

Magnetic field induced polarization effects in intrinsically granular superconductors

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Based on the previously suggested model of nanoscale dislocations 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 chemomagnetolectricity, the model predicts also a few other interesting effects, including charge analogues of Meissner paramagnetism (at low fields) and "fishtail" anomaly (at high fields). The conditions under which these effects can be experimentally measured in non-stoichiometric high- T_c superconductors are discussed.

I. INTRODUCTION

Both granular superconductors and artificially prepared arrays of Josephson junctions (JJAs) proved useful in studying the numerous quantum (charging) effects in these interesting systems, including blockade of Cooper pair tunneling [1], Bloch oscillations [2], propagation of quantum ballistic vortices [3], spin-tunneling related effects using 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 the recent review on charge and spin effects in mesoscopic 2D Josephson junctions).

More recently, it was realized that JJAs can be also used as quantum channels to transfer quantum information between distant sites [9–11] through the implementation of the so-called superconducting qubits which take advantage of 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 (GS) in high- T_c superconductors (HTS). As was shown earlier [14], GS based phenomena can shed some light on the origin and evolution of the so-called paramagnetic Meissner effect (PME) which manifests itself both in high- T_c and conventional superconductors [15,16].

In this paper, within a previously suggested [14] model of JJAs created by a regular 2D network of twin-boundary (TB) dislocations with strain fields acting as an insulating barrier between hole-rich domains in underdoped crystals, we address yet another class of interesting phenomena which are actually dual to the chemomagnetic effects described in Ref. [14]. Specifically, we discuss a possible existence of a non-zero electric polarization $P(B, \delta)$ (chemomagnetolectric effect) and the related change of the charge balance in intrinsically granular non-stoichiometric 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 phase and a charge analog of "fishtail"-like anomaly at high magnetic fields.

II. THE MODEL

Recall that the observed [13,17–20] in HTS single crystals regular 2D dislocation networks of oxygen depleted regions with the size d_0 of a few Burgers vectors can provide quite a realistic possibility for existence of 2D Josephson network within CuO plane [21,22]. In this regard, it is also important to mention the pioneer works by Khaikin and Khlyustikov [23–25] on twinning-induced superconductivity in dislocated crystals.

At the same time, in underdoped crystals there is a real possibility to facilitate oxygen transport via the so-called osmotic mechanism [14,19,20,26] which relates a local value of the chemical potential $\mu(\mathbf{x}) = \mu(0) + \nabla\mu \cdot \mathbf{x}$ with a

local concentration of point defects as follows $c(\mathbf{x}) = e^{-\mu(\mathbf{x})/k_B T}$, and allows us to explicitly incorporate the oxygen deficiency parameter δ into our model by relating it to the excess oxygen concentration of vacancies $c_v \equiv c(0)$ as follows $\delta = 1 - c_v$. Assuming the following connection between the variation of mechanical and chemical properties of planar defects, namely $\mu(\mathbf{x}) = K\Omega_0\epsilon(\mathbf{x})$ (where $\epsilon(\mathbf{x}) = \epsilon_0 e^{-|\mathbf{x}|/d_0}$ is screened strain field produced by tetragonal regions in d -wave orthorhombic $YBCO$ crystal, Ω_0 is an effective atomic volume of the vacancy, and K is the bulk elastic modulus), we can study the properties of TB induced JJs under intrinsic chemical pressure $\nabla\mu$ (created by the variation of the oxygen doping parameter δ). More specifically, a single *SIS* type junction (comprising a Josephson network) is formed around TB due to a local depression of the superconducting order parameter $\Delta(\mathbf{x}) \propto \epsilon(\mathbf{x})$ over distance d_0 producing thus a weak link with Josephson coupling $J(\delta) = \epsilon(\mathbf{x})J_0 = J_0(\delta)e^{-|\mathbf{x}|/d_0}$ 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 a resistance of the junction). Notice that, in accordance with the observations, for stoichiometric situation (when $\delta \simeq 0$), the Josephson coupling $J(\delta) \simeq 0$ and the system loses its explicitly granular signature.

To describe the influence of chemomagnetic effects on charge balance of an intrinsically granular superconductor, we employ a model of 2D overdamped Josephson junction array which is based on the well known Hamiltonian

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

and introduces a short-range (nearest-neighbor) interaction between N junctions (which are formed around oxygen-rich superconducting areas with phases ϕ_i), arranged in a two-dimensional (2D) lattice with coordinates $\mathbf{x}_i = (x_i, y_i)$. The areas are separated by oxygen-poor insulating boundaries (created by TB strain fields $\epsilon(\mathbf{x}_{ij})$) producing a short-range Josephson coupling $J_{ij} = J_0(\delta)e^{-|\mathbf{x}_{ij}|/d}$. 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 an average junction size). As usual, the second term in the rhs 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 $\epsilon(\mathbf{x}_{ij})$ will be responsible for dielectric properties of oxygen-depleted regions as well via the δ -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 reads

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

where ϕ_{ij}^0 is the initial phase difference (see below), $\mathbf{n}_{ij} = \mathbf{X}_{ij}/|\mathbf{X}_{ij}|$ with $\mathbf{X}_{ij} = (\mathbf{x}_i + \mathbf{x}_j)/2$, and $w = 2\lambda_L(T) + l$ with λ_L being the London penetration depth of superconducting area and l an insulator thickness (which, within the discussed here scenario, is simply equal to the TB thickness [26]).

As usual, to safely neglect the influence of the self-field effects in a real material, the corresponding Josephson penetration length $\lambda_J = \sqrt{\Phi_0/2\pi\mu_0 j_c w}$ must be larger than the junction size d . Here j_c is the critical current density of superconducting (hole-rich) area. As we shall see below, this condition is rather well satisfied for HTS single crystals.

III. CHEMOMAGNETOELECTRICITY

In what follows, we shall be interested in the behavior of magnetic field induced electric polarization (chemomagnetolectricity) in chemically induced GS described by a 2D JJA. Recall that a conventional (zero-field) pair polarization operator within the model under discussion reads [27,28]

$$\mathbf{p} = \sum_{i=1}^N q_i \mathbf{x}_i \quad (3)$$

In view of Eqs.(1)-(3), and taking into account a 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 via the equation of motion

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

Resolving the above equation, we arrive at the following net value of the magnetic-field induced longitudinal (along x -axis) electric polarization $P(\delta, \mathbf{B}) \equiv \langle p_x(t) \rangle$ and the corresponding effective junction charge

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

where $S = 2\pi d^2$ is properly defined normalization area, τ is a characteristic time (see Discussion), and we made a usual substitution $\frac{1}{N} \sum_{ij} A_{ij}(t) \rightarrow \frac{1}{S} \int d^2x A(\mathbf{x}, t)$ valid in the long-wavelength approximation [28].

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

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

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

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, $b = B/B_0$, $\tilde{b} = b - b_\mu$, and $b_\mu = B_\mu/B_0 \simeq (k_B T \tau / \hbar) \delta$ where $B_\mu(\delta) = (\mu_v \tau / \hbar) B_0$ is the chemically-induced contribution (which disappears in optimally doped systems with $\delta \simeq 0$), and $B_0 = \Phi_0/wd$ is a characteristic Josephson field.

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

At the same time, Fig.2 presents a true *chemoelectric* effect with concentration (deficiency) induced effective junction charge $Q(\delta, 0)$ in zero magnetic field. Notice that $Q(\delta, 0)$ exhibits a maximum around $\delta_c \simeq 0.2$ (in agreement with the classical percolative behavior observed in non-stoichiometric $YBa_2Cu_3O_{7-\delta}$ samples [17]).

It is of interest also to consider the magnetic field behavior of the concomitant effective flux capacitance $C \equiv \tau dQ(\delta, B)/d\Phi$ which in view of Eq.(6) reads

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

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

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

IV. CHARGE ANALOG OF "FISHTAIL" ANOMALY

So far, we neglected a possible field dependence of the chemical potential μ_v of oxygen vacancies. Recall, however, that in high enough 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 end up with a superconducting analog of the so-called *magnetoconcentration* effect [14] with field induced creation of oxygen vacancies $c_v(B) = c_v(0) \exp(-\Delta\mu_v(B)/k_B T)$ which in turn brings about a "fishtail"-like behavior of the high-field chemomagnetization (see Ref. [14] for more details).

Fig.4 shows the field behavior of the effective junction charge in the presence of the above-mentioned magnetoconcentration effect. As it is clearly seen, $Q(\delta(B), B)$ exhibits a "fishtail"-like anomaly typical for 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 an intrinsic chemomagnetic field $B_\mu(\delta(B))$ (which now also depends on B via the magnetoconcentration effect). Notice that a "fishtail" structure of $Q(\delta(B), B)$ manifests itself even at zero values of

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 applied magnetic field B/B_0 in the presence of magnetoconcentration effect (Cf. Fig.3).

V. DISCUSSION

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

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

Furthermore, the characteristic frequencies $\omega \simeq \tau^{-1}$ needed to probe the suggested here effects are related to the processes governed by tunneling relaxation times $\tau \simeq \hbar/J_0(\delta)$. Since for oxygen deficiency parameter $\delta = 0.1$ the chemically-induced zero-temperature Josephson energy in non-stoichiometric *YBCO* single crystals is of the order of $J_0(\delta) \simeq k_B T_C \delta \simeq 1meV$, we arrive at the required frequencies of $\omega \simeq 10^{13}Hz$ and at the following estimates of the effective junction charge $Q_0 \simeq e = 1.6 \times 10^{-19}C$ and flux capacitance $C_0 \simeq 10^{-18}F$. Notice that the above estimates fall into the range of parameters used in typical experiments for studying the single-electron tunneling effects both in JJs and JJAs [1,2,12,31] suggesting thus quite an optimistic possibility to observe the above-predicted field induced effects experimentally in non-stoichiometric 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].)

And 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

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

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

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 check this phenomenon of field-induced weakening of the Coulomb blockade experimentally.

VI. CONCLUSION

In conclusion, within a realistic model of 2D Josephson junction arrays created by 2D 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 intrinsically granular material under applied magnetic fields were predicted, including a phenomenon of chemomagnetolectricity, an anomalous low-field magnetic behavior of the effective junction charge (and flux capacitance) in paramagnetic Meissner phase and a charge analog of "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 predicted here effects in non-stoichiometric high- T_c superconductors were discussed.

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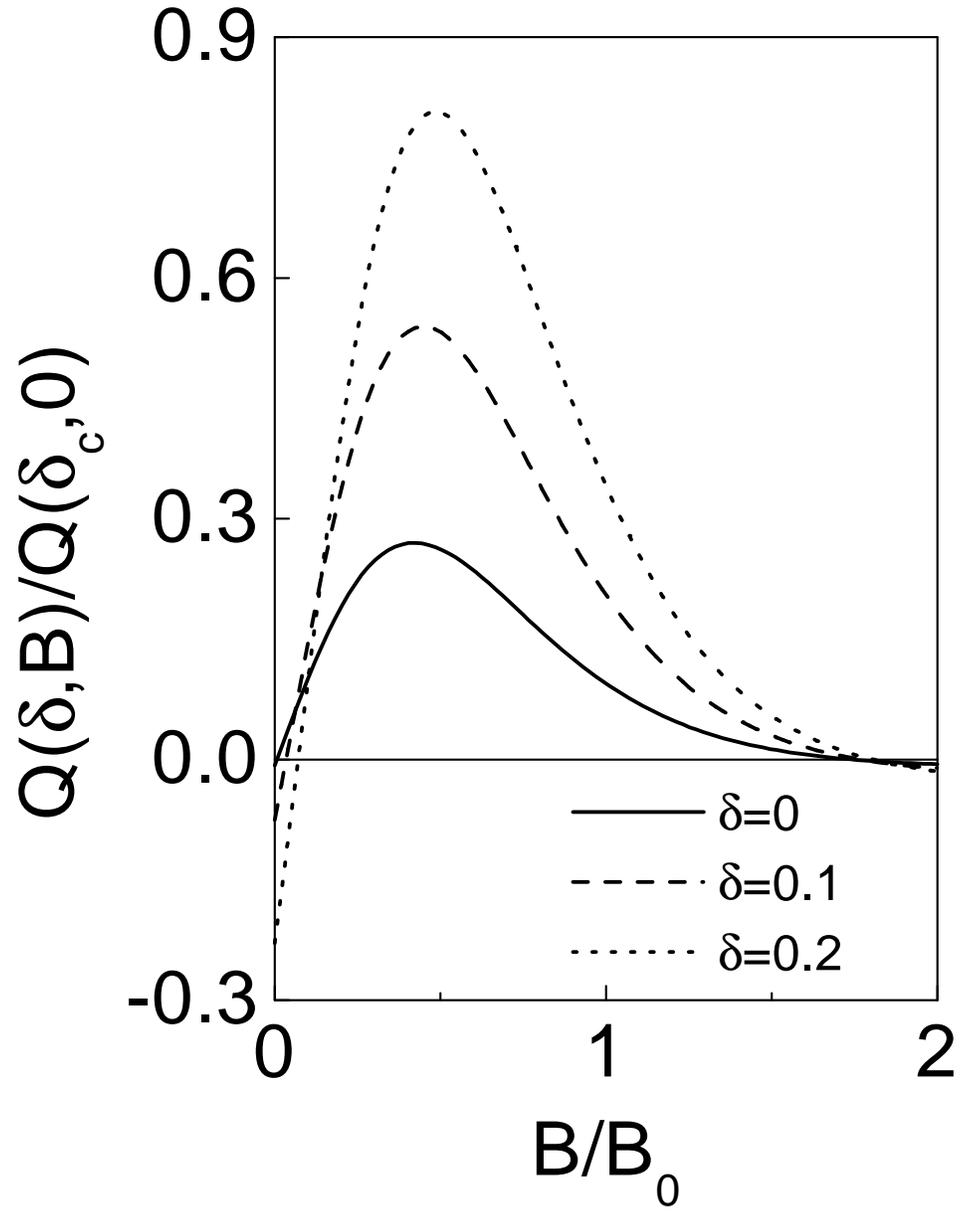


FIG. 1. The effective junction charge $Q(\delta, B)/Q(\delta_c, 0)$ (chemomagnetoelectric effect) as a function of applied magnetic field B/B_0 , according to Eq.(6), for different values of oxygen deficiency parameter: $\delta \simeq 0$ (solid line), $\delta = 0.1$ (dashed line), and $\delta = 0.2$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

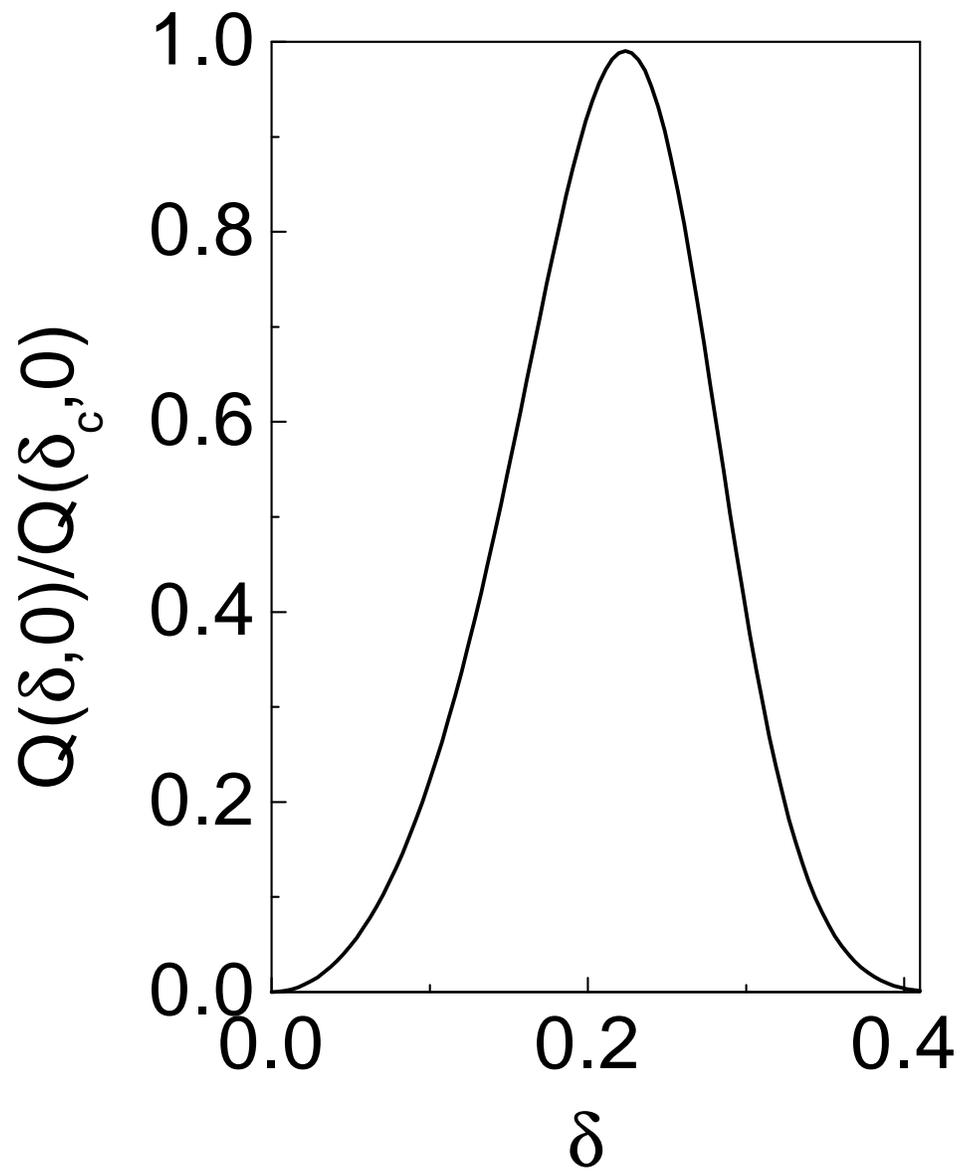


FIG. 2. Chemically induced effective junction charge $Q(\delta, 0)/Q(\delta_c, 0)$ in a zero applied magnetic field (true chemoelectric effect).

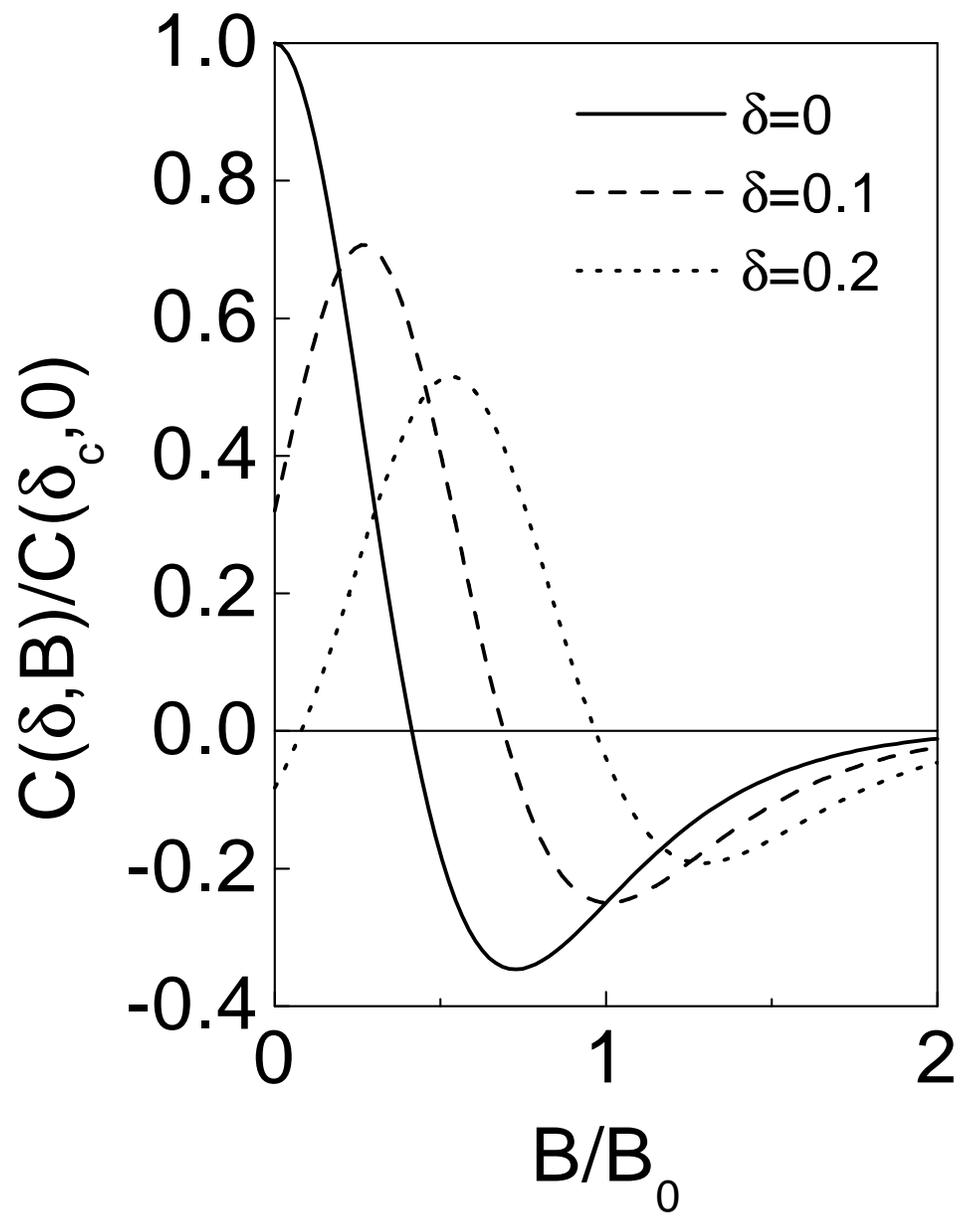


FIG. 3. The effective flux capacitance $C(\delta, B)/C(\delta_c, 0)$ as a function of applied magnetic field B/B_0 , according to Eq.(7), for different values of oxygen deficiency parameter: $\delta \simeq 0$ (solid line), $\delta = 0.1$ (dashed line), and $\delta = 0.2$ (dotted line).

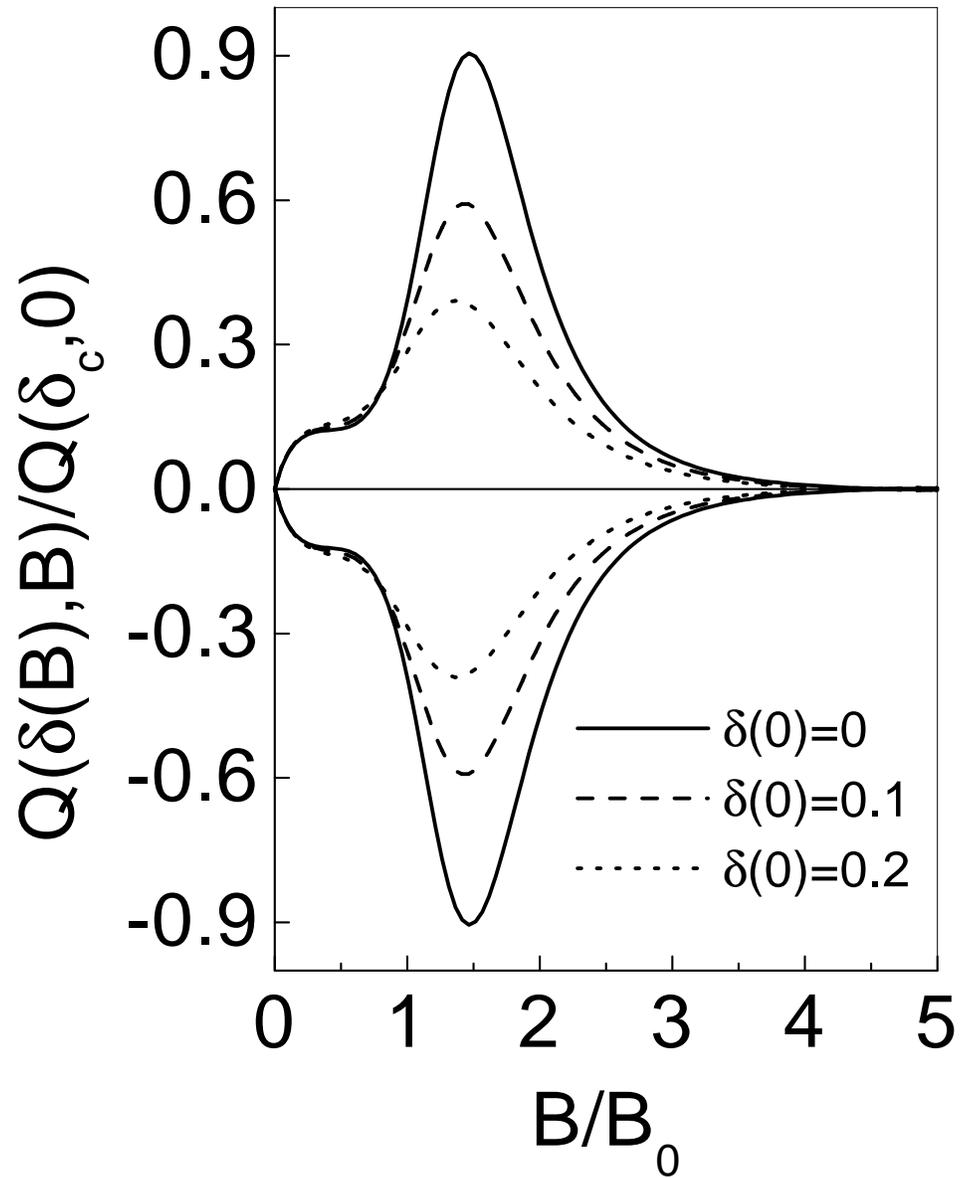


FIG. 4. A "fishtail"-like behavior of effective charge $Q(\delta(B), B)/Q(\delta_c, 0)$ in applied magnetic field B/B_0 in the presence of magnetoconcentration effect (with field-induced oxygen vacancies $\delta(B)$) for three values of field-free deficiency parameter: $\delta(0) \simeq 0$ (solid line),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

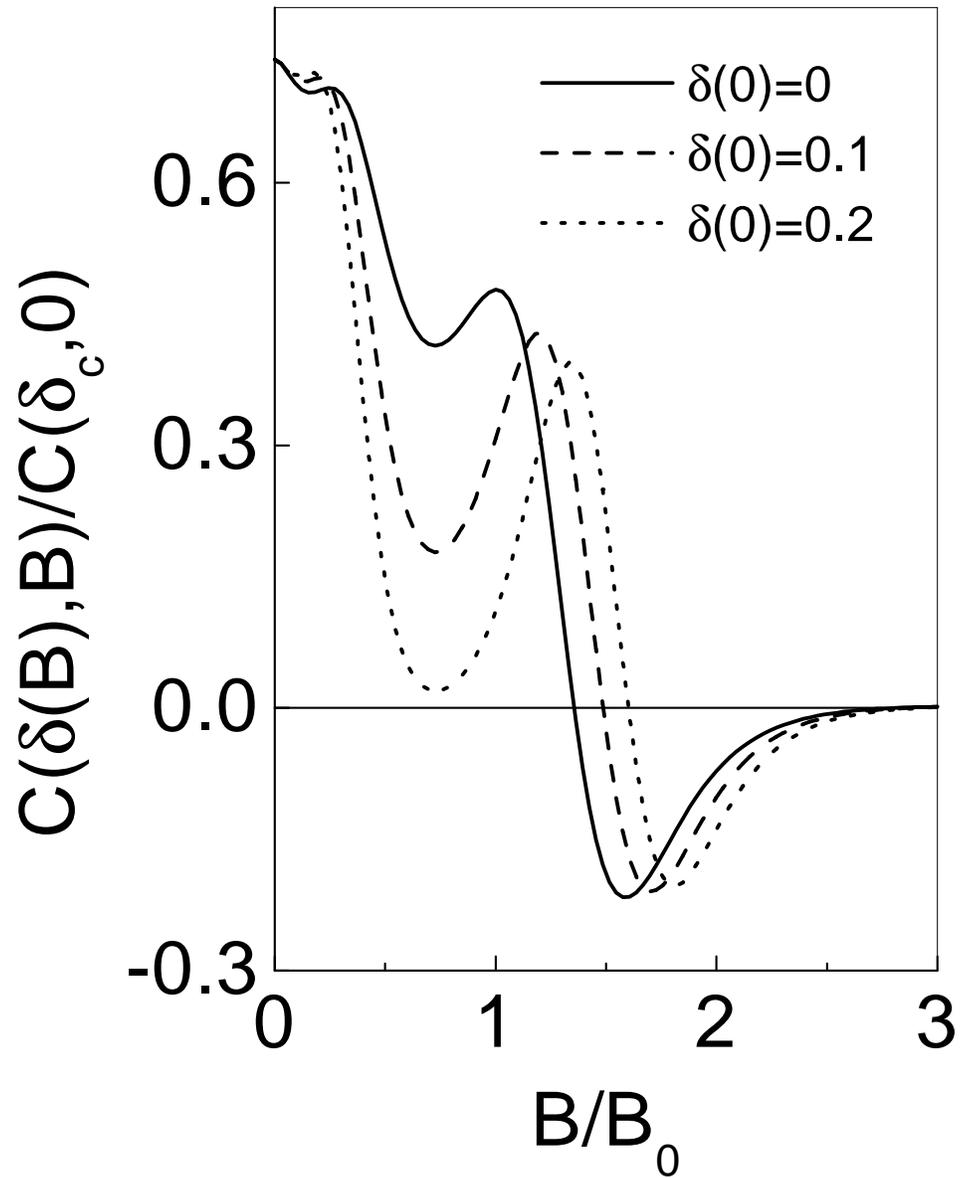


FIG. 5. The behavior of the effective flux capacitance $C(\delta(B), B)/C(\delta_c, 0)$ in applied magnetic field B/B_0 in the presence of magnetoconcentration effect for three values of field-free deficiency parameter: $\delta(0) \simeq 0$ (solid line), $\delta(0) = 0.1$ (dashed line), and $\delta(0)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