Analog of Magnetoelectric Effect in High-$T_c$ Granular Superconductors

Sergei A. Sergeenkov and Jorge V. José

1 Instituto de Física, Universidade Federal do Rio Grande do Sul, 91501-970 Porto Alegre, Brazil
2 Physics Department and Center for the Interdisciplinary Research on Complex Systems, Northeastern University, Boston, MA, 02115, USA
3 Instituto de Física, Universidad Nacional Autónoma de México, Apdo. Postal 20-364, 01000 México D. F., México

We propose the existence of an electric-field induced nonlinear magnetization in a weakly coupled granular superconductor due to time-parity violation. As the field increases the induced magnetization passes from para- to dia-magnetic behavior. We discuss conditions under which this effect could be experimentally measured in high temperature superconductors.

By applying a constant external electric field through an insulating layer, recent experimental studies have found an enhancement in the critical current of ceramic high-$T_c$ superconductors (HTCS). It has been shown that for any field-induced effects to be experimentally measurable, the electrostatic screening length should be larger than the superconducting coherence length. This happens to be the case in HTCS, which have a reduced carrier number density. To account for the unusual behavior of the field dependent critical current observed in HTCS ceramics, however, it was argued that some other mechanisms (in addition to the conventional carrier number density modification) have to be taken into consideration. In particular, it was suggested that an applied electric field may actually induce a substantial change into the original weak-links structure of granular samples.

In this letter we predict yet another interesting effect which can be produced by an external electric field applied to a granular material. This effect is similar to (but physically different from) the magnetoelectric effect (MEE) seen in magnetic ferroelectrics (like antiferromagnetic Bi$_2$FeO$_4$). It has also been discussed in the context of some exotic nonmagnetic normal metal conductors, with the symmetry of a mirror isomer, and pyroelectric superconductors (where the supercurrent passing through a metal of a polar symmetry is assumed to be accompanied by spin polarization of the carriers). The effect discussed here entails the electric field generation of a magnetic moment (paramagnetic in small to diamagnetic for large electric fields) due to superconducting currents that circulate between grains. The basic physical reason for the appearance of this effect is that the applied electric field induces a magnetization $M$, that changes its sign ($M \rightarrow -M$) under a time-parity transformation. There have been other microscopic mechanisms proposed to have time-parity violation effects in HTCS. One that attracted significant experimental attention was a theoretical consequence of the anyon picture of HTCS. Another proposition was based on a spin-orbit coupling. Up to now, however, the experimental search for their signatures have not bared fruit.

In the case of weakly-coupled superconducting grains, the phenomenological reason for the time-parity violation comes from the fact that the total free energy $F$ for the material exhibiting both magnetic and electric properties contains the term $\alpha_{mn}(E,H)$, with the coefficients $\alpha_{mn} \neq 0$ (here $\{m,n\} = \{x,y,z\}$). In the standard MEE, the function $f_{mn} \equiv E_m H_n$, which leads to the corresponding linear effect, which is either an electric-field induced magnetization $M_m(E) \equiv \partial F/\partial H_m = \alpha_{mn} E_n$ (in zero magnetic field) or a magnetic-field induced polarization $P_m(H) \equiv \partial F/\partial E_m = \alpha_{mn} H_n$ (in zero electric field). Since $H$ (and $M$) changes its sign under time-parity transformation while $E$ (and $P$) remains unchanged, an electric field induced magnetization will break time-parity symmetry even in zero magnetic field applied. As we show below, in our case the symmetry breaking term in $F$, represented by a nonzero coefficient $\alpha_{mn}$, has a more general nonlinear form for $f_{mn}$. We recall that the standard linear MEE can appear when an external electric field $E$ interacts with an inner magnetic field $H_{DM}$ of the Dzyaloshinskii-Moriya (DM) type. The DM interaction leads to a term, apart from the standard isotropic term in the Heisenberg Hamiltonian, of the form $H_{DM} = \sum_{i,j} \vec{D}_{i,j} \cdot (\vec{S}_i \wedge \vec{S}_j)$, where $\vec{S}_i$ is a Heisenberg spin and the constant vector $\vec{D}_{i,j}$ arises from the spin-orbit coupling. An analogous situation occurs in our case, as we describe below. To see how we can get a nonzero $\alpha$, or equivalently a DM type interaction in a granular superconductor, we model a HTCS ceramic sample by a random three-dimensional (3D) overdamped Josephson junction array. This model has proven to be useful in describing the metastable magnetic properties of HTCS. In thermodynamic equilibrium, this model has a Boltzmann factor with a random 3D-XY model Hamiltonian. Specifically, the general form of the Hamiltonian (describing both DC and AC effects) reads

$$H_{XY}(t) = - \sum_{i,j} E_J(r_{i,j}) \cos \phi_{i,j}(t).$$

Here $\{i\} = r_i$ is a 3D lattice vector; $E_J(r_{i,j})$ is the Josephson coupling energy, with $r_{i,j} = r_i - r_j$ the separation between the grains; the gauge invariant phase difference is defined as

$$\phi_{i,j}(t) = \phi_{i,j}(0) - A_{i,j}(t),$$

1
where \( \phi_{i,j}(0) = \phi_i - \phi_j \) with \( \phi_i \) being the phase of the superconducting order parameter; \( A_{i,j}(t) \) is (time-dependent, in general) frustration parameter, defined as

\[
A_{i,j}(t) = \frac{2\pi}{\Phi_0} \int_t^J \vec{A}(\vec{r}', t) \cdot d\vec{r}',
\]

with \( \vec{A}(\vec{r}', t) \) the (space-time dependent) electromagnetic vector potential which involves both external fields and the electric and magnetic possible self-field effects (see below); \( \Phi_0 = \hbar/2e \) is the quantum of flux, with \( \hbar \) Planck’s constant, and \( e \) the electronic charge. Expanding the cosine term, and using trigonometric identities we can explicitly rewrite the above Hamiltonian as

\[
H_{\text{XY}} = -\sum_{i,j} E_J [\cos(A_{i,j}) \vec{S}_i \cdot \vec{S}_j - \sin(A_{i,j}) \vec{k} \cdot \vec{S}_i \wedge \vec{S}_j],
\]

where the two-component \( \text{XY} \) spin vector is defined as \( \vec{S}_i \equiv (\cos \phi_i, \sin \phi_i) \), and \( \vec{k} \) is a unit vector along the \( z \)-axis [14]. We see that the second term in this Hamiltonian has the same form as in the DM contribution, and thus we can surmise that the time parity will be broken by applying an external field (electric or magnetic) to the granular system. To bring to the fore this possibility, we show below in a simple but yet nontrivial model that this is indeed the case.

One important property of the \( H_{\text{XY}} \) Hamiltonian is that it is random because \( A_{i,j} \) itself is a random variable. There are different types of \( A_{i,j} \) randomness that can be considered [13]. For simplicity, in the present paper, we consider a long-range (Sherrington-Kirkpatrick-like) interaction between grains (assuming \( E_J(r_{i,j}) = E_J \)) and model the true short-range behavior of a ceramics sample through the randomness in the position of the superconducting grains in the array (using the exponential distribution law \( P_r(r_{i,j}) \), see below). Here we restrict our consideration to the case of an external electric field only but it can be shown that the scenario suggested in this paper will also carry through when applying an external magnetic field (which will induce another time-parity breaking phenomenon in the granular material; namely, magnetic field induced electric polarizability [15]). Besides, in what follows we also ignore the role of Coulomb interaction effects assuming that the grain’s charging energy \( E_c \ll E_J \) (where \( E_c = e^2/2C \), with \( C \) the capacitance of the junction).

In the case of a granular material, we show here that the corresponding time-parity breaking DM internal field can be related to the electric field induced magnetic moment produced by the circulating Josephson currents between the grains. As is known [16][17], a constant electric field \( \vec{E} \) applied to a single Josephson junction (JJ) causes a time evolution of the phase difference. In this particular case Eq.(2) reads

\[
\phi_{i,j}(t) = \phi_{i,j}(0) + \frac{2e}{\hbar} \vec{E} \cdot \vec{r}_{i,j} t.
\]

The resulting AC superconducting current in the junction is

\[
I_{i,j}(t) = \frac{2e}{\hbar} \vec{E} \cdot (\vec{r}_{i,j}/2).
\]

If, in addition to the external electric field \( \vec{E} \), the network of superconducting grains is under the influence of an applied magnetic field \( \vec{H} \), the frustration parameter \( A_{i,j}(t) \) in Eq.(3) takes the following form

\[
A_{i,j}(t) = \frac{\pi}{\Phi_0} (\vec{H} \wedge \vec{r}_{i,j}) \cdot \vec{r}_{i,j} - \frac{2\pi}{\Phi_0} \vec{E} \cdot \vec{r}_{i,j} t.
\]

Here, \( \vec{r}_{i,j} = (\vec{r}_i + \vec{r}_j)/2 \), and we have used the conventional relationship between the vector potential \( \vec{A} \) and a constant magnetic field \( \vec{H} = \text{rot} \vec{A} \) (with \( \text{rot} \vec{H} = 0 \)), as well as a homogeneous electric field \( \vec{E} = -\partial \vec{A}/\partial t \) (with \( \text{rot} \vec{E} = 0 \)). In the type II HTCS the magnetic self-field effects for the array as a whole are expected to be negligible [13]. The grains themselves are in fact larger than the London penetration depth and we must then have that the corresponding Josephson penetration length must be much larger than the grain size (since the self-induced magnetic fields can in principle be quite pronounced for large-size junctions even in zero applied magnetic fields [17]). Specifically, this is justified for short junctions with the size \( d \ll \lambda_j \), where \( \lambda_j = \sqrt{\Phi_0/4\pi \mu_0 J_c} \lambda_L \) is the Josephson penetration length with \( \lambda_L \) being the grain London penetration depth and \( J_c \) its Josephson critical current density. In particular, since in HTCS \( \lambda_L \approx 150 \text{nm} \), the above condition will be fulfilled for \( d \simeq 1 \mu \text{m} \) and \( j_c \simeq 10^4 \text{A/m}^2 \) which are the typical parameters for HTCS ceramics [16]. Likewise, to ensure the uniformity of the applied electric field, we also assume that \( d \ll \lambda_E \), where \( \lambda_E \) is an effective electric field penetration depth [14].

When the AC supercurrent \( I_{i,j}(t) \) (defined by Eqs.(2), (5) and (6)) circulates around a set of grains, that form a random area plaquette, it induces a random AC magnetic moment \( \vec{m}(t) \) of the Josephson network [2]

\[
\vec{m}(t) \equiv \left[ \frac{\partial H_{\text{XY}}}{\partial \vec{H}} \right]_{\vec{H}=0} = \pi \sum_{i,j} I_{i,j}(t)(\vec{r}_{i,j} \wedge \vec{R}_{i,j}).
\]

Notice that in the MEE-like effect discussed here for a granular superconductor, the electric-field induced magnetic moment in the system is still present in zero applied magnetic field due to the phase coherent currents between the weakly-coupled superconducting grains.

To consider the essence of the superconducting electric field-induced MEE, we assume that in a zero electric field the phase difference between the adjacent grains \( \phi_{i,j}(0) = 0 \) which corresponds to a fully coherent state of the array. In this particular case, the electric-field induced averaged magnetization reads

\[
\vec{M}_s(\vec{E}) \equiv \overline{\vec{m}(t)} = \frac{1}{\tau} \int_0^\tau dt \int_0^\infty d\vec{r}_{i,j} d\vec{R}_{i,j} \mathcal{S}(\vec{r}_{i,j}, \vec{R}_{i,j}) \vec{m}(t),
\]

where

\[
\mathcal{S}(\vec{r}_{i,j}, \vec{R}_{i,j}) = \frac{2\pi}{\Phi_0} \left[ \frac{\partial H_{\text{XY}}}{\partial \vec{H}} \right]_{\vec{H}=0} (\vec{r}_{i,j}, \vec{R}_{i,j}) \equiv \frac{2\pi}{\Phi_0} (\vec{r}_{i,j} \wedge \vec{R}_{i,j})(\vec{r}_{i,j} \wedge \vec{R}_{i,j})
\]

with the DM contribution, and

\[
\vec{M}_{\text{DM}}(\vec{E}) = \frac{2\pi}{\Phi_0} (\vec{r}_{i,j} \wedge \vec{R}_{i,j})(\vec{r}_{i,j} \wedge \vec{R}_{i,j})
\]
where $\tau$ is the electronic relaxation scattering time, and $S$ is the joint probability distribution function (see below).

To obtain an explicit expression for the electric-field dependent magnetization, we consider a site positional disorder that allows for small random radial displacements. Namely, the sites in a 3D cubic lattice are assumed to move from their equilibrium positions according to the normalized (separable) distribution function

$$S(\vec{r}_{i,j}, \vec{R}_{i,j}) \equiv P_r(\vec{r}_{i,j})P_R(\vec{R}_{i,j})$$  \hspace{1cm} (9)

It can be shown that the main qualitative results of this paper do not depend on the particular choice of the probability distribution function. For simplicity here we assume an exponential distribution law for the distance between grains, $P_r(\vec{r}) = P(x_1)P(x_2)P(x_3)$ with $P_r(x_j) = (1/d)e^{-x_j/d}$, and a short range distribution for the dependence of the center-of-mass probability $P_R(\vec{R})$ (around some constant value $D$). The specific form of the latter distribution will not affect the qualitative nature of the final result. (Notice that in fact the former distribution function $P_r(\vec{r})$ reflects a short-range character of the Josephson coupling in granular superconductor. Indeed, according to the conventional picture the Josephson coupling $E_{ij}(\vec{r}_{ij})$ can be assumed to vary exponentially with the distance $\vec{r}_{ij}$ between neighboring grains, i.e., $E_{ij}(\vec{r}_{ij}) = E_0 e^{-\vec{k} \cdot \vec{r}_{ij}}$. For isotropic arrangement of identical grains, with spacing $d$ between the centers of adjacent grains, we have $\vec{k} = (\frac{\pi}{d}, \frac{\pi}{d}, \frac{\pi}{d})$ and thus $d$ is of the order of an average grain size.) Taking the applied electric field along the $x$-axis, $\vec{E} = (E_x, 0, 0)$, we get finally

$$M_z(E_x) = \frac{B_z(E_x)}{\mu_0} - H_z(E_x),$$  \hspace{1cm} (10)

for the induced transverse magnetization (along the $x_3 = z$-axis), where

$$B_z(E_x) = \mu_0 M_0 \frac{E_x/E_0}{1 + (E_x/E_0)^2},$$  \hspace{1cm} (11)

and

$$H_z(E_x) = M_0 \left( \frac{E_0}{E_x} \right) \log \sqrt{1 + \left( \frac{E_x}{E_0} \right)^2},$$  \hspace{1cm} (12)

stand for the electric-field induced magnetic induction $B_z(E_x)$ and magnetic field $H_z(E_x)$, respectively. The induced Josephson current $I(E_x)$ is simply given by Ampere’s law $I(E_x) = H_z(E_x)d$. In these equations, $M_0 = 2\pi e J_z N d D / h$, with $N$ the total number of grains and $E_0 = h / 2 d e \tau$. Eq.(10) is the main result of this paper, which we proceed to analyze below.

As is seen from Eq.(10), the behavior of the magnetization in the applied electric field is determined by the competition between the two contributions, the magnetic induction $B_z(E_x)$ and the current induced magnetic field $H_z(E_x)$ (or the corresponding Josephson current $I(E_x)$). Namely, below a critical (threshold) field $E_c \approx 1.94 E_0$ where $B_z(E_x) > \mu_0 H_z(E_x)$, a paramagnetic phase of the MEE is due to the modification of the magnetic induction in the applied electric field. On the other hand, above the threshold field (when $E_c$ becomes larger than $E_c$) the Josephson current $I(E_x)$ induced contribution starts to prevail, leading to the appearance of the diamagnetic signal (seen as a small negative part of the induced magnetization in Fig.1). Such an electric field induced paramagnetic-to-diamagnetic transition has been actually observed for behavior of the critical current in ceramic HTCS and it was attributed to a proximity-mediated enhancement of the superconductivity in a granular material in a strong enough electric field. Explicitly, the critical current in a YBCO sample was found to reach a maximum at $E = 4 \times 10^7 V/m$. To relate this experimental value with the model parameters, first of all, we need to estimate an order of magnitude of the relaxation time $\tau$ in a zero applied electric field. This will provide an upper limit for the relevant $\tau$-distribution in our system. It is reasonable to connect zero-field $\tau = \tau(0)$ with the Josephson tunneling time $\tau_j = (R_0 / R_n)(h / E_j)$ (where $R_0 = h / 4 e^2$, and $R_n$ is the normal state resistance between grains). Typically, for HTCS ceramics $E_j / k_B \approx 90 K$ and $R_n / R_0 \approx 10^{-3}$, so that $\tau_j \approx 10^{-10} s$. At the same time, at high enough elec-
electric fields where the MEE becomes strongly nonlinear, we can expect quite a tangible decrease of the relaxation time. Indeed, for an average grain size $d \approx 1 \mu m$, the characteristic field $E_0 = 4 \times 10^5 V/m$ (which corresponds to the region where a prominent enhancement of the critical current was observed \cite{1–3}) introduces a substantially shorter relaxation time $\tau(E_0) = \hbar/2deE_0 \approx 10^{-16} s$, in agreement with observations \cite{4}.

To estimate the relative magnitude of the superconducting analog of the MEE predicted here, we can compare it with the normal (Ohmic) contribution to the magnetization

$$\vec{M}(E) \equiv \vec{\mu}(E)/\int_0^\infty \mu dt = \frac{1}{E} \int_0^\infty \frac{d\vec{r}_i,j d\vec{R}_i,j S(\vec{r}_i,j, \vec{R}_i,j) \vec{\mu}_n}{0},$$

(13)

where

$$\vec{\mu}_n = \pi \sum_{i,j} I_{i,j}^n (\vec{r}_i,j \wedge \vec{R}_i,j).$$

(14)

Here $I_{i,j}^n = V_{i,j}/R_n$ is the normal current component due to the applied electric field $\vec{E}$, with $V_{i,j} = \vec{E} \cdot \vec{r}_i,j$ being the induced voltage, and $R_n$ the normal state resistance between grains. As a result, the normal state contribution (for $\vec{E}$ along the x-axis) reads $M_n = \alpha_n E_x$, with $\alpha_n = \pi e^2 dN/R_n$. Similarly, according to Eq.(10), the low field contribution to the superconducting MEE gives $M_s \simeq \alpha_s E_x$ with $\alpha_s = 2\pi e^2 E_s N_\tau(0)d^2D/h^2$. Thus, at low enough applied fields (when $E_x \ll E_0$)

$$\frac{\alpha_s}{\alpha_n} \simeq \frac{\tau(0)}{\tau_J},$$

(15)

where $\tau_J = (R_0/R_n)(h/E_J)$ with $R_0 = h/4e^2$. According to our previous discussion on the relevant relaxation-time distribution spectrum in our model system, we may conclude that $\tau(0) \leq \tau_J$. So, we arrive at the following ratio between the coefficients of the superconducting to normal MEEs, namely $\alpha_s/\alpha_n \leq 1$. The above estimate of the weak-links induced MEE (along with its rather specific field dependence, see Fig.1) suggests quite an optimistic possibility to observe the predicted effect experimentally in HTCS ceramics or in a specially prepared system of arrays of superconducting grains.

We note that in the present analysis we have not explicitly considered the polarization effects (due to the interaction between the applied electric field and the grain’s charges) which may become important at high enough fields (or for small enough grains), leading to more subtle phenomena (like Coulomb blockade and reentrant-like behavior) that will demand the inclusion of charging energy effects in the analysis.

In summary, we have used a zero-temperature random 3D XY model to predict the appearance of a novel electric-field induced magnetization in a granular superconductor (an analog of the magnetoelectric effect). The induced magnetization has a very distinctive nonlinear and para- to dia-magnetic behavior as the field is increased. We have also estimated the possible parameter range conditions to observe this effect experimentally.

We thank N. Israeloff, A. Goldman and P. Martinoli for informative communications on the subject. SAS acknowledges the financial support from the Brazilian funding agency CNPq. The work of JVJ was partially funded by NSF Grant DMR-9521845.

* Permanent address: Bogoliubov Laboratory of Theoretical Physics, Joint Institute for Nuclear Research, Dubna, Russia

[1] Smirnov B.I., Krishtopov S.V. and Orlova T.S., Phys. Solid State, 34 (1992) 1331.

[2] Smirnov B.I., Orlova T.S. and Krishtopov S.V., Phys. Solid State, 35 (1993) 1118.

[3] Orlova T.S. and Smirnov B.I., Supercond. Sci. Techn., 7 (1994) 899.

[4] For a comprehensive review see Mannhart, J., Supercond. Sci. Techn., 9 (1996) 49.

[5] Mannhart J. et al., Phys. Rev. Lett., 67 (1991) 2099; Xi X.X. et al., ibid, 68 (1992) 1240.

[6] Rakmanov A.L. and Rozhkov A.V., Physica C, 267 (1996) 233.

[7] Freeman A.J. and Schmid H., Eds. Magnetoelectric Interaction Phenomena in Crystals (Gordon and Breach, New York) 1975. See also, Landau L.D. and Lifshitz E.M., Electrodynamics of Continuous Media (Pergamon Press, Oxford) 1960.

[8] Levitov L.S., Nazarov Yu.V. and Eliahsberg G.M., Sov. Phys. JETP, 61 (1985) 133.

[9] Edelstein V.M., Phys. Rev. Lett., 75 (1995) 2004.

[10] Halperin B.I. et al., Phys. Rev. B, 340 (1989) 8726.

[11] Dzyaloshinskii I.E., Sov. Phys. JETP, 37 (1959) 881; Moriya T., Phys. Rev., 120 (1960) 91.

[12] Ebner C. and Stroud D., Phys. Rev. B, 31 (1985) 165.

[13] Choi J. and José J.V., Phys. Rev. Lett., 62 (1989) 320. See also, Blatter G., Feigel’man M.V., Geshkenbein V.B., Larkin A.I. and Vinokur V.M., Rev. Mod. Phys., 66 (1994) 1125.

[14] Girgis M.J.P., Phys. Rev. B, 45 (1992) 7547; Kopeć T.K. and José J.V., ibid, 52 (1995) 16140.

[15] Sergeenkov S., J. Phys. I France, 7 (1997) 1175.

[16] Tinkham M., Introduction to Superconductivity Second edition (McGraw-Hill, New York) 1996.

[17] Lebeau C., Rosenblatt J., Robotou A. and Peyral P., Europhys. Lett., 1 (1986) 313.

[18] Domínguez D. and José J.V., Phys. Rev. B, 53 (1996) 11692.

[19] Mühlschlegel B. and Mills D.L., Phys. Rev. B, 29 (1985) 159.