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Comment on "Precision global measurements of London penetration depth in FeTe_{0.58}Se_{0.42}"

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Cho *et al.* [Phys. Rev. B **84**, 174502 (2011)] have reported on the temperature dependence of the London penetration depth deduced from tunnel diode oscillator (TDO) measurements in optimally doped Fe(Se,Te) single crystals. According to their analysis, these measurements could suggest a nodeless two-gap pairing symmetry with strong pair-breaking effects. However, to reach this conclusion, the authors fit the temperature dependence of the superfluid density with a two band *clean* limit model, which is incompatible with the presence of strong pair-breaking effects, deduced from the T^n temperature dependence of the London penetration depth below $T_c/3$. Moreover, they claim that their results are also ruling out the suggestion that surface conditions can significantly affect the TDO data, but this conclusion is based on one very specific damaging process and completely ignores the large dispersion in the previously published TDO data.

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In a recent article, Cho *et al.*¹ reported on the temperature dependence of the London penetration depth deduced from tunnel diode oscillator (TDO) measurements in a series of Fe(Se,Te) crystals close to optimal doping. TDO is a powerful technique to very accurately determine the temperature dependence of the variation in the penetration depth $[\Delta\lambda(T)]$, especially for $T \rightarrow 0$ and, hence, to obtain valuable information on the low-energy excitations present in the system. All the measurements performed by several groups on Fe(Se,Te) samples from different origins agree, for instance, on the fact that, at low temperatures, $\Delta\lambda(T)$ can be well described by a power law $\Delta\lambda(T) \propto T^n$ with $n \sim 2.0-2.3$.^{1–4} This power law suggests the presence of pair-breaking effects as expected in dirty *d*-wave superconductors or in the case of interband scattering in $s\pm$ superconductors.⁵

However, to obtain a complete description of the gap structure, it is necessary to determine the temperature dependence of the normalized superfluid density (ρ_s) on the entire temperature range. As $\rho_s(T) = 1/[1 + \Delta\lambda(T)/\lambda_0]^2$, both the amplitude of $\Delta\lambda(T)$ and the zero-temperature penetration depth λ_0 have to be determined precisely to obtain reliable $\rho_s(T)$ data. Nevertheless, very different $\Delta \lambda$ values have been reported in samples with very similar T_c values (optimally doped samples) by the different groups involved in the study of Fe(Se,Te). Indeed, taking, for instance, T = 5 K for comparison purposes, the different $\Delta\lambda(5 \text{ K})$ values vary from \sim 35 nm for the Bristol group³ to \sim 130 nm for the Grenoble group,⁴ and values ranging from ~ 30 to 50 nm in Ref. 1 to \sim 110 nm in Ref. 2 have been reported by the Ames group. It is, hence, of fundamental importance to understand the origin of this dispersion to obtain unambiguous $\rho_s(T)$ data.

A possible influence of edge roughness in iron-based superconductors has been pointed out by Hashimoto *et al.*⁶ noting that $\Delta\lambda(T)$ can vary by a factor of 2 from one sample to another in KFe₂As₂. In Ref. 1, the authors claim that they have ruled out the possibility that surface roughness can significantly affect the amplitude of $\Delta\lambda(T)$ and that they have shown that the temperature dependence of the superfluid density is consistent with a nodeless two-gap pairing symmetry in the presence of strong pair-breaking effects. However, we believe that the data have been overinterpreted as: (a) only one specific kind of disorder has been investigated, (b) they used the same λ_0 value to obtain ρ_s in Refs. 1 and 2, even though the $\Delta\lambda(T)$ values differ by a factor of ~ 3 , and (c) the two-gap *clean* limit model used to fit the $\rho_s(T)$ data is incompatible with the presence of strong pair-breaking effects.

I. ON THE INFLUENCE OF EDGE ROUGHNESS ON $\Delta\lambda(T)$

In Ref. 1, this possibility has been rejected on the basis of measurements on sample "2-R" in which some roughness has been introduced by razor damaging and for which $\Delta\lambda(5 \text{ K})$ (only) increases by $\sim 60\%$ (and A is, subsequently, only marginally modified when the edges are cut back "as clean as possible"). Unfortunately, no structural information on the roughness introduced by the razor damaging is given, but it is hard to believe that such a procedure could be characteristic of all kinds of edge roughness. Indeed, the amplitude of the TDO signal will be sensitive to a roughness on the scale of λ_0 but would be only marginally affected by defects on much larger scales. Moreover, even if the dispersion in the $\Delta\lambda(T)$ values measured in Ref. 1 remains reasonable (\sim 30–50 nm), the authors completely ignore the large dispersion in the values previously published (including their own data, which differ by a factor \sim 3) stating that the values obtained in Ref. 1 are "similar to other reports." The origin of this large dispersion remains an open question. The amplitude of $\Delta\lambda$ might be affected by various parameters, such as sample inhomogeneities or disorder (see discussion below), microcracks, etc., . . ., but the influence of edge roughness cannot be excluded from the data presented in Ref. 1.

II. ON THE CHOICE OF THE PARAMETERS

In a clean *s*-wave superconductor (that is, for a mean-free path *l* larger than the coherence length ξ), $\lambda(0)$ depends only on the Fermi surface properties⁷ and is, hence, independent of the sample quality. However, it is now well established that strong pair-breaking effects are present in iron-based superconductors, and Fe(Se,Te) cannot be discussed within the framework of those standard materials. Indeed, in contrast to those later systems, all scattering events (and not only



FIG. 1. Temperature dependence of the superfluid density deduced from TDO in Ref. 2 (closed circles) and Ref. 1 (open circles) together with the one deduced from H_{c1} measurements in Ref. 4 (open squares) and muon spin rotation (μ SR) data in Ref. 9 (open crosses). The solid line is a $1 - t^2$ dependence (with $t = T/T_c$), and the shaded area corresponds to the ρ_s values expected from the specific heat jump at T_c (see text for details).

spin-flip scattering) are expected to be detrimental,⁸ and the absolute value of the superfluid density is reduced even for $T \rightarrow 0. \lambda(0)$ is then expected to depend strongly on T_c (see, for instance, Ref. 5) and, concomitantly, $\Delta\lambda(5 \text{ K})$ will be directly proportional to $\lambda(0)/T_c^2$ (for strongly reduced T_c values⁵). This close relationship between $\Delta\lambda$ and λ_0 has been recently established experimentally in a series of $Ba(Fe_xNi_{1-x})_2As_2$ crystals.⁹ A factor ~ 3 variation in $\Delta\lambda(5 \text{ K})$ (for samples with similar T_c values) should, hence, be directly related to a similar variation in $\lambda(0)$. However, the Ames group introduced the same $\lambda(0)$ value (measured in Ref. 2) to obtain $\rho_s(T)$ in Refs. 1 and 2, even though they measured a $\Delta\lambda(5 \text{ K})$ value \sim 3 times smaller in Ref. 1 than in Ref. 2. This inconsistent choice led, by construction, to very different temperature dependences of the superfluid density (see Fig. 1) and forced the authors to drastically change their conclusion from " $\rho_s(T)$ at temperatures on the order of T_c is fully described by only one component, determined by the band with a smaller gap" in Ref. 2 to "this result indicates that a 75% contribution of superfluid density comes from the band which has the larger gap" in Ref. 1, shedding doubts on the validity of any of those two conclusions (see also discussion below).

Note that the $\lambda(0)$ value used in Refs. 1 and 2 is actually consistent with those obtained by other groups (using different techniques): $\lambda_0 \sim 500 \text{ nm} \pm 15\%$ (Refs. 4 and 10), and all samples (close to optimal doping) presented very similar T_c values. As $\lambda(0)$ and T_c (Ref. 8) are expected to be very sensitive to scattering, this suggests a rather good homogeneity in the different Fe(Se,Te) crystals. Similarly, a large number of specific heat measurements^{4,11} also led to very similar values of the jump at $T_c: \Delta C_p/T_c \sim 40 \text{ mJ mol}^{-1} \text{ K}^{-2} \pm 20\%$, again confirming this homogeneity. It is, hence, difficult to attribute the dispersion in the $\Delta\lambda$ values to the sample bulk quality. However, as TDO measurements are sensitive to surfaces (for $T_c \rightarrow 0$), the influence of edge roughness-or any other surface inhomogeneity-cannot be excluded. Note also that the slope of $\rho_s(t)$ for $t \rightarrow 1$ (with $t = T/T_c$) is thermodynamically related to ΔC_p and to the slope of the upper critical field (B'_{c2}) $\Delta C_p = (\mu_0 T_c) \cdot (dH_c/dT)_{|T \to T_c}^2 \sim (B_{c2}'/\mu_0).$ through $(\Phi_0/4\pi\lambda_0^2) \cdot (d\rho_s/dt)_{|t\to 1}$. Introducing $B'_{c2} \sim 13 \text{ T/K}$ (Ref. 4), one obtains $(d\rho_s/dt)_{|t\to 1} \sim 2.6 \ (\pm 40\%)$ (see shaded area in Fig. 1). As shown in Fig. 1, the $\rho_s(T)$ data obtained in Ref. 1 are in good agreement with those deduced from H_{c1} measurements⁴ and μ SR data¹⁰ and are thermodynamically consistent with the specific heat data, whereas, those previously obtained in Ref. 2 (and TDO data in Ref. 4) clearly deviate from the thermodynamical cone.

III. ON THE FITTING PROCEDURE

It is, hence, in principle, possible to perform an analysis of the temperature dependence of the ρ_s values obtained in Ref. 1 (or, more generally speaking, on all the open symbols in Fig. 1). However, the T^n dependence measured at low temperatures by all the groups strongly supports the presence of pair-breaking effects. It is, hence, physically incorrect to fit the full temperature range dependence of this superfluid density using a two-gap model in the *clean* limit. Indeed, in this case, $\Delta \lambda$ should vary exponentially for $k_B T < \Delta_{\min}/5$, (where Δ_{\min} is the smallest gap), that is, for $T \leq 3$ K taking the Δ_{\min} value obtained in Ref. 1 in clear disagreement with their own data displaying a T^n law in this temperature range. Note, however, that the influence of scattering on the temperature dependence of the superfluid density in a two-gap system has been investigated in Ba(Fe,Co)₂As₂ by the Ames group in Ref. 12 and, since the temperature dependence observed in this latter system is very close to the one observed in Fe(Se,Te), it would have been much more valid to perform a similar analysis in Ref. 1.

Finally, note that $1/\lambda^2(T)$ is expected to vary as $1 - t^2$ in the pair-breaking model close to critical scattering (closed gap⁵). As shown in Fig. 1, this dependence (solid line) reproduces the experimental dependence very well without any adjustable parameter, whereas, *six parameters* (including T_c) have been used in Ref. 1 to "self-consistently" fit the data.

In conclusion, the origin of the large dispersion in the amplitude of $\Delta\lambda(T)$ obtained in Fe(Se,Te) crystals by TDO is still an open question, but the possibility that edge roughness might alter the amplitude of $\Delta\lambda(5 \text{ K})$ (Ref. 6) cannot be ruled out from the measurements presented in Ref. 1. Using the same λ_0 values to reconstruct the superfluid density in samples presenting very different $\Delta\lambda(T)$ (as performed in Refs. 1 and 2) is incorrect and leads, by construction, to very different temperature dependences for ρ_s . Finally, the power-law dependence of the penetration depth at low temperatures ($\Delta\lambda \propto T^n$) is not compatible with the *clean* limit two-gap model used to describe the temperature dependence of strong scattering here hinders any determination of the gap values from the temperature dependence of the superfluid density.

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