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# POLARIZATION EFFECTS INDUCED BY A MAGNETIC FIELD IN INTRINSICALLY GRANULAR SUPERCONDUCTORS

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Based on the previously suggested model of nanoscale dislocation-induced Josephson junctions and their arrays, we study the magnetic-field-induced electric polarization effects in intrinsically granular superconductors. In addition to a new phenomenon of chemomagnetoelectricity, the model also predicts a few other interesting effects, including charge analogues of Meissner paramagnetism (at low fields) and a «fishtail» anomaly (at high fields). The conditions under which these effects can be experimentally measured in nonstoichiometric high-$T_c$ superconductors are discussed.

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## 1. INTRODUCTION

Both granular superconductors and artificially prepared arrays of Josephson junctions (JJAs) proved useful in studying the numerous quantum (charging) effects, including blockade of Cooper pair tunneling [1], Bloch oscillations [2], propagation of quantum ballistic vortices [3], spin-tunneling related effects with specially designed SFS-type junctions [4, 5], novel Coulomb effects in SINIS-type nanoscale junctions [6], and recently observed geometric quantization phenomena [7] (see, e.g., Ref. [8] for a recent review on charge and spin effects in mesoscopic two-dimentional Josephson junctions).

More recently, it was realized that JJAs can also be used as quantum channels to transfer quantum information between distant sites [9–11] through the implementation of the so-called superconducting qubits, which involve both charge and phase degrees of freedom (see, e.g., Ref. [12] for a review on quantum-state engineering with Josephson-junction devices).

At the same time, imaging of the granular structure in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystals [13] revealed an apparent charge segregation of its electronic structure into superconducting domains (of the order of a few nanometers) located in an electronically distinct background. In particular, it was found that at low levels of hole doping ($\delta \leq 0.2$), the holes become concentrated at certain hole-rich domains. Tunneling between such domains leads to intrinsic granular superconductivity in high-$T_c$ superconductors (HTS). As was shown earlier [14], granular superconductivity based phenomena can shed some light on the origin and evolution of the so-called paramagnetic Meissner effect (PME) which manifests itself in both high-$T_c$ and conventional superconductors [15, 16].

In this paper, within a previously suggested [14] model of JJAs created by a regular two-dimentional network of twin-boundary dislocations with strain fields acting as an insulating barrier between hole-rich domains in underdoped crystals, we address another class of interesting phenomena that are actually dual to the chemomagnetic effects described in Ref. [14]. Specifically, we discuss a possible existence of a nonzero electric polarization $P(B, \delta)$ (chemomagnetoelectic effect) and the related change of the charge balance in an intrinsically granular nonstoichiometric material under the influence of an applied magnetic field. In particular, we predict an anomalous low-field magnetic behavior of the effective junction charge $Q(B, \delta)$ and concomitant magnetocapacitance $C(B, \delta)$ in paramagnetic Meissner

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phase and a charge analog of a «fishtail»-like anomaly at high magnetic fields.

## 2. THE MODEL

We recall that the regular two-dimentional dislocation networks of oxygen depleted regions with the size $d_0$ of a few Burgers vectors, observed in HTS single crystals [13, 17–20], can provide quite a realistic possibility for the existence of a two-dimentional Josephson network within the CuO plane [21, 22]. In this regard, it is also important to mention the pioneering works by Khaikin and Khlyustikov [23–25] on twinning-induced superconductivity in dislocated crystals.

At the same time, in underdoped crystals, there is a realistic possibility to facilitate oxygen transport via the so-called osmotic mechanism [14, 19, 20, 26], which relates the local value of the chemical potential

$$\mu(\mathbf{x}) = \mu(0) + \nabla\mu \cdot \mathbf{x}$$

with the local concentration of point defects as

$$c(\mathbf{x}) = \exp(-\mu(\mathbf{x})/k_B T),$$

and allows explicitly incorporating the oxygen deficiency parameter $\delta$ into our model by relating it to the excess oxygen concentration of vacancies $c_v \equiv c(0)$ as

$$\delta = 1 - c_v.$$

Assuming the relation between the variation of mechanical and chemical properties of planar defects,

$$\mu(\mathbf{x}) = K\Omega_0\epsilon(\mathbf{x}),$$

where

$$\epsilon(\mathbf{x}) = \epsilon_0 \exp(-|\mathbf{x}|/d_0)$$

is the screened strain field produced by tetragonal regions in a $d$-wave orthorhombic YBCO crystal, $\Omega_0$ is an effective atomic volume of the vacancy, and $K$ is the bulk elastic modulus), we can study the properties of twin-boundary induced JJs under the intrinsic chemical pressure $\nabla\mu$ (created by the variation of the oxygen doping parameter $\delta$). More specifically, a single SIS-type junction (comprising a Josephson network) is formed around the twin-boundary due to a local depression of the superconducting order parameter $\Delta(\mathbf{x}) \propto \epsilon(\mathbf{x})$ over distance $d_0$, thus producing a weak link with the Josephson coupling

$$J(\delta) = \epsilon(\mathbf{x})J_0 = J_0(\delta)\exp(-|\mathbf{x}|/d_0),$$

where

$$J_0(\delta) = \epsilon_0 J_0 = (\mu_v/K\Omega_0)J_0$$

(here, $J_0 \propto \Delta_0/R_n$, with $R_n$ being the resistance of the junction). We note that in accordance with the observations, for a stoichiometric situation (when $\delta \approx 0$), the Josephson coupling $J(\delta) \approx 0$ and the system loses its explicitly granular signature.

To describe the influence of chemomagnetic effects on charge balance of an intrinsically granular superconductor, we use the model of two-dimentional overdamped Josephson junction array based on the well-known Hamiltonian

$$\mathcal{H} = \sum_{i,j}^{N} J_{ij}(1 - \cos\phi_{ij}) + \sum_{i,j}^{N} \frac{q_i q_j}{2C_{ij}}. \qquad (1)$$

We introduce a short-range (nearest-neighbor) interaction between $N$ junctions (which are formed around oxygen-rich superconducting areas with phases $\phi_i$), arranged in a two-dimensional lattice with coordinates $\mathbf{x_i} = (x_i, y_i)$. The areas are separated by oxygen-poor insulating boundaries (created by twin-boundary strain fields $\epsilon(\mathbf{x}_{ij})$) producing a short-range Josephson coupling

$$J_{ij} = J_0(\delta)\exp(-|\mathbf{x}_{ij}|/d).$$

Thus, typically for granular superconductors, the Josephson energy of the array varies exponentially with the distance $\mathbf{x}_{ij} = \mathbf{x}_i - \mathbf{x}_j$ between neighboring junctions (with $d$ being the average junction size). As usual, the second term in the right-hand side of Eq. (1) accounts for Coulomb effects, where $q_i = -2en_i$ is the junction charge with $n_i$ being the pair number operator. Naturally, the same strain fields $\epsilon(\mathbf{x}_{ij})$ are also responsible for dielectric properties of oxygen-depleted regions via the $\delta$-dependent capacitance tensor

$$C_{ij}(\delta) = C[\epsilon(\mathbf{x}_{ij})].$$

If, in addition to the chemical pressure

$$\nabla\mu(\mathbf{x}) = K\Omega_0\nabla\epsilon(\mathbf{x}),$$

the network of superconducting grains is under the influence of an applied frustrating magnetic field $\mathbf{B}$, the total phase difference through the contact is given by

$$\phi_{ij} = \phi_{ij}^0 + \frac{\pi w}{\Phi_0}(\mathbf{x}_{ij} \wedge \mathbf{n}_{ij}) \cdot \mathbf{B} + \frac{\nabla\mu \cdot \mathbf{x}_{ij} t}{\hbar}, \qquad (2)$$

where $\phi_{ij}^0$ is the initial phase difference (see below),

$$\mathbf{n}_{ij} = \frac{\mathbf{X}_{ij}}{|\mathbf{X}_{ij}|}, \quad \mathbf{X}_{ij} = \frac{\mathbf{x}_i + \mathbf{x}_j}{2},$$

and

$$w = 2\lambda_L(T) + l,$$

with $\lambda_L$ being the London penetration depth of the superconducting area and $l$ the insulator thickness (which, within the scenario discussed here, is simply equal to the twin-boundary thickness [26]).

As usual, to safely neglect the influence of the self-field effects in a real material, the corresponding Josephson penetration length

$$\lambda_J = \sqrt{\frac{\Phi_0}{2\pi\mu_0 j_c w}}$$

must be larger than the junction size $d$. Here, $j_c$ is the critical current density of the superconducting (hole-rich) area. As we see below, this condition is rather well satisfied for HTS single crystals.

## 3. CHEMOMAGNETOELECTRICITY

In what follows, we are interested in the behavior of the magnetic-field-induced electric polarization (chemomagnetoelectricity) in chemically induced granular superconductivity described by a two-dimentional JJA. We recall that a conventional (zero-field) pair polarization operator within the model under discussion is given by [27, 28]

$$\mathbf{p} = \sum_{i=1}^{N} q_i \mathbf{x}_i. \tag{3}$$

In view of Eqs. (1)–(3) and the usual «phase–number» commutation relation

$$[\phi_i, n_j] = i\delta_{ij},$$

it can be shown that the evolution of the pair polarization operator is determined by the equation of motion

$$\frac{d\mathbf{p}}{dt} = \frac{1}{i\hbar}[\mathbf{p}, \mathcal{H}] = \frac{2e}{\hbar}\sum_{i,j}^{N} J_{ij}\sin\phi_{ij}(t)\mathbf{x}_{ij}. \tag{4}$$

Solving this equation, we obtain the net value of the magnetic-field-induced longitudinal electric polarization

$$P(\delta, \mathbf{B}) \equiv \langle p_x(t)\rangle$$

(along the $x$ axis) and the corresponding effective junction charge

$$Q(\delta, \mathbf{B}) = \frac{2eJ_0}{\hbar\tau d}\int_0^{\tau} dt \int_0^{t} dt' \times \\ \times \int \frac{d^2x}{S}\sin\phi(\mathbf{x}, t')x\exp(-|\mathbf{x}|/d), \tag{5}$$

where $S = 2\pi d^2$ is the properly defined normalization area, $\tau$ is a characteristic time (see Discussion), and we made a usual substitution

$$\frac{1}{N}\sum_{i,j} A_{ij}(t) \to \frac{1}{S}\int d^2x\, A(\mathbf{x}, t)$$

valid in the long-wavelength approximation [28].

To capture the very essence of the superconducting analog of the chemomagnetoelectric effect, we assume for simplicity in what follows that a stoichiometric sample (with $\delta \approx 0$) does not have any spontaneous polarization at zero magnetic field, that is, $P(0,0) = 0$. According to Eq. (5), this condition implies that $\phi_{ij}^0 = 2\pi m$ for the initial phase difference with $m = 0, \pm 1, \pm 2, \ldots$

Choosing the applied magnetic field along the $c$ axis (and normal to the CuO plane), that is, $\mathbf{B} = (0, 0, B)$, we finally obtain

$$Q(\delta, B) = Q_0(\delta)\frac{2\tilde{b} + b(1 - \tilde{b}^2)}{(1 + b^2)(1 + \tilde{b}^2)^2} \tag{6}$$

for the magnetic field behavior of the effective junction charge in chemically induced granular superconductors. Here,

$$Q_0(\delta) = e\tau J_0(\delta)/\hbar$$

with $J_0(\delta)$ defined earlier,

$$b = B/B_0, \quad \tilde{b} = b - b_\mu,$$

and

$$b_\mu = B_\mu/B_0 \approx (k_B T\tau/\hbar)\delta,$$

where

$$B_\mu(\delta) = (\mu_v\tau/\hbar)B_0$$

is the chemically induced contribution (which disappears in optimally doped systems with $\delta \approx 0$), and

$$B_0 = \Phi_0/wd$$

is a characteristic Josephson field.

Figure 1 shows changes of the initial (stoichiometric) effective junction charge $Q$ (solid line) with the oxygen deficiency $\delta$. We note a sign change of $Q$ (dotted and dashed lines) driven by nonzero values of $\delta$ at low magnetic fields (a charge analog of a chemically induced PME). According to Eq. (6), the effective charge changes its sign as soon as the chemomagnetic contribution $B_\mu(\delta)$ exceeds the applied magnetic field $B$ (see Discussion).

At the same time, Fig. 2 presents a true chemoelectric effect with the concentration (deficiency) induced effective junction charge $Q(\delta, 0)$ in zero magnetic

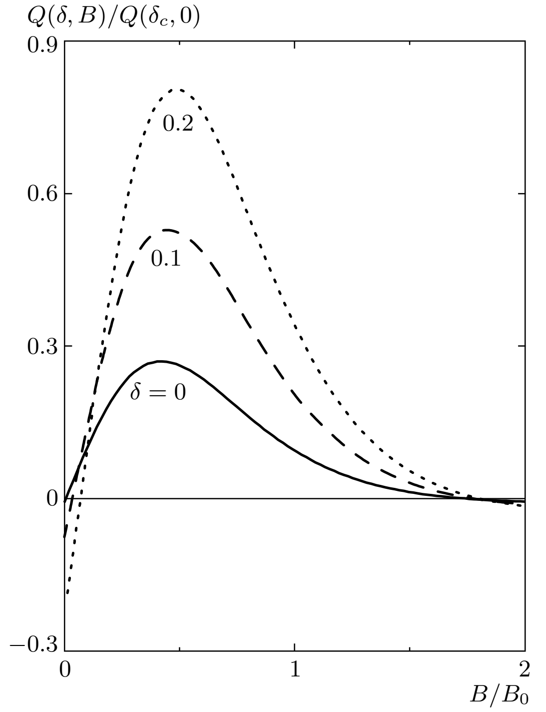


**Fig. 1.** The effective junction charge $Q(\delta, B)/Q(\delta_c, 0)$ (chemomagnetoelectric effect) as a function of the applied magnetic field $B/B_0$, according to Eq. (6), for different values of the oxygen deficiency parameter: $\delta \approx 0$ (solid line), $\delta = 0.1$ (dashed line), and $\delta = 0.2$ (dotted line)

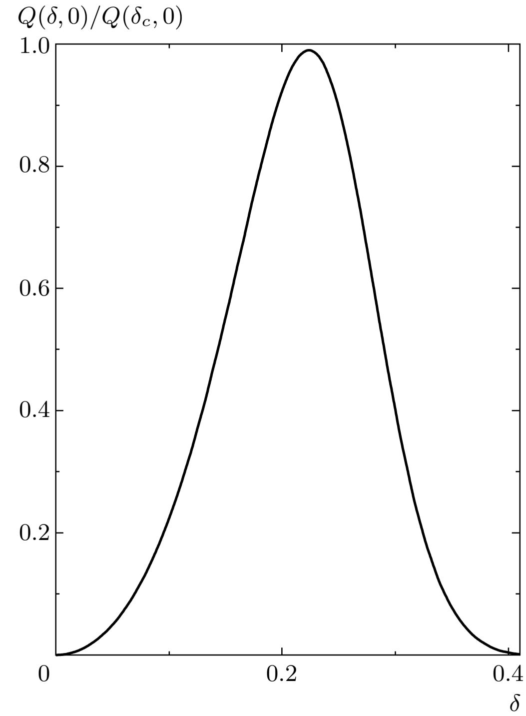


**Fig. 2.** Chemically induced effective junction charge $Q(\delta, 0)/Q(\delta_c, 0)$ in zero applied magnetic field (true chemoelectric effect)

field. We note that $Q(\delta, 0)$ exhibits a maximum around $\delta_c \approx 0.2$ (in agreement with the classical percolative behavior observed in nonstoichiometric $YBa_2Cu_3O_{7-\delta}$ samples [17]).

It is also of interest to consider the magnetic field behavior of the concomitant effective flux capacitance

$$C \equiv \frac{\tau\, dQ(\delta, B)}{d\Phi},$$

which, in view of Eq. (6), is given by

$$C(\delta, B) = C_0(\delta)\frac{1 - 3b\tilde{b} - 3\tilde{b}^2 + b\tilde{b}^3}{(1 + b^2)(1 + \tilde{b}^2)^3}, \qquad (7)$$

where

$$\Phi = SB, \quad C_0(\delta) = \tau Q_0(\delta)/\Phi_0.$$

Figure 3 depicts the behavior of the effective flux capacitance $C(\delta, B)$ in an applied magnetic field for different values of the oxygen deficiency parameter: $\delta \approx 0$ (solid line), $\delta = 0.1$ (dashed line), and $\delta = 0.2$ (dotted line). We note a decrease of the magnetocapacitance amplitude and its peak shifting with increase of $\delta$ and a sign change at low magnetic fields, which is another manifestation of the charge analog of a chemically induced PME (cf. Fig. 1).

## 4. CHARGE ANALOG OF THE «FISHTAIL» ANOMALY

So far, we neglected a possible field dependence of the chemical potential $\mu_v$ of oxygen vacancies. We recall, however, that in sufficiently high applied magnetic fields $B$, the field-induced change of the chemical potential

$$\Delta\mu_v(B) \equiv \mu_v(B) - \mu_v(0)$$

becomes tangible and should be taken into account [14, 29, 30]. As a result, we obtain a superconducting analog of the so-called magnetoconcentration effect [14] with field-induced creation of oxygen vacancies

$$c_v(B) = c_v(0)\exp(-\Delta\mu_v(B)/k_BT),$$

which in turn leads to a «fishtail»-like behavior of the high-field chemomagnetization (see Ref. [14] for more details).

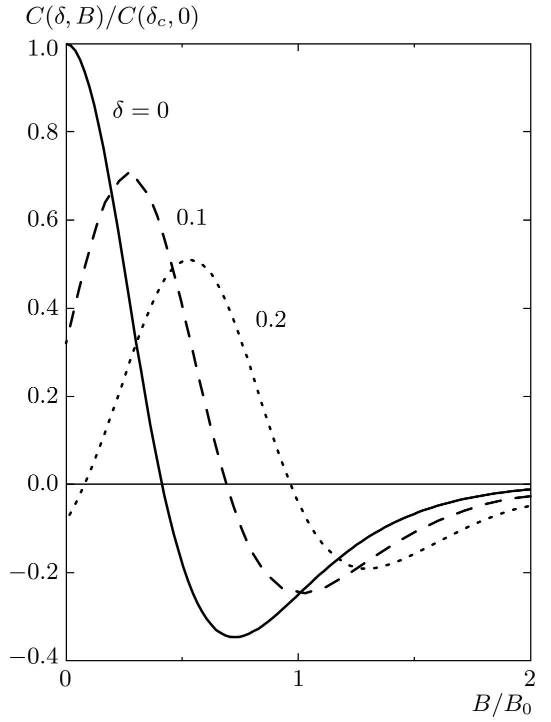


**Fig. 3.** The effective flux capacitance $C(\delta,B)/C(\delta_c,0)$ as a function of the applied magnetic field $B/B_0$, according to Eq. (7), for different values of the oxygen deficiency parameter: $\delta \approx 0$ (solid line), $\delta = 0.1$ (dashed line), and $\delta = 0.2$ (dotted line)

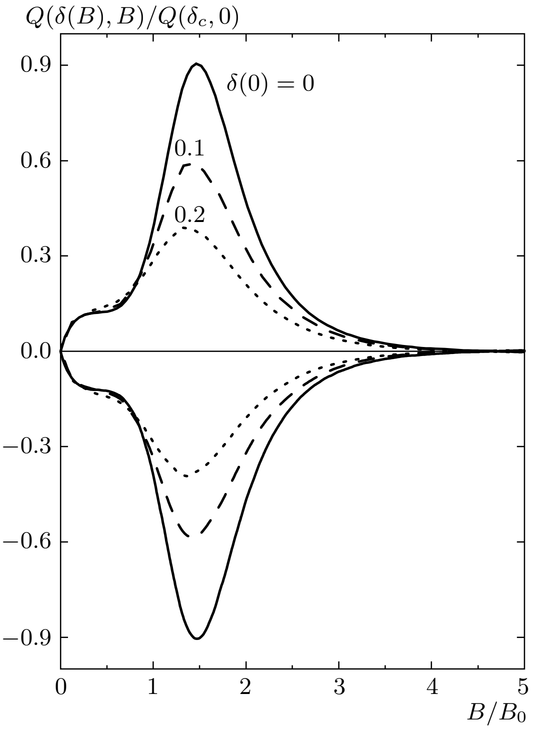


**Fig. 4.** A «fishtail»-like behavior of the effective charge $Q(\delta(B),B)/Q(\delta_c,0)$ in the applied magnetic field $B/B_0$ in the presence of a magnetoconcentration effect (with field-induced oxygen vacancies $\delta(B)$) for three values of the field-free deficiency parameter: $\delta(0) \approx 0$ (solid line), $\delta(0) = 0.1$ (dashed line), and $\delta(0) = 0.2$ (dotted line)

Figure 4 shows the field behavior of the effective junction charge in the presence of the abovementioned magnetoconcentration effect. As is clearly seen, $Q(\delta(B),B)$ exhibits a «fishtail»-like anomaly typical of the previously discussed [14] chemomagnetization in underdoped crystals with intragrain granularity (for symmetry and better visual effect, we also plotted $-Q(\delta(B),B)$ in the same figure). This more complex structure of the effective charge appears when the applied magnetic field $B$ matches the intrinsic chemomagnetic field $B_\mu(\delta(B))$ (which now also depends on $B$ via the magnetoconcentration effect). We note that a «fishtail» structure of $Q(\delta(B),B)$ manifests itself even at zero values of the field-free deficiency parameter $\delta(0)$ (solid line in Fig. 4), thus confirming a field-induced nature of intrinsic granularity [13, 17–20]. Likewise, Fig. 5 depicts the evolution of the effective flux capacitance $C(\delta(B),B)$ in the applied magnetic field $B/B_0$ in the presence of a magnetoconcentration effect (cf. Fig. 3).

## 5. DISCUSSION

Thus, the present model predicts the appearance of two interrelated phenomena (dual to the previously discussed behavior of chemomagnetism [14]), a charge analog of Meissner paramagnetism at low fields and a charge analog of the «fishtail» anomaly at high fields. To see whether these effects can be actually observed in a real material, we estimate the order of magnitude of the main model parameters.

Using the values $\lambda_L(0) \approx 150$ nm, $d \approx 10$ nm, and $j_c \approx 10^{10}$ A/m$^2$ typical [17, 19] for HTS single crystals, we estimate the characteristic field as $B_0 \approx 0.5$ T and the chemomagnetic field as $B_\mu(\delta) \approx 0.5B_0$. Therefore, the predicted charge analog of PME should be observable for applied magnetic fields $B < 0.25$ T. We note that for the above set of parameters, the Josephson length is of the order of $\lambda_J \approx 1$ $\mu$m, which means that the small-junction approximation assumed in this paper is valid and the «self-field» effects can be safely neglected.

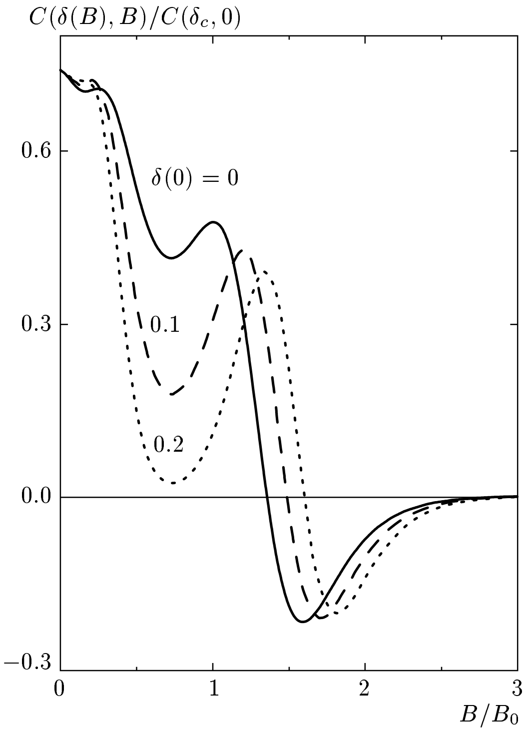


**Fig. 5.** The behavior of the effective flux capacitance $C(\delta(B),B)/C(\delta_c,0)$ in the applied magnetic field $B/B_0$ in the presence of a magnetoconcentration effect for three values of the field-free deficiency parameter: $\delta(0)\approx 0$ (solid line), $\delta(0)=0.1$ (dashed line), and $\delta(0)=0.2$ (dotted line)

Furthermore, the characteristic frequencies $\omega \approx \tau^{-1}$ needed to probe the effects suggested here are related to the processes governed by tunneling relaxation times $\tau \approx \hbar/J_0(\delta)$. Because for the oxygen the deficiency parameter $\delta = 0.1$, the chemically induced zero-temperature Josephson energy in nonstoichiometric YBCO single crystals is of the order of $J_0(\delta) \approx k_B T_C \delta \approx 1$ meV, we obtain the required frequencies $\omega \approx 10^{13}$ Hz and the estimates of the effective junction charge $Q_0 \approx e = 1.6\cdot 10^{-19}$ C and flux capacitance $C_0 \approx 10^{-18}$ F. We note that the above estimates fall into the range of parameters used in typical experiments for studying single-electron tunneling effects both in JJs and JJAs [1, 2, 12, 31], thus suggesting quite an optimistic possibility to observe the predicted field-induced effects experimentally in nonstoichiometric superconductors with pronounced networks of planar defects or in artificially prepared JJAs. (It is worth mentioning that a somewhat similar behavior of the magnetic-field-induced charge and related flux capacitance has been observed in 2D electron systems [32].)

Finally, it can be easily verified that in view of Eqs. (1)–(5), the field-induced Coulomb energy of the oxygen-depleted region within our model is given by

$$E_C(\delta,B) \equiv \left\langle \sum_{i,j}^{N} \frac{q_i q_j}{2C_{ij}} \right\rangle = \frac{Q^2(\delta,B)}{2C(\delta,B)} \qquad (8)$$

with $Q(\delta,B)$ and $C(\delta,B)$ defined by Eqs. (6) and (7).

A thorough analysis of the above expression reveals that in the PME state (when $B \ll B_\mu$), the chemically induced granular superconductor is always in the so-called Coulomb blockade regime (with $E_C > J_0$), while in the «fishtail» state (for $B \geq B_\mu$), the energy balance tips in favor of tunneling (with $E_C < J_0$). In particular,

$$E_C(\delta, B = 0.1B_\mu) = \frac{\pi}{2} J_0(\delta)$$

and

$$E_C(\delta, B = B_\mu) = \frac{\pi}{8} J_0(\delta).$$

It would be also interesting to verify this phenomenon of field-induced weakening of the Coulomb blockade experimentally.

## 6. CONCLUSION

In conclusion, within a realistic model of two-dimentional Josephson junction arrays created by two-dimentional network of twin boundary dislocations (with strain fields acting as an insulating barrier between hole-rich domains in underdoped crystals), a few novel electric polarization related effects expected to occur in an intrinsically granular material under applied magnetic fields were predicted, including a phenomenon of chemomagnetoelectricity, an anomalous low-field magnetic behavior of the effective junction charge (and flux capacitance) in the paramagnetic Meissner phase, and a charge analog of a «fishtail»-like anomaly at high magnetic fields as well as field-dependent weakening of the chemically induced Coulomb blockade. The experimental conditions needed to observe the effects predicted here in nonstoichiometric high-$T_c$ superconductors were discussed.

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