

# Temperature dependence of the superfluid density as a probe for multiple gaps in Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub>: Manifestation of three weakly interacting condensates

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The knowledge about the gap size and structure is of utmost importance for a theory of the superconducting pairing mechanism. The number of superconducting gaps is an important part of the description and modeling of multiband systems such as the iron-based superconductors. Here, we present a study on the temperature dependence of the superfluid density,  $\rho_s(T)$ , in Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub> obtained from terahertz spectra of conductivity and dielectric permittivity of thin film samples with critical temperatures  $T_c \approx 20$ –22 K. We demonstrate that the temperature dependence of the superfluid density,  $\rho_s(T)$ , can be explained best by a model of three interacting superconducting condensates. Our results refine the standard two-band approach for Co-doped BaFe<sub>2</sub>As<sub>2</sub>.

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## I. INTRODUCTION

Iron-based superconductors display an extreme diversity in their physical properties which is intimately related to their multiband structure (up to five bands) as well as with a subtle dependence of these properties on tiny impurity concentrations [1–3]. Enormous experimental efforts are being undertaken to obtain information about fundamental properties such as the number of superconducting (sc) energy gaps, their symmetry, or the type and strength of interband interaction. No consensus has been reached today on the symmetry of the sc order parameter: there are experiments that report *s-wave* [7–9] as well as *d-wave* [14–16] symmetry (see also the review article given in Ref. [17]). Theoretical approaches to the problem of superconductivity in these compounds can be therefore considered as rather diverse [18–26,28]. Reliable experimental data about the aforementioned issues will profoundly support a theoretical understanding of high temperature superconductivity in multiband systems.

Many experiments reveal two distinct gap sizes while other gaps were not resolved, called *clustering* of gap sizes (see Fig. 1 in Inosov *et al.* [27]). In order to reduce complexity it turned out to be sufficient to consider mainly two gaps or two effective bands as a *minimal model*. This leads to a description of the superconducting state with two effective gaps that are merely a composition of gaps and not necessarily coinciding with single gaps around each Fermi sheet (compare, e.g., Ref. [28]). However, strictly speaking, a model of superconductivity in Co-doped BaFe<sub>2</sub>As<sub>2</sub> should account for at least three gaps—around the inner and outer hole pockets at  $\Gamma$  (center of the Brillouin zone) and a gap around the electron pockets at the *M*-point [4,29] that slightly differs

from the gap size around the inner hole pockets. Based on optical spectroscopy and ellipsometry two gaps with sizes in the range of  $\Delta = 20$ –26.5 cm<sup>-1</sup>, and above 55 cm<sup>-1</sup> (see Table I), were found. A very small gap around  $\Delta = 15$  cm<sup>-1</sup> (corresponding to  $2\Delta = 2.1k_B T_c$ ) was identified in Co-doped BaFe<sub>2</sub>As<sub>2</sub> thin films by taking efforts in the terahertz regime [5]. Recent experiments point towards a more complicated structure of the superconducting state in Co-doped BaFe<sub>2</sub>As<sub>2</sub> [30], but it is still challenging to resolve the smallest gap in Co-doped BaFe<sub>2</sub>As<sub>2</sub> even by high resolution angle-resolved photoemission spectroscopy (ARPES).

In this regard, important information on the sc gaps can be obtained from terahertz (THz) measurements that allow one, in one experiment, to obtain the value of a small gap, optical characteristics, and the temperature dependence of the London penetration depth  $\lambda_L(T)$ . The self-consistent analysis of all data obtained with the terahertz spectroscopy allows one to significantly clarify the structure of the superconducting state of a multigap superconductor.

## II. TWO- VS THREE-BAND MODEL

In an earlier paper [31] we have discussed the normal state conductivity of Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub> as being determined by two subsystems I and II of charge carriers with substantially different conductivities,  $\sigma_{II}^n/\sigma_I^n \approx 0.1$ . We have determined the optical characteristics of these subsystems and resolved a small energy gap  $\Delta \approx 15$  cm<sup>-1</sup> with *s-wave* symmetry. It was shown that this gap is related to the highly conducting subsystem I. The available ARPES data do not allow one to unambiguously identify the structure of the superconducting subsystems I and II. According to the data of Brouet *et al.* (see Fig. 7 in Ref. [32]) and the discussion in Ref. [31], two scenarios are possible for Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub>. In the first one, just one highly conducting electron band is assigned to subsystem I

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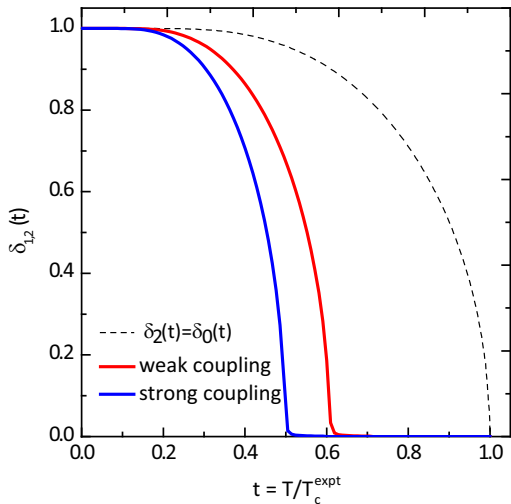


FIG. 1. (Color online) Calculated temperature dependence of the smaller gap  $\delta_1$  of a two-band superconductor with very weak ( $10^{-5}$ ) interband coupling. Blue line,  $\tilde{\alpha}_1 \approx 0.9$ ; red line,  $\tilde{\alpha}_1 \approx 1.1$ . Dashed line shows the large gap  $\delta_2(t)$  that coincides in this case with the universal function  $\delta_0(t)$  of the BCS theory.

and two hole bands to subsystem II. In this case, the detailed structure of the hole subsystem II (gap values and their symmetries), due to relatively low conductivity, has only small influence on the optical properties of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ . Following the analysis  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  must be considered a two gap superconductor. According to the second scenario, subsystem I has two bands: an electronic band and an outer hole band. Then,  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  must be considered a three-gap superconductor. We further demonstrated that the temperature dependency of  $\lambda_L(T)$  could be described in general in a simple two-band approach by considering a strongly coupled subsystem II within the  $\alpha$  model [33] and subsystem I within a weak coupling BCS model. Values  $\Delta_1(0) \approx 15 \text{ cm}^{-1}$  in subsystem I [ $\alpha_1 = \Delta_1(0)/T_c \approx 1.1$ ],  $\Delta_2(0) \approx 30 \text{ cm}^{-1}$  [ $\alpha_2 = \Delta_2(0)/T_c \approx 2.2$ ] in subsystem II, along with a weak interband coupling were found.

However, this simple model was not able to fully reproduce the dependence of the superfluid density  $\rho_s(t) = \lambda_L^2(0)/\lambda_L^2(t)$

TABLE I. Experimentally obtained gap values for  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  in comparison (selected publications).

$x$	$T_c$ (K)	Gap values ( $\text{cm}^{-1}$ )				Ref.
		$\Delta_1$	$\Delta_2$	$\Delta_3$	$\Delta_4$	
0.065	24.5	$<20^a$	26.5	40	78.5	[6]
0.07	23.0		25		56.5	[10]
0.075	25.5			$43 \pm 1^b$	$53 \pm 4^c$	[4]
0.08	25.0		20		60.5	[11]
0.08	22.5		25			[12]
0.1	22.0		24		64	[13] <sup>d</sup>
0.1	20.0	$15.5 \pm 0.5$				[5] <sup>d</sup>

<sup>a</sup>Speculative.

<sup>b</sup>Measured for the electron bands around the  $M$  point.

<sup>c</sup>Measured for the inner hole bands around the  $\Gamma$  point.

<sup>d</sup>Measured on a thin film.

on the reduced temperature  $t = T/T_c$ , especially at temperatures  $t \approx 0.4$ – $0.7$  [ $T = 7$ – $14 \text{ K}$ —close to the BCS critical temperature of the isolated subsystem I:  $T_c = \Delta_1(0)/1.76 \approx 12 \text{ K}$ ].

In contrast to the studied temperature dependence of the penetration depth from Ref. [31] the superfluid density  $\rho_s$  is proportional to a linear combination of the order parameters  $\Delta_J$  of the bands

$$\rho_s \sim \Delta_1(T) \tanh \frac{\Delta_1(T)}{2T} + \frac{\sigma_2^n}{\sigma_1^n} \Delta_2 \tanh \frac{\Delta_2(T)}{2T}. \quad (1)$$

We want to point our attention here to the superfluid density since its temperature dependence is more sensitive to the detailed behavior of  $\Delta_J(T)$  and, therefore, more appropriate in the analysis of subtle deviations from the common two-band approach. As we will show by using  $\rho_s(t)$  differences between two- and three-band models can be better recognized.

In the present work, to clarify the above mentioned possibilities, we rely on additional measurements of terahertz response on  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  films with similar sc characteristics and comparable critical temperatures (see Appendix for further information). The obtained temperature behavior of the superfluid density was thus reproduced. Details of the terahertz measurements are described in Refs. [5].

We have investigated both scenarios mentioned above. First, based on a more general approach that considers strong coupling corrections in both subsystems, we have analyzed a possibility of describing the properties of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  within a two-band model with  $\Delta_{\min} \approx 15 \text{ cm}^{-1}$ . Secondly, we have considered a three-gap model of superconductivity of this compound. Our analysis shows that three bands are needed (scenario 2) to satisfactorily describe the temperature dependence of  $\rho_s(t)$  of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  assuming sc gap values of  $\Delta_1 \approx 15 \text{ cm}^{-1}$ ,  $\Delta_2 \approx 21 \text{ cm}^{-1}$ , and  $\Delta_3 \approx 30$ – $35 \text{ cm}^{-1}$ . Our findings are in accordance with experimental data on infrared spectra of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  films [30].

### III. SUPERFLUID DENSITY STRUCTURE IN $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$

Properties of multiband superconductors are usually studied within the BCS formalism since their consistent analysis in a standard strong coupling theory is rather complicated. The main drawback of the BCS approach, however, is an underestimation of the normal quasiparticle density as a consequence of neglecting retardation and damping effects of the electron-boson interaction (EBI). As a result the critical temperature  $T_c^{\text{BCS}}$  of strongly coupled superconductors is significantly overestimated [34,35]. This discrepancy is removed by empirically introducing effective temperatures into the BCS distribution function that provide the same critical temperatures (calculated and measured  $T_c^{\text{expt}}$ ) that take into account the increased number of quasiparticles in strongly coupled superconductors. For a single-band superconductor this temperature is given by  $T_c^{\text{expt}}$  ( $\alpha$  model) [33,36].

#### A. Two bands

Consider first a two-band system: besides  $T_c^{\text{expt}}$  also an effective temperature  $T^*$  for the band with the smaller gap

(denoted here by  $\Delta_1$ ) is necessary. The BCS-like equations formally coincide with regular BCS equations. In particular, the BCS-like expressions for reduced gaps  $\delta_J(t) = \Delta_J(t)/\Delta_J(0)$  as a function of the reduced temperature  $t = T/T_c^{\text{expt}}$  are written as ( $J = 1, 2$ ) [36]

$$\ln \delta_1(t) = -\tilde{n}_1(t) - \tilde{\Lambda}_{12}[1 - \delta_2(t)/\delta_1(t)], \quad (2)$$

$$\ln \delta_2(t) = -\tilde{n}_2(t) - \tilde{\Lambda}_{21}[1 - \delta_1(t)/\delta_2(t)], \quad (3)$$

where  $\tilde{n}_J(t)$  is the contribution of intraband quasiparticles of the  $J$ th band,

$$\tilde{n}_J(t) = 2 \int_0^\infty d\omega \frac{f[\tilde{\alpha}_J \epsilon_J(\omega)/t]}{\epsilon_J(\omega)}, \quad (4)$$

$$\epsilon_J(\omega) = \sqrt{\omega^2 + \delta_J^2(t)}. \quad (5)$$

Here,  $f$  is the Fermi distribution function and  $J$  is the reduced spectrum of a superconductor. The effective constants of interband interaction,  $\tilde{\Lambda}_{12}$  and  $\tilde{\Lambda}_{21}$ , are given by

$$\tilde{\Lambda}_{12} = \tilde{\lambda}_{12}/\theta(0), \quad \tilde{\Lambda}_{21} = \tilde{\lambda}_{21}\theta(0), \quad (6)$$

where  $\theta(0) = \Delta_1(0)/\Delta_2(0)$  is connected with the renormalized EBI constants  $\tilde{\lambda}_{IJ}$  [36]. It can be shown that the effective parameters  $\tilde{\alpha}_{1,2}$  of the distribution function in (4) are determined by the condition of equal critical temperatures in the bands

$$\ln \frac{\tilde{\alpha}_2}{\alpha_0} = \frac{\tilde{\Lambda}_{21}}{\tilde{\Lambda}_{12} + \ln \frac{\alpha_0}{\tilde{\alpha}_1}} \ln \frac{\alpha_0}{\tilde{\alpha}_1}, \quad (7)$$

where  $\alpha_0 = \pi/\gamma_E \approx 1.764$ , and  $\tilde{\alpha}_1 = \Delta_1(0)/T^*$  is experimentally obtained.

### B. Application to Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub>

The temperature dependence of the superfluid density  $\rho_s(t)$  of strongly coupled superconductors [37] written for a multiband superconductor reads as

$$\rho_s(t) = \frac{\sum \tilde{\rho}_s^J(t)}{\sum \tilde{\rho}_s^J(0)}, \quad (8)$$

where the contribution of the  $J$ th band is [31]

$$\tilde{\rho}_s^J(t) = \sigma_n^J \alpha_J^{\text{expt}} \delta_J(t) \left\{ \tanh \frac{\alpha_J^{\text{expt}} \delta_J(t)}{2t} - \frac{2}{\pi} \int_0^\infty d\omega \frac{\delta_J(t) \tilde{\gamma}_{\text{imp}}^J / \alpha_J^{\text{expt}}}{\omega^2 + (\tilde{\gamma}_{\text{imp}}^J / \alpha_J^{\text{expt}})^2} \times \frac{\tanh[\alpha_J^{\text{expt}} \epsilon_J(\omega)/2t]}{\epsilon_J(\omega)} \right\}, \quad (9)$$

with  $\alpha_J^{\text{expt}} = \Delta_J(0)/T_c^{\text{expt}}$ ,  $\sigma_n^J$  is the normal state static (dc) conductivity,  $\tilde{\gamma}_{\text{imp}}^J = \gamma_{\text{imp}}^J/T_c^{\text{expt}}$  and  $\gamma_{\text{imp}}^J = 1/2\tau_J$  is the intraband relaxation rate of elastic impurity scattering and the sc gaps  $\delta_J(T)$  are determined by BCS-like equations (2)–(4).

The value  $\Delta_2(0)/T_c^{\text{expt}} \approx 2.2$  found for Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub> in Ref. [31] points towards strong coupling.

The coupling in subsystem I can be determined only indirectly from the BCS-like estimate of the ratio of  $\Delta_1(0)$  to the

critical temperature  $T_c^{(i)}$  of the isolated ( $\tilde{\Lambda}_{12} = 0$ ) subsystem I that is determined from Eqs. (2) and (4) via the condition  $\tilde{\alpha}_1 T_c^{\text{expt}}/T_c^{(i)} = \alpha_0$  or by an equivalent relation  $\Delta_1(0)/T_c^{(i)} = \alpha_0 T^*/T_c^{\text{expt}}$ . The latter equation fully determines the interval of possible values of  $T^*$ : from  $T_c^{\text{expt}}$  [ $\Delta_1(0)/T_c^{(i)} = \alpha_0$ , weak intraband EBI] up to the BCS critical temperature of the two-band superconductor  $T_c^{\text{BCS}} \approx \Delta_2(0)/\alpha_0$  [ $\Delta_1(0)/T_c^{(i)} \approx \Delta_2(0)/T_c^{(\text{expt})} = 2.2$ , strong intraband EBI].

Figure 1 demonstrates the possible behaviors of the gaps  $\delta_{1,2}(t)$  in a superconductor with *two-gap* parameters, Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub>, for a weak intraband EBI in the subsystem with the small gap ( $\tilde{\alpha}_1 \approx 1.1$ ) and for a strong intraband EBI ( $\tilde{\alpha}_1 \approx 0.9$ ). The specific values of the effective temperature  $T^*$  and of  $\tilde{\alpha}_1$  for this superconductor can be found from experiments only.

In our calculations the experimentally determined parameters of the subsystems were varied within the measurement uncertainties and the values of  $\tilde{\alpha}_1$  within physically allowable ranges and it was found that the temperature dependence  $\rho_s(t)$  cannot be satisfactorily interpreted by only a two-band model assuming a gap  $\Delta_{\text{min}} \approx 15 \text{ cm}^{-1}$  (Fig. 2). We stress that the dependence shown in Fig. 2 is the most favorable for the two-band model that cannot be improved any more and that our statement about the insufficiency of that model is *precise*.

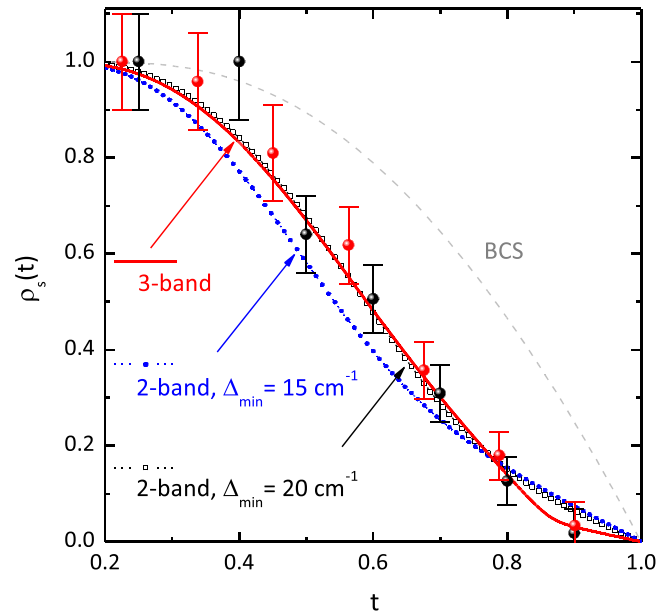


FIG. 2. (Color online) Experimental values for the superfluid density  $\rho_s(t)$  for Ba(Fe<sub>0.9</sub>Co<sub>0.1</sub>)<sub>2</sub>As<sub>2</sub> determined in a thin film with  $T_c = 20 \text{ K}$  (black symbols, thickness  $\approx 90 \text{ nm}$ ) and in a thin film with  $T_c = 22 \text{ K}$  (red symbols, thickness  $\approx 50 \text{ nm}$ ). Differences in the two-band fit (with parameters from Ref. [31], dotted blue line) and in the three-band fits for an anisotropic, i.e.,  $s + d$ -wave (red line) scenario with parameters from Table II. Within the interval  $t = 0.4\text{--}0.7$  the discrepancy between the two-band and the three-band models exceeds the experimental uncertainty (see the Appendixes). The solutions according to the two-band fit with  $\Delta \approx 20 \text{ cm}^{-1}$  (black dotted line) and a BCS one-band approach (dashed gray line) are shown as well.

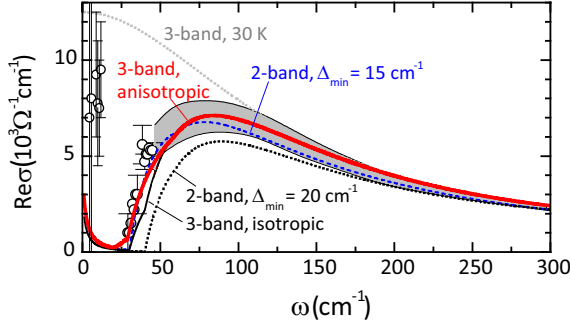


FIG. 3. (Color online) Optical conductivity  $\text{Re } \sigma(\omega)$  of the  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  films (open circles at terahertz frequencies and gray area at the infrared; data from Ref. [5]). Although a two-band model (dashed blue line) would fit, it is not able to fully reproduce  $\rho_s(t)$  given in Fig. 2. The anisotropic three-band model (red line) fits  $\text{Re } \sigma(\omega)$  better than the isotropic three-band model (black line). The black dotted line corresponds to a two-band calculation with  $\Delta \approx 20 \text{ cm}^{-1}$  that is fitting the superfluid data equally well as the anisotropic three-band model (Fig. 2), but drastically fails in a description of  $\text{Re } \sigma(\omega)$ . In contrast, the three-band model is capable of describing both  $\rho_s(t)$  and  $\text{Re } \sigma(\omega)$ . All calculations are done for  $T = 5 \text{ K}$  except the normal state (gray dotted line) that was taken from the spectrum at  $30 \text{ K}$ .

Increasing the value of  $\Delta_{\min}$  to  $20 \text{ cm}^{-1}$  would give a much better fit for  $\rho_s(t)$ . (We present this scenario also in Figs. 2 and 3.) However, the solution with  $\Delta_{\min}$  to  $20 \text{ cm}^{-1}$  within a two-band model does not confirm the real part of the optical conductivity,  $\text{Re } \sigma(\omega, T)$  (Fig. 3). This means that the first scenario practically cannot hold because subsystem I of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  is actually already a two-band (electron and hole) subsystem with the spectrum having a minimal gap  $\Delta_1(0) \approx 15 \text{ cm}^{-1}$  and one more gap—an intermediate one with the value  $> 20 \text{ cm}^{-1}$  (second scenario).

This intermediate gap cannot be isotropic since the behavior of the conductivity,  $\text{Re } \sigma(\omega, T = 5 \text{ K})$ , in such a case would conflict with the experimentally observed behavior (Fig. 3). Since the interband interaction in  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  is rather small, the origin of the anisotropy of the gap in the intermediate band 2 can be connected only with the anisotropy of the intraband interaction  $w_{22}$ . Such interaction can exist in an electronic band that has an *intraband* scattering with vectors comparable with the reciprocal lattice vectors. Our assumption that the intermediate gap relates to the electronic subsystem is confirmed by recent ARPES experiments, where a very small gap is found around the outer  $d_{xy}$  hole pocket (Borisenko and Evtushinsky [38]).

According to the second scenario, three gaps should appear in the electronic spectrum of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ : minimal gap  $\Delta_{\min}(0) \approx 15 \text{ cm}^{-1}$  (possibly around the outer hole pocket,  $J = 1$ ), intermediate gap (possibly around the electron pocket,  $J = 2$ ), and maximal gap  $\Delta_{\max}(0) \approx 30 \text{ cm}^{-1}$  (around the inner hole pocket,  $J = 3$ ) that determines the critical temperature  $T_c$ . To estimate the empirical strong coupling parameters  $\tilde{\alpha}_j$  in the three-band model, we have used the results obtained from tunnel spectra of  $\text{Mg}_{1-x}\text{Al}_x\text{B}_2$  [35,39].

Because of rather large experimental uncertainties in  $\rho_s(t)$  of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$ , it is enough for our purpose to use a

TABLE II. Parameters for the anisotropic three-band model of superconductivity in  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  with  $\Delta_2(0) = \Delta_2(\pi/4, 0)$ .

$J$	$\sigma_J^s (\text{cm}^{-1})$	$\gamma_{\text{imp}}^J (\text{cm}^{-1})$	$\tilde{\Lambda}_{J3}$	$\Delta_J(0) (\text{cm}^{-1})$	$\alpha_J^{\text{expt}}$
1	6000	75	0.05–0.1	15	1.1
2	6000	55	0.05–0.1	21	1.46
3	500	200		30	2.2

simplified model that takes into account interband coupling constants  $\tilde{\lambda}_{13}, \tilde{\lambda}_{23}$  of the bands  $J = 1, 2$  with the inner hole pocket ( $J = 3$ ) and an anisotropic spectrum in band 2 assuming weak impurity induced renormalization of the sc gap. For the dependence of polar angles  $\varphi, \varphi'$  on the two-dimensional circular Fermi surface interaction  $w_{22}(\varphi, \varphi') = u_\varphi \lambda_{22} u_{\varphi'}$ , we get

$$\Delta_2(\omega, \varphi, t) \approx \bar{\Delta}(0) \delta_2(t) \beta(\varphi, t), \quad (10)$$

where  $\beta(\varphi, t) = u_\varphi + \lambda_{22} \tilde{\Lambda}_{23} \delta_3(t) / \delta_2(t)$ , and  $\lambda_{22} \approx 0.3$  is the intraband coupling constant [31,36]. The equation for the reduced gap  $\delta_2(t)$  can be obtained in full analogy with Eqs. (11)–(13) from Ref. [36] taking into account an anisotropic electronic spectrum [see Eq. (21) in Ref. [36]]:

$$\ln \delta_2(t) + \langle \bar{\beta}(\varphi, t) \ln \beta(\varphi, t) - \bar{\beta}(\varphi, 0) \ln \beta(\varphi, 0) \rangle = -2\tilde{n}_2(t) + \frac{\tilde{\Lambda}_{23}}{\langle u_\varphi \rangle \bar{\beta}(\varphi, 0)} \left\{ \frac{\delta_3(t) \bar{\beta}(\varphi, 0)}{\delta_2(t) \bar{\beta}(\varphi, t)} - 1 \right\}, \quad (11)$$

$$\tilde{n}_2(t) = \left\langle \bar{\beta}(\varphi, t) \int_0^\infty d\omega \frac{f(\tilde{\alpha}_2 \varepsilon_2(\omega, \varphi, t) / t)}{\varepsilon_2(\omega, \varphi, t)} \right\rangle, \quad (12)$$

$$\varepsilon_2(\omega, \varphi, t) = \sqrt{\omega^2 + \delta_2^2(t) \beta^2(\varphi, t)}, \quad (13)$$

where the averages are  $\langle F \rangle \Rightarrow \frac{1}{2\pi} \left\langle \int_0^{2\pi} u_\varphi F d\varphi \right\rangle$  and  $\bar{\beta}(\varphi, t) = \beta(\varphi, t) / \langle \beta(\varphi, t) \rangle$ . The equations for bands 1 and 3 in our model are

$$\delta_3(t) = \delta_0(t), \quad (14)$$

$$\ln \delta_1(t) = -\tilde{n}_1(t) - \tilde{\Lambda}_{13} \{1 - \delta_0(t) / \delta_1(t)\}. \quad (15)$$

The results of the calculations of the optical conductivity  $\text{Re } \sigma(\omega)$  for temperatures  $T = 5 \text{ K}$  and  $30 \text{ K}$  and of the superfluid density  $\rho_s(t)$  in  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  with anisotropic  $w_{22}(\varphi, \varphi')$  mixed  $s + d$  wave interaction in which the  $d$  part has a standard Monthoux-Pines form [20,21,40],  $u_\varphi = 1 + k_d \cos 2\varphi$ , are shown in Figs. 2 and 3, for the values given in Table II and with  $\tilde{\alpha}_{1,2} = \alpha_{1,2}^{\text{expt}}$ ,  $k_d = 0.5$ , together with the experimental data.

### C. Insufficiency of the two-band approach

The insufficiency of the two-band approach is not based on the empirical parameters but on the precise result in  $\rho_s(t)$ . Using the two-band approach, a reasonable description for the full temperature regime can only be achieved with  $\Delta_{\min} = 20 \text{ cm}^{-1}$ , that is, however, in disagreement with the experimental observation of a gap with  $\Delta_{\min} = 15 \text{ cm}^{-1}$  (Fig. 3).

A two-band model with  $\Delta_{\min} = 15 \text{ cm}^{-1}$  that guarantees a fit to  $\text{Re } \sigma(\omega)$  does not accurately enough describe the temperature dependence of  $\rho_s(t)$ . Therefore, a third band is introduced in order to obtain reasonable fits for both  $\rho_s(t)$  and  $\sigma(\omega)$ .

The best fit to the experiment [ $\rho_s(t)$  and simultaneously  $\text{Re } \sigma(\omega)$ ] is obtained in the case of not one, but *two* superconducting condensates in subsystem I [ $\Delta_1(0) \approx 15 \text{ cm}^{-1}$  and  $\Delta_2(0) \approx 21 \text{ cm}^{-1}$ ] that are weakly ( $\tilde{\Lambda}_{I,II} \approx 0.1$ ) interacting with the inner hole pocket superconducting condensate [ $\Delta_3(0) \approx 30 \text{ cm}^{-1}$ ]. The  $\sigma(\omega, T)$  data (Fig. 3) demonstrates that a two-band model with  $\Delta_{\min} = 20 \text{ cm}^{-1}$  falls outside the experimental data points. A three-band model approach is thus favored. Note that the intermediate gap  $\Delta_2$  (presumably in the electron bands) must be weakly (nodeless) anisotropic and, in particular, may have a mixed  $s + d$  symmetry. We also want to point out that this result is in accordance with the recent infrared reflectivity experiments in Ref. [30] performed on  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  film with  $T_c \approx 20 \text{ K}$ . In Ref. [30] signatures of two small gaps  $\Delta_1(0) \approx 15 \text{ cm}^{-1}$  and  $\Delta_2(0) \approx 21 \text{ cm}^{-1}$  have been detected.

#### IV. CONCLUSIONS

The temperature dependence of the superfluid density,  $\rho_s(t)$ , obtained from THz-IR measurements of the optical conductivity and the dielectric permittivity of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  thin films revealed a refinement of the standard two-gap scenario. Overdoped  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  must be considered as a system with three BCS-like condensates with significantly different superconducting gaps. The refinement of the two-band model becomes necessary because of conflicting results when applying a two-band model to  $\text{Re } \sigma(\omega)$  and  $\rho_s(t)$  simultaneously: best fit values for  $\Delta_{\min}(0)$  differ in the description of the temperature dependence of the superfluid density,  $\rho_s(t)$ , from the description of  $\text{Re } \sigma(\omega)$ . This problem can be resolved by using a three-band model. Based on the analysis of  $\rho_s(t)$  three weakly coupled condensates with gaps of  $\Delta_1 \approx 15 \text{ cm}^{-1}$ ,  $\Delta_2 \approx 21 \text{ cm}^{-1}$ , and  $\Delta_3 \approx 30\text{--}35 \text{ cm}^{-1}$  were identified. The used model parameters are in agreement with  $\text{Re } \sigma$ . We would like to once again emphasize that in our case above the minimalistic three-band model provides a good description of the optical data  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  and thus can be used for a qualitative study of the properties of Co-doped  $\text{BaFe}_2\text{As}_2$ .

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#### APPENDIX A: EXPERIMENTAL METHODS

Thin films of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  used in this study were grown by pulsed laser deposition (KrF laser) on  $(\text{La,Sr})(\text{Al,Ta})\text{O}_3(100)$  substrates at a temperature of  $700^\circ\text{C}$  under vacuum conditions of  $p = 10^{-8}$  mbar. The thickness of the films is about 40 nm and 90 nm. The critical transitions have been found by resistive measurements at  $T_c \approx 20$  and 22 K, respectively. For more detailed information on thin film growth of Fe-pnictide superconductors, see Ref. [41].

The optical experiments based on the measurements of complex transmissivity allow one to determine the minimal energy gap from the optical conductivity spectra (real part) and the superfluid density from the imaginary part of the optical conductivity  $\sigma(\omega, T)$ . The technique is unique in the sense that it allows one to mark absolute error bars for the superfluid density, and these bars are in fact within standard limits. Differences between the two-band and the three-band models (as given in Fig. 2) can be found in  $\rho_s(t)$  in the temperature range  $t = 0.4\text{--}0.7$ . Numerically the differences are 0.1 (at  $t = 0.5$ ) with an error bar of  $\pm 0.08$  (total 0.16), 0.09 (at  $t = 0.56$ ) with an error bar of  $\pm 0.08$  (total 0.16), and 0.1 (at  $t = 0.6$ ) with an error bar of  $\pm 0.07$  (total 0.14).

#### APPENDIX B: BELOW SC-GAP ABSORPTION

The problem of the below-sc gap (low frequencies) absorption is well known, not only in pnictides, but also in cuprate superconductors. At these frequencies the real part of the complex conductivity  $\text{Re } \sigma$ , that is proportional to the absorptivity, is much smaller than the imaginary part  $\text{Im } \sigma$ . This leads to very large error bars in  $\text{Re } \sigma$ . In Fig. 3 we reproduce the full set of corresponding experimental data for  $\text{Re } \sigma$  (including from Ref. [5]) that shows that the uncertainty can reach values of up to  $\pm 100\%$ . At present, no theory can account for this below-gap absorption that can be of intrinsic but also of extrinsic origin. However, most importantly, the low frequency region does not influence the conclusions drawn above.

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