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Finite gap behaviour in the superconductivity of the 'infinite layer' n-doped high-T_c superconductor $Sr_{0.9}La_{0.1}CuO_2$

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Abstract
We report on the first small-angle neutron scattering measurements from the flux line lattice (FLL) in the high-T_c cuprate superconductor $Sr_{0.9}La_{0.1}CuO_2$. Using a polycrystalline sample, the scattered intensity decreases monotonically with scattering angle away from the undiffracted beam, independently of the azimuthal angle around the beam. The absence of clear peaks in the intensity suggests the establishment of a highly disordered FLL within the grains. We find that the intensity distribution may be represented by the form factor for a single flux line in the London approximation, with some contribution from crystal anisotropy. Most interestingly however, we find that, over the observed field range, the temperature dependence of the diffracted intensity is best represented by s-wave pairing, with lower limits of the gap values being very similar to the Bardeen–Cooper–Schrieffer value of $\Delta(0) = 1.76\, k_B T_c$. However, a qualitative consideration of corrections to the observed intensity suggests that these gap values are likely to be higher, implying strong-coupling behaviour.

1. Introduction

Current experimental evidence suggests the electron (n-)doped cuprates may possess a conventional s-wave component to their pairing symmetry (see for example, [1–4]). This is in stark contrast to the hole (p-)doped cuprates that exhibit overwhelming evidence for d-wave pairing symmetry (see for example, [5, 6]). As the pairing symmetry reflects the underlying superconducting mechanism, the experimental evidence collected thus far suggests that it may not be possible to establish a general model describing the superconductivity for all cuprates. An important member of the n-doped cuprates is $Sr_{0.9}La_{0.1}CuO_2$ (SLCO), which exhibits the highest T_c of any of the n-doped compounds (43 K) and a relatively short in-plane penetration depth, $\lambda_{ab}(0) \sim 1160$ Å [7]. The crystal also possesses an 'infinite layer' crystal structure [8] referring to the fact that the CuO_2 planes are separated only by Sr and La atoms, with no charge reservoir region common to the T and T' structures of other cuprates. Also in sharp contrast to other cuprates, the c-axis coherence length $\xi_c(0) > c$ [9], suggesting strong interplanar coupling, and three-dimensional behaviour all the way to zero temperature. Recent interest has been renewed in SLCO due to phase-pure preparation with large superconducting fraction [10], coinciding with suggestions of the dependence of pairing symmetry on doping [11, 12]. Since then, SLCO has been under intense scrutiny, with various techniques providing conflicting evidence for the nature of the pairing. Tunnelling

spectroscopy [3] and heat capacity measurements [13] have given support for strongly coupled s-wave pairing, whilst a thermopower study on similarly structured compounds [14] has suggested the pairing is s-wave and phonon-mediated. Meanwhile, evidence for d-wave pairing is provided by NMR and Knight shift measurements [15]. Clearly, the nature of the pairing remains controversial.

The technique of small-angle neutron scattering (SANS) from the flux line lattice (FLL) is a powerful bulk probe of the microscopic mixed state in type-II superconductors. The FLL symmetry and coordination often reflect the underlying superconducting state, as well as the intrinsic physics of flux lines themselves (see for example, [16–19]). Here, we present the first SANS study of SLCO. We find that the FLL established in our sample possesses very little longitudinal and translational order. We also interpret the temperature dependence of the diffracted intensity as being well represented by the requirement of an s-wave-like component in the order parameter. This is the case for the entire investigated field range, and adds further evidence for s-wave behaviour.

2. Experimental details

As single-crystal samples are unavailable at present, the sample was composed of unoriented polycrystalline specimens prepared using high-pressure techniques described elsewhere [20]. It consisted of two approximately right circular cylinders of dimensions 3–4 mm, total mass 0.45 g, with cylinder axes parallel to the applied field. The size of the grains was variable and estimated to be of the order of ∼1 to ∼100 μm. Neutron Laue measurements revealed no observable grain alignment. SQUID magnetometry measurements revealed a $T_c \sim 43$ K showing the optimal doping of our sample. Other previous measurements on SLCO have deduced $\lambda_{ab}(0) = 1160$ Å [7] and $\xi_{ab}(0) = 48.6$ Å [9] yielding a Ginzburg–Landau parameter $\kappa \sim 24$, showing the strong type-II nature of the material. The SANS measurements reported here were performed using the D11 instrument at the Institut Laue-Langevin. Neutrons of wavelengths 6–10 Å were used with a FWHM spread in wavelength of 10%. In typical SANS experiments, the sample is mounted in a cryomagnet with the ability to rock the sample and applied magnetic field together. This allows the rotation of the FLL reciprocal lattice, which subsequently allows a specific reciprocal lattice vector to form a chord of the Ewald sphere. When this occurs, the Bragg condition is met for this order of diffraction and the diffracted intensity associated with this is observed on the detector. Resolution effects, both instrumental and intrinsic to the sample, give the Bragg reflection a finite size in reciprocal space. Hence it is normal to rotate the FLL reciprocal lattice through a range of angles, with typically the Bragg angle at the mid-point, to observe all of the diffracted intensity associated with a specific reflection. The resulting angular dependence of the observed diffracted intensity is termed a 'rocking curve'. In all cases the FLL is prepared in the sample by field cooling through T_c. Background small-angle scattering from the crystallites was subtracted using zero-field cooled measurements, leaving only the signal from the FLL.

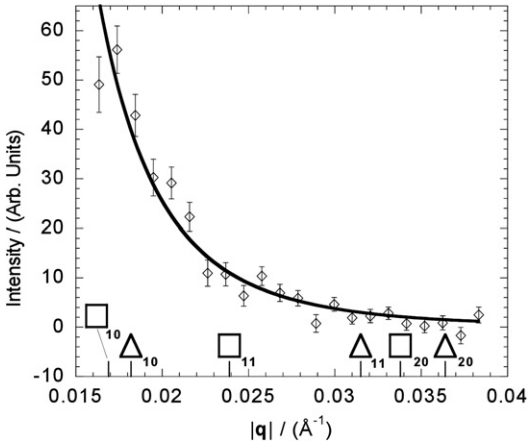

Figure 1. The dependence of intensity as a function of q for the FLL signal at 1.5 T. The line is a fit for the exponent n where $I \propto q^{-n}$. The markers along the horizontal axis show the theoretical value of q where a Bragg peak for different order isotropic triangular and square FLL coordinations would be expected to be observed. Data below $q \sim 0.015$ Å^{-1} are neglected due to poor subtraction effects caused by small-angle scattering from the crystallites.

3. Results and discussion

In an applied field of 1.5 T, rocking curve measurements were carried out over a range of $\omega = 0°$–$1.6°$ rotation about the vertical axis. Here ω is the angle between the neutron beam and applied field. Hence, for the chosen neutron wavelength, $\lambda_n = 10$ Å and detector distance of 10 m, the instrument was well set up for the observation of a peak in intensity due to a FLL. From the rocking curve measurements we observed the FLL scattered intensity varied little with rocking angle, and the observed scattering distribution over the entire detector showed no clear peak in the intensity, in any direction radially from the centre of the detector. Instead, the intensity was distributed approximately isotropically in azimuthal angle around the detector, and decreased radially from the diffracted beam. Figure 1 presents the summed intensity from within a 2π annular sector centred on the undiffracted beam, as a function of $|q|$. Before discussing the lack of a clear peak in the intensity, we discuss the disordered nature of the FLL, as evidenced by estimations of the longitudinal and translational flux line correlation lengths. Using 1.6° as a lower bound of any rocking curve FWHM, we estimate the upper bound of the longitudinal correlation length of the flux lines in the direction of the applied field to be $\ell_l \sim 1$ μm. This assumes that an intensity peak would be observed at the q for an isotropic triangular FLL coordination, $q_\triangle$. Similarly from figure 1, assuming isotropic triangular FLL domains, we estimate an upper bound for the translational order of the flux lines in the direction of the reciprocal lattice vector to be $\ell_t \sim 0.2$ μm. These very short correlation lengths suggest that the FLL structure is highly uncorrelated, with disorder inherent within the crystallites, and not limited as a consequence of the crystallite size in the sample. We suggest the origin of this disorder to be due to strong flux line pinning to structural defects within the grains.

Bearing in mind this disorder, we explain the absence of the intensity peak through two possible scenarios. Firstly, we consider an application of the simple London model to explain the absence of a diffraction peak. Within the London theory for high-κ materials, the integrated intensity for a single (h, k) Bragg reflection from a correlated FLL is $I_{hk} \propto |F_{hk}|^2/q_{hk}$ [21]. Here F_{hk} is the form factor, or spatial Fourier component, describing the field profile of the FLL within the sample, and we have retained the geometrical $(1/q)$ factor in the denominator. For the uncorrelated FLL however, the neutron phase adds incoherently at the detector, resulting in the absence of intense Bragg reflections, consistent with our observations shown in figure 1. For uniaxial superconductors, the dependence of the form factor on angle of applied field and effective mass anisotropy within a single-flux-line model has been extensively studied [22–24]. As our sample is polycrystalline, we account for the fact the applied field direction will be different for each grain and hence average over the in-plane directions. Overall, this reduces the dependence on q of the intensity for the ideal single flux line to $I \propto q^{-5}$. Figure 1 shows the fit of the intensity as a function of q^{-n} with the fit for the index n found to be 4.7(2). This is comparable to the ideal index of 5, lending support to the single-flux-line form factor model of the data.

A second possible scenario is that, despite the apparent disorder revealed by the short correlation lengths, an intensity peak exists but is not clearly observed due to poor statistics on subtraction at low q. This is due to a large amount of small-angle background scattering from the crystallites around the beamstop. However, we can infer the presence of a peak by modelling the higher-q data shown in figure 1. To do this we use the model of Cubitt *et al* [25], designed specifically to analyse SANS data recorded on a polycrystalline sample of MgB_2. The model predicts for a specific anisotropy, the intensity distribution as a function of q away from the predicted Bragg peak, using anisotropic London theory. For the case of SLCO, we use the anisotropy derived from the coherence length of $\gamma_\xi = 9.3$ [9]. The model integrates over all the possible orientations a FLL can adopt when formed in the individual crystallites. Taking into account appropriate instrumental parameters, for our sample of SLCO, the model predicts a dependence on q of the diffracted intensity, away from the diffraction peak, of $I \propto q^{-5.5}$. This result agrees less well with the index of 4.7(2) shown in figure 1, and suggests that the better description lies with the form factor of a single flux line within the London approximation. In any case, anisotropic effects are already included in our use of the single-flux-line London model, and it is clear from the flat rocking curves that the FLL established in our sample can be characterized by strong pinning induced disorder.

We now turn to our principal results of the field and temperature dependence of the diffracted intensity. Due to the large angular width of the rocking curves, we assume that strong pinning holds the rocking curve width constant with temperature, and prevents the thermal motion or melting of flux lines. This allows the measurement of intensity at fixed sample angle. In the London regime for high-κ materials, the square root of the diffracted intensity $\sqrt{I}$, is proportional to the temperature dependent superfluid density $n_s(T)$. Using the approach of Bonalde *et al* [26] we numerically compute the superfluid density as a function of temperature within a quasiparticle framework, by using the expression

$$n_s(T) \propto \frac{1}{\lambda^2(T)} \propto 1 + 2\left\langle \int_0^\infty d\varepsilon \frac{\partial f}{\partial E(k,T)} \right\rangle. \qquad (1)$$

In equation (1), f is the Fermi function and $E(k, T) = (\varepsilon^2 + |\Delta_k(\phi, T)|^2)^{\frac{1}{2}}$ defines the excitation energy spectrum, with ε being the single-particle excitation energy. The gap function is assumed to be separable into momentum and temperature dependent factors $\Delta_k(\phi, T) = \Delta_k(\phi)\Delta(0)\tanh(1.74(1 - T/T_c)^{\frac{1}{2}})$, where the temperature dependent factor fits the predictions of weak-coupling BCS theory. For d-wave pairing, the variation of $\Delta_k(\phi)$ can be taken as $\Delta_k(\phi) = \sqrt{2}\cos(2\phi)$, with ϕ describing the azimuthal angle about a cylindrical Fermi surface. For the case of isotropic s-wave pairing $\Delta_k(\phi) = 1$. $\langle \cdots \rangle$ indicates an angular average over the Fermi surface. Figure 2 shows the temperature dependence of the superfluid density at 1.6 T fitted with various models, with the quality of the fit in each case evaluated using least squares.

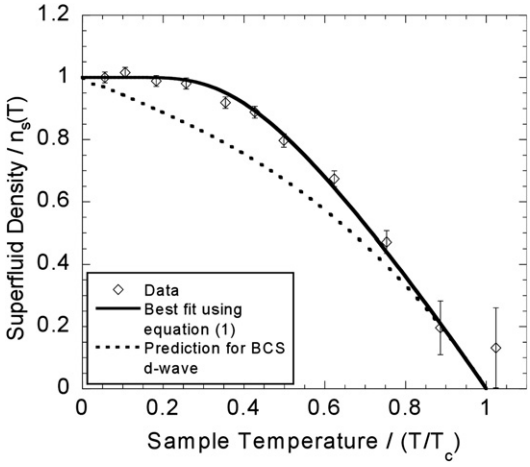

Figure 2. The temperature dependence of the superfluid fraction in an applied magnetic field of 1.6 T. The temperature scale is normalized using the observed $T_c(B) = 38(2)$ K. The solid line represents the best fit for the zero-temperature gap parameter within the isotropic framework. The dotted line is the d-wave BCS weak-coupling model, with $\Delta(0) = 1.76\, k_B T_c$.

Figure 2 shows the fit of equation (1) to the data at 1.6 T, allowing the zero-temperature gap value to vary. Within the s-wave framework, the best fit zero-temperature gap value is $\Delta(0) = 1.70(8)\, k_B T_c$. This value is remarkably similar to the ideal BCS value $\Delta(0) = 1.76\, k_B T_c$. For comparison, figure 2 also shows the prediction for the d-wave BCS weak-coupling model. This is clearly a poor fit to the data. Indeed, as the model is sensitive to the nodes in the gap, large variations in the magnitude of the gap parameter do not improve the line-node gap model significantly. Modelling further data taken at 0.6 T gives a best fit within the s-wave framework of $\Delta(0) = 1.76(10)\, k_B T_c$ ($T_c(B) = 40(3)$ K) and at 4.8 T gives $\Delta(0) = 1.69(6) k_B T_c$ ($T_c(B) = 35(1)$ K). At all fields we find good agreement with a BCS picture, lending strong support from

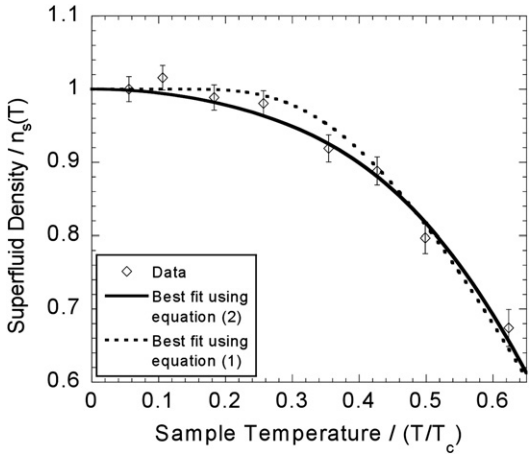

Figure 3. The superfluid density as a function of temperature at 1.6 T. The temperature scale is normalized with the same T_c as in figure 2. The solid line represents the fit using equation (2) fitting for $\Delta(0)$ and $\xi(0)$. The dotted line is the fit returned by equation (1), without the core correction, and is included for comparison.

our data for an s-wave-like behaviour of the order parameter. Also, the reduction in T_c with increasing field confirms the FLL origin of the signal. The result of s-wave behaviour is consistent with previous tunnelling spectroscopy and heat capacity measurements [3, 13]. However, these measurements deduced a strong-coupling nature to the superconductivity of $\Delta(0) \sim 3.5\, k_B T_c$ in sharp contrast to our values.

This discrepancy could be taken into account by considering various corrections to the observed intensity. One such correction arises from accounting for the finite size of the flux line cores. For comparison to the uncorrected data in figure 2, we introduce Yaouanc *et al*'s [27] core-correction term derived from an approximation to Clem's [28] solutions of the Ginzburg–Landau theory. They choose the form of the correction to be $\exp(-\sqrt{2}q\xi(T))$. Here q is the reciprocal space wavevector, which we assume to be $\boldsymbol{q}_\triangle$. This is reasonable considering the majority of the scattered intensity lies close to $\boldsymbol{q}_\triangle$, before rapidly falling off with increasing q. With $\xi(T)$ being the effective temperature dependent coherence length, equation (1) is modified to become

$$n_s(q,T) \propto \left[1 + 2\left\langle \int_0^\infty \mathrm{d}\varepsilon \frac{\partial f}{\partial E(k,T)} \right\rangle\right] \exp(-\sqrt{2}q\xi(T)). \tag{2}$$

In (2) we use an empirical $(1-(T/T_c)^2)^{-1/2}$ temperature dependence of the coherence length and fit up to $\sim$0.6 T_c in order to avoid the effect of the divergence of the temperature dependence as $T \to T_c$.

As depicted in figure 3, our best fit to the 1.6 T data incorporating the correction factor gives $\Delta(0) = 2.7(1)\, k_B T_c$ and $\xi \sim 40$ Å. The value of ξ is reasonable as $\xi_c < \xi < \xi_{a,b}$, where ξ_c (= 5.2 Å [9]) and $\xi_{a,b}$ represent the lower and upper bounds respectively. Similarly at 0.6 T the best fit gap returned is $\Delta(0) = 2.2(2)\, k_B T_c$ and $\xi \sim 40$ Å. However, at 4.8 T the best fit gap value is $\Delta(0) = 3.8(2)\, k_B T_c$ with $\xi \sim 30$ Å. At this high an induction, the use of the simple exponential correction term is now invalid, and a higher-field correction such as Hao *et al*'s [29] high-field extension to the Clem model is more suitable. Nevertheless, we see that incorporating a core correction results in a significant increase in the estimated coupling strength of the superconductivity within the s-wave model.

Further corrections to the intensity might also be necessary due to the effect of a temperature independent 'static' Debye–Waller-like factor[5] which is dependent on the root mean square displacement of a flux line from its mean position, and the d-spacing of the flux lines. Such a correction is likely to be significant considering the observed disorder of the FLL. However, as the static Debye–Waller-like factor is temperature independent, the intensity correction will be an absolute numerical adjustment and will not result in a different deduced gap value. Such a correction may be important in the determination of other length scale parameters, such as the penetration depth.

The absence of detailed FLL structural information within the data inhibits the determination of precise numerical corrections to our results. We conclude however, that in the limit of no intensity corrections, the fits returned from modelling the data with equation (1) can be considered as lower bounds for gap values at all fields.

Our deduction of s-wave behaviour is consistent with previously observed behaviour upon doping SLCO with impurities [3, 30]. It is a well-known result that doping exclusively d-wave superconductors with a small concentration of nonmagnetic impurities results in a strong suppression of T_c. The previous work on SLCO investigating the effect of doping with impurities at Cu sites has deduced strong evidence for conventional s-wave behaviour, with no suppression of T_c with increasing impurity concentration. This s-wave-like behaviour contradicts theories that attempt to reconcile the conflicting observed s-wave behaviour in the n-doped cuprates. Models by Luo *et al* [31] and a similar one by Yuan *et al* [32] attempt to describe the conflicting behaviour within an entirely d-wave framework. Luo *et al* use a two band BCS-like weak-coupling model assuming a $d_{x^2-y^2}$ symmetry pairing of carriers within each band. One of the bands, band 1, appears around $(\pi, 0)$, whilst band 2 appears around $(\frac{\pi}{2}, \frac{\pi}{2})$. Due to the positions of pockets at these Fermi surface locations, the d-wave nodes do not intersect the Fermi surface in band 1. Hence, all the carriers in this band have an anisotropic but non-zero gap while maintaining d-wave symmetry. However, if the pairing were exclusively d-wave in SLCO, then strong impurity effects would still be observed in band 1. The authors also claim the s-wave behaviour should be largely apparent when the sample is underdoped. We clearly observe s-wave behaviour at optimal doping, with our sample possessing the highest T_c for this compound. These theories require a knowledge of the band dispersion around the Fermi surface. However, the band dispersions exhibited by SLCO may be quite different to other n-doped cuprates due to the infinite layer crystal structure, and strong interplanar coupling. This strong coupling could make

[5] The static Debye–Waller factor has the form $\exp(\frac{-q^2\langle u^2\rangle}{2})$ where $\langle u^2\rangle$ is the component of the root mean square displacement of the flux line in the direction of q. As $q = 2\pi/d$, where d is the average flux line spacing, the factor can thus be expressed $\exp(\frac{-4\pi^2}{2}\frac{\langle u^2\rangle}{d^2})$, showing a clear field dependence. Neither q nor $\langle u^2\rangle$ are temperature dependent.

a conventional pairing mechanism energetically favourable, as has been previously suggested [3]. Subsequently, for the proposed theoretical models [31, 32] to describe adequately the behaviour seen in SLCO, important modifications are required to account for the larger c-axis dispersion and strong interplanar interaction.

4. Summary

We have performed the first SANS study on a polycrystalline sample of SLCO. We find that the FLL structure is highly disordered with FLL correlation lengths of the order of micrometre and below. As these correlation lengths are smaller than the dimensions of the crystallite sizes, we conclude that FLL disorder is due to strong flux line pinning caused by structural defects within the crystallites, as opposed to being limited by crystallite size. The dependence of the intensity on q is well modelled by the form factor of a single flux line, which would be expected for a strongly disordered FLL. However, we cannot completely rule out some influence of crystal anisotropy in this dependence.

Our measurements of the FLL diffracted intensity as a function of temperature at three different fields lends support to the presence of s-wave-like pairing behaviour. In the limit of perfect FLL structure and large flux line spacings, we find lower limits for gap values over the entire investigated field range to be well modelled by values very close to the BCS gap value of $\Delta(0) = 1.76\, k_B T_c$. However, corrections to the observed intensity due to the finite size of the flux line cores leads to a significant increase of the gap value for all fields, indicating SLCO is likely to exhibit strong-coupling behaviour. This would also be the case when accounting for corrections due to static FLL structural disorder. Limitations in the data prevent a numerical estimate of the strongly coupled gap values. However, our data are unlikely to be well described by proposed theories describing s-wave behaviour in the n-doped cuprates [31, 32], with modifications to these theories required for SLCO due to its unique crystal structure. Future SANS measurements on this compound would benefit from sizable single-crystal samples.

Acknowledgments

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