TF-\(\mu^+\)SR in overdoped Tl\(_2\)Ba\(_2\)CuO\(_{6+x}\): simplicity still elusive

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Abstract. Encouraged by predictions of simple Fermi liquid behavior in the overdoped versions of high temperature superconductors (HT\(_{SC}\)) based on CuO\(_2\) planes, we have performed TF-\(\mu^+\)SR experiments on several high quality oriented crystal mosaics of Tl\(_2\)Ba\(_2\)CuO\(_{6+x}\) ("Tl-2201") in the vortex-lattice state where the magnetic penetration depth \(\lambda\) can be extracted from the characteristic superconducting (SC) lineshape by \(\chi^2\)-minimization fitting in the time domain. The results show evidence for a surprising degree of disorder, causing some “smearing” of the characteristic lineshape features, but we were able to extract \(\lambda\) as a function of temperature, revealing a remarkably consistent \(T\)-dependence of the SC carrier density \(n_s \propto \lambda^{-2}\) that suggests this “role-model cuprate” is anything but “simple”. The extracted superfluid density for all dopings exhibits a clear inflection point with temperature, suggesting a vortex lattice transition. Such a transition would have important implications for other measurements.

In the cuprate HT\(_{SC}\)s, charge doping of the CuO\(_2\) planes engenders superconductivity (SC) between the limits of an undoped insulating antiferromagnet (AFM) and what is believed to be a conventional Fermi liquid (FL) state at high dopings. One would like to understand how the unconventional SC state evolves out of these more conventional electronic ground states. However, overdoped cuprates are rare and few have highly ordered CuO\(_2\) planes. The Tl\(_2\)Ba\(_2\)CuO\(_{6+x}\) (Tl-2201) system offers tunability throughout the overdoped regime with highly-ordered, flat CuO\(_2\) planes, doped \(via\) dilute interstitial oxygen atoms in the distant TlO layers [1]; the overdoping arises from substitution of copper on the thallium site at the \(\sim 8\%\) level [2, 3, 4] or self-doping to eliminate a predicted electron pocket at the \(\Gamma\) point [5].

Previous data on powder samples [6] was used to extend the “Uemura plot” [7] to overdoped cuprates, but such samples do not produce characteristic lineshapes allowing detailed analysis.

We recently reported the first TF-\(\mu^+\)SR results on high-quality single crystal mosaics of Tl-2201 for a range of dopings[8]; additional details are provided here. From the magnetic field distribution in the vortex state, we extract the temperature dependence of the SC order parameter and the magnetic penetration depth \(\lambda\). Surprisingly, these do not appear to follow the simple \(d\)-wave temperature dependence found in under- and optimally-doped cuprates[9], exhibiting instead an unexpected inflection point in a qualitatively different screening response.
Single crystals of Tl-2201 were grown in gold-sealed alumina crucibles by an encapsulated copper-rich self-flux method as described elsewhere [10]. It was necessary to vary the degree of sealing during the growth process, to allow evolved gases to escape, then to contain volatile and toxic Tl₂O during growth. The oxygen content was set by annealing under controlled oxygen partial pressures and temperatures [11]: two different annealing schemes were employed depending on the desired oxygen content [10]. Crystals were assembled in mosaics on substrates of aluminized mylar or GaAs, with the c-axis perpendicular to the substrate.

![Image of a graph showing complex TF-μ⁺SR time spectrum on a T<sub>c</sub> ≈ 56 K Tl-2201 mosaic at 0.1 T and 10 K, shown in a rotating reference frame (RRF), including the time-domain best fit, residual errors of which are shown in (b) for the most sensitive early times. (c) Fourier transforms at 0.1 T and several temperatures.](image)

**Figure 1.** (color online) Example of μ⁺SR data. (a) Complex TF-μ⁺SR time spectrum on a T<sub>c</sub> ≈ 56 K Tl-2201 mosaic at 0.1 T and 10 K, shown in a rotating reference frame (RRF), including the time-domain best fit, residual errors of which are shown in (b) for the most sensitive early times. (c) Fourier transforms at 0.1 T and several temperatures.

The main advantage of TF-μ⁺SR in studies of Type-II SC is its ability, under optimal circumstances, to determine the absolute value of the magnetic penetration depth λ and its inverse square, which is proportional to the density n<sub>s</sub> of SC carriers[9]. Unfortunately, “optimal circumstances” do not yet apply in the case of Tl₂Ba₂CuO<sub>6+x</sub>, where only small improvements of global χ<sup>2</sup>-minimization fits distinguish between the broadening σ<sub>d</sub> due to vortex lattice disorder [which should scale with λ<sup>-2</sup>(T)] and the T-independent broadening σ<sub>0</sub> due to nuclear dipoles and crystal defects. The amplitude A<sub>B</sub> of the background signal due to muons stopping outside the sample is also known only by finding the best global fit; like σ<sub>0</sub>, the value of this parameter is subtly coupled to that of λ<sup>-2</sup>.

An example of the data collected, in this case at 10 K on a T<sub>c</sub> = 56 K mosaic, is shown in Fig. 1, including the time domain fit in panel (a) and its residuals in panel (b). Time-domain fits to a numerically generated vortex lattice field distribution [9] converged very well and fully reproduce the data on all mosaics at all fields and temperatures. Fourier transforms corresponding to the field distribution are also shown for a variety of temperatures — an additional peak just above
the “cusp” is attributable to muons stopping outside the sample; the time-domain fits account for this contribution. The high-frequency “cutoff” is indistinct in these mosaics, precluding a quantitative analysis of the coherence length $\xi$, but the absolute magnetic penetration depth $\lambda$, manifested here in the linewidth, may be reliably extracted to within $\sim 10\%$.

**Figure 2.** Field dependence of TF-$\mu^+$SR lineshape in a $T_c \approx 46$ K Tl-2201 mosaic at 10 K. At lower fields the lineshape develops a low-frequency tail, possibly due to stronger pinning disorder. At high fields the high-frequency “tail” is suppressed. At all fields the sharp features such as the “cusp” (van Hove singularity from saddle points between vortices) and high-frequency “cutoff” (due to the finite size of vortex cores) are “smeared out”, presumably due to disorder in the vortex lattice.

**Figure 3.** (color online) LEFT: temperature dependence of frequency spectra at $H = 0.1$ T for a $T_c = 75$ K Tl-2201 mosaic. RIGHT: normalized $\lambda^{-2}(T)/\lambda^{-2}(0)$ (squares and triangles) and similarly normalized empirical linewidth (circles) vs. reduced temperature $T/T_c$ for the same sample.
For the $T_c = 75$ K mosaic at $H = 0.1$ T, fits were also made to an empirical lineshape model composed of a Gaussian “cusp” signal, an exponential “tail” signal and an exponential “breadth” signal whose positions and widths are scaled in fixed ratios. This model gives excellent fits in a tiny fraction of the time required for the numerical simulations, but (a) the overall width (scaling factor) cannot be directly interpreted in terms of $\lambda^{-2}$; and (b) the normalized $T$-dependence of the “linewidth” usually has a different shape than that of $\lambda^{-2}$. Here (see Fig. 3) the two match perfectly, suggesting that in some cases this economical method may have validity; but it must always be “spot-checked”! All results shown in later Figures (including those for the $T_c = 75$ K mosaic) are from rigorous numerical lineshape simulation fits, not from the empirical model.

Figure 4. (color online) LEFT: temperature dependence of fitted $\lambda^{-2}$ values at $H = 0.1$ T for all Ti-2201 mosaics studied. The smooth curve is the Meissner phase microwave data[12] on a $T_c = 25$ K Ti-2201 crystal at 2.497 GHz. RIGHT: same data plotted as normalized $\lambda^{-2}(T)/\lambda^{-2}(0)$ vs. reduced temperature $T/T_c$. All dopings exhibit essentially the same $T/T_c$-dependence of the normalized superfluid density, with an inflection point at $T \sim 0.5T_c$. The fact that the susceptibility shows no such inflection point indicates that it is intrinsic to the vortex phase.

Table 1. Zero-temperature magnetic penetration depths in 0.1 T for overdoped Ti-2201 mosaics having various $T_c$s, from a linear extrapolation of $\lambda^{-2}(T)$ at low temperatures, with estimated uncertainties in parentheses.

| $T_c$ (K)   | $\lambda_{ab}(0)$ (nm) |
|------------|------------------------|
| 46(1) (A)  | 187(2)                 |
| 46(1) (B)  | 165(2)                 |
| 56(1)      | 166(1)                 |
| 60(1)      | 175(1)                 |
| 72(1)      | 182(2)                 |
| 75(1)      | 153(2)                 |

Figure 4 (left) shows the extracted $\lambda^{-2}(T)$ for the six mosaics measured. A rather similar, and highly unusual, $T$-dependence is immediately apparent. The extent of this similarity is more striking in Fig. 4 (right), where $\lambda^{-2}$ is normalized to its linearly extrapolated $T = 0$ value (see Table 1) and plotted against the reduced temperature $T/T_c$ — the relative temperature dependence is almost identical. Its most intriguing feature, exhibited clearly in all six mosaics, is upward curvature around $\frac{1}{2}T_c$. This unusual temperature dependence is essentially model independent and is evident in any measure of the linewidth. The $T$-dependence of the SC carrier density in a single-gap $s$- or $d$-wave superconductor exhibits downward curvature over the entire temperature range $0 - T_c$. 
The $d$-wave symmetry of the SC order parameter in Tl-2201 has been conclusively established by observation of half-integer flux quanta at crystal boundaries in films[13], and the presence of line nodes is evident in microwave measurements[14, 15, 12] and thermal transport[16]. Indeed, our data also show linear $T$-dependence below $\sim 0.2 T_c$, characteristic of line nodes. Near $T_c$, our $T$-dependence is also not unusual.

The scaling of the $T$-dependence over a wide doping range on different mosaics strongly suggests that it is intrinsic to the vortices, rather than a phase transition within the SC dome, such as an extension of the $T^*$ pseudogap transition; moreover, the similarity with much more disordered overdoped LSCO[17] and cleaner, slightly underdoped YBa$_2$Cu$_4$O$_8$[18] implies that it is not peculiar to Tl$_2$Ba$_2$CuO$_{6+x}$.

A number of possible explanations for the self-similar $T$-dependence of $\lambda^{-2}$ are explored in a forthcoming publication[8]: multiband SC with independent gaps[19], dilute paramagnetic impurities[20] and phase transitions in the vortex lattice [21] are all ruled out by various arguments; a transition from a triangular to a square vortex lattice [22] remains a possibility, but preliminary fits to a square lattice show no improvement of $\chi^2$ relative to the usual triangular lattice model.

A 3D-2D “vortex lattice melting” transition would produce much more dramatic lineshape changes then are seen in these data; thermal “depinning”, on the other hand, would produce a small drop in the apparent linewidth, if anything.

We conclude that the upward curvature arises from some fundamental property intrinsic to the $d$-wave vortices themselves. Franz et al.[23] predicted a small but ubiquitous admixture of $s$-wave order in the vicinity of a $d$-wave vortex core where $d$-wave order varies spatially due to the vortex lattice. Because the shielding current is only gapless in certain directions, the supercurrent path cannot be cylindrical about the vortex core, altering the field distribution. Intervortex interactions and the fraction of muons near vortex cores both increase with temperature as $\xi$ and $\lambda$ expand, and with field as the vortex density increases, but the reduced gradients of the order parameter mean that less $s$-wave component should be induced. An anomalous excess broadening of the $\mu^+\text{SR}$ lineshape would indeed be introduced, but detailed modeling is required to determine whether this can successfully explain the observed temperature dependence.

The nature of SC in the cuprates remains an open question; the overdoped regime offers the promising prospect of understanding the normal state from which SC emerges. However, our $\mu^+\text{SR}$ data indicate that the story here is not simple. An unusual $T$-dependence of the penetration depth, seen now in at least three distinct material families and inconsistent with microwave measurements of nominally the same quantity on equivalent crystals of Tl-2201[12], is most readily explained by a generic phase transition of the vortex lattice. Aside from the vortex physics and what we may learn about the gap symmetry of the material, this has important ramifications for other techniques. $\mu$SR is uniquely suited to extracting the absolute penetration depth, and the values obtained from this technique underpin results in other techniques which can’t measure it in absolute terms (or at all). These absolute values, however, are model-dependent. A square vortex lattice at low temperature, for instance, would require recalculating a number of previous results throughout the field of HT$_2$SC, and would serve as a caution to researchers working on other systems that may have line nodes.

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