Superconducting and Ferromagnetic Phases in SrTiO_{3}/LaAlO_{3} Oxide Interface Structures: Possibility of Finite Momentum Pairing

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Department of Physics, Massachusetts Institute of Technology,
77 Massachusetts Avenue, Cambridge, MA 02139

We introduce a model to explain the observed ferromagnetism and superconductivity in LAO/STO oxide interface structures. Due to the polar catastrophe mechanism, 1/2 charge per unit cell is transferred to the interface layer. We argue that this charge localizes and orders ferromagnetically via exchange with the conduction electrons. Ordinarily this ferromagnetism would destroy superconductivity, but due to strong spin-orbit coupling near the interface, the magnetism and superconductivity can coexist by forming an FFLO-type condensate of Cooper pairs at finite momentum, which is surprisingly robust in the presence of strong disorder.

Introduction – It is known that a conducting electronic state can form at the interface between two insulating oxides[1]. A particularly well studied example is the TiO_2 interface between $SrTiO_3$ and $LaAlO_3$. The carrier density can be controlled by a backgate on the $SrTiO_3$ side and superconductivity (SC) has been discovered over a range of densities with maximum T_c of about 0.3 K[2]. Recently, signs of ferromagnetism (FM) have also been reported[3–7]. In particular, Li *et al.*[5] showed that SC and FM coexist in the same sample and that the FM moment is large, $\approx 0.3 - 0.4\mu_B$ per interface unit cell. Assuming that the FM and SC arise from the interface, these observations raise the question of whether the SC has to be unconventional in order to coexist with FM. Before addressing this question we need to understand first the nature of the electronic state at the interface and up to now no clear picture has emerged[8, 9, 11]. Are most of the electrons localized or extended? Does the FM come from local moments or the mobile electrons and what is its origin? In this paper we propose a model for the interface electrons which is consistent with existing transport data. Based on this model we explain the existence of FM and the coexistence of SC and FM. For the latter, the key idea is that a large Rashba-type spin-orbit coupling exists at the interface [12]. Such a Rashba term is particularly favorable for the formation of a condensate at finite momentum, called a Fulde-Ferrell-Larkin-Ovchinikov (FFLO) state which coexists with FM[13, 14]. This general idea was pointed out earlier by Barzykin and Gorkov[15]. However, they considered only the clean case and their solution is quickly destroyed by disorder. Surprisingly, with increasing disorder the FFLO state is revived[16]. We suggest that the SC observed at the interface is described by this disordered stabilized helical FFLO state. This state is sometimes referred to as a "helical" FFLO state[16] since, pairing occurs at a single momentum $\mathbf{q}$ so that $\Delta(\mathbf{r}) = \Delta e^{i\mathbf{q}\cdot\mathbf{r}}$ unlike the usual FFLO state where pairing occurs at both $\pm\mathbf{q}$ so that $\Delta(\mathbf{r}) = \Delta \cos\mathbf{q}\cdot\mathbf{r}$.

The Model – As shown in Fig. 1, $LaAlO_3$ consists of layers with alternating charged, while $SrTiO_3$ has
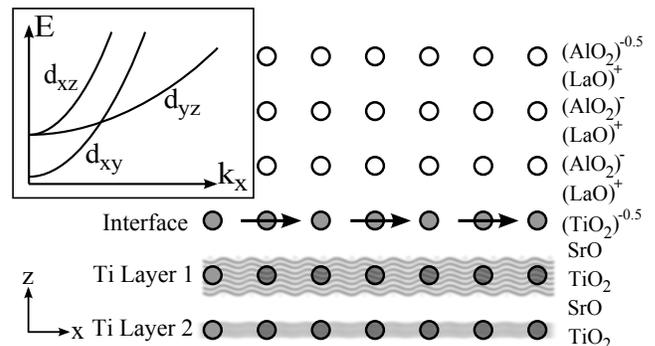

FIG. 1. Schematic depiction of the $SrTiO_3/LaAlO_3$ oxide interface structure. Filled and empty circles depict Ti and Al ions. The half electron charge per unit cell is transferred to the interface TiO_2 layer localizes and orders magnetically (shown as arrows on the interface layer) via exchange polarization of conduction electrons on the subsequent Ti layers (shown as wavy cloud in Ti Layers 1 and 2). Inset: Dispersion of electron bands arising from 3d orbitals on Ti layers near the interface.

charged-neutral layers. As a result of the charge discontinuity at the interface, an electric potential proportional to the number of $LaAlO_3$ layers is built up. This phenomenon is termed the polar catastrophe. Since the Ti ions allow for mixed valence charge compensation, to avoid the polar catastrophe, half an electron per unit cell is transferred from the surface AlO_2 layer to the TiO_2 across the interface. The electrons are expected to occupy the d_{xy} orbital on the Ti atoms. Due to the relatively narrow bandwidth, an on-site repulsion U and a nearest-neighbor Coulomb repulsion V will cause these electrons to be localized on every other interface site. This picture of local moment formation at the interface has been proposed before [9]. Super-exchange via the oxygen is expected to provide a weak antiferromagnetic exchange.

The application of a back gate or the presence of defects forces more electrons to the interface. We assume that the effect of U and V makes it too costly to place these electrons at the interface layer. Instead, the additional electrons mainly occupy the Ti layer next to it

(called layer 1), and their wavefunctions spill over to layers 2 and 3 as well. We model these electrons as occupying two dimensional conducting bands. These are the electrons seen in transport measurements, with a typical areal density of $3 \times 10^{13} \text{cm}^{-2}$, or about 10% of the density compared with the localized electrons. These electrons initially go into the d_{xy} band [10]. Hall effect measurements show nonlinearity in magnetic field H with increasing gate voltage, which has been interpreted as the appearance of a second carrier with lower mobility[10, 17, 18]. We assume that these are the d_{xz} and d_{yz} bands, illustrated in the inset of Fig. 1. These bands are highly anisotropic with a heavy mass in one direction which may be responsible for the lower mobility. The d_{xz} and d_{yz} bands are higher in energy because their lobes point towards the negative charge at the interface and because their bandwidths are narrower.

In addition, transport measurements show that the elastic scattering rate $1/\tau$ drops rapidly with increasing carrier density [18]. Furthermore, from the analysis of magnetoresistance, it was found that a Rashba term $H_{\mathrm{R}} = \alpha \hat{z} \cdot (\boldsymbol{\sigma} \times \boldsymbol{k})$ grows rapidly with the gate voltage [12], reaching a spin splitting $\Delta_{so} = 2\alpha k_F$ of 10 meV near the peak of the superconducting dome, a value comparable to the Fermi energy of ≈ 40 meV. Since a back gate voltage tends to pull carriers from the interface, the decrease of $1/\tau$ is reasonable but the increase of Δ_{so} is counterintuitive. We believe this trend is a consequence of increasing admixture of the d_{xz} and d_{yz} bands with increasing carrier density. The Rashba energy is determined by the polarization of the electron wave-function due to the asymmetric environment at the interface and the contributions come mainly from near the atomic core, where the electron is subject to a large electric field $-\partial V/\partial z$:

$$\Delta_{so} \propto |k_{\parallel}| \frac{2}{c^2} \int d\boldsymbol{r} \frac{dV}{dz} |\psi_{\boldsymbol{k}_{\parallel}}(\boldsymbol{r})|^2 \qquad (1)$$

where the wavefunction $\psi_{\boldsymbol{k}_{\parallel}}(\boldsymbol{r}) = \sum_{\ell} a_{\ell} \phi_{\boldsymbol{k}_{\parallel}}^{\ell}(\boldsymbol{r})$ and ℓ denote various angular momenta which are admixed due to the asymmetric environment of the interface [19]. Let us restrict ourselves to admixtures between d, p and s states. Since $\nabla V \propto \hat{z}$, non-vanishing contributions involving the d bands in Eq.(1) come only from the cross term between d_{xz} and p_x and between d_{yz} and p_y. Furthermore, the d_{xy} band can have a nonzero Δ_{so} only via the admixture of either s and p or, more importantly of d_{xz} with p_y and d_{yz} with p_x. We expect that the latter hybridization between d and p orbitals gives rise to Δ_{so} that increases with chemical potential.

The final ingredient of our model is the exchange coupling between the local moments and the conduction electrons. We write the standard phenomenological form

$$H_J = J_K \sum_i \int d\boldsymbol{r} \, \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{s}}(\boldsymbol{r}) \delta(\boldsymbol{R}_i - \boldsymbol{r}) \qquad (2)$$

where $\hat{\boldsymbol{s}}(r) = \frac{1}{2} \psi_{\alpha}^{\dagger} \boldsymbol{\sigma}_{\alpha\beta} \psi_{\beta}(r)$ is the electron spin density operator in the d_{xy} band, and $\hat{\boldsymbol{S}}_i$ is the local spin operator on site i. We introduce a similar coupling J_K' for the d_{xz} and d_{yz} bands. It is useful to introduce $J_0 = J_K/n_0$ where n_0 is the inverse of the interface unit cell area, and similarly $J_0' = J_K'/n_0$. The Schrieffer-Wolff expression is $J_0 = 2\tilde{t}^2(\frac{1}{U+\varepsilon_d} - \frac{1}{\varepsilon_d})$ where $\tilde{t}$ is the hybridization between the local moment and the conduction band orbital and $\varepsilon_d < 0$ is the orbital energy of the local moment relative to the chemical potential. We note that the d_{xz} and d_{yz} orbitals in layer 1 are orthogonal to the localized d_{xy} orbital in the interface layer, so that the hybridization $\tilde{t}'$ vanishes except for the admixture of other orbitals in the d_{xz} and d_{yz} bands. We therefore expect $J_0' \ll J_0$.

The Origin of Ferromagnetism – We note that the present problem is the opposite limit to the familiar problem of dilute Kondo impurities, where the Kondo screening of the local moments competes with RKKY interactions between them. Here the density of local moments $n_i = \frac{1}{2} n_0$ is much greater than the conduction electron density n_c, i.e., the separation between local moments $1/\sqrt{n_i}$ is much smaller than k_F^{-1}. We can still view the local moments as interacting via RKKY interactions, but this interaction will be ferromagnetic and with a relatively long range of $(2k_F)^{-1}$. The FM ordering temperature T_F can be worked out [20] and apart from a numerical constant, the result was shown to be equivalent to a mean field treatment of H_J which we shall adopt below. In this picture, which was introduced by Zener [21] and referred to as the Zener kinetic exchange mechanism, the local moments order by polarizing the conduction electrons. This mechanism has been applied successfully to explain the FM of Mn substitution in GaAs and we borrow the results here [22]. We introduce the average localized spin order per site $\boldsymbol{\mathcal{S}} = \frac{1}{N_i} \sum_i \langle \hat{\boldsymbol{S}}_i \rangle$ and the average electron density $\mathbf{s} = \frac{1}{\text{vol}} \int d\boldsymbol{r} \langle \hat{\boldsymbol{s}}(\boldsymbol{r}) \rangle$. To quadratic order the total free energy density takes the form

$$E_{\text{tot}} = \frac{1}{2} \frac{|\mu_0 \boldsymbol{\mathcal{S}}|^2}{\chi_0} n_i + \frac{1}{2} \frac{|\mu_0 \mathbf{s}|^2}{\chi_c} + J_0 \frac{n_i}{n_0} \boldsymbol{\mathcal{S}} \cdot \mathbf{s}. \qquad (3)$$

The last term is the mean field decoupling of Eq.(2). In the first term $\chi_0 = \frac{1}{3} \frac{\mu_0^2 S(S+1)}{T+\theta}$ where $\mu_0 = g\mu_B$ and $S = 1/2$, $g = 2$ in what follows, $\theta > 0$ is the Weiss term due to the weak AF super-exchange exchange which we shall ignore below. In the second term $\chi_c = \frac{1}{4} \mu_0^2 \nu(0)$ where $\nu(0) = m^*/\pi\hbar^2$ is the density of states including spin of a free electron gas. (The presence of a Rashba term does not change the spin susceptibility of a free electron gas [15, 23].) Minimizing Eq.(3) with respect to $\boldsymbol{\mathcal{S}}$ leads to a purely quadratic term in $|\boldsymbol{\mathcal{S}}|^2$, and the sign change of its coefficient determines the FM transition temperature

$$k_B T_F = \frac{S(S+1)}{12} J_0^2 \frac{n_i}{n_0} \frac{\nu(0)}{n_0}. \qquad (4)$$

We find that $\nu(0)/n_0 = 0.64(m_{xy}^*/m_{\mathrm{e}})$ eV^{-1}. For

$m_{xy}^*/m = 0.7$, $J_0 = 1.3$ eV will give the observed $T_F \approx 300$ K. By comparison, for Mn/GaAs, J_0 is ≈ 1 eV. Here $\tilde{t}$ is smaller, but $|\varepsilon_d|$ is also smaller because the same orbital is involved in the local moment and the conduction electron, so the estimated J_0 appears reasonable. Thus we conclude that the Zener kinetic exchange mechanism can account for a robust FM state.

Next we estimate the polarization of the conduction electron. In the mean field theory, the effect of $\mathcal{S}$ on the conduction electrons is described by an effective Zeeman field $H_J \approx \int d\mathbf{r}\, \mu_0 \mathbf{H}_{\mathrm{MF}} \cdot \mathbf{s}(\mathbf{r})$ where $\mathbf{H}_{\mathrm{MF}} = J_0 \frac{n_i}{n_0} \mathcal{S}$. Li et al.[5] reported an ordered moment of $= 0.3\mu_B$ per interface unit cell, i.e., $0.6\mu_B$ per local moment in our picture, which implies $|\mathcal{S}| = 0.3$. Using $J_0 = 1.3$ eV, we estimate a Zeeman spin splitting $|\mu_0 \mathbf{H}_{\mathrm{MF}}| \approx 200$ meV, which is comparable to or exceeds the Fermi energy. Thus the d_{xy} band is largely spin polarized. Since $J_0' \ll J_0$, we expect a smaller (but still significant) polarization of the d_{xz} and d_{yz} bands.

Nature of the Superconducting State – We assume that the superconductivity originates from a conventional electron-phonon coupling mechanism, which is modeled by an attractive short range interaction g with a cut-off given by the Debye frequency ω_D. Since the superconducting transition temperature is $\mathrm{T}_c \approx 0.3$ K, the pairing gap ≈ 0.04meV is the lowest energy scale in the problem. In the d_{xy} band we estimate a Zeeman splitting of 0.2 eV, which exceeds the Pauli limit by more than 3 orders of magnitude, and precludes the possibility of pairing in the d_{xy} band. The d_{xz} and d_{yz} bands will also be partially polarized due to the exchange interaction. However, the exchange splitting, $\mu_0 B$, in these bands is expected to be much smaller (although likely still $\mu_0 B \gg \Delta$). Moreover, as we argue above, we expect that the Rashba spin-orbit coupling, Δ_{so}, is even larger in the d_{xz} and d_{yz} bands than that observed in the d_{xy} bands. It is natural to look to strong spin-orbit coupling to preserve pairing in the d_{xz} and d_{yz} bands despite large Zeeman splitting.

The enhancement of B_c due to Rashba spin-orbit coupling was first demonstrated by [15] the case of weak Rashba coupling ($\Delta_{so} \ll \varepsilon_F$) and no disorder, and later by [16] for the case of weak Rashba coupling and moderate disorder. They showed that an FFLO state is favored, where the pairing occurs with a finite center of mass momentum[24]. Here we extend their analysis to treat arbitrarily strong values of Δ_{so} and disorder. We begin by neglecting disorder and find the susceptibility to form Cooper pairs at finite pair momentum $\mathbf{q} = q\hat{y}$. The dispersion for the $\pm$ Rashba branches is: $\varepsilon_{\mathbf{k}+\mathbf{q}/2}^{\pm} = \frac{(\mathbf{k}+\mathbf{q}/2)^2}{2m} - \mu \pm \alpha\sqrt{k_x^2 + (k_y + q/2 + \mu_0 B/\alpha)^2}$.[25] In the physically relevant limit: $v_F q, B \ll \Delta_{so}$, and we can expand in q and B:

$$\varepsilon_{\mathbf{k}+\mathbf{q}/2}^{\pm} \approx \varepsilon_k^{\pm}(B=0) + (v_F q/2 \pm \mu_0 B)\sin\phi_k + \mathcal{O}(v_F q^2, B^2) \tag{5}$$

where $v_F = \sqrt{\alpha^2 + 2\mu/m}$ is the Fermi-velocity for the

Rashba bands and $\phi_k = \tan^{-1}(\frac{k_y}{k_x})$. The key is that choosing $q = \frac{2\mu_0 B}{v_F}$[26] ensures $\varepsilon_{\mathbf{k}+\mathbf{q}/2}^{-} = \varepsilon_{-\mathbf{k}+\mathbf{q}/2}^{-} + \mathcal{O}(\frac{B^3}{\Delta_{so}^2})$ for *all* angles ϕ_k. This should be contrasted with the usual FFLO case without spin-orbit coupling, where the linear terms cannot be cancelled for all angles for any choice of q. However, we cannot prevent an energy mismatch in both bands simultaneously. By choosing $\mathbf{q} = 2\mu_0 B\hat{y}/v_F$ we optimize for the ε_- branch, which has a larger density of states, ν_-, and find:

$$\mu_0 B_c \approx \Delta_0 \left(\Delta_{so}/\Delta_0\right)^{\frac{1+\alpha/v_F}{2+\alpha/v_F}} \tag{6}$$

where $\Delta_0 = \omega_D \exp\left[\frac{-1}{(\nu_+ + \nu_-)g}\right]$ is the superconducting gap in the absence of the Zeeman field $\mu_0 B$. In the limit $\alpha \ll v_F$ we recover the results of Barzykin and Gor'kov: $\mu_0 B_c \approx \Delta_0\sqrt{\Delta_{so}/\Delta_0}$. For the oxide interface system we expect stronger spin-orbit coupling, $\Delta_{so} \approx \varepsilon_F$, and find an even larger enhancement: $\mu_0 B_c \approx \Delta_0(\Delta_{so}/\Delta_0)^{2/3}$.

The above calculation is only valid in very clean systems for which $\Delta_0 \tau \gg 1$. In practice, we expect to be in the dirty limit, $\Delta_0 \tau \ll 1$. To incorporate impurity scattering, we consider spin-less, short-ranged impurities and compute the disorder averaged Cooper-channel susceptibility in the $\varepsilon_F \tau \gg 1$ limit, by summing the ladder diagrams for impurity scattering (called the Cooperon). As shown in Fig. 2, there are 3 regimes. First, in the weak disorder regime ($\tau^{-1} < \Delta_0$) we find that the critical field drops rapidly to the Pauli limit $B_c \approx \Delta_0(\Delta_0\tau)^{\frac{1+\alpha/v_F}{1-\alpha/v_F}}$. This can be understood as follows: as in the clean case, the pair momentum minimizes the effect of the magnetic field in the ε_- branch, while keeping the pair breaking in the ε_+ branch. However, impurities can scatter Cooper pairs from the $-$ band to the $+$ band where they rapidly decohere. Thus, disorder enhances the dephasing effects of the Zeeman field, which becomes fully pair breaking as $\tau^{-1} \to \Delta_0$. On the other hand, for very strong disorder, $\tau^{-1} \gg \Delta_{so}$, the Rashba bands $\varepsilon^{\pm}$ lose their identifies due to the rapid impurity scattering. Here, spin and momentum become decoupled and the problem reduces to that of conventional parabolic bands with effective spin-orbit scattering rate $\tilde{\tau}_{so}^{-1} = \Delta_{so}^2\tau \ll \tau^{-1}$. This is the D'yakonov-Perel limit where the spin diffuses in small steps between rapid impurity scattering[27]. In this limit, it was demonstrated in [28] that $\mu_0 B_c \approx \Delta_0/\sqrt{\Delta_0\tilde{\tau}_{so}}$, and SC occurs at $q = 0$.

The interesting limit is the intermediate regime $\Delta_0 \ll \tau^{-1} \ll \Delta_{so}$[16]. Here the disorder is weak enough that the Rashba bands maintain their identity, but a pair in the ε^+ band can readily be scattered to a pair in the ε^- band. Unlike the weak disorder case, the pairing is strongly admixed and it is not possible to sacrifice the pair breaking of one band in favor of the other. On the other hand, due to the spin-orbit coupling both spin (the Zeeman energy) and momentum show diffusive behavior. As a consequence, the pair breaking effect of the magnetic
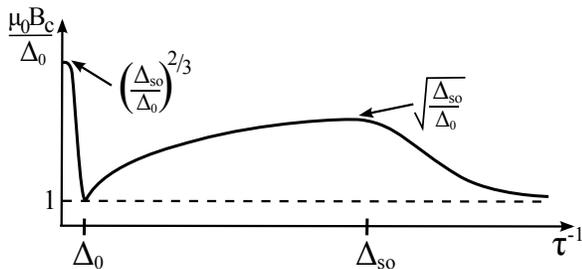
FIG. 2. Critical Zeeman splitting, B_c, measured with respect to the bare SC gap Δ_0 as a function of disorder strength τ^{-1}. The FFLO state identified in [15] is rapidly destroyed as $\Delta_0\tau \to 1$. Remarkably, for stronger disorder the FFLO state re-emerges and B_c is enhanced beyond the Pauli limit (shown as a dashed line).

field is weaker. The pairing interaction is dominated by the Cooperon, $\mathcal{C}$, which ordinarily develops a diffusion pole $\mathcal{C} = \frac{1}{i\omega + Dq^2}$ with $D = \frac{v_F^2\tau}{2}$, but in the present case q is replaced by $q \pm \frac{2\mu_0 B}{v_F}$ for the $\pm$ bands respectively. Due to the strong mixing of the pairing channels, the effective pair breaking strength is given by the combination: $\sum_{\lambda=\pm} \frac{\nu_\lambda}{4m} \left(v_F q + \lambda 2\mu_0 B\right)^2$. Since $\nu_+ \neq \nu_-$ this combination is minimized by finite momentum $q = \frac{4\alpha B}{\alpha^2 + v_F^2}$, and we predict that an FFLO state exists in the intermediate regime, but with a different q from the clean case. The corresponding B_c is: $\mu_0 B_c \approx \frac{\Delta_0}{\sqrt{\Delta_0\tau}}$. Physically, on this limit $\tau_B \ll \tau$ and, unlike the weak disorder, the pair dephasing time τ_B^2/τ grows with increasing disorder.

Discussion – To summarize, we propose a model to explain the coexistence of SC and FM observed in STO/ALO oxide interface structures. In this model, a half-charge per unit cell is transferred to the interface layer and forms a lattice of local moments due to Coulomb repulsion. These local moments then order ferromagnetically via exchange with lower-density bands of mobile electrons residing in Ti layers near the interface. The large FM exchange would ordinarily kill SC in these mobile bands. However, the presence of a large spin-orbit coupling enables the formation of an FFLO state which can coexist with strong magnetism. In this FFLO state, Cooper pairs form with finite pair momentum perpendicular to the direction of magnetic ordering. Unlike the usual FFLO state without spin-orbit coupling [13, 14], spin-orbit coupling parametrically enhances B_c beyond the Pauli limit and enables FFLO pairing to survive to much stronger disorder (up to $\Delta_{so}\tau \gtrsim 1$). Experimentally, τ^{-1} decreases with back-gate voltage V_G while simultaneously Δ_{so} increases. In our picture, the observed T_c vs V_G dome is related to the behavior of B_c shown in Fig. 2 sweeping from $\tau^{-1} > \Delta_{so}$ to $\tau^{-1} < \Delta_{so}$.

We wish to emphasize that our model suggests that the FM and SC occur in different bands, and are uniformly distributed at the interface. Another possible explanation of the coexistence is in the spatial separation of the two phases. Indeed, a recent experiment [6] shows that the FM is arrange in domains and might not occupy the entire interface layer. However, the large total magnetic moment of the system [5], implies that the domains should occupy most of the area at the interface. The SC order parameter, in contrast, does not go to zero at any point in the plane.

Before concluding, we briefly discuss some experimental signatures of our model. First, if the exchange coupling for the d_{xz} and d_{yz} bands is such that the conduction electrons polarize in the opposite direction of the interface moments, then the external in-plane field $H_\parallel$ would align the local moments along $H_\parallel$ but would reduce the total Zeeman field seen by the d_{xz} and d_{yz} bands to $\mu_0(B - H_\parallel)$. This leads to the unusual prediction that T_c should exhibit a maximum at finite $H_\parallel$. Second, one can look for $\mathbf{q} \neq 0$ pairing by creating a Josephson tunneling junction with a conventional SC film. By applying a magnetic field parallel to the junction, Cooper pairs tunnel at finite momentum $\Delta\mathbf{k}$, and the Josephson current would peak when $\Delta\mathbf{k} = \mathbf{q}$.

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[26] Here and throughout we have assumed $\alpha > 0$, for $\alpha < 0$ one should replace $\mathbf{q} \to -\mathbf{q}$.

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