Induced paramagnetic states by localized π loops in grain boundaries

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Recent experiments on high-temperature superconductors show a paramagnetic behavior localized at grain boundaries (GBs). This paramagnetism can be attributed to the presence of unconventional d-wave induced π junctions. By modeling the GBs as an array of π and conventional Josephson junctions we determine the conditions of the occurrence of the paramagnetic behavior.

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The discovery of spontaneous currents in granular high-T_c superconductors[1–5] was a strong indication that a d-wave symmetry of the order parameter is present in these materials. Indeed the d-wave scenario implies the possibility of the existence of so-called π junctions, i.e., Josephson junction formed between superconductors with unconventional pairings which cause a π shift in the phase-current relation.[6] A π loop is an unconventional superconducting loop which contains an odd number of π junctions. In zero field the ground state of a π loop shows two energy degenerate magnetization states corresponding to two spontaneous current states, clockwise and counterclockwise. In a nonzero magnetic field these spontaneous currents act like orbital currents in paramagnetism.[7] Therefore, if the sample were field cooled, to permit the inner loops to ''feel'' the magnetic field, the response would be paramagnetic, as indeed was found in early works on the paramagnetic Meissner effect (PME) in $Bi_2Sr_2CaCu_2O_8$ (BSSCO) by Braunisch *et al.*[8] The PME was also observed in different high-T_c ceramic materials.[9]

However, the presence of the PME in conventional low-T_c samples shows that it cannot always be attributed to d-wave pairing.[10] Recently experiments and simulations were devised to test the relation between multiple connectiveness and the PME in conventional systems. A square array of low-T_c junctions was field cooled and shown to be paramagnetic over a large interval of the magnetic field.[11–13] These papers also proposed a qualitative explanation for the effect based on the array multiple connectiveness rather than the presence of a π-junction. The effect of adding a π junction in square arrays was analyzed in Ref. 14.

The observation of spontaneous currents in $YBa_2Cu_3O_{7-x}$ (YBCO) biepitaxial 0°-90° tilt-tilt and twist-tilt grain boundary (GB) junctions[5] indicates that paramagnetic effects due to d-wave pairing could be observed in GBs. In Ref. 4 spontaneous magnetic moments was observed both in high-T_c films, where granularity or defects pin some vortices, and along the GBs. Nevertheless, the sample response in field cooling was diamagnetic. A recent experiment by Il'ichev *et al.*[15] found that YBCO biepitaxial 45° asymmetric GB junctions in (nominally) zero field cooling show a paramagnetic response at low field. The origin of this paramagnetism could be debated. Is this simply due to the presence of localized π loops that will act similarly to two-dimensional systems,[14] or can it be explained by means of paramagnetic quasiparticle currents due to the existence of midgap states?[16] Here we want explore the first alternative in detail.

In general a loop containing p Josephson junctions will have different magnetization states when a magnetic field is applied. If the junctions are identical the loop current I_n is the solution of the following equation:[17]

$$\frac{I_n}{I_0}=\sin\left[\frac{1}{p}\left(2\pi n-k\pi-2\pi f-\beta\frac{I_n}{I_0}\right)\right], \tag{1}$$

where $n=0,1,\ldots,p-1$ is the quantum number in the flux quantization expression. f is the frustation equal to the external flux normalized to flux quantum Φ_0 and β is the superconducting quantum interference devios parameter $2\pi I_0 L/\Phi_0$, with L the loop inductance and I_0 the critical current of junctions in the loop. Varying n gives different families of independent solutions within a 2π phase change.[18] k is an index which is equal to 1 if there are an odd number of π junctions in the loop, and equal to zero otherwise.

For any p the lowest energy solutions of Eq. (1) are diamagnetic for conventional loops and paramagnetic for π loops.[17] When $\beta<1$ we have only one solution in the $p=1$ loop which is diamagnetic in the conventional loop and paramagnetic in the π loop without spontaneous currents.[3] But in multijunctions loops $(p>1)$ we can have more states due to the presence of nontrivial solutions when changing the quantum number n. This implies that π loops with, e.g., $p=2$ will also show spontaneous currents for low β's. Indeed for small β the solutions of Eq. 1 can be written as $\gamma_\pm \simeq \sin(\pm\pi/2-\pi f)[1-\cos(\pm\pi/2-\pi f)\beta_L/2]$. So, for $f=0$, we have two opposite spontaneous currents. For $0<f<1/2$ the solution γ_+ is positive (paramagnetic) and γ_- is negative (diamagnetic). Moreover, $\gamma_+<\gamma_-$, giving a lower energy for the paramagnetic solution.[19]

Small β π loops could likely be localized between GBs with different orientations along a junction[20] or where faceting causes an imperfect not completely flat GB passing from a conventional junction to a π junction, or vice versa. Recently engineered ''zigzag'' arrays of mixed π/conventional junctions have also been realized and measured.[21,22] These can be described as an array of π loops separated by all conventional or all π regions.[23]

In the following we will describe the GB as $1d$ array of $N+1$ Josephson junctions placed along it. The π additional phase is supposed to vary along the array giving arise to π and conventional sections separated by localized π loops (see Fig. 1).[24,25] We assume that system is not disordered.

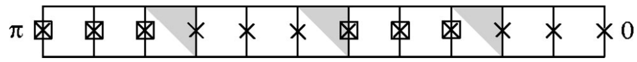

FIG. 1. Mixed π/conventional one-dimensional Josephson junction array with localized π-loops (half-gray).

The magnetization dynamics of this N-loop system can be described using the discrete sine-Gordon equation:[26]

$$\varphi_{j,tt}+\alpha\varphi_{j,t}+(-1)^{k(j)}\sin\varphi_j=\frac{1}{\beta}(\varphi_{j+1}-2\varphi_j+\varphi_{j-1})+\frac{2\pi}{\beta}(f_{j^+}-f_{j^-}), \qquad (2)$$

where φ_j is the phase of the jth junction in the GB, $f_{j^\pm}=\Phi_{ext,j^\pm}/\Phi_0$ is the frustation in the $j^\pm$th loop preceding $(-)$ or following $(+)$ the jth junction; the index $k(j)$ will be 0 for conventional junctions and 1 for π junctions. Times are normalized with respect to the Josephson plasma frequency ω_J, and α is the normalized conductance. To include boundaries we set $\varphi_0=\varphi_1$, $\varphi_{N+1}=\varphi_{N+2}$, and $f_0=f_{N+1}=0$. We assume f_j is a constant equal to f for $1<j<N$. This implies that the magnetic field enters as boundary conditions on the two side loops of the array. The term $2\pi f/\beta^{1/2}$ is equal to the normalized magnetic field at boundary $\eta=2\pi/\Phi_0\cdot\lambda_L\lambda_J B_{ext}$ (see Ref. 26). Equation (2) is analogous to that deduced in the continuous limit by Goldobin *et al.* in Ref. 23 in the context of analysis of ''zigzag'' arrays. We note that it can be shown that Eq. (2) for N equal to 1 implies Eq. (1) for p equal to 2.

To evaluate model parameters we now use experimental data on YBCO GB junctions.[4,15] We remark that the model will apply mainly to these high-T_c systems with critical current densities given in Ref. 5. The results can be equivalently valid for artificial zigzag arrays.[21,22] In YBCO GB junctions the Josephson length λ_J is smaller than the GB physical dimension L, thus the normalized length $l=L/\lambda_J$ is larger than 1.[4] The grain dimension along the GB Δx is usually smaller than L, being roughly of 1 μm for GB of Ref. 15 or also less in other circumstances.[27] The GB faceting is even smaller, ranging around 0.1–0.01 μm.[2,15] A rough estimate of β can be made identifying $\beta^{1/2}$ with the normalized length of grain $\Delta x/\lambda_J$.[28] From Refs. 4 and 5 we found $\lambda_J\sim 5$ μm, which gives $\beta\simeq 0.04$. GB faceting will give also a smaller β.

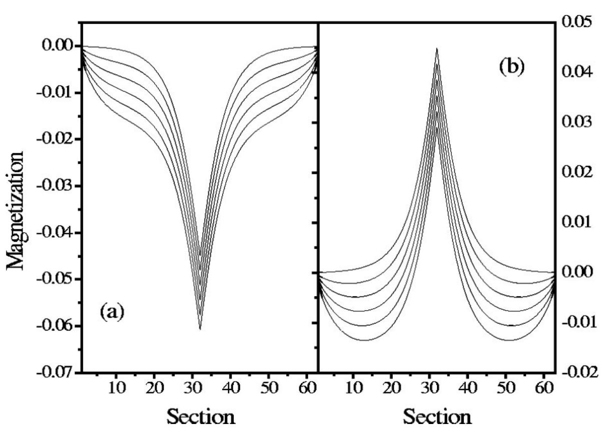

FIG. 2. Simulated magnetization of an $N=63$ Josephson junction array with a single π loop in the middle with $\beta_L=0.04$ and $\alpha=0.25$: (a) Diamagnetic solution with progressively increasing magnetic field η top to bottom 0, 0.1, 0.2, 0.3, 0.4, 0.5 (b) Paramagnetic solution with progressively increasing magnetic field [same values as in (a)].

By integrating Eq. (2) we find the phases for all junctions. Initially the phases of conventional junctions are set to zero and the phases of the π junction to π or $-\pi$, which are the stable equilibrium points of the single junction potential. These two possible choices correspond to two different signs of the spontaneous current circulating around π loops. α was set to 0.25, which is within the interval proposed in Ref. 23. We do not use a field cooling process as in Ref. 13 because initial conditions naturally set out diamagnetic or paramagnetic solutions as in the single loop. In the absence of a bias current, the system naturally sets in a static equilibrium solution (ground state[23]) after a few plasma periods. Then the local magnetization is evaluated by

$$m_j=\frac{\Phi_{tot,j}}{\Phi_0}-\frac{\Phi_{ext}}{\Phi_0}=\frac{\Delta\varphi_j}{2\pi}-f, \qquad (3)$$

where $\Delta\varphi_j=\varphi_{j+1}-\varphi_j$, and the mean magnetization by

$$m=\frac{1}{N}\sum m_j=\frac{1}{N}\frac{\sum\Delta\varphi_j}{2\pi}-f=\frac{\Delta\varphi}{2\pi N}-f, \qquad (4)$$

where $\Delta\varphi=\varphi_{N+1}-\varphi_1$. In the absence of an external magnetic field the magnetization for a single localized π loop in the array center (asymmetric $0-\pi$ junction[24]) has the shape reported in Fig. 2 (topmost curves), where the two spontaneous magnetizations are shown for a $N=63$ loop array with $\beta=0.04$. The shape is very similar to that of the ''half-fluxon'' obtained in the continuous approach[23] due to relatively small β. In Fig. 2 the effect of the magnetic field increase on the spontaneous magnetizations is also shown. The magnetic field breaks the symmetry of two solutions: one is paramagnetic and the other diamagnetic. With the increase of the magnetic field the magnetization of the paramagnetic state is progressively reduced due to the screening diamagnetic currents that are generated at the boundary. The same currents add to the magnetization of the diamagnetic state, giving a larger diamagnetic magnetization.

In Fig. 3 the mean magnetization for an array with a single π loop is reported (circles). We note that magnetization of paramagnetic state is zero at a threshold field $\eta^*\simeq 0.29$. The linear decrease of mean is similar to that observed for (large β) single loops.[17] For the parameters of Fig. 2 the physical threshold field is $B^*\sim 38$ mG with the λ_L given in Ref. 4.

In Fig. 4(a) we report the magnetization pattern in an array of $N=255$ loops with 15 localized π loops. According to Ref. 23 flux quanta are sufficiently separated here to stay stable, the (minimum) length of conventional or π sections

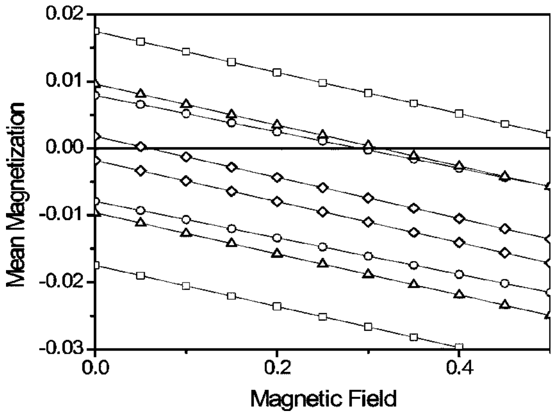

FIG. 3. Mean magnetizations of both paramagnetic (upper curve) and diamagnetic (lower curve) solutions for Josephson junction mixed arrays. For all curves $\alpha=0.25$ and $\beta=0.04$: ○ $N=63$ with a single π-loop; ◇ $N=255$ with 15 π loops and one odd paramagnetic half flux quantum; △ $N=255$, with 15 π loops and ten paramagnetic half flux quanta; □ $N=255$, with 15 π loops and 12 paramagnetic half flux quanta.

being $\Delta x/\lambda_J \simeq 4.64$. The solution shows seven positive-negative pairs of half flux quanta plus an unpaired half flux quantum. In Fig. 4(a) the unpaired half flux quantum is positive, so the solution is paramagnetic. An analogous diamagnetic solution exists when the unpaired half flux quantum is negative. Even π-loop configurations have zero spontaneous magnetization and are diamagnetic in small fields. Unpaired paramagnetic half flux quanta can be induced in the sample by a (moderate) field cooling process in a small field, similar to Ref. 4. The behavior of the mean magnetization is reported Fig. 3. Both the spontaneous magnetization and threshold field are very small in this case. With the above data we find $B^* \sim 7.6$ mG. In the same Fig. 4 we also report the case in which ten [Fig. 4(b)] and 12 (Fig. 4c) π loops have initial paramagnetic magnetizations, which correspond to a stronger field cooling effect.[29] The corresponding mean magnetizations are again reported in Fig. 3. The mean magnetization for 12 paramagnetic π loops becomes zero at $\eta^* \simeq 0.6$, which corresponds to $B^* \sim 80$ mG.

For the sake of clarity and brevity, the results shown above have been obtained in the absence of disorder. Disorder has to be taken into account when we aim to describe high-T_c materials, and this will be the subject of future investigations. Here we just observe that disorder can locally change the penetration length altering the section length $\Delta x/\lambda_J$ and/or permitting larger screening currents in the sample. A small $\Delta x/\lambda_J$ implies that "currentless" (constant phase) states can occur[23,25] without spontaneous currents. These facts, together with the small values of the above threshold fields, imply that it should be not surprising that, also in moderate fields, the state is diamagnetic.[4] Therefore, the presence (or absence) of spontaneous currents would no longer be strictly correlated with paramagnetism. In Ref. 15 paramagnetism actually appears without measurable spontaneous currents in scanning SQUID microscope images. However independently of the presence or absence of spontaneous currents, the above paramagnetic states should be different from the quasiparticle paramagnetism induced by midgap states. Indeed the midgap induced paramagnetism is independent on the system dimension, so it would also appear for very small submicron GB junctions, where π loops likely will not appear. On the other hand, the presence of π loops can be revealed in other ways, using, for example, transport properties of GB junctions.[30]

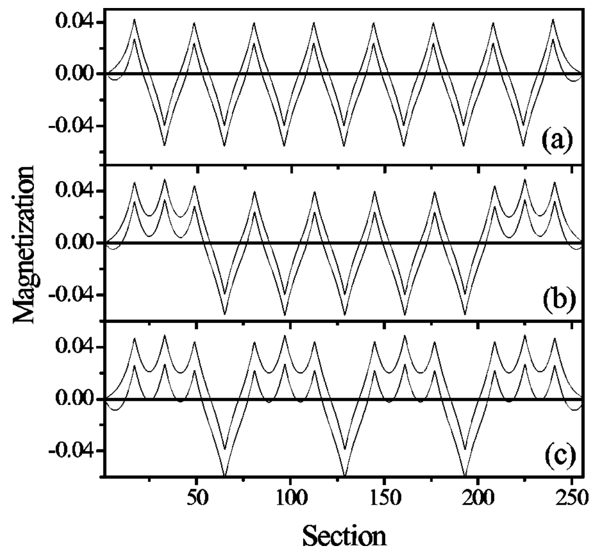

FIG. 4. Simulated magnetization of a $N=255$ Josephson junction 15 π-loop array with $\beta_L=0.04$ and $\alpha=0.25$: (a) Solution with one unpaired paramagnetic half flux quantum; top curve $\eta=0$; bottom curve $\eta=0.1$. (b) Solution with ten paramagnetic half flux quanta; top curve $\eta=0$, bottom curve $\eta=0.5$; (c) Solution with 12 paramagnetic half flux quanta; top curve $\eta=0$ bottom curve $\eta=0.7$.

In conclusion localized π loops in GBs can show both spontaneous magnetization and a paramagnetic behavior. For samples large with respect to the penetration depth, implying a low β for each loop, paramagnetism exists in a relatively narrow region just near the zero field. In the absence of significant field cooling effects the energy difference between diamagnetic and paramagnetic fundamental state solutions can be very small, so the observation of paramagnetism can be difficult or strictly depend on the particular sample. Moreover in high-T_c materials disorder can easily hinder the above picture. It is simpler to probe paramagnetic and diamagnetic states similar to that reported in Fig. 2 for engineered systems of π loops, as recently reported in Ref. 22 for two-dimensional systems.

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[29] We have tested that this ''field cooling at hand'' is equivalent, except for the location of half-fluxons, to take a random distribution of phases and cooling increasing β to its final value for a given *f*. This procedure was used in $2d$ systems where it is not easy to choose an initial phase distribution Ref. 14.
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