Two-order-parameter GL fits

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This addendum to Ref.[1] describes the procedures used to fit the two-order-parameter GL model to the data and discusses the resulting parameters.

For a ring consisting of two Josephson-coupled layers, the most natural form for a GL-functional in the 1D approximation is

$$\begin{aligned} F[\psi_1,\psi_2,\varphi] &= F_1[\psi_1,\varphi]+F_2[\psi_2,\varphi] \\ &+ \frac{w\gamma}{2}\int_0^L dx|\psi_1-\psi_2|^2 \quad \text{with} \qquad (1) \\ F_i[\psi_i,\varphi] &= wd_i\int_0^L dx\left\{\frac{\hbar^2}{2m}\left|\left(-i\nabla+\frac{\varphi}{R}\right)\psi_i\right|^2\right. \\ &+ \left.\frac{\alpha_i}{2}|\psi_i|^2+\frac{\beta_i}{4}|\psi_i|^4\right\} \qquad (2) \end{aligned}$$

(cf. Eqn. (3), (4) of Ref. [1]). Phenomenologically, the coupling term can be motivated in the weak coupling limit by the requirement that it should yield a $\cos(\phi_1-\phi_2)$ dependence on the phase difference and not change the free energy if two identical superconductors with the same phase are coupled to each other. For small γ and near T_c, it also reproduces the temperature dependence of the critical current density $J_c=\gamma 2\pi/\Phi_0|\psi_1||\psi_2|$ near T_c, which was calculated from microscopic theory in Refs. [2, 3]. For identical $T_{c,1}=T_{c,2}$, one obtains $J_c\propto|\psi|^2\propto 1-T/T_c$. For $T\lessapprox T_{c,1}<T_{c,2}$ on the other hand, the temperature dependence near $T_{c,1}$ is dominated by $|\psi_1|\propto\sqrt{1-T/T_c}$.

By absorbing the terms $\gamma|\psi_i|^2$ in the functional (1) into the quadratic terms in F_i and renormalizing the order parameter, it can be brought into the form

$$\begin{aligned} \tilde{F}[\tilde{\psi}_1,\tilde{\psi}_2,\varphi] &= F_1[\tilde{\psi}_1,\varphi]+F_2[\tilde{\psi}_2,\varphi] \\ &- \frac{wd\tilde{\gamma}}{2}\int_0^L dx(\tilde{\psi}_1^*\tilde{\psi}_2+\tilde{\psi}_1\tilde{\psi}_2^*) \quad \text{with} \quad (3) \\ \tilde{F}_i[\tilde{\psi}_i,\varphi] &= wd\int_0^L dx\left\{\frac{\hbar^2}{2m}\left|\left(-i\nabla+\frac{\varphi}{R}\right)\tilde{\psi}_i\right|^2\right. \\ &+ \left.\frac{\tilde{\alpha}_i}{2}|\tilde{\psi}_i|^2+\frac{\tilde{\beta}_i}{4}|\tilde{\psi}_i|^4\right\} \qquad (4) \end{aligned}$$

which has been shown to emerge from microscopic theory for two-band superconductors [4]. We are not aware of a direct derivation of the functional (1) for the tunneling case from microscopic theory. However, it has been shown [5] that a tunneling BCS Hamiltonian leads to the same Greens functions as a two band BCS model [6] to second order in the tunneling matrix elements. Thus, it appears that the equivalence between (1) and (3) holds much beyond GL theory. Although one may expect higher order terms to become significant at stronger coupling, the above functionals should still give a qualitatively correct approximation.

In principle, either form could be used as a starting point to obtain fit models, as the parameters of one are convertible into those of the other with the following transformations:

$$\begin{aligned} \tilde{\psi}_i &= \nu_i^{1/2}\psi_i \qquad (5) \\ \tilde{\alpha}_i(T) &= \alpha_i(T)+d_i^{-1}\gamma \qquad (6) \\ \tilde{\beta}_i &= \nu_i^{-1}\beta_i \qquad (7) \\ \tilde{\gamma} &= (d_1d_2)^{-1/2}\gamma \qquad (8) \end{aligned}$$

where $\nu_i=d_i/(d_1+d_2)$ is the volume fraction occupied by ψ_i in a layered structure.

Not counting the known geometric dimensions and $\nu_{1,2}$, which are not independent from other parameters, there are five GL parameters at any given T and seven if modeling a linear temperature dependence of the $\alpha_{1,2}$, as customary for GL theory. It turns out that in practice, fits of the model to our data are effectively underdefined, so that a wide range of parameter combinations reproduce individual Φ_a-I curves or $\xi(T)$ and $\lambda^{-2}(T)$ fairly well. To overcome this problem, we have fixed some parameters in each individual fit and made an effort to choose the fixed values such that a physically reasonable dependence of all parameters on w is obtained. Although one might expect more accurate results from fitting all data curves at once, this computationally less complex procedure is sufficient to obtain a consistent overall picture.

Naturally, it is most appropriate to fix parameters that are expected to be independent of T and/or w and physically meaningful. In the functional (1),(2), $T_{c,i}$ as defined by $\alpha_i(T_{c,i})=0$, is the actual bare T_c of ψ_i. In the second form [Eqns. (3) and (4)] on the other hand, the temperatures at which $\tilde{\alpha}_i(T)=0$ will be smaller than the bare $T_{c,i}$ because of the γ/ν_i term in Eqn. (6). Furthermore, the values of $\tilde{\beta}_{1,2}$ and $\tilde{\gamma}$ do not correspond to their physical values for a Josephson coupled bilayer system due to the absorption of ν_i into the normalization of $\tilde{\psi}_i$. Thus, the form (1) and (2) is more adequate for our purposes.

We have parametrized the temperature dependence of $\alpha_{1,2}$ by $\alpha_i(T)=(T/T_{c,i}-1)\overline{\alpha}_i$, and assumed all other coefficients to be independent of T. The complete set

of fit model parameters consists of $\overline{\alpha}_{1,2}$, $\nu_1 = 1 - \nu_2$, $\epsilon_{1,2} = \overline{\alpha}_{1,2}^2/\beta_{1,2}$, and γ/d. ν_1 had to be included because the individual layer thicknesses $d_{1,2}$ appearing in the prefactors of Eqn. (2) are unknown in our samples. The condensation energy densities extrapolated to $T = 0$, $\epsilon_{1,2}$, were used instead of, say, $\beta_{1,2}$, as they should be independent of the mean free path and thus are reasonable to keep fixed.

For the purpose of fitting, we set $\hbar^2/m$ to 1 and normalize $\psi_{1,2}$ and consequently $\beta_{1,2}$ such that one would obtain $|\psi|^2 = \alpha/\beta = \lambda^{-2}$ in the absence of any applied flux or phase winding for a single OP. For this choice of units, it is natural to measure the coupling in terms of γ/d so that the cross section wd only appears in the prefactor of the GL functional (1), which has to be multiplied by $\Phi_0^2/4\pi^2\mu_0$ to convert to physical energy. For equal phase winding numbers $n_1 = n_2 = n$ in both OPs, one can make the ansatz $\psi_i(x) = |\psi_i|e^{inx}$, where x is the spatial coordinate along the ring circumference. Variation of Eqn. (1) w.r.t. $|\psi_{1,2}|$ leads to

$$(\alpha_i + (\varphi - n)^2/R^2)|\psi_i| + \beta|\psi_i|^3 + \gamma(|\psi_i| - |\psi_{2-i}| = 0$$

for $i = 1, 2$ which can be solved numerically for $|\psi_1|$ and $|\psi_2|$. With the above normalization of $|\psi_{1,2}|$, the supercurrent circulating the ring is

$$I = -\frac{wd\Phi_0}{\mu_0 2\pi R}(\varphi - n)(|\psi_1|^2 + |\psi_2|^2) \qquad (9)$$

In order to extract an effective λ^{-2} and ξ_{GL} to be used as fit model from a $\Phi_a - I$ curve computed numerically using Eqn. (9), we have approximated it by a cubic polynomial at small φ according to Eqn. (1) of Ref. [1]: λ^{-2} is proportional to the linear coefficient , and ξ_{GL}^2/R^2 the ratio of the cubic and linear coefficients. The same procedure was used to obtain the experimental values of λ^{-2} and ξ_{GL}, shown in Fig. 2 of [1], from fits to measured curves like the one in shown Fig. 3(d) of Ref. [1], which show two component effects but no fluxoid transitions. While the interpretation of those values as a physical coherence length and superfluid density is somewhat questionable at least for weak coupling, this provides an operational definition that is consistent with the 1D GL result in Eqn. (1) of Ref. [1].

For the simultaneous fits to the temperature dependence of λ^{-2} and ξ_{GL} shown in Fig. 2 of Ref. [1], we fixed $T_{c,1}$, $T_{c,2}$ and $\epsilon_{1,2}$ and kept $\overline{\alpha}_{1,2}$, $\nu_1 = 1 - \nu_2$ and γ/d as free parameters. The fit results are shown in the first 4 rows of table I.

	Data type	w (nm)	R (μm)	$T_{c,1}$ (K)	$T_{c,2}$ (K)	$\overline{\alpha}_1$ (μm^{-2})	$\overline{\alpha}_2$ (μm^{-2})	ϵ_1 (μm^{-4})	ϵ_2 (μm^{-4})	γ/d (μm^{-2})	ν_1	J_c (kA/cm^2)
1	T-dep.	370	1.2	1.53	1.98	-205	-414	4545	4545	64	0.78	196
2	T-dep.	320	2.0	1.53	1.98	-241	-389	4545	4545	22	0.76	63
3	T-dep.	250	1.2	1.52	1.98	-189	-366	4545	4545	7.6	0.80	26
4	T-dep.	190	0.8	1.52	1.98	-219	-728	4545	4545	2.2	0.87	5
5	T-dep.	190	2.0	1.53	1.98	-136	-736	4545	4545	0.96	0.81	3
6	$\Phi_a - I$	190	2.0	1.56	1.80	-94	-1325	4783	4475	1.2	0.37	3
7	simul.	190	2.0	1.53	1.90	-123	-362	2278	2274	0.24	0.66	1

TABLE I: The first four rows show the parameters from the fits in Fig. 2 of Ref. [1]. Row 5 and 6 contain parameters from the ring in Fig. 3, obtained from the T-dependence of λ^{-2} and ξ_{GL} and $\Phi_a - I$ curves as in Fig 3(d), respectively. The parameters in the last row were used for the simulation of hysteretic $\Phi_a - I$ curves.

In the 1-OP case, λ^{-2} and ξ_{GL} give a complete description of the $\Phi_a - I$ for a given n and geometry. If two-OP effects are manifest in a $\Phi_a - I$ curve on the other hand, its more complex shape contains additional information, which can be used to check the consistency with the 2-OP GL-coefficients obtained from the temperature dependence. We have done this by fitting all the relevant $\varphi - I$ curves from the one ring presented in Fig. 3 of Ref. [1] with fixed values for $\beta_{1,2}$, obtained from the temperature dependence fits. This constraint was necessary as otherwise there would be a wide range of parameter combinations consistent with the data. From the results, we have extracted the parameters listed in row 6 of table I by fitting the temperature dependences of $\alpha_{1,2}$ with a linear model and converting all parameters to the definitions used to model the temperature dependence. The results differ by up to a factor of about 2 from the values obtained from $\xi_{GL}(T)$ and $\lambda^{-2}(T)$ of the same ring (row 5). It is possible that the disagreement could be reduced by a better choice of the fixed parame-

ters. However, we speculate that it may partly be due to fluctuation effects in ψ_1 at large φ, where $|\psi_1|$ is strongly suppressed, which go beyond the scope of our model.

Row 7 of table I shows the input parameters for the numerical modeling of hysteretic $\Phi_a - I$ curves with different phase winding numbers, shown in Fig. 3 (g)-(i) of Ref. [1]). The phenomenological parameters $\kappa_1 \approx 6$ and $\kappa_2 \approx 25$ were chosen for best similarity between the simulations and data. Our transition criterion $E_{act}(\varphi - n) < \kappa_i k_B T$ was motivated as a condition for the thermally activated transition rate $1/\tau_0 e^{-E_{act}(\varphi)/k_B T}$ to become comparable to the experimental time scale $t_{exp} \sim 0.1$ s [7, 8]. Identifying κ with $\log(t_{exp}/\tau_0)$, this corresponds to attempt rates of $1/\tau_0 \sim 5000$ s^{-1} and 10^{11} s^{-1} for ψ_1 and ψ_2, respectively. Given that those rates depend exponentially on the $\kappa_{1,2}$, the unphysical values (particularly for ψ_1) do not represent an excessively large inconsistency. Using row 5 as input parameters, the similarity of the simulation results with the data was less convincing.

The line width dependence of the parameters in table I shows a clear trend in γ, corresponding to the crossover from intermediate coupling to nearly complete proximitization. There is also evidence for an increase of $\overline{\alpha}_2 = \xi_{GL,2}^{-2}$ with decreasing w, indicating a shorter mean free path in the narrower rings. This is likely due to the same effect that eliminates ψ_2 altogether in rings with $w \leq 120$ nm. However, the opposite trend in $\overline{\alpha}_1$ suggests that the sampling of both layers by electronic wave functions at stronger coupling may also play a role in determining the $\overline{\alpha}_{1,2}$. All other parameters show no strong dependence on w.

Near T_c, the thermodynamic critical field H_c is given by the relations $8\pi^2 H_c(T)^2 \Phi_0^2 = \lambda^{-2}\xi_{GL}^{-2} = \alpha(T)^2/\beta$. Due to deviations from the linear temperature dependence of ξ_{GL}^{-2} and λ^{-2} implied by our GL model, the second equality breaks down at low T. Assuming a phenomenological temperature dependence $H_c(T) = H_c(0)(1 - (T/T_c)^2)$, and matching it to the GL result near T_c, one obtains $\epsilon = \overline{\alpha}^2/\beta = 32\pi^2 H_c(0)^2 \Phi_0^2$. $\epsilon = 4500\mu\text{m}^{-4}$, as used in most of the fits, thus corresponds to $H_c(0) = 80$ G. Given the extrapolation involved in this calculation, this is in reasonable agreement with the bulk value $H_c(0) = 100$ G. Taking into account that the enhanced T_c should also increase the condensation energy makes the agreement slightly worse.

To estimate the critical interlayer Josephson current density J_c at low T from the coupling constant γ, we use the relation $J_c(T) = 2.67 J_c(0)(1 - T/Tc)$ [2, 3], which is valid near T_c for symmetric junctions between identical superconductors. The dissimilarity between the two layers in our rings should only lead to correction factors of order unity. Including prefactors and approximating the amplitudes by their values in the uncoupled system, the contribution of the coupling term to Eqn.(1) per unit area is

$$E_J(T) = \frac{\Phi_0^2}{8\pi^2\mu_0}\gamma|\psi_1||\psi_2| = \frac{\Phi_0^2}{8\pi^2\mu_0}\gamma\sqrt{\frac{\alpha_1(T)\alpha_2(T)}{\beta_1\beta_2}}$$

for $T \lessapprox Tc$, thus giving a critical current density

$$J_c(0) = \frac{\Phi_0}{2.67 \cdot 4\pi\mu_0}\gamma\sqrt{\frac{\overline{\alpha}_1(T)\overline{\alpha}_2(T)}{\beta_1\beta_2}}$$

The last column in table I shows the values obtained from the fit parameters.This can be compared to known junction fabrication processes: Extrapolating the Nb/Al/Al0_x/Nb trilayer process characterization in Ref. [9] to a 10 min, 10^{-6} mBar exposure, one obtains a critical current density on the order of 100 kA/cm^2, which compares well to the largest extracted values. Given that according to Ref. [9], a factor 100 reduction in critical current corresponds to a four orders of magnitude larger exposure, it seems unlikely that the oxidization process for smaller line widths can be thought of as analogous to the oxygen exposure of a thin film after deposition. It is more plausible that the co-deposition of aluminum and possible direct contact with the resist plays a role.

In conclusion, we have shown that different ways of analyzing and modeling our data in a two-OP GL framework give physically reasonable values for the GL coefficients that agree within about a factor 2. Thus, we find that the modeling supports the two-OP interpretation.

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