Tunneling properties of an orbital antiferromagnet

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Tunneling between two orbital antiferromagnets (systems in which the magnetic ordering is produced by spontaneous currents in the ground state) is studied. It is shown that in contrast to tunneling between ordinary antiferromagnets (in which the magnetization results from spin ordering), where the spin-dependent part of the tunneling current is proportional to the relative orientation of the magnetizations in them, for orbital antiferromagnets the spin-dependent part of the current depends on the relative local orientation of the spontaneous currents. Unfortunately, because a semi-infinite crystal (a crystal bounded by a surface) does not possess a center of inversion, it is impossible to distinguish between the contributions to the tunneling current from spontaneous currents and spin polarization. The lack of a center of inversion in the presence of initial spin ordering results in the induction of spin currents, and the initial spontaneous currents inevitably result in magnetization of the electron spins. These contributions can only be distinguished numerically. © *1995 American Institute of Physics.*

1. INTRODUCTION

Magnetic ordering in solids can result from the ordering of electron spins. For elements with d and f shells, it is sometimes said that the atomic spins are ordered, but the appearance of an atomic moment is nonetheless of an electronic nature. In the case of spin ordering in a crystal, there is a spin density distribution $\mathbf{s}(\mathbf{r})$ and an associated magnetization distribution. In the case of spin ordering, the electron density matrix and the free energy of the system are functionals of the spin density, which can be chosen to be the order parameter. In the presence of spin ordering, there is no microscopic current in a unit cell of the crystal.

At the same time, states of the crystal in which the spin density is identically zero ($\mathbf{s}(\mathbf{r})\equiv 0$) but a current $\mathbf{j}(\mathbf{r})\neq 0$ circulates in the unit cell in the ground state of the crystal (this can be termed an orbital magnetic material) are not forbidden, in principle . The total current averaged over the volume of the crystal is then zero: $\int \mathbf{j}(\mathbf{r})d\mathbf{r}\equiv 0$; otherwise, a macroscopic magnetic moment would arise, and this could not correspond to the ground state of the crystal.[1] The current distribution produces a nonuniform magnetization distribution in the crystal. In contrast to spin ordering, however, the magnetization cannot be chosen as an order parameter in terms of which one can express the density matrix and the free energy of the elctrons

At the same time, the current also cannot be chosen as an order parameter, because the free energy would then depend on the gauge of the vector potential. A different quantity must be chosen as the true order parameter in terms of which the current and magnetization are expressed.

The Possible of the existence of states of a crystal with nonzero current in the ground state was pointed out in Ref. 3 on the basis of a model with electron-hole pairing.[2] The nature of this state was then investigated in detail in a number of papers.[4,5] It was found in these papers that the true order parameter is the toroidal moment $\mathbf{T}(\mathbf{r})$, which is invariant under simultaneous time reversal and coordinate inversion. The microscopic current distribution can be expressed in terms of the toroidal moment $\mathbf{j}(\mathbf{r})=[\nabla[\nabla\mathbf{T}(\mathbf{r})]]$.[5]

Subsequently, states of a crystal carrying current in the ground state were rediscovered on the basis of the two-dimensional Hubbard model (the so-called flux phase).[6,7] In this case, the boson fields at lattice bonds (the latter can be expressed in terms of electron averages) can be chosen to be the order parameter. In the model considered below, there is no toroidal moment.

It would be interesting to know what kind of exerpiments can distinguish magnetic order that results from spin ordering from the case when the magnetization is produced by a nonuniform current distribution with no spin ordering. It turns out that tunneling measurements are such experiments. Our objective in the present paper is to clarify the characteristics of tunneling in a crystal in the flux-phase state.

2. QUALITATIVE ANALYSIS

We first examine qualitatively tunneling in the flux-phase state. For definiteness, we have in mind the flux-phase on a two-dimensional square lattice.[6,7] This phase can be described in the language of effective single-particle states. In the flux-phase, half of the elementary magnetic flux quantum penetrates into each square unit cell. The electron spectrum can be calculated as in the single-particle problem, where the hopping integrals between nearest neighbors in the lattice have phase factors with specially selected phases $\tilde{t}_{ij}=t_{ij}\exp(i\theta_{ij})$, (where t_{ij} is the hopping integral between sites i and j).[8] The flux penetrating a unit cell is given by the phase increment around the cell perimeter:

$$\Phi=\sum_{\langle ij\rangle}\theta_{ij}. \tag{1}$$

The current along the $\langle ij\rangle$ bond is

$$j_{ij}=e\ \mathrm{Im}\{\tilde{t}_{ij}\}. \tag{2}$$

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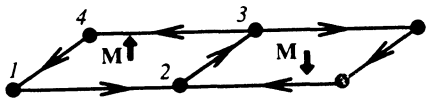

FIG. 1.

With this choice of the phases

$$\tilde{t}_{12}=\tilde{t}_{23}=\tilde{t}^*_{43}=\tilde{t}^*_{14}=t\exp\left(i\frac{\pi}{4}\right), \tag{3}$$

half of the elementary flux penetrates a cell (the increment of the phase ϕ to π in units of $\hbar c/e$), and the currents flowing along opposite sides of the square are equal in magnitude and oppositely directed ($j_{12}=-j_{34}$, $j_{23}=-j_{41}$). The current-induced magnetizations are oppositely directed at the centers of neighboring cells (Fig. 1). There is no spin ordering; the currents for electrons with different spin directions flow identically. This magnetic state cannot be observed in tunneling experiments with an ordinary magnetic needle (in the case of scanning tunneling microscopy) with spin ordering. Indeed, the density matrix for the electrons in a needle with spin magnetic ordering can be represented in the form[9]

$$\hat{\rho}_t=\rho_0\hat{I}+\rho_s(\hat{\sigma}\mathbf{M}_t), \tag{4}$$

where $\rho_{0,s}$ are the spin-independent and the spin-dependent parts of the density of states; $\hat{I}$ and $\hat{\sigma}$ are respectively the unit matrix and the vector of Pauli matrices, and $\mathbf{M}_t$ is the magnetization in the needle (we assume that the needle is single-domain, and $\mathbf{M}_t$ does not depend on the site number).

For a system in the flux-phase the density matrix is diagonal in the spins and is a function of the hopping integrals and their phases

$$\hat{\rho}_{fp}=\rho_{fp}(\theta_{ij})\hat{I}, \tag{5}$$

since $\hat{\rho}$ is the matrix inverse of the effective single-particle Hamiltonian, which is diagonal in the spins

$$\hat{H}_{fp}=\sum_{\langle ij\rangle\sigma}\tilde{t}_{ij}(\theta)c^+_{i\sigma}c_{j\sigma}. \tag{6}$$

The tunneling current is proportional to the trace of the local densities of states

$$I\propto \mathrm{Tr}\{\hat{\rho}_t\hat{\rho}_{fp}\}, \tag{7}$$

where Tr denotes the trace over the spin indices. Because $\hat{\rho}_{fp}$ is diagonal in the spin, the current is insensitive to the orientation of the magnetization in the needle.

In tunneling from a surface with spin ordering, however, the tunneling current would contain a term that depends on the relative orientation of the magnetizations in the needle and at the surface[9]

$$I=I_0+I_s(\mathbf{M}_t\mathbf{M}_c), \tag{8}$$

where $\mathbf{M}_{t,c}$ are the magnetizations in the needle and the crystal, and $I_{0,s}$ are constants which depend on the details of the electronic structure. This term in the current makes it possible, in scanning tunneling microscopy experiments with a magnetic needle, to identify individual magnetic ions on the surface.[10]

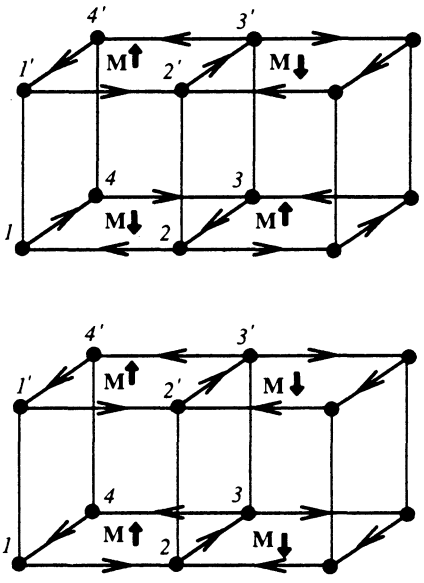

FIG. 2.

In summary, the spin magnetization in the needle is insensitive to a change in the orientation of the magnetic moment in an orbital magnet from one cell to a neighboring cell. In delete a spin antiferromagnet, however, the spin-dependent part of the tunneling current would have different signs in neighboring cells. A change of the sign of the magnetic moment in an orbital antiferromagnet from cell to cell can be observed if tunneling occurs in a crystal in the flux-phase state.

3. TUNNELING BETWEEN ORBITAL ANTIFERROMAGNETS

To describe tunneling between two systems in the flux-phase state, we consider two planar square lattices between which a weak tunneling bond exists. In one case the relative arrangement of the lattices is such that the magnetization in the cells lying below one another is oriented in the same direction, and in the second case it is oriented in opposite directions (Fig. 2; the fine lines represent the weak tunneling bonds). To describe the flux-phase, it is convenient to use the approach of Ref. 7. The flux-phase state can be described on the basis of the Hamiltonian

$$H=\sum_{\langle ij\rangle\sigma}tc^+_{i\sigma}c_{j\sigma}-\frac{J}{2}\sum_{\langle ij\rangle\alpha\beta}c^+_{i\alpha}c_{i\beta}c^+_{j\beta}c_{j\alpha}+\frac{U}{2}\sum_{i\sigma}(c^+_{i\sigma}c_{i\sigma}-1)^2, \tag{9}$$

where the summation extends over nearest neighbors $\langle ij\rangle$, t is a hopping integral, J is the exchange interaction constant at neighboring sites, and U is the intrasite Coulomb repulsion constant. Such a hybrid model gives the Hubbard model in the limit $J\to 0$ and the Heisenberg model in the limits $U\to 0$ and $t\to 0$.

The interaction terms in the Hamiltonian are formally eliminated by a Hubbard–Stratonovich transformation. Completing the square with auxiliary boson fields does not

change the partition function. In so doing, an additional integration over these degrees of freedom arises. This supplementation of the Hamiltonian formally reduces the original problem to one with a quadratic Hamiltonian in the fermion degrees of freedom plus additional couplings for the fermion and boson fields. The transformed Lagrangian corresponding to the Hamiltonian (9) has the form

$$\hat{L}=\sum_{\langle ij\rangle\sigma}\left[\frac{2}{J}|\chi_{ij}|^2+(t+\chi_{ij})c_{i\sigma}^{+}c_{j\sigma}\right]+\sum_{i\sigma}\left[\frac{1}{2U}\phi_i^2-i\phi_i+c_{i\sigma}^{+}\left(\frac{d}{dt}+i\phi_i\right)c_{i\sigma}\right], \tag{10}$$

and the couplings are

$$i\phi_i=U(c_{i\sigma}^{+}c_{i\sigma}-1), \tag{11}$$

$$\chi_{ij}=-\frac{J}{2}c_{i\sigma}^{+}c_{j\sigma}, \tag{12}$$

where ϕ_i and χ_{ij} are the additional boson fields.

In calculating the partition function, the integration over the fermion degrees of freedom can be evaluated exactly and the integration over the boson fields can be performed, as a rule, in the saddle-point approximation. In calculating the different averages in this approximation the fermion part of the problem is effectively a single-particle problem, and the boson fields are actually replaced by their averages at the saddle point.

We are interested in calculating the tunneling current between two weakly coupled systems (L and R), described by the Hamiltonians (9) and located in the flux-phase state.

The tunneling-current operator can be expressed in terms of the tunneling-coupling operator (the Heisenberg representation is used for the operators), and they have the following form:

$$\hat{T}(t)=\sum_{ij\sigma}[T_{LRij}c_{iL\sigma}^{+}(t)c_{jR\sigma}(t)+\text{h.c.}], \tag{13}$$

and

$$\hat{I}(t)=ie\sum_{ij\sigma}[T_{LRij}c_{iL\sigma}^{+}(t)c_{jR\sigma}(t)-\text{h.c.}], \tag{14}$$

where subscripts i and j refer to the L and R systems, respectively, and T_{LRij} is the tunneling matrix element between sites i and j in the L and R systems.

The tunneling current is defined as the average

$$I=\langle\hat{I}(t)\rangle=\int[dc_L][dc_L^{+}][d\chi_L][d\phi_L][d_{cR}][dc_R^{+}]\times[d\chi_R][d\phi_R]\hat{I}(t)\times\exp\left\{-i\int_p[\hat{L}_L(t_1)+\hat{L}_R(t_1)+\hat{T}(t_1)]dt_1\right\}, \tag{15}$$

where the integration (15) extends over a closed time contour.[11,12] For weak tunneling coupling, the argument of the exponential can be expanded to first order in the tunneling-coupling operator. One can then average independently over the L and R states of the systems (the tunneling current is then proportional to T_{LR}^2). We have

$$I=e\sum_{ij\sigma\sigma'}\int dt_1[T_{LRij}\langle c_{iL\sigma}^{+}(t)c_{i'L\sigma'}(t_1)\rangle_L T_{RLj'i'}\times\langle c_{j'R\sigma'}^{+}(t_1)c_{jR\sigma}(t)\rangle_R+\text{h.c.}], \tag{16}$$

where the symbols $\langle\ldots\rangle_{L,R}$ denote averaging over the states of the noninteracting L and R systems,

$$\langle\ldots\rangle_{L,R}=\int[dc_{L,R}][dc_{L,R}^{+}][d\chi_{L,R}][d\phi_{L,R}](\ldots)\times\exp\left\{-i\int_p L_{L,R}(t_1)dt_1\right\}. \tag{17}$$

In what follows, these indices are suppressed. The expression (16) can be put into the form

$$I=e\int_{-\infty}^{\infty}[\hat{T}_{LR}\hat{g}_L^{<}(t-t_1)\hat{T}_{RL}\hat{g}_R^{>}(t_1-t)-\text{h.c.}], \tag{18}$$

where the matrix notation $\hat{T}_{LR}\equiv\{T_{LRij}\}$ has been introduced for convenience, and $\hat{g}^{<,>}$ are the Keldysh Green's functions (GFs) for the fermions. To calculate them, the boson degrees of freedom are replaced by c numbers at the saddle point. By definition, we have

$$\hat{g}_R^{<}(t-t_1)=-\langle c_{j'R\sigma'}^{+}(t_1)c_{jR\sigma}(t)\rangle, \tag{19}$$

$$\hat{g}_R^{>}(t-t_1)=\langle c_{jR\sigma}(t_1)c_{j'R\sigma'}^{+}(t_1)\rangle,$$

and similarly for the L system.

The Keldysh Green's functions can be expressed in terms of retarded Green's functions and the single-particle distribution function

$$\hat{g}_{L,R}^{>,<}(\varepsilon)=\hat{\rho}_{L,R}(\varepsilon)\begin{cases}f_{L,R}(\varepsilon)\\ f_{L,R}(\varepsilon)-1\end{cases}, \tag{20}$$

$$\hat{\rho}_{L,R}(\varepsilon)=-\frac{1}{\pi}\text{Im}\{\hat{g}_{L,R}^{r}(\varepsilon)\}.$$

It is convenient first to find the retarded Green's function in the momentum representation, and then transform to the site representation. The retarded Green's function is the matrix inverse of the effective single-particle Hamiltonian

$$H=\sum_{\langle ij\rangle\sigma}(t+\chi_{ij})c_{i\sigma}^{+}c_{j\sigma}+i\sum_{i\sigma}\phi_i c_{i\sigma}^{+}c_{i\sigma}. \tag{21}$$

The boson fields on the couplings χ_{ij} in the flux-phase are given by the relations (3). The field ϕ shifts the reference energy, which is important for our purposes, so that in what follows such terms will be dropped.

The current along the bonds (for example, 1 and 2), taking into account the equations of motion (12), has the form

$$j_{12}=i\sum_{\sigma}\langle(c_{1\sigma}^{+}c_{2\sigma}-c_{2\sigma}^{+}c_{1\sigma})\rangle=\frac{4}{J}\text{Im}\{\chi_{12}\}=-j_{34},$$

and the Green's function in the momentum representation has the form

$$\hat{g}^{-1,r}(\varepsilon,\mathbf{k})=\begin{pmatrix}\varepsilon+i0 & t(\mathbf{k})\\ t^*(\mathbf{k}) & \varepsilon+i0\end{pmatrix}, \tag{22}$$

where

$$t(\mathbf{k})=2t(\cos k_x+\cos k_y)+\chi_{12}\exp(ik_x)+\chi_{34}\exp(-ik_x)$$
$$+\chi^*_{23}\exp(ik_y)+\chi^*_{41}\exp(-ik_y),$$

and the Green's function in the site representation is obtained via Fourier transformation,

$$\hat{g}^r_{\mathbf{n},\mathbf{n}'}(\varepsilon)=\int\frac{dk_x dk_y}{(2\pi)^2}\exp(i\mathbf{k}(\mathbf{n}-\mathbf{n}'))\hat{g}^r(\varepsilon,\mathbf{k}), \tag{23}$$

where the indices $\mathbf{n}$ and $\mathbf{n}'$ run over the sites of a square lattice.

When nearest neighbors are taken into account in the tunneling between the L and R systems (this is not for our results, but greatly simplifies the discussion), the expression for the tunneling current assumes the form

$$I=e|T_{LR}|^2$$
$$\times\int d\varepsilon\,\mathrm{Tr}\{\hat{\rho}^L_{11}(\varepsilon)\hat{\rho}^R_{1'1'}(\varepsilon)+\ldots+\hat{\rho}^L_{44}(\varepsilon)\hat{\rho}^R_{4'4'}(\varepsilon)$$
$$+\hat{\rho}^L_{12}(\varepsilon)\hat{\rho}^R_{2'1'}(\varepsilon)+\hat{\rho}^L_{23}(\varepsilon)\hat{\rho}^R_{3'2'}(\varepsilon)+\ldots\}$$
$$\times[f_L(\varepsilon)-f_R(\varepsilon)], \tag{24}$$

where $T_{LR}=T_{11'}=\ldots=T_{44'}$ and $f_{L,R}(\varepsilon)$ are the single-particle distribution functions with the chemical potentials shifted by the amount of the applied voltage ($\mu_L-\mu_R=eV$).

All nontrivial information about the structure of the flux-phase is contained in the off-diagonal elements of the density matrix in the tunneling current. Modulating the magnitude of the tunneling current when one system is shifted relative to the other by the lattice constant (Fig. 2) yields the quantity (per unit cell of the square lattice)

$$\Delta I=I_+-I_-=e|T_{LR}|^2\int_{\mu_L}^{\mu_R}d\varepsilon\,d_L(\varepsilon)d_R(\varepsilon)$$
$$\times\mathrm{Re}\{[\chi^L_{12}\chi^{R*}_{1'2'}-\chi^L_{12}\chi^R_{1'2'}]+[\chi^L_{23}\chi^{R*}_{2'3'}-\chi^L_{23}\chi^R_{2'3'}]$$
$$+[\chi^L_{34}\chi^{R*}_{3'4'}-\chi^L_{34}\chi^R_{3'4'}]+[\chi^L_{41}\chi^{R*}_{4'1'}-\chi^L_{41}\chi^R_{4'1'}]\}, \tag{25}$$

where the currents I_+ and I_- correspond to situations when the currents along the neighboring edges (Fig. 2) in the L and R systems flow in the same and opposite directions, respectively. We have introduced the notation

$$d_{L,R}(\varepsilon)=\int\frac{dk_x dk_y}{(2\pi)^2}$$
$$\times 2\,\frac{[\delta(\varepsilon-\mu_{L,R}-E_+(\mathbf{k}))+\delta(\varepsilon-\mu_{L,R}-E_-(\mathbf{k}))]}{[E_+(\mathbf{k})-E_-(\mathbf{k})]}.$$

The fermion spectrum has the form

$$E_\pm(\mathbf{k})=\pm|t(\mathbf{k})|.$$

Therefore, according to Eqs. (25), the modulation of the tunneling current accompanying a displacement of the lattices with respect to one another by the lattice constant has the form

$$\Delta I=I_+-I_-\propto\mathrm{Im}\{\chi^L_{12}\}\mathrm{Im}\{\chi^R_{12}\}\propto j^L_{12}\,j^R_{1'2'}. \tag{26}$$

The structure of the tunneling current can be interpreted as follows. The off-diagonal elements of the density matrix $\rho^L_{\mathbf{n},\mathbf{n}'}$ (similarly for the R system) represent the transition amplitude from site $\mathbf{n}$ to site $\mathbf{n}'$. The off-diagonal matrix elements (for example, $T_{RL}\rho^L_{12}T_{LR}\rho^R_{2'1'}$) describe transitions from a site $\{1L\to2L\to2'R\to1'R\to1L\}$ (Fig. 2). Transitions from sites $\{1'R\to1L\}$ and $\{2L\to2'R\}$ are and depend on the relative phase at sites 1,2 and 1′,2′. This same value of the relative phase at the sites determines the presence and the direction of the current flowing along the bonds. Under displacement by the lattice constant, the relative phase is reversed, and accordinglythe direction of the currents flowing along the bonds located beneath one another changes. Therefore, tunneling electrons feel the relative phase (direction of the currents) between sites in the L and R systems.

4. CONCLUSIONS

Modulation of the tunneling current under a relative displacement by the lattice constant formally results from the off-diagonal elements of the density matrix, which are proportional to the hopping integrals between sites within each system (the latter are present in the single-particle problem). However, such modulation does not occur in the single-particle problem. The single-particle hopping integrals are given by the matrix elements associated with the overlap of the site orbitals

$$t_{ij}\propto\int d\mathbf{r}\phi_i H\phi^*_j.$$

All single-particle overlap integrals have the same phase.[13,14] since all single-particle atomic orbitals ϕ_i at sites have the same phase. The phase ϕ_i should not be confused with the phase of the characteristic Bloch wave function of a single-particle state, which is associated with translations. The change in the hopping integral as a result of many-particle effects (as follows from Eq. (10), χ_{ij} is an additive correction to the single-particle hopping integral) is given by

$$\chi_{ij}=\langle c^+_{i\sigma}c_{j\sigma}\rangle.$$

The choice of phase for different pairs of neighboring sites is not unique, and is fixed by the interaction, so that this choice would correspond to minimum energy (the conditions of an extremum with respect to χ_{ij} at the saddle point). The phases χ_{ij} become "frozen in", and there is no latitude in choosing them. The interaction (in the language of the effective single-particle states) results in additional entanglement of the single-particle Bloch wave functions with specially chosen phases inside the cells.

Therefore, in tunneling between two orbital antiferromagnets, the tunneling current is sensitive to the relative di-

rection of the spontaneous currents in the L and R systems and not to the relative orientation of the magnetizations, as would be the case for spin antiferromagnets.

In a pure spin magnetic ordering in the needle and the crystal, the tunneling current contains a component proportional to the scalar product of the magnetizations. As shown in Ref. 15, however, because a semi-infinite crystal (bounded by a surface) does not possess a center of inversion, the magnetic ordering and exchange magnetization of the electron spins result, to the extent of the spin–orbit interaction, in the appearance of a toroidal moment and a spontaneous current related to it as $\mathbf{j}=[\nabla[\nabla\mathbf{T}]]$. This fact will inevitably lead to the appearance of terms in the tunneling current proportional to the local product of the currents. This makes it impossible to distinguish the pure current contribution from the pure spin contribution to the tunneling current, and the relative magnitude of these contributions differs only numerically. The impossibility of making such a separation is of fundamental significance. Magnetic spin ordering generates a magnetic field that influences not only the spin of an electron but also its orbital motion. In addition, in contrast to the effect on the spectrum via the spins and the spin–orbit interaction, the orbital contribution is not relativistically small. Correctly taking into accountthe effect of a magnetic field on the orbital motion of Bloch electrons (electrons in the periodic potential of the lattice) leads to the problem of Ref. 8, where the electron spectrum consists of an infinite number of energy bands (the number of bands is finite only for special values of the magnetic field). Taking the effect of the magnetic field on the orbital motion into account should also result in the appearance of spontaneous currents. The question of the symmetry of such currents is still not completely clear. However, the answer for free electrons is known (the periodic potential is equal to zero). In this limit, in the absence of a center of inversion, the magnetic field introduces an asymmetry in the spectrum for a wave vector perpendicular to the magnetic field.[16] The appearance of the toroidal moment is associated with the asymmetry of the electron spectrum. In addition, the asymmetry of the spectrum in a magnetic field is a general property of surface groups.[17] For a nonuniform distribution of the magnetic field (for example, for antiferromagnetic ordering), the spontaneous currents will be nonuniform on the scales of a cell.

For orbital spontaneous currents in a cell in the flux-phase state, these currents will inevitably result in magnetization of the electron spins and a spin-dependent contribution to the tunneling current. Even in this case, therefore, it is impossible to distinguish the "spin" and "current" contributions to the tunneling current.

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