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Nonuniform superconducting phases in a layered ferromagnetic superconductor

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Abstract. – We determine the phase diagram of the ferromagnetic (F)/superconducting (S) atomic superlattice with arbitrary coupling between the layers. The exchange field acting on F layers may induce different kinds of modulated superconducting phases with either in-plane modulation or a phase shift between adjacent S layers. When the exchange field is increased, the re-entrance of the superconductivity emerges and the "π phase" where the superconducting order parameter changes its sign in successive S layers is realized.

The singlet superconductivity and ferromagnetic order cannot coexist in bulk samples due to their antagonistic characters. However, the ferromagnetic order (with the Curie temperature Θ) is transformed into a spiral or domainlike structure in the presence of superconductivity (with the critical temperature T_c) when $\Theta \ll T_c$ [1]. So, the recent discovery of the ruthenocuprates $RuSr_2RCu_2O_8$ (with R = Gd, Eu, Y), which exhibit some kind of ferromagnetic order below $\Theta \simeq 130$ K and superconductivity below $T_c \simeq 30$–45 K [2], was rather surprising. The structure of these compounds comprises RuO_2 monolayers, where the magnetism is attributed to Ru ions and CuO_2 bilayers, where the singlet superconductivity is expected to settle as in the cuprates of the high-T_c superconductors' family. At the moment, it is not clear whether the ferromagnetic component in RuO_2 layers is due to the canting of the antiferromagnetically aligned magnetic moments located at Ru sites or the uncompensated magnetic moments of two antiferromagnetic subsystems of Ru^{4+} and Ru^{5+} ions. Whatever its origin, this small ferromagnetic component is expected to lie in the layers and should interact with the delocalized electrons.

Thus the electronic properties of the ruthenocuprates seem to be well described by the model of ferromagnetic (F)/superconducting (S) atomic multilayer first considered by Andreev *et al.* [3]. These authors use a microscopic theory to describe a system of thin (atomic-scale) F and S layers, both metallic. In S layers, the electrons feel an isotropic (s-wave) attractive interaction of amplitude Λ [4], which would result in the critical temperature T_{c0} for individual layers; in F layers, they feel the exchange field h, t is the transfer integral describing the coupling between the layers in the tight-binding model. The electronic anisotropy is characterized by the ratio $t/\varepsilon_{\mathrm{F}} \ll 1$, where ε_{F} is the Fermi energy. Their main result is the study of the (T, h)-phase diagram when layers are in the regime of Josephson coupling: $t \ll T_{\mathrm{c}0} \ll \varepsilon_{\mathrm{F}}$, or equivalently $\xi_{\mathrm{c}} \ll d \ll \xi_{ab}$, d is the interlayer distance, $\xi_{\mathrm{c}} \sim dt/T_{\mathrm{c}0}$ is the coherence length in the direction perpendicular to the layers, ξ_{ab} is the in-plane coherence length. They found that i) the critical temperature T_{c} of the superlattice is sligthly decreased at low fields owing to the small proximity effect of F layers on S layers, ii) T_{c} returns to $T_{\mathrm{c}0}$ at large fields because the exchange field in F layers strongly decouples S layers, and iii) the transition from "0 phase" to "π phase" takes place at the critical field $h_{\mathrm{c}}(T)$ ($h_{\mathrm{c}}(0) = 0.87\ T_{\mathrm{c}0}$, $h_{\mathrm{c}}(T_{\mathrm{c}}) = 3.77\ T_{\mathrm{c}0}$). While in the former state the order parameter is the same in all layers, in the latter state it has an alternating sign in successive S layers which corresponds to π shift in the superconducting phase. Further theoretical studies later demonstrated that such a transition is proved to manifest through nonmonotonic behaviour of the critical current [5] and Josephson penetration depth [6]. The spontaneous vortex phase existence detected in some experiments on $\mathrm{RuSr_2GdCu_2O_8}$ [7] was attributed to the possibility of (or proximity to) a "0 phase" to "π phase" transition with the estimated parameter h = 10–20 K and $T_{\mathrm{c}0} \approx T_{\mathrm{c}}$ [6].

It is of interest to compare the "π phase" in the F/S multilayer with the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase which appears in the presence of a uniform exchange field in bulk superconductors [8, 9]. In singlet superconductors, the Zeeman splitting due to such a field is unfavourable to the Cooper-pair formation. Indeed, in the presence of the exchange field h electrons with opposite spins and momenta $\pm\boldsymbol{k}$ acquire different energies $\varepsilon(\boldsymbol{k}) \pm h$, where $\varepsilon(\boldsymbol{k})$ is the kinetic energy, whereas the Cooper instability would take place for electrons with equal energies. Meanwhile, the instability may take place for electrons with nonopposite momenta $\pm\boldsymbol{k} + \boldsymbol{q}/2$ having nearly equal energies $\varepsilon(\boldsymbol{k} + \boldsymbol{q}/2) - h \approx \varepsilon(-\boldsymbol{k} + \boldsymbol{q}/2) + h$ provided that $q \approx 2h/v_{\mathrm{F}}$, where v_{F} is the Fermi velocity. This effect would result in the formation of Cooper pairs with finite momentum $\boldsymbol{q}$, corresponding to the nonuniform FFLO phase [8, 9]. Such a phase would appear at low temperatures $T < 0.56T_{\mathrm{c}}$; the region of its existence is between the normal (N) state and the uniform superconducting state, that is for exchange fields of the same order as T_{c} [10]. Thus, the period of the modulation $(2\pi/q)$ would be comparable to the superconducting coherence length $\xi_0 = v_{\mathrm{F}}/2\pi T_{\mathrm{c}}$. It is very difficult to obtain this phase in real compounds and only a few hints for its existence have been received in the experiment [11]. The reasons are that it is very sensitive to the impurities, and the orbital effect which provides another mechanism for the Cooper-pair destruction is usually much stronger. In the previoulsy described F/S multilayer, the "π phase" can also be interpreted as the FFLO state with the modulation perpendicular to the layers, but its period is now determined by the crystal structure rather than ξ_{c} which is too small. In addition, with the microsopic separation of F and S order, this provides a more robust condition for such FFLO state existence.

One may wonder what occurs in the case of intermediate coupling between the layers when $T_{\mathrm{c}0} \lesssim t \ll \varepsilon_{\mathrm{F}}$ (or $d \lesssim \xi_{\mathrm{c}} \ll \xi_{ab}$). New properties in the S/F superlattice are expected to be induced. i) It was already known that in the absence of exchange field in F layers ($h = 0$), $T_{\mathrm{c}}(t)$ decreases fast [12]. Indeed, the highly coupled system feels the effective coupling constant $\Lambda_{\mathrm{eff}} = \Lambda/2$, which strongly suppresses superconductivity in the Bardeen-Cooper-Schrieffer theory. ii) At $t \gg T_{\mathrm{c}0}$, the phase diagram is expected to exhibit similar features to that of two-dimensional (2D) superconductors with the effective exchange field $h_{\mathrm{eff}} = h/2$ and $\Lambda_{\mathrm{eff}} = \Lambda/2$. That is, the tricritical point (h^*, T^*) below which the FFLO state appears should obey $T^* \approx 0.56T_{\mathrm{c}}(h = 0)$ and $h^*/4\pi T^* \approx 0.304$ [10, 13]. In this case, the FFLO state presents the in-plane modulation of the order parameter which could not appear when $t \ll T_{\mathrm{c}0}$. iii) Yet, the argument on magnetic-field–induced decoupling of S layers still holds, and we also expect

T_c to return to T_{c0} at large fields, thus showing a strong re-entrance of the superconducting phase. Moreover, as $d \lesssim \xi_c$, the out-of-plane FFLO modulation is no more restricted to "0 phase" and " π phase" only, while the effective exchange field acting on S layers becomes too small again to induce the in-plane modulation. From now on we will call incommensurate (IC) phase the modulated phase between the "0 phase" and "π phase" in order to distinguish it from the in-plane FFLO phase. Its name is chosen in analogy with the terminology of phase transitions in ferroelectrics. Let us notice that some hint for the FFLO state appearance in the ruthenocuprate was given in ref. [14] on the basis of a band structure calculation, but the effective field acting in S layers was obtained assuming a ferromagnetic alignement of the Ru ions, whereas later it proved to be predominantly antiferromagnetic [15].

Below, we determine the phase diagram which reveals all these features. We show that the different kinds of IC and FFLO modulations can appear at large interlayer coupling, however they are never present simultaneously. The transition line $T_c(h)$ from N to the different S states is nonmonotonic and, at large interlayer coupling, the superconductivity is even destroyed at intermediate exchange fields, then being restored at larger ones.

We use the same model as in ref. [3] and we consider the superlattice with the elementary unit cell which consists in one superconducting and one ferromagnetic layer, both metallic. For simplicity, it is supposed here that quasiparticles in both layers have the same energy spectrum $\xi(\boldsymbol{p})$. The Hamiltonian of the system is given by

$$\begin{aligned} H &= \sum_{\boldsymbol{p},n,i,\sigma} \xi(\boldsymbol{p}) a^\dagger_{n,i,\sigma,\boldsymbol{p}} a_{n,i,\sigma,\boldsymbol{p}} + t\left[a^\dagger_{n,1,\sigma,\boldsymbol{p}} a_{n,-1,\sigma,\boldsymbol{p}} + a^\dagger_{n+1,-1,\sigma,\boldsymbol{p}} a_{n,1,\sigma,\boldsymbol{p}} + \text{h.c.}\right] + \\ &\quad + H_{\text{int1}} + H_{\text{int2}}\,, \\ H_{\text{int1}} &= \frac{\Lambda}{2} \sum_{\boldsymbol{p}_1,\boldsymbol{p}_2,\boldsymbol{q},n,\sigma} a^\dagger_{n,1,\sigma,\boldsymbol{p}_1} a^\dagger_{n,1,-\sigma,-\boldsymbol{p}_1+\boldsymbol{q}} a_{n,1,-\sigma,-\boldsymbol{p}_2+\boldsymbol{q}} a_{n,1,\sigma,\boldsymbol{p}_2}\,, \\ H_{\text{int2}} &= -\sum_{\boldsymbol{p},n,\sigma} h\sigma a^\dagger_{n,-1,\sigma,\boldsymbol{p}} a_{n,-1,\sigma,\boldsymbol{p}}\,, \end{aligned} \tag{1}$$

where $a^\dagger_{n,i,\sigma,\boldsymbol{p}}$ is the creation operator of an electron with spin σ in the n-th elementary cell and momentum $\boldsymbol{p}$ in the layer i is parallel to the plane, where $i = 1$ for the S layer, and $i = -1$ for the F layer. In ref. [3], the order parameter was assumed to change from cell to cell in the form $\Delta_n = |\Delta| e^{iqn}$. The quasimomentum q lies in the direction perpendicular to the layers and quasiparticle Green's functions are obtained in the standard way. At the second-order N/S transition, the order parameter is vanishingly small and we may look at the solution in the form

$$\Delta_n(\boldsymbol{r}_\parallel) = |\Delta| e^{iqn} e^{i\boldsymbol{q}_\parallel \cdot \boldsymbol{r}_\parallel}, \tag{2}$$

where $\boldsymbol{r}_\parallel$ is the in-plane coordinate, $\boldsymbol{q}_\parallel$ is the in-plane modulation wave vector. The linearized self-consistency equation for the order parameter reads

$$|\Delta| = -\Lambda T N(0) \sum_{\omega_n} \int \mathrm{d}\xi \int_0^{2\pi} \frac{\mathrm{d}k}{2\pi} \int_0^{2\pi} \frac{\mathrm{d}\theta}{2\pi} \frac{|\Delta|(\widetilde{\omega}_{n+} + h)(\widetilde{\omega}_{n-} + h)}{[\widetilde{\omega}_{n-}(\widetilde{\omega}_{n-} + h) - |\mathcal{T}_{q+k}|^2][\widetilde{\omega}_{n+}(\widetilde{\omega}_{n+} + h) - |\mathcal{T}_k|^2]}, \tag{3}$$

where $N(0)$ is the electron density of state at the Fermi level in the normal state, the angle $\theta = (\widehat{\boldsymbol{v}_F, \boldsymbol{q}_\parallel})$ between $\boldsymbol{q}_\parallel$ and the velocity $\boldsymbol{v}_F$ on the Fermi surface, $\omega_{n\pm} = i\omega_n \pm \xi(\boldsymbol{p})$, $\widetilde{\omega}_{n\pm} = \omega_{n\pm} + \boldsymbol{v}_F \cdot \boldsymbol{q}_\parallel/2$, $\omega_n = (2n+1)\pi T$ are Matsubara frequencies at temperature T, $\mathcal{T}_k = 2t\cos(k/2)e^{ik/2}$. Naturally, when $t = 0$, this equation defines the critical temperature T_{c0} for 2D S layer. By

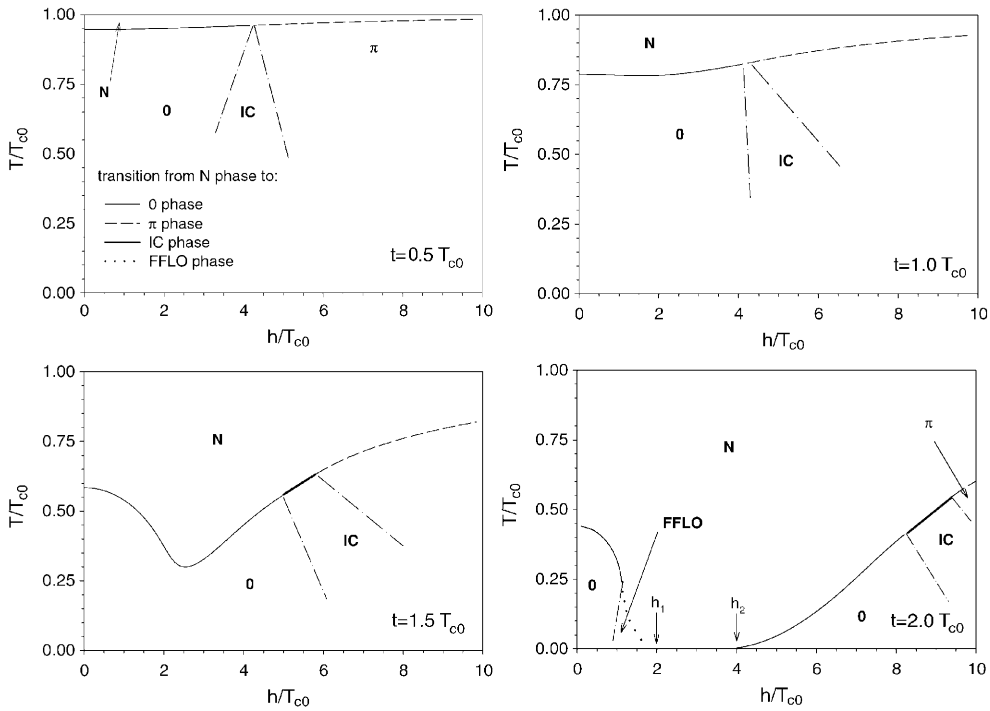

Fig. 1 – The (h, T)-phase diagram for different values of the coupling parameter $t/T_{c0} = 0.5; 1.0; 1.5; 2.0$. The dash-dotted lines indicate the transitions between different kinds of superconducting phases (not calculated).

introducing T_{c0} we now renormalize eq. (3) in the standard way [10]. We obtain

$$\ln\frac{T_{c0}}{4\gamma T} = \frac{1}{4}\mathrm{Re}\sum_{s_1,s_2=\pm}\int\frac{\mathrm{d}k}{2\pi}\int\frac{\mathrm{d}\theta}{2\pi}\left(1-\frac{s_1 h}{2\mathcal{R}_k}\right)\left(1-\frac{s_2 h}{\mathcal{R}_{k+q}}\right)\psi\times \\ \times\left(\frac{1}{2}+i\frac{h}{4\pi T}\left[1+\frac{\boldsymbol{v}_\mathrm{F}\cdot\boldsymbol{q}_\parallel+s_1\mathcal{R}_k+s_2\mathcal{R}_{k+q}}{h}\right]\right), \tag{4}$$

where ψ is the digamma function, $\psi(1/2) = -\ln(4\gamma)$, $\gamma \approx 1.78$, $\mathcal{R}_k = \sqrt{h^2/4+\mathcal{T}_k^2}$, summation is on the signs $s_1, s_2 = \pm$. Equation (4) gives the transition temperatures of different possible superconducting phases which are discussed in more details below.

The transition from N state to the "0 phase" corresponds to $(q = 0, \boldsymbol{q}_\parallel = 0)$. While T_c increases with the exchange field at small interlayer coupling (see, for instance, the case $t = 0.5$ T_{c0} in fig. 1), it starts to behave nonmontonously at larger interlayer coupling, showing reentrance at low temperatures (see $t = 1.0T_{c0}$ and $t = 1.5T_{c0}$ in fig. 1). It may eventually vanish at some critical field h_1, then the superconducting state is only recovered above some critical field h_2, and the critical temperature then increases from 0 to T_{c0} at very large exchange field (see $t = 2.0T_{c0}$ in fig. 1). The values h_1 and h_2 are calculated by taking the limit $T \to 0$ in eq. (4):

$$\ln\frac{\Delta_0}{h} = \frac{1}{4}\sum_{s_1,s_2=\pm}\int\frac{\mathrm{d}k}{2\pi}\int\frac{\mathrm{d}\theta}{2\pi}\left(1-\frac{s_1 h}{2\mathcal{R}_k}\right)\left(1-\frac{s_2 h}{\mathcal{R}_{k+q}}\right)\ln\left|1+\frac{\boldsymbol{v}_\mathrm{F}\cdot\boldsymbol{q}_\parallel+s_1\mathcal{R}_k+s_2\mathcal{R}_{k+q}}{h}\right|, \tag{5}$$

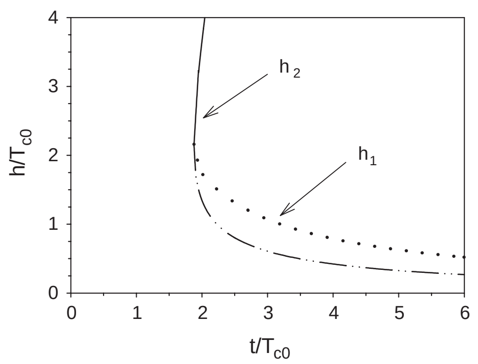

Fig. 2

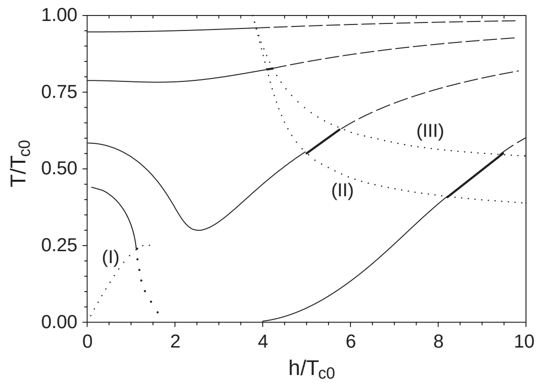

Fig. 3

Fig. 2 – The (t, h)-phase diagram at $T = 0$. $h_1(t)$ (thick dotted line) corresponds to the N/FFLO transition. It substitutes to the dash-dot-dotted line which would have corresponded to the less favorable N/"0 phase" transition. $h_2(t)$ (thin straight line) corresponds to the N/"0 phase" transition.

Fig. 3 – Critical lines $T_c(h)$ of fig. 1 are shown on the same phase diagram. In addition, the dotted line (I) corresponds to the location of the tricritical point $(h^*(t), T^*(t))$ as t is varied, (II) corresponds to the location of $(h_a(t), T_c(h_a(t)))$, (III) corresponds to the location of $(h_b(t), T_c(h_b(t)))$.

where $\Delta_0 = (\pi/\gamma)\, T_{c0}$. At low temperatures, higher critical field may be obtained when considering the in-plane FFLO modulation. From eq. (5) we obtain the critical fields at $T = 0$: $h_1(t)$ and $h_2(t)$, the former actually corresponds to the FFLO state, while the latter corresponds to the "0 phase", both are shown in fig. 2. Expanding eq. (4) in the small values of the modulation vector $\boldsymbol{q}_\parallel$, we find that the change of sign of the coefficient in front of the term $\sim \boldsymbol{q}_\parallel^2$:

$$\mathrm{Re} \sum_{s_1, s_2 = \pm} \int \frac{\mathrm{d}k}{2\pi} \left(1 - \frac{s_1 h}{2\mathcal{R}_k}\right) \left(1 - \frac{s_2 h}{\mathcal{R}_k}\right) \psi_2 \left(\frac{1}{2} + i \frac{h}{4\pi T} \left[1 + \frac{(s_1 + s_2)\mathcal{R}_k}{h}\right]\right) = 0 \qquad (6)$$

(where ψ_2 is the second derivative of ψ) along the critical line $T_c(h)$ occurs at the tricritical point $(h^*(t), T^*(t))$. Such a point indicates that for $T < T^*(t)$, the FFLO modulation is favoured. The second-order critical line between N and FFLO state is then obtained by finding the value $\boldsymbol{q}_\parallel$ which gives the highest critical field at every temperature. Numerical results for $t = 2.0 T_{c0}$ are shown in fig. 1. The location of the tricritical points when the interlayer coupling parameter is varied is represented in fig. 3. It is interesting to note that they are situated near the line $h/4\pi T \approx 0.304$, as was expected. This portion of the phase diagram is very similar to that calculated for 2D superconductors [13]. In this case, the N/FFLO transition is known to compete with the transition of the first order into the uniform superconducting state [10]. Here it is not the case: by expanding the free energy given in ref. [3] in powers of $|\Delta|$ we find that the coefficient in front of the term $\sim |\Delta|^4$ changes its sign only at some temperature lower than T^* for all t.

At larger exchange fields we also obtain the transition into the IC phase (see fig. 1). The transition from N to "0 phase" takes place when $h_1 < h < h_a$. At $h > h_a$, the system prefers to develop the IC modulation. The location of the critical point $(h_a(t), T_c(h_a(t)))$ is obtained by expanding eq. (4) in the small parameter q (and $\boldsymbol{q}_\parallel = 0$) and determining the change of sign of the coeffcient in front of the term $\sim q^2$. Similarly, the "π phase" corresponds to $h_b < h$ when the modulation wave vector is cut off by the crystalline structure once again. The location of the critical point $(h_b(t), T_c(h_b(t)))$ is obtained in the same way by expanding eq. (4) in the small parameter $[\pi - q]$ (and $\boldsymbol{q}_\parallel = 0$). The location of both critical points is represented in fig. 3 when the coupling is varied. The IC phase happens in between when $h_a < h < h_b$. Thus, the transition from "0 phase" to "π phase" occurs continuously. In fact, it was also noted in ref. [3] where this transition was found to be second order, but would take place on a vanishingly small exchange field interval $h_b - h_a \sim t^4/T_{c0}^3$ in the small-coupling limit.

For completeness, let us note that the "0 phase" to "π phase" transition was also predicted in the case of artificially grown multilayers with finite thicknesses [16]. However, in this case, the dirty limit is assumed and no in-plane modulation for the order parameter can take place in the S layers. Recently, however, it was proposed that a superconducting state with a three-dimensionnal structure could be favorable for some range of parameters. In this case, the in-plane modulation would occur on the anomalous Green function in F layers induced by proximity effect [17]. This is a quite different situation from the one we predict in this paper. Moreover no re-entrance of superconductivity was predicted in such models.

In conclusion, we have calculated the transition line from the normal state into different kinds of superconducting states in the strongly coupled F/S superlattice on the basis of a microscopic model. A quite rich variety of phase diagrams is obtained for different values of the interlayer coupling. We find that such model is expected to behave like a quasi-2D superconductor in the presence of effective exchange field at small fields, thus exhibiting the in-plane FFLO modulation at low temperatures. Superconductivity is also induced at large exchange fields and then the "0 phase" to "π phase" transition occurs continuously with the modulation which grows to its saturated value as the exchange field is increased. These results may apply to layered ferromagnetic superconductors with $T_c \ll \Theta$, that is when the magnetism is hardly affected by the superconductivity and the exchange field takes a definite value. It is interesting to note that in such case the superconducting modulated phases may appear as soon as the critical temperature is crossed, but different kinds of modulations cannot coexist in the same sample. Such a situation may be realized in the ruthenocuprates. However, it is easy to imagine still richer behavior when $\Theta \lesssim T_c$ and the temperature dependence of the exchange field can no longer be neglected. Our results may also apply to antiferromagnetic superconductors in the paramagnetic phase when the application of the magnetic field induces a weak ferromagnetic component.

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