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Josephson-junction thermodynamics and the superconducting phase transition in a SQUID device

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In a model of two ideal BCS superconductors coupled by a tunneling Hamiltonian the nonvanishing of the Josephson internal energy (and entropy) for $T \rightarrow T_c^-$ is shown to be a consequence of superconducting correlations, which persist in the thermodynamic limit even in the mean-field approximation. The ensuing rapid increase of the Josephson free energy as the temperature of a tunneling junction drops below the superconducting bulk transition temperature T_c makes this transition of first order whenever the phase difference across the junction is fixed to a nonzero value. Taking this into account results in an availability potential governing the nonequilibrium thermodynamics of the junction which, in contrast with previously published results, has no unphysical features like latent heat released upon entering (or a superconducting phase dependent value in) the normal state. The analysis *inter alia* predicts a lowering of the critical temperature (to T_c') for the junction, which has meanwhile been observed in high-quality superconducting quantum interference devices. [S0163-1829(97)51714-0]

I. INTRODUCTION

The Josephson energy $G_J(\phi, T)$ has the thermodynamic significance of a (Gibbs) free energy, for changes $dG_J(\phi, T)$ are equal to electrical work $V(t)I_J(\phi, T) dt$ done at constant temperature. Given an expression for $G_J(\phi, T)$, one should thus be able to calculate the associated entropy $S_J(\phi, T) = -\partial_T G_J(\phi, T)|_\phi$ and internal energy $U_J = G_J + TS_J$. These quantities play an important role in the nonequilibrium thermodynamic theory of Josephson devices.¹⁻⁴

The Josephson entropy does not vanish as the temperature of the system approaches the bulk critical temperature T_c of the superconducting phase transition. Indeed, for one of the simplest formulas for G_J [Eq. (13)] this entropy tends to a constant value, as the gap $\Delta(T) \propto \sqrt{T_c - T}$ if $T \rightarrow T_c^-$. S_J vanishes for $T > T_c$, and therefore it has a finite jump δS_J at $T = T_c$. The case is even more dramatic for a junction between unequal superconductors a and b [we take $(T_c)_a < (T_c)_b$], where the phase-dependent part of $G_J(\phi, T)$ is proportional to $\Delta_a \Delta_b$ when $\Delta_a \ll T$, and hence S_J even diverges if $T \rightarrow (T_c)_a^-$.

While an entropy jump in itself is familiar from first-order transitions, presently S is greater in the low- than in the high-temperature phase, for $\partial \Delta(T) / \partial T < 0$. Hence, latent heat would be released upon entering the normal state, implying an unphysical instability.

The scenario can be studied in a superconducting quantum interference device (SQUID)¹⁻³ by incorporating the junction in a superconducting ring. Indeed, the phase difference $\phi = 2\pi\Phi/\Phi_0$ (with $\Phi_0 = h/2e$) is externally controlled in a ring with negligible self-inductance so that the flux Φ

equals Φ_{ext} . We explain the mentioned instability, and show that for $\phi \neq 0$ the Josephson coupling lowers the actual T_c , while the transition becomes first instead of second order.

In Sec. II we investigate the nonvanishing of the Josephson internal energy for $T \rightarrow T_c^-$ in a model of two BCS superconductors coupled by a tunneling Hamiltonian. In Sec. III we explain the effect of the Josephson coupling on the superconducting transition, and examine the consequences for a theory of nonisothermal flux dynamics. The predicted decrease in T_c is estimated in Sec. IV. A detailed account is available in Ref. 5.

II. MICROSCOPIC THEORY

The system is modeled by the Hamiltonian

$$H = H_0 + H_T = H_a + H_b + H_T, \quad (1)$$

with $(c = a, b; V_c > 0)$

$$H_c = H_{c,\text{kin}} + H_{c,\text{int}} \\ = \sum_{p\alpha} \xi_p c_{p\alpha}^\dagger c_{p\alpha} - V_c \sum_{pp'}' c_{p\uparrow}^\dagger c_{-p\downarrow}^\dagger c_{-p'\downarrow} c_{p'\uparrow}, \quad (2)$$

$$H_T = \sum_{pq\alpha} T_{pq} a_{p\alpha}^\dagger b_{q\alpha} + \text{H.c.} \quad (3)$$

We work in the grand canonical ensemble and the term $-\mu N_c$ is included in H_c , i.e., $\xi_p = p^2/2m - \mu$. The prime on the sum implies restriction to $p^{(\prime)}$ with $|\xi_{p^{(\prime)}}| < \omega_D$, the Debye frequency. Neglecting the cutoff amounts to taking the

limit $\Delta_c, T \ll \omega_D \ll \mu$ (throughout $\hbar = k_B = 1$) and is allowed in all steps except when using the BCS gap equation to arrive at the last line of Eq. (18).

To introduce the Green's functions \mathbf{G}_c of the uncoupled superconductors we define the Nambu field as⁶

$$\Psi_c(\mathbf{p}, \tau) = (c_{p\uparrow}(\tau), -\bar{c}_{-p\downarrow}(\tau))^T, \quad (4)$$

so that

$$\mathbf{G}_c(\mathbf{p}, \tau) = -\langle T_\tau \{ \Psi_c(\mathbf{p}, \tau) \bar{\Psi}_c(-\mathbf{p}) \} \rangle_0 \quad (5)$$

$$= \begin{pmatrix} \mathcal{G}_c(\mathbf{p}, \tau) & \mathcal{F}_c(\mathbf{p}, \tau) \\ \mathcal{F}_c^+(\mathbf{p}, \tau) & -\mathcal{G}_c(-\mathbf{p}, -\tau) \end{pmatrix} \quad (6)$$

yields

$$\mathbf{G}_c(\mathbf{p}, \omega) = \frac{1}{\omega^2 + \xi_p^2 + \Delta_c^2} \begin{pmatrix} -i\omega - \xi_p & \Delta_c e^{i\phi_c} \\ \Delta_c e^{-i\phi_c} & -i\omega + \xi_p \end{pmatrix}, \quad (7)$$

Δ_c being the magnitude of the gap.

In Nambu notation the tunneling Hamiltonian reads

$$H_T = \sum_{pq} \bar{\Psi}_a(-\mathbf{p}) \mathbf{T}_{pq} \Psi_b(\mathbf{q}) + \text{H.c.}, \quad (8)$$

where

$$\mathbf{T}_{pq} = \text{diag}(T_{pq}, -T_{-p-q}^*). \quad (9)$$

Evaluating the change in grand canonical potential due to tunneling in lowest-order perturbation theory one finds

$$\begin{aligned} \delta G &\equiv G(\{T_{pq}\}) - G(\{T_{pq}=0\}) \\ &= -\frac{1}{2} T \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \langle T_\tau \{ H_T(\tau_1) H_T(\tau_2) \} \rangle_0 \\ &= T \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \\ &\quad \times \sum_{pq} \text{Tr} \{ \mathbf{G}_a(\mathbf{p}, \tau_2 - \tau_1) \mathbf{T}_{pq} \mathbf{G}_b(\mathbf{q}, \tau_1 - \tau_2) \mathbf{T}_{pq}^\dagger \} \\ &= T \sum_{pq\omega} \text{Tr} \{ \mathbf{G}_a(\mathbf{p}, \omega) \mathbf{T}_{pq} \mathbf{G}_b(\mathbf{q}, \omega) \mathbf{T}_{pq}^\dagger \}, \end{aligned} \quad (10)$$

where we used Eq. (8) for H_T and applied Wick's theorem. To evaluate Eq. (10), we assume that the tunneling amplitudes obey time-reversal symmetry $\mathbf{T}_{pq} = T_{pq} \sigma_3$ and are energy independent (on the scale of T_c). Decomposing the sum over \mathbf{p} as $\sum_{\mathbf{p}} = N_a(0) \int_{-\infty}^{+\infty} d\xi_p \int (d\hat{p}/4\pi)$ [where the approximation $N_a(\xi_p) \approx N_a(0)$ is correct to first order in $(T_c)_a/\mu$], the integrals over $\xi_{p,q}$ can be performed with $\mathbf{G}_{a,b}$ as in Eq. (7). This yields

$$\delta G = -2T\gamma \sum_{\omega} Q P^{-1}, \quad (11)$$

with $P \equiv \sqrt{\omega^2 + \Delta_a^2} \sqrt{\omega^2 + \Delta_b^2}$, $Q \equiv \omega^2 + \Delta_a \Delta_b \cos \phi$ ($\phi \equiv \phi_a - \phi_b$), and the dimensionless conductance

$\gamma = \pi^2 \langle |T_{pq}|^2 \rangle N_a(0) N_b(0) = \pi / (4e^2 R^N)$, the relation between the tunneling amplitudes and the normal-state resistance R^N being standard.⁷

The sum in Eq. (11) diverges for δG , but converges for its difference between the superconducting and normal states $\delta G^{S-N} \equiv \delta G^S - \delta G^N$, which is the quantity of interest. A general property is its nonnegativity: $\delta G^{S-N} \propto \sum_{\omega} (1 - Q/P) \geq 0$ because

$$\begin{aligned} P^2 - Q^2 &\geq (\omega^2 + \Delta_a^2)(\omega^2 + \Delta_b^2) - (\omega^2 + \Delta_a \Delta_b)^2 \\ &= \omega^2 (\Delta_a - \Delta_b)^2 \geq 0. \end{aligned} \quad (12)$$

For equal gaps $\Delta_a = \Delta_b = \Delta$ one finds (see Ref. 7)

$$\begin{aligned} G_J &\equiv \delta G^{S-N} = 2T\gamma \Delta^2 (1 - \cos \phi) \sum_{\omega} (\omega^2 + \Delta^2)^{-1} \\ &= \gamma \Delta \tanh(\Delta/2T) (1 - \cos \phi), \end{aligned} \quad (13)$$

consistent with Anderson's theorem,⁸ implying that $G_J(\phi=0) = 0$ (to lowest order in T_c/μ), which will be crucial in Sec. III.

For unequal gaps $\Delta_{a,b} \ll T$ one obtains

$$G_J = \frac{\gamma}{4T} (\Delta_a^2 + \Delta_b^2 - 2\Delta_a \Delta_b \cos \phi) + O[(\Delta/T)^4], \quad (14)$$

while for two superconductors with very different T_c near the lower of these temperatures, i.e., for $\Delta_a \ll T \ll \Delta_b$, the result reads⁵ (γ_E is Euler's constant)

$$\begin{aligned} \gamma^{-1} G_J &= \frac{2}{\pi} \Delta_b - \frac{\pi T^2}{3\Delta_b} \\ &\quad + \frac{1}{\pi} \left\{ \frac{\Delta_a^2}{\Delta_b} - 2\Delta_a \cos \phi \right\} \left\{ \ln \left(\frac{4\Delta_b}{\pi T} \right) + \gamma_E \right\} \\ &\quad + T \{ O[(T/\Delta_b)^3] + O[T\Delta_a/\Delta_b^2] + O[(\Delta_a/T)^3] \}. \end{aligned} \quad (15)$$

Since $\Delta_{a,b} \propto \sqrt{(T_c)_{a,b} - T}$ for $T \rightarrow (T_c)_{a,b}^-$, with $U = \partial_\beta (\beta G)$ the above formulas for G_J predict a constant [Eq. (13)] or even divergent [Eqs. (14) and (15)] internal energy (and entropy) difference δU^{S-N} upon approaching the (lowest) T_c from below. This property is a consequence of deviations from BCS mean-field theory (even though these superconducting correlations are of order $N_{a,b}^-$), as is demonstrated by an explicit calculation of δU in which such correlations are neglected. Consider

$$\delta U = \delta \langle H_a \rangle + \delta \langle H_b \rangle + \langle H_T \rangle, \quad (16)$$

where $\delta \langle \cdot \rangle = \langle \cdot \rangle_\gamma - \langle \cdot \rangle_0$. In first order $\langle H_T \rangle = 2\delta G$, with δG as in Eq. (10). For $\langle H_a \rangle$ one has

$$\langle H_a \rangle = \sum_{p\alpha} \xi_p \langle a_{p\alpha}^\dagger a_{p\alpha} \rangle - V_a \sum_{pp'}' \langle a_{p\uparrow}^\dagger a_{-p\downarrow}^\dagger a_{-p'\downarrow} a_{p'\uparrow} \rangle. \quad (17)$$

Expanding the second term in terms of two-point functions, the Hartree-Fock contribution to $\delta \langle H_a \rangle_{\text{MF}}$ vanishes in the thermodynamic limit. One is left with

$$\begin{aligned}
\langle H_a \rangle_{\text{MF}} &= \sum_{p\alpha} \xi_p \langle a_{p\alpha}^\dagger a_{p\alpha} \rangle - V_a \sum_{pp'}' \langle a_{p\uparrow}^\dagger a_{-p\downarrow}^\dagger \rangle \langle a_{-p'\downarrow} a_{p'\uparrow} \rangle \Rightarrow \\
\delta \langle H_a \rangle_{\text{MF}} &= \sum_{p\alpha} \xi_p \delta \langle a_{p\alpha}^\dagger a_{p\alpha} \rangle \\
&\quad - V_a \sum_{pp'}' 2 \operatorname{Re}[\mathcal{F}_a(\mathbf{p}', \tau=0) \delta \langle a_{-p\downarrow}^\dagger a_{p\uparrow}^\dagger \rangle] \\
&= \sum_{p\alpha} \xi_p \delta \langle a_{p\alpha}^\dagger a_{p\alpha} \rangle \\
&\quad - \Delta_a \sum_p 2 \operatorname{Re}[e^{i\phi_a} \delta \langle a_{-p\downarrow}^\dagger a_{p\uparrow}^\dagger \rangle] \\
&= \sum_p \operatorname{Tr}\{(\Delta_a e^{i\phi_a} \sigma_3 \sigma_1 - \xi_p \sigma_3) \delta \langle \Psi_a(\mathbf{p}) \bar{\Psi}_a(-\mathbf{p}) \rangle\},
\end{aligned} \tag{18}$$

where we used $\delta \langle a_{p\uparrow}^\dagger a_{-p\downarrow}^\dagger \rangle = O[N_a^{-1}]$ so that only the linear term contributes to $\delta \langle H_a \rangle$. To evaluate the rhs of Eq. (18) in lowest order transform to frequencies, and use $\Delta_a e^{i\phi_a} \sigma_3 \sigma_1 - \xi_p \sigma_3 = \mathbf{G}_a(\mathbf{p}, \omega)^{-1} - i\omega$ to arrive at

$$\begin{aligned}
\delta \langle H_a \rangle_{\text{MF}} &= T \sum_{pq\omega} \operatorname{Tr}\{[i\omega - \mathbf{G}_a(\mathbf{p}, \omega)^{-1}] \\
&\quad \times \mathbf{G}_a(\mathbf{p}, \omega) \mathbf{T}_{pq} \mathbf{G}_b(\mathbf{q}, \omega) \mathbf{T}_{pq}^\dagger \mathbf{G}_a(\mathbf{p}, \omega)\} \\
&= -T \sum_{pq\omega} \omega \operatorname{Tr}\{(\partial_\omega \mathbf{G}_a(\mathbf{p}, \omega)) \mathbf{T}_{pq} \mathbf{G}_b(\mathbf{q}, \omega) \mathbf{T}_{pq}^\dagger\} \\
&\quad - \frac{1}{2} \langle H_T \rangle,
\end{aligned} \tag{19}$$

$\delta \langle H_b \rangle_{\text{MF}}$ following by symmetry. Substitution into Eq. (16) shows that $\langle H_T \rangle$ cancels in δU_{MF} , viz.,

$$\begin{aligned}
\delta U_{\text{MF}} &= -T \sum_{pq\omega} \omega \partial_\omega \operatorname{Tr}\{\mathbf{G}_a(\mathbf{p}, \omega) \mathbf{T}_{pq} \mathbf{G}_b(\mathbf{q}, \omega) \mathbf{T}_{pq}^\dagger\} \\
&= \partial_\beta |_{\Delta_{a,b}} \beta \left[T \sum_{pq\omega} \operatorname{Tr}\{\mathbf{G}_a(\mathbf{p}, \omega) \mathbf{T}_{pq} \mathbf{G}_b(\mathbf{q}, \omega) \mathbf{T}_{pq}^\dagger\} \right] \\
&= \partial_\beta (\beta \delta G)_{\Delta_{a,b}}.
\end{aligned} \tag{20}$$

As our derivation is valid in the normal (N) and the superconducting (S) phase, this relation also holds between $\delta U_{\text{MF}}^{S-N}$ and δG^{S-N} . By Eqs. (13)–(15) and (20), $\delta U_{\text{MF}}^{S-N}$ vanishes as $T \rightarrow T_c^-$, which is what we set out to show.

III. THERMODYNAMICS

The free energy of the system (with respect to the N state) also contains the phase (or SQUID flux) independent bulk condensation energy, which is quadratic in $T_c - T$ near T_c . It usually completely dominates the Josephson term $G_J [\geq 0$ by

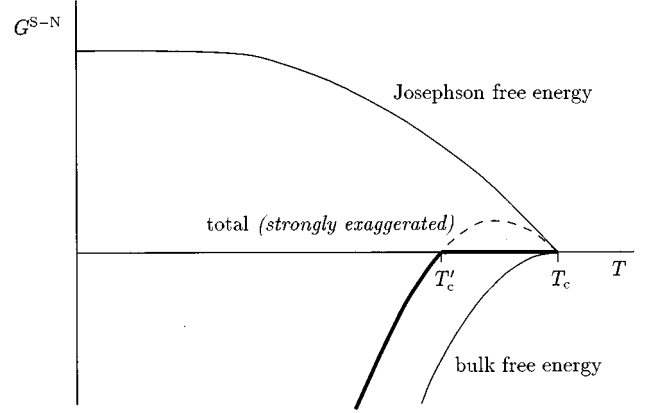


FIG. 1. The Josephson, bulk and total free energy of a junction with $\phi \neq 0$ (arbitrary units). The dashed line indicates the free energy of the superconducting phase, the thick line the equilibrium free energy $G(T)$.

virtue of Eq. (12)]. Accounting for the bulk energy is crucial for resolving the thermodynamic instability of the junction below T_c (see Sec. I).

For a fixed $\phi \neq 0$, G^{S-N} varies with temperature as in Fig. 1. Just below T_c the total free energy in the S phase is higher than in the N phase, so that the system remains in the state $\Delta = 0$ (always a solution of the gap equation) until $G^{S-N} = 0$ again, at T'_c [see Eq. (24)]. At this $T'_c < T_c$, Δ jumps to a finite value. This first-order transition is indeed accompanied by latent heat (see Sec. I), as follows from the kink in $G(T)$. The significance of Anderson's theorem (see Sec. II) is that the transition remains of second order if no flux is applied externally.

Now consider the nonequilibrium thermodynamics of the system (see Refs. 1–3). Equation (19) of Ref. 3 and the one below it yield

$$\begin{aligned}
A &= -E_J \cos\left(2\pi \frac{\Phi}{\Phi_0}\right) \left\{ 1 - \frac{3\tilde{T}^2 T_r - 2\tilde{T}^3}{T_c^3} \right\} + \frac{(\Phi - \Phi_{\text{ext}})^2}{2L} \\
&\quad + \alpha \frac{\frac{1}{3} T^3 - \frac{1}{2} T^2 T_r}{T_c^3}
\end{aligned} \tag{21}$$

for the availability $A = G + (T - T_r)S$ of a SQUID in contact with a bath at temperature T_r . Here $E_J = I_c(T=0)/2e$, α arises in a Debye-type bulk free energy $G_{\text{bulk}} = -\frac{1}{6}\alpha(T/T_c)^3$, and $\tilde{T} \equiv \min(T, T_c)$.

Following Eq. (21), an availability barrier persists in the N state near $\Phi = \frac{1}{2}\Phi_0$. However, this barrier is entirely due to the Josephson coupling and related to the unphysical negative latent heat discussed in Sec. I. This artifact is remedied by accounting for the bulk condensation energy, which we now model *à la* Refs. 1–3. Since Eq. (21) implies $S_{\text{bulk}} = \frac{1}{2}\alpha T^2/T_c^3$, we take $S_{\text{bulk}}^S = \frac{1}{2}(\alpha/T_c)(T/T_c)^{2+\eta}$ ($\eta > 0$). For the S⁻ phase one obtains

$$\begin{aligned}
A^S = E_J & \left[1 - \cos\left(2\pi \frac{\Phi}{\Phi_0}\right) \right] \left[1 - \frac{3T^2 T_r - 2T^3}{T_c^3} \right] + \frac{(\Phi - \Phi_{\text{ext}})^2}{2L} \\
& + \alpha \left\{ \frac{1}{2(3+\eta)} - \frac{1}{6} + \left(\frac{1}{2} - \frac{1}{2(3+\eta)} \right) \left(\frac{T}{T_c} \right)^{3+\eta} - \frac{1}{2} \left(\frac{T}{T_c} \right)^{2+\eta} \frac{T_r}{T_c} \right\}, \quad (22)
\end{aligned}$$

while A^N follows from Eq. (22) with $E_J = \eta = 0$. The free energy $G^{S,N}$ results from $A^{S,N}$ by setting $T_r = T$. Our final result for $A(T)$ then follows from⁹

$$\begin{aligned}
A(T) = & \{ \theta(T - T_c) + \theta(T_c - T) \theta(G^S(T) - G^N(T)) \} A^N(T) \\
& + \theta(T_c - T) \theta(G^N(T) - G^S(T)) A^S(T). \quad (23)
\end{aligned}$$

A calculation of the junction's actual critical temperature T'_c (see above in this section) is now easy in the limit $E_J \ll \alpha$, in which the effect of the Josephson coupling is small and hence $\varepsilon \equiv (T_c - T'_c)/T_c \ll 1$. Expansion of $G^S = G^N$ in $E_J/\alpha \ll 1$ yields

$$\varepsilon = \frac{12E_J}{\alpha\eta} \left[1 - \cos\left(2\pi \frac{\Phi}{\Phi_0}\right) \right]. \quad (24)$$

IV. FINAL REMARKS

The thermodynamic analysis of Sec. III can also be motivated in the context of a dynamical theory of nonisothermal behavior, which has potential relevance for a wide class of physical systems.⁴ The relevant ‘‘reaction coordinate’’ (here the flux) and the temperature then become fluctuating quantities, the values of which spread over the entire accessible state space. Hence, there is no *a priori* possibility to restrict the variables to a bounded region such as $T < T_c$ (or $T > T_c$), so that covering the phase transition becomes crucial.

Of course, the phenomenology of Sec. III is no substitute

for solving Gor'kov equations for a finite-size SQUID. Also, the model (1) obscures the fact that tunneling is an interface phenomenon. However, this should allow a Ginzburg-Landau description near T_c , e.g., to determine the effective interaction volume V (see below).

To estimate T'_c , take $\Phi = \frac{1}{2}\Phi_0$ and $E_J = h\Delta(0)/8e^2R^N \approx 10^{-19}/R^N$ J (for Al) in Eq. (24). From Eq. (22) we find $G_{\text{bulk}}^{N-S} = \frac{1}{4}\alpha\eta(1 - T/T_c)^2$, which yields $\alpha\eta \approx 6VB_c(0)^2/\mu_0$ upon comparison with $G_{\text{bulk}}^{N-S} \approx (V/2\mu_0)\{1.73B_c(0)(1 - T/T_c)\}^2$ [Ref. 6, Eq. (36.12)], where $\mu_0 = 4\pi \times 10^{-7}$ H m⁻¹ and $B_c(0) \approx 10^{-2}$ T [for Al (Ref. 10)], so that $\varepsilon \approx 5 \times 10^{-21}/VR^N$. For $V = 0.1 \mu\text{m}^3$ and $R^N = 50 \Omega$, one arrives at $\varepsilon \approx 10^{-3}$. With $T_c \approx 1$ K this means that the effect is in the mK range, and hence observable in state-of-the-art devices of sufficiently small dimensions and of a high material quality such that T_c is sharply defined.

In conclusion, this work highlights hitherto neglected consequences of the phase-independent part of the Josephson coupling [the first term in Eq. (13)], which near T_c is not an arbitrary constant. In the meantime, its predictions concerning the oscillations of T_c versus the applied magnetic flux in a SQUID device have indeed been confirmed experimentally.¹¹

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⁹The relative stability of phases can also be determined in terms of the availability (instead of G), but then A has to be considered as a function of the enthalpy and the analysis becomes more involved; see further Ref. 5.

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