maxwell relation in terms of the effective electrical and magnetic dipole moments for the system and comparing coupling strengths at the same electrical and magnetic d.c. biases. Our findings therefore apply to magnetolectric systems comprising more than two phases and correct the two widely held but opposing views that are compromised by incorrect parameterization or inconsistent bias conditions. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.3693375>]

Magnetolectric (ME) coupling between electrical and magnetic variables has been observed in both multiferroic and non-multiferroic materials,¹ but it is stronger and thus more technologically promising in two-phase systems comprising strain-coupled piezoelectric and magnetostrictive materials.¹⁻⁵ Direct ME effects are typically parameterized in terms of a coupling coefficient $\alpha_D = dP/dH$ that represents a change in electrical polarization P due to applied magnetic field H . Converse ME effects are typically parameterized in terms of a coupling coefficient $\alpha_C = \mu_0 dM/dE$ that represents a change in magnetization M due to applied electric field E (μ_0 is the permeability of free space). But the relationship between the strength of direct and converse ME effects for two-phase systems is controversial.

One view is that the two-phase systems can be described by the Maxwell relation $dP/dH = \mu_0 dM/dE$ for a single phase,^{1,2} i.e., that the direct and converse ME effects as parameterized above are of equal strength. However, this assumes that the appropriate thermodynamic potential per unit volume contains parent terms $-PE$ and $-\mu_0 MH$ that describe the same phase, so the Maxwell relation is only valid by accident for the special case where both phases occupy equal volume fractions. The alternative view is that direct and converse ME coupling strengths are unrelated in two-phase systems, but inequivalent conditions of electrical and magnetic bias render supporting experimental observations invalid.¹⁻⁵ Both views are therefore incorrect.

Instead, direct and converse coupling strengths may be simply related for two-phase systems via the above Maxwell relation, provided it is rewritten as $dp/dH = \mu_0 dm/dE$, in order to describe the entire system via effective magnetic dipole moment m and effective electrical dipole moment p . The direct effect parameterized as $\alpha_d = dp/dH$, and the converse effect parameterized as $\alpha_c = \mu_0 dm/dE$, are then of equal strength such that $\alpha_d = \alpha_c$. For a simple geometry, m and p correspond directly to M and P for each phase, but

more generally we may write $dQ/dH = \mu_0 dm/dV$ using the experimental variables of charge Q and voltage V .

To confirm the above, we studied direct and converse ME effects in a ME heterostructure comprising a 5 mm \times 10 mm \times 0.5 mm single-crystal slab of piezoelectric $(\text{PbZn}_{1/3}\text{Nb}_{2/3}\text{O}_3)_{93}-(\text{PbTiO}_3)_7$ (PZN-PT, from Microfine Materials Technologies Pte Ltd.) whose (011) faces were sandwiched using epoxy glue between two polycrystalline 75 μm -thick layers of magnetostrictive $\text{Fe}_{82}\text{Ga}_{18}$ (Galfenol, from ETREMA Products, Inc.).

The measurement system that we developed is presented in Fig. 1. The d.c. electrical bias is generated by the high-voltage amplifier that is controlled by the signal generator. The d.c. magnetic bias is generated by an external coil (not shown). Direct ME effects were measured by integrating the current passing through the sample using the current-to-voltage converter whose output is read by the lock-in amplifier. Converse ME effects were measured via the pick-up coil, which was calibrated using our sample by equating the response to an a.c. magnetic field with the response of a vibrating-sample magnetometer to an a.c. magnetic field of the same strength.

At selected electrical and magnetic d.c. bias fields in the range $H = 0-300$ Oe and $E = 0-10$ kV/cm, direct ME measurements were performed using a small a.c. excitation field of amplitude 1 Oe, and converse ME measurements were performed using a small a.c. excitation field of amplitude

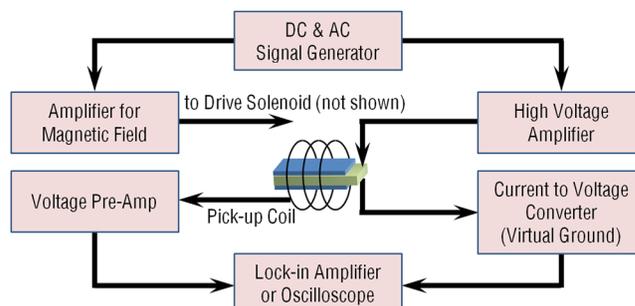


FIG. 1. (Color online) Diagram of the measurement set-up.

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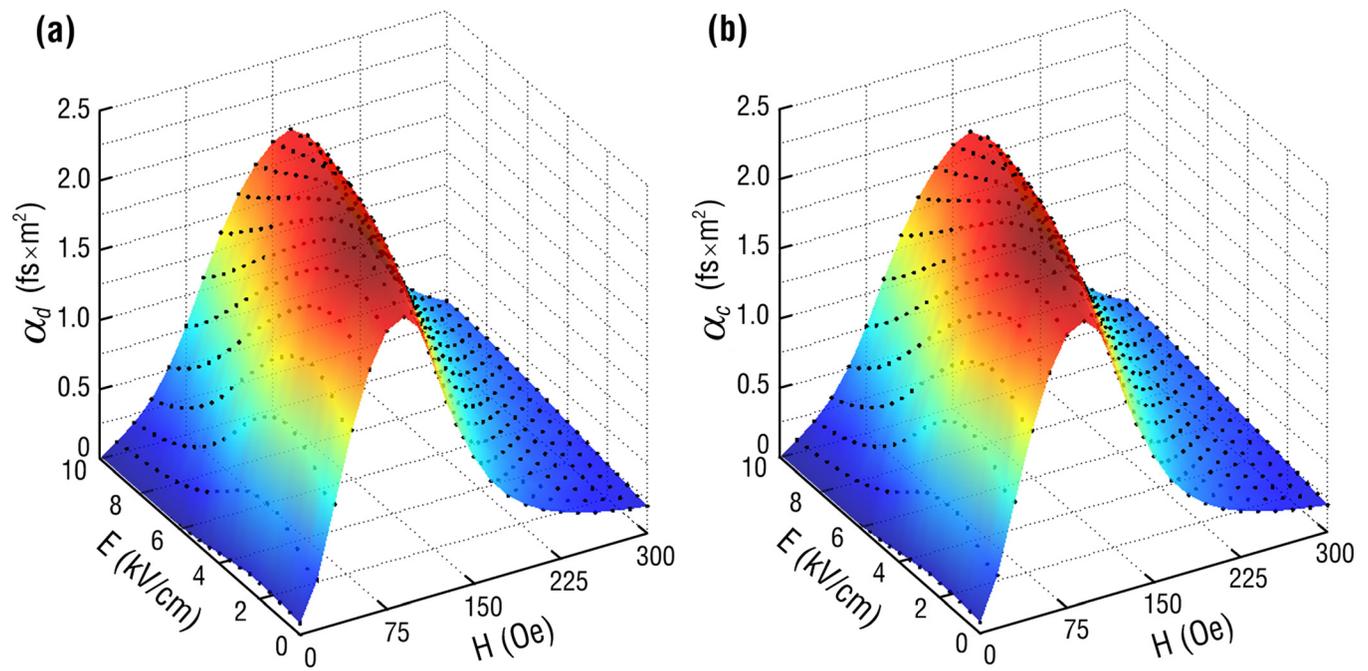


FIG. 2. (Color online) (a) Direct and (b) converse ME coefficients for Galfenol/PZN-PT/Garfenol at selected bias fields E and H . Each black dot represents a data point. The variation with H is consistent with the saturation of Galfenol magnetostriction (see Ref. 6).

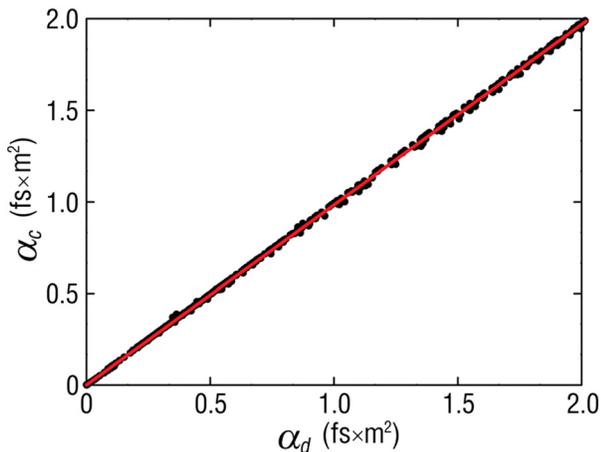


FIG. 3. (Color online) We plot the value of α_c against α_d for each bias condition (H , E). The linear fit gives $\alpha_c = 0.9841\alpha_d$. The coefficient of determination $R^2 = 0.99994$.

20 V/cm. Both a.c. excitations were performed at 200 Hz, which is slow enough to avoid any significant electromechanical resonance losses and fast enough to approximate adiabatic conditions.

As expected from the insight presented earlier, we find that α_d (Fig. 2(a)) and α_c (Fig. 2(b)) vary with bias fields E and H in a very similar manner (Fig. 3) and typically differ by just $\sim 1.6\%$. This small discrepancy rises to $\sim 5\%$ at low

H , likely due to hysteresis or non-linear behavior when the bias and excitation fields are of comparable strength. We therefore confirm $\alpha_d = \alpha_c$ here, and also for $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3/\text{metglas}$ in a separate study to be published elsewhere.

For strain-coupled two-phase ME systems, we have predicted and verified that the direct and converse coupling coefficients possess equal strength when parameterized using variables that describe the entire system. Our findings, therefore, also apply to ME systems comprising more than two phases. This work corrects the two widely held opposing views that are compromised by incorrect parameterization or inconsistent bias conditions and has practical relevance as it permits both converse and direct ME coefficients to be determined even when only one can be measured conveniently.

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