

Piezomagnetism and Stress Induced Paramagnetic Meissner Effect in Mechanically Loaded High- T_c Granular Superconductors

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Two novel phenomena in a weakly coupled granular superconductor under an applied stress are predicted which are based on recently suggested piezophase effect (a macroscopic quantum analog of the piezoelectric effect) in mechanically loaded grain boundary Josephson junctions. Namely, we consider the existence of stress induced paramagnetic moment in zero applied magnetic field (piezomagnetism) and its influence on a low-field magnetization (leading to a mechanically induced paramagnetic Meissner effect). The conditions under which these two effects can be experimentally measured in high- T_c granular superconductors are discussed.

Despite the fact that granular superconductors have been actively studied (both experimentally and theoretically) for decades, they continue contributing to the variety of intriguing and peculiar phenomena (both fundamental and important for potential applications) providing at the same time a useful tool for testing new theoretical concepts [1]. To give just a few recent examples, it is sufficient to mention paramagnetic Meissner effect [2–5] (PME) originated from a cooperative behavior of weak-links mediated orbital moments and found to be responsible for unusual aging effects [6] in high- T_c granular superconductors (HTGS). Among others are also recently introduced thermophase [7,8] and piezophase [9] effects suggesting, respectively, a direct influence of a thermal gradient and an applied stress on phase difference between the adjacent grains. Besides, using a model of random overdamped Josephson junction arrays, two dual time-parity violating effects in HTGS have been predicted [10,11]. Namely, an appearance of magnetic field induced electric polarization along with the concomitant change of the junction capacitance (magneto-electric effect [10]) and existence of electric field induced magnetization (converse magnetoelectric effect [11]) via a Dzyaloshinski-Moria type interaction mechanism.

In this Letter we discuss a possibility of two other interesting effects expected to occur in a granular material under sufficient mechanical loading. Specifically, we predict the existence of stress induced paramagnetic moment in zero applied magnetic field (piezomagnetism) and its influence on a low-field magnetization (leading to a mechanically induced PME).

The possibility to observe tangible piezoeffects in mechanically loaded grain boundary Josephson junctions (GBJJs) is based on the following arguments. It is well known [12–14] that the grain boundaries (GBs) are the natural sources of weak links (or GBJJs) in granular superconductors. Under plastic deformation, GBs were found [15] to move rather rapidly via the movement of the grain boundary dislocations (GBDs) comprising these GBs. As a matter of fact, using the so-called method of acoustic emission, the plastic flow of GBDs with the maximum rate of $v_0 = 1mm/s$ has been registered [16]

in *YBCO* ceramics at $T = 77K$ under the external load of $\sigma = 10^7 N/m^2$. Using the above evidence, in Ref.9 a *piezophase* response of a single GBJJ (created by GBDs strain field ϵ_d acting as an insulating barrier of thickness l and height U in a *SIS*-type junction with the Josephson energy $J \propto e^{-l\sqrt{U}}$) to an externally applied mechanical stress was considered. The resulting stress-strain and stress-current diagrams were found [9] to exhibit a quasi-periodic (Fraunhofer-like) behavior typical for Josephson junctions (JJs). To understand how piezoeffects can manifest themselves through GBJJs, let us invoke an analogy with the so-called *thermophase effect* suggested originally by Guttman *et al.* [7] (as a quantum mechanical alternative for the conventional thermoelectric effect) to occur in a single JJ and later applied to HTGS [8]. In essence, the thermophase effect assumes a direct coupling between an applied temperature drop ΔT and the resulting phase difference $\Delta\phi$ through a JJ. When a rather small temperature gradient is applied to a JJ, an entropy-carrying normal current $I_n = L_n\Delta T$ (where L_n is the thermoelectric coefficient) is generated through such a junction. To satisfy the constraint dictated by the Meissner effect, the resulting supercurrent $I_s = I_c \sin[\Delta\phi]$ (with $I_c = 2eJ/h$ being the Josephson critical current) develops a phase difference through a weak link. In other words, the temperature gradient stimulates a superconducting phase gradient which in turn drives the reverse supercurrent. The normal current is locally canceled by a counterflow of supercurrent, so that the total current through the junction $I = I_n + I_s = 0$. As a result, supercurrent $I_c \sin[\Delta\phi] = -I_n = -L_n\Delta T$ generates a nonzero phase difference via a transient Seebeck thermoelectric field leading to the linear thermophase effect [7] $\Delta\phi = -\arcsin(L_{tp}\Delta T) \simeq -L_{tp}\Delta T$ with $L_{tp} = L_n/I_c(T)$.

By analogy, we can introduce a *piezophase effect* (as a quantum alternative for the conventional piezoelectric effect) through a JJ [9]. Indeed, a linear conventional piezoelectric effect relates induced polarization P_n to an applied strain ϵ as [17] $P_n = d_n\epsilon$, where d_n is the piezoelectric coefficient. The corresponding normal piezocurrent density is $j_n = dP_n/dt = d_n\dot{\epsilon}$ where $\dot{\epsilon}(\sigma)$ is a rate of plastic deformation (under an applied stress σ) which

depends on the number of GBDs of density ρ and a mean dislocation rate v_d as follows [18] $\dot{\epsilon}(\sigma) = b\rho v_d(\sigma)$ (where b is the absolute value of the appropriate Burgers vector). In turn, $v_d \simeq v_0(\sigma/\sigma_m)$ with σ_m being the so-called ultimate stress. To meet the requirements imposed by the Meissner effect, in response to the induced normal piezocurrent, the corresponding Josephson supercurrent of density $j_s = dP_s/dt = j_c \sin[\Delta\phi]$ should emerge within the contact. Here $P_s = -2enb$ is the Cooper pair's induced polarization with $n = N/V$ the pair number density, and $j_c = 2ebJ/\hbar V$ is the critical current density. The neutrality conditions ($j_n + j_s = 0$ and $P_n + P_s = \text{const}$) will lead then to the linear piezophase effect $\Delta\phi = -\arcsin[d_{pp}\dot{\epsilon}(\sigma)] \simeq -d_{pp}\dot{\epsilon}(\sigma)$ (with $d_{pp} = d_n/j_c$ being the piezophase coefficient) and the concomitant change of the pair number density under an applied strain, viz., $\Delta n(\epsilon) = d_{pn}\epsilon$ with $d_{pn} = d_n/2eb$. Given the markedly different scales of stress induced changes in defect-free thin films [19] and weak-links-ridden ceramics [20], it should be possible to experimentally register the suggested here piezophase effects.

To adequately describe magnetic properties of a granular superconductor, we employ a model of *random* three-dimensional (3D) overdamped Josephson junction array which is based on the well known tunneling Hamiltonian [21–24]

$$\mathcal{H} = \sum_{ij}^N J(r_{ij})[1 - \cos\phi_{ij}], \quad (1)$$

where $\{i\} = \vec{r}_i$ is a 3D lattice vector, N is the number of grains (or weak links), $J(r_{ij})$ is the Josephson coupling energy with $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ the separation between the grains; the gauge invariant phase difference is defined as $\phi_{ij} = \phi_{ij}^0 - A_{ij}$, where $\phi_{ij}^0 = \phi_i - \phi_j$ with ϕ_i being the phase of the superconducting order parameter, and $A_{ij} = \frac{2\pi}{\Phi_0} \int_i^j \vec{A}(\vec{r}) \cdot d\vec{l}$ is the frustration parameter with $\vec{A}(\vec{r})$ the electromagnetic vector potential which involves both external fields and possible self-field effects (see below); $\Phi_0 = h/2e$ is the quantum of flux.

In the present paper, we consider a long-range interaction between grains [8,10,11,24] (with $J(r_{ij}) = J$) and model the true short-range behavior of a HTGS sample through the randomness in the position of the superconducting grains in the array (see below). For simplicity, we shall ignore the role of Coulomb interaction effects assuming that the grain's charging energy $E_c \ll J$ (where $E_c = e^2/2C$, with C the capacitance of the junction). As we shall see, this condition is reasonably satisfied for the effects under discussion.

According to the above-discussed scenario, under an applied stress the superconducting phase difference will acquire an additional contribution $\delta\phi_{ij}(\sigma) = -B\vec{\sigma} \cdot \vec{r}_{ij}$, where $B = d_n\dot{\epsilon}_0/\sigma_m j_c b$ with $\dot{\epsilon}_0 = b\rho v_0$ being the maximum deformation rate and the other parameters defined earlier. If, in addition to the external loading,

the network of superconducting grains is under the influence of an applied frustrating magnetic field \vec{H} , the total phase difference through the contact reads (where $\vec{R}_{ij} = (\vec{r}_i + \vec{r}_j)/2$)

$$\phi_{ij}(\vec{H}, \vec{\sigma}) = \phi_{ij}^0 + \frac{\pi}{\Phi_0} (\vec{r}_{ij} \wedge \vec{R}_{ij}) \cdot \vec{H} - B\vec{\sigma} \cdot \vec{r}_{ij}. \quad (2)$$

It is well known [1,10,11,24] that the self-induced Josephson fields can in principle be quite pronounced for large-size junctions even in zero applied magnetic fields. So, to safely neglect the influence of these effects in a real material, the corresponding Josephson penetration length λ_J must be much larger than the junction (or grain) size. Specifically, this condition will be satisfied for short junctions with the size $d \ll \lambda_J$, where $\lambda_J = \sqrt{\Phi_0/4\pi\mu_0 j_c \lambda_L}$ with λ_L being the grain London penetration depth and j_c its Josephson critical current density. In particular, since in HTGS $\lambda_L \simeq 150nm$, the above criterium will be rather well met for $d \simeq 1\mu m$ and $j_c \simeq 10^4 A/m^2$ which are the typical parameters for HTGS ceramics [1]. Likewise, to ensure the uniformity of the applied stress σ , we also assume that $d \ll \lambda_\sigma$, where λ_σ is a characteristic length over which σ is kept homogeneous.

When the Josephson supercurrent $I_{ij}^s = I_c \sin\phi_{ij}$ circulates around a set of grains (that form a random area plaquette), it induces a random magnetic moment $\vec{\mu}_s$ of the Josephson network [21]

$$\vec{\mu}_s \equiv -\frac{\partial\mathcal{H}}{\partial\vec{H}} = \sum_{ij} I_{ij}^s (\vec{r}_{ij} \wedge \vec{R}_{ij}), \quad (3)$$

which results in the stress induced net magnetization

$$\vec{M}_s(\vec{H}, \vec{\sigma}) \equiv \frac{1}{V} \langle \vec{\mu}_s \rangle = \int_0^\infty d\vec{r}_{ij} d\vec{R}_{ij} f(\vec{r}_{ij}, \vec{R}_{ij}) \vec{\mu}_s, \quad (4)$$

where V is a sample's volume and f the joint probability distribution function (see below). To capture the very essence of the superconducting piezomagnetic effect, in what follows we assume for simplicity that an *unloaded sample* does not possess any spontaneous magnetization at zero magnetic field (that is $M_s(0, 0) = 0$) and that its Meissner response to a small applied field H is purely diamagnetic (that is $M_s(H, 0) \simeq -H$). According to Eq.(2), this condition implies $\phi_{ij}^0 = 2\pi m$ for the initial phase difference with $m = 0, \pm 1, \pm 2, \dots$. Incidentally, this is also a requirement for current conservation at zero temperature [21].

In order to obtain an explicit expression for the piezomagnetization, 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 $f(\vec{r}_{ij}, \vec{R}_{ij}) \equiv$

$f_r(\vec{r}_{ij})f_R(\vec{R}_{ij})$. As usual [8,10,11], 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, $f_r(\vec{r}) = f(x)f(y)f(z)$ with $f(x_j) = (1/d)e^{-x_j/d}$, and some short range distribution for the dependence of the center-of-mass probability $f_R(\vec{R})$ (around some constant value D). While the specific form of the latter distribution is not important for the effects under discussion, it is worthwhile to mention that the former distribution function $f_r(\vec{r})$ reflects a short-range character of the Josephson coupling in granular superconductors where [25] $J(\vec{r}_{ij}) = Je^{-\vec{\kappa}\cdot\vec{r}_{ij}}$. For isotropic arrangement of identical grains, with spacing d between the centers of adjacent grains, we have $\vec{\kappa} = (\frac{1}{d}, \frac{1}{d}, \frac{1}{d})$ and thus d is of the order of an average grain size.

Taking the applied stress along the x -axis, $\vec{\sigma} = (\sigma, 0, 0)$, normally to the applied magnetic field $\vec{H} = (0, 0, H)$, we get finally

$$M_s(H, \sigma) = -M_0 \frac{H_{tot}(H, \sigma)/H_0}{[1 + H_{tot}^2(H, \sigma)/H_0^2]^2}, \quad (5)$$

for the induced transverse magnetization (along the z -axis), where $H_{tot}(H, \sigma) = H - H^*(\sigma)$ is the total magnetic field with $H^*(\sigma) = (\sigma/\sigma_0)H_0$ being a stress-induced contribution. Here, $M_0 = I_cSN/V$ with $S = \pi dD$ being a projected area around the Josephson contact, $H_0 = \Phi_0/S$, and $\sigma_0 = \sigma_m(j_c/j_d)(b/d)$ with $j_d = d_n\dot{\epsilon}_0$ and $\dot{\epsilon}_0 = b\rho v_0$ being the maximum values of the dislocation current density and the plastic deformation rate, respectively.

Fig.1 presents the stress induced magnetization at different applied magnetic fields, calculated according to Eq.(5). As is seen, in practically zero magnetic field the piezomagnetization is purely paramagnetic (solid line), exhibiting a strong nonlinear behavior. With increasing the stress, it first increases reaching a maximum, and then rather rapidly dies away. Under the influence of small applied magnetic fields (dotted and dashed lines), the piezomagnetism turns diamagnetic (for low external stress) with its peak shifting toward higher loading. At the same time, Fig.2 shows changes of the initial stress-free diamagnetic magnetization (solid line) under an applied stress. As we see, already relatively small values of an applied stress render a low field Meissner phase strongly paramagnetic (dotted and dashed lines) simultaneously shifting the peak toward higher magnetic fields. According to Eq.(5), the initially diamagnetic Meissner effect turns paramagnetic as soon as the piezomagnetic contribution $H^*(\sigma)$ exceeds an applied magnetic field H . To see whether this can actually happen in a real material, let us estimate the typical values of the piezomagnetic field H^* . By definition, $H^*(\sigma) = (\sigma/\sigma_m)(j_d/j_c)(d/b)H_0$ where $H_0 = \Phi_0/S$

is a characteristic magnetic field, and σ_m is an ultimate stress field. Typically [3–5], for HTGS ceramics $S \approx 10\mu m^2$, leading to $H_0 \simeq 1G$. To estimate the needed value of the dislocation current density j_d , we turn to the available experimental data. According to Ref.14, a rather strong polarization under compressive pressure $\sigma/\sigma_m \simeq 0.1$ was observed in *YBCO* ceramic samples at $T = 77K$ yielding $d_n = 10^2 C/m^2$ for the piezoelectric coefficient. Usually [12–14,16,20], for GBJJs $\dot{\epsilon}_0 \simeq 10^{-2} s^{-1}$, and $b \simeq 10nm$ leading to $j_d = d_n\dot{\epsilon}_0 \simeq 1A/m^2$ for the maximum dislocation current density. Using the typical values of the critical current density $j_c = 10^4 A/m^2$ and grain size $d \simeq 1\mu m$, we arrive at the following estimate of the piezomagnetic field $H^* \simeq 10^{-2} H_0$. Thus, the predicted stress induced paramagnetic Meissner effect (PME) should be observable for applied magnetic fields $H \simeq 10^{-2} H_0 \simeq 0.01G$ which correspond to the region where the original PME was first registered [2–6]. In turn, the piezoelectric coefficient d_n is related to an effective charge Q in the GBJJ as [26] $d_n = (Q/S)(d/b)^2$. Given the above-obtained estimates, we get a reasonable value of $Q \simeq 10^{-13} C$ for the charge accumulated at a GBJJ. It is interesting to notice that the above values of the applied stress σ and the resulting effective charge Q correspond (via the so-called electroplastic effect [26]) to an equivalent applied electric field $E = b^2\sigma/Q \simeq 10^7 V/m$ at which rather pronounced electric-field induced effects in HTGS were either observed (like an increase of the critical current in *YBCO* ceramics [27]) or predicted to occur (like a converse magnetoelectric effect [11]).

In conclusion, let us briefly discuss the contribution of the so-called striction effects [23] (which usually accompany any stress related changes). According to Ref.28 the Josephson projected area S was found to slightly decrease under pressure thus leading to some increase of the characteristic field $H_0 = \Phi_0/S$. In view of Eq.(5), it means that a smaller compression stress will be needed to actually reverse the sign of the induced magnetization M_s . Furthermore, if an unloaded granular superconductor already exhibits the PME, due to the orbital currents induced spontaneous magnetization resulting from an initial phase difference $\phi_{ij}^0 = 2\pi r$ in Eq.(2) with fractional r (in particular, $r = 1/2$ corresponds to the so-called [2–6] π -type state), then according to our predictions this effect will either be further enhanced by applying a compression (with $\sigma > 0$) or will disappear under a strong enough extension (with $\sigma < 0$) able to compensate the pre-existing effect. Given a very distinctive nonlinear character of $M_s(H, \sigma)$ (see Figs.1 and 2), the above-estimated range of accessible parameters suggests quite an optimistic possibility to observe the predicted effects experimentally either in HTGS ceramics or in a specially prepared system of arrays of superconducting grains. Finally, it is worth noting that a rather strong nonlinear response of the transport properties in *HgBaCaCuO* ceramics was observed [20] under compressive pressure

with $\sigma/\sigma_m \simeq 0.8$. Specifically, the critical current at $\sigma = 9kbar$ was found to be three times higher its value at $\sigma = 1.5kbar$, clearly indicating a weak-links-mediated origin of the phenomenon (in the best defect-free thin films this ratio never exceeds a few percents [19]).

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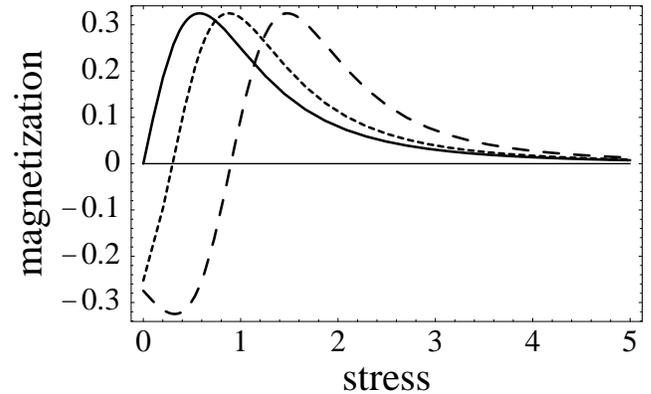


FIG. 1. The induced magnetization M_s/M_0 as a function of the reduced applied stress σ/σ_0 , according to Eq.(5) for different values of reduced applied magnetic field: $H/H_0 = 0.001$ (solid line), $H/H_0 = 0.01$ (dotted line), and $H/H_0 = 0.1$ (dashed line).

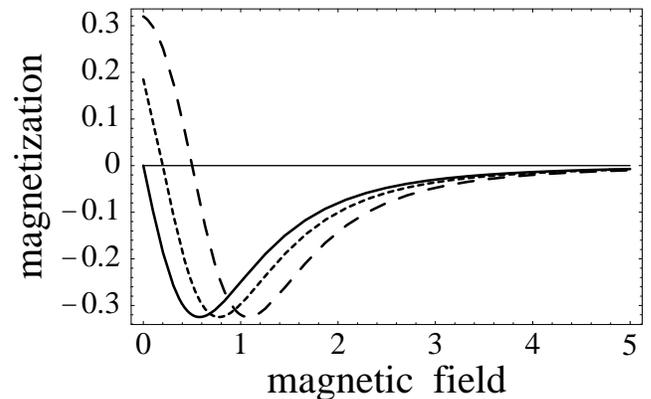


FIG. 2. The magnetization M_s/M_0 as a function of the reduced applied magnetic field H/H_0 , according to Eq.(5) for different values of reduced applied stress: $\sigma/\sigma_0 = 0$ (solid line), $\sigma/\sigma_0 = 0.01$ (dotted line), and $\sigma/\sigma_0 = 0.1$ (dashed line).