Spin density wave induced disordering of the vortex lattice in superconducting La$_{2-x}$Sr$_x$CuO$_4$

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We use small angle neutron scattering to study the superconducting vortex lattice in La$_{2-x}$Sr$_x$CuO$_4$ as a function of doping and magnetic field. We show that near optimally doping the vortex lattice coordination and the superconducting coherence length $\xi$ are controlled by a van-Hove singularity crossing the Fermi level near the Brillouin zone boundary. The vortex lattice properties change dramatically as a spin-density-wave instability is approached underdoping. The Bragg glass paradigm provides a good description of this regime and suggests that SDW order acts as a novel source of disorder on the vortex lattice.

I. INTRODUCTION

A commonality across the borocarbides, cuprates, ferro-pnictides, heavy-fermion and organic superconductors is the coexistence of magnetism and superconductivity. The corresponding order parameters typically compete and often a small perturbation is sufficient to tip the balance between the two. For example, the magnetism carried by the rare earth ions R in the borocarbides RNi$_2$B$_2$C can lead to nearly reentrant superconducting phase diagrams, and spontaneously forming superconducting vortices at zero applied field. Equally, vortices induced under applied fields may permit enhanced magnetic correlations in the core regions where the superconducting order parameter is suppressed. This idea was put forward to explain field-induced and enhanced magnetic correlations observed in the cuprate superconductor La$_{2-x}$Sr$_x$CuO$_4$ (LSCO).

Although the effect of static magnetism on moving vortices was recently considered theoretically, little is known about how the presence of magnetic correlations affects the arrangement of vortices. Here we address the problem from an experimental point of view. When magnetism and superconductivity coexist there are at least four relevant length scales: the penetration depth $\lambda$, the vortex core size $\zeta$, the vortex spacing $a_0$, and the magnetic correlation length $\xi$. The vortex spacing $a_0 \propto H^{-0.5}$ scales with the applied magnetic field $H$ and in LSCO the magnetic correlation length can be tuned by varying the doping concentration. Using small angle neutron scattering (SANS) we have studied two different regimes (see Fig. 1): (i) far away from the magnetic ordering where $\xi, \zeta \ll a_0$ and (ii) entering the magnetic phase where $\zeta \sim a_0$. In the first regime, where static magnetism is absent, the vortex lattice (VL) structure and core size are understood from pure fermiological considerations. In the second regime with static long-range magnetism, the vortex arrangement exhibits increasing disorder. We find these regimes to be well-described within the topical ‘Bragg glass’ paradigm, where disorder results in an algebraic decay of the translational order of the vortices. VL disordering is usually driven by effects extrinsic to superconductivity such as rare earth magnetism in RNi$_2$B$_2$C or sample impurities and crystalline defects. In contrast, magnetic and SC order parameters are intertwined in LSCO; we show that this provides a novel and tunable source of VL disorder.

As shown in Fig. 1, the appearance of magnetism is, in essence, concomitant with the suppression of SANS intensity with field and underdoping. Drawing upon results from the literature and new observations reported herein, we are also able to plot the VL structure at low temperature $T$ versus magnetic field $H$ and doping $x$.

II. EXPERIMENTAL METHODS

Single crystals of La$_{2-x}$Sr$_x$CuO$_4$ with $x = 0.105-0.22$ were grown by the traveling solvent floating zone method. The static and dynamic magnetic properties of the samples were characterized using both neutron diffraction and neutron spectroscopy and good agreement was found between our data and previously published results.

The SANS experiments reported here were carried out over a series of experiments using the SANS-I instrument.
at SINQ, and the D11 and D22 instruments at ILL. In all experiments we adopted the experimental geometry where external magnetic fields $\mu_B H$ are applied parallel to the crystal $c$-axis, and almost parallel to the neutron beam. The scattered neutrons are recorded using a position-sensitive-detector placed behind the sample. For each doping, up to three different neutron wavelengths spanning the range $\lambda_n = 5$–$16$ Å were used in order to cover the applied field $\mu_B H$ range of 0.03–10 T. In all cases, a zero-field cooled background was subtracted from the field-cooled data in order to leave just the VL signal.

Our experimental setup is shown in Fig. 2 where we also illustrate the relationship between the VL in the sample and the quantities extracted at the position-sensitive-detector. Typically, to observe the signal due to the VL, the sample and cryomagnet are rotated together by angles (such as that shown by $\omega$ in Fig. 2) in order to bring a reciprocal lattice vector onto the Bragg condition at the detector. Due to both the finite resolution of the instrument, and the mosaic spread or imperfection of the VL, the Bragg spots occupy a finite volume in reciprocal space, and can be described, in a first approximation, by three widths $\tau_\omega$, $\tau_r$, and $\tau_A$ within the detector plane, and $\tau_\perp$ perpendicular to the detector plane. $\tau_\omega$ is determined experimentally by recording the rocking curve, and corresponds to the rocking curve width.

![Image of a schematic diagram illustrating the experimental geometry chosen for our experiments. A square VL in real space forms a two-dimensional reciprocal VL, the properties of which are recorded by SANS using a position-sensitive detector. The Bragg spots in reciprocal space exhibit finite widths $w_f$, $w_q$ and $w_\perp$ that are dependent on both the instrumental resolution and properties of the VL. These three lengths are estimated at the detector by recording the angular widths $\tau_\omega$, $\tau_r$ and $\tau_A$ within the detector plane, and $\tau_\perp$ perpendicular to the detector plane. $\tau_\omega$ is determined experimentally by recording the rocking curve, and corresponds to the rocking curve width.](image-url)
III. RESULTS AND DISCUSSION

A. Vortex lattice morphology

Our observations of the VL structure and coordination can be quantified in terms of a dimensionless parameter
\[ \sigma = 4\pi^2\mu_0 H/|G|^2 \]
where |G| is the magnitude of the reciprocal VL vector. For a regular hexagonal VL coordination \( \sigma = \sqrt{3}/2 \), while for a square coordination \( \sigma = 1 \). The definition of \( \sigma \) is useful because it does not require details of the positions \( G \) of Bragg peaks; only the magnitude |G| is needed. At low fields, the VL is susceptible to orientational disorder due to impurities or defects in the sample, but |G| can still be measured. For example, in Fig. 3(a) the diffracted intensity measured in LSCO \( x = 0.145 \) at 0.2 T does not show well-defined Bragg spots. This indicates a large \( \tau_0 \) and poor orientational order of the VL about the c-axis. Nevertheless we can determine the VL coordination by averaging over the azimuthal angle \( \varphi \) so that the diffracted intensity \( I(|q|) \) becomes a function of |q| only, see Fig. 3(c). |G| is determined from the peak position in the |q|-dependence. Fitting a Lorentzian lineshape to these 0.2 T data, yields \( \sigma = 0.88(2) \) very close to the value expected for a hexagonal VL. At \( \mu_0 H = 1.3 \) T in Fig. 3(b), we find four Bragg spots with G along the Cu-O bond directions and \( \sigma = 0.99(1) \), indicating not only an improved VL orientational order but moreover a square coordination. As shown quantitatively in Fig. 4(a), the VL coordination in underdoped LSCO \( x = 0.145 \) changes steadily from hexagonal to square over the range \( \mu_0 H = 0.2 \) to 0.8 T. In contrast, on the optimally- and over-doped side of the phase diagram \( x \geq 0.17 \), the VL structure becomes square by \( \mu_0 H \approx 0.4 \) T (Fig. 4(a)).

A minimum in either the Fermi velocity \( v_F(k) \) or the superconducting gap \( \Delta(k) \) are well-known sources of field-driven hexagonal-to-square VL transitions. In both LSCO and YBa\(_2\)Cu\(_3\)O\(_y\) (YBCO) the band structure is predominantly two-dimensional but with some c-axis dispersion near the \( k = (\pi,0) \)-point. A crucial difference between the band structures of LSCO and YBCO is that in LSCO, a van Hove singularity crosses the Fermi level \( \epsilon_F \) at \( (\pi,0) \) somewhere between \( x = 0.17 \) and 0.22, leading to a huge \( v_F \)-anisotropy. This offers an explanation as to why the square VL is oriented along the \( (\pi,0) \) direction in LSCO as opposed to the nodal \( (\pi,\pi) \)-direction in YBCO. As LSCO is underdoped, the van Hove singularity is pushed further away from \( \epsilon_F \) leading to a smaller \( v_F \)-anisotropy and consequently a larger field is required to form the square VL, as indeed observed for LSCO \( x = 0.145 \) (Fig. 4a).
FIG. 4. (Color online) (a) Dimensionless constant $\sigma$, defined in the text, as a function of magnetic field for LSCO $x = 0.145, 0.16, 0.17$ (Ref. 23), 0.20 (Ref. 24), and 0.23. $\sigma = \sqrt{3}/2$ is expected for a hexagonal lattice and a square vortex lattice has $\sigma = 1$. The change from $\sigma = \sqrt{3}/2$ to 1 therefore reveals the a hexagonal-to-square transition of the vortex lattice structure (see also Fig. 1). (b) SANS intensity $I$ versus applied magnetic field for NCCO $x = 0.15$ (Ref. 37) and La$_{2-x}$Sr$_x$CuO$_4$ with $x = 0.105$ (Ref. 25), and 0.145–0.23, (this work). For clarity, the intensities for each of the compositions have been given an arbitrary vertical offset. Solid lines are fits to the Clem model form factor where the superconducting coherence length $\xi$ is the only parameter (see Fig. 5).

Dashed lines indicate power law dependencies; $I \sim H^{-0.5}$ and $I \sim H^{-2}$. Notice that the Bragg glass paradigm for vortices in the presence of disorder is consistent with a cross over from $I \sim H^{-0.5}$ to $I \sim H^{-2}$, see text.

B. Diffracted SANS intensity

We define the VL intensity of the first order diffraction peak $I$ as the sum of the area under $I(|q|)$, which is itself a sum over rocking angles. Overlap measurements of $I$ vs $\mu_0 H$, shown in Fig. 4(b) for $x = 0.105$–0.23, were done whenever the neutron wavelength $\lambda_n$ was changed. For $x \geq 0.16$, intensity could be observed up to the highest applied field 10 T. By contrast, for $x = 0.145$ no intensity was observed above the quantum critical field ($\mu_0 H = 7 \pm 1$ T in our sample) for SDW order.25,26,31 It is, however, still possible that the VL extends slightly into the SDW ordered phase. This is the case in LSCO $x = 0.105$,22 where a 3D vortex lattice exhibits $I \propto H^{-2}$ (Fig. 3(b)) over two decades of intensity and co-exists with short range SDW order at very low fields $H_{c1} < \mu_0 H \lesssim 0.2$ T. VL intensity is also observed in LSCO $x = 0.12$; a compound where long-range SDW order exists already in zero field.25 At $\mu_0 H = 0.05$ T, the $|q|$-dependence of the intensity $I(|q|)$ (Fig. 5(d)) suggests a hexagonal VL coordination. To the best of our knowledge, this provides the first evidence by SANS of a VL co-existing with SDW-order at the 1/8-anomaly. The field range of co-existence is small; on increasing the field to just $\mu_0 H = 0.1$ T, Fig. 5(d) shows that the VL signal has already fallen to the background level. Notice that the fields 0.1–0.2 T are much smaller than those required to decouple 3D superconductivity.51
C. Coherence length from the VL form factor

In the case of perfect crystalline VL order, the observed intensity of the first order diffraction peak \( I(H) \propto \sum F^2 / |G| \), where \( F \) is the form factor of a single vortex and has units of field. The sum is over all the \( q \)-vectors contributing to the intensity near wave-vector \( |G| \), and we have assumed that the rocking curve width remains constant with field in obtaining \( I(H) \). A variational solution to the Ginzburg-Landau model, namely the Clem model for the VL form factor \( \mathcal{F} \propto G K_1(\xi) \), where \( K_1 \) denotes the modified Bessel function of first order, \( G = |G| = 2\pi / \sigma \Phi_0 \), and the vortex core size \( \xi \) is the only fit parameter.\textsuperscript{22} We point out that the application of the Clem model to our data yields an upper bound for \( \xi \); disorder effects are, for example, not included.\textsuperscript{18} With increasing vortex lattice disorder, the degree by which the Clem model overestimates the coherence length is larger. By comparing the doping dependence of the extracted \( \xi \) with the Ginzburg-Landau coherence length \( \xi_{GL} = \sqrt{\Phi_0 / 2\pi H_2} \), estimated indirectly from specific heat\textsuperscript{50} and high-field magneto-resistance experiments\textsuperscript{49} a reasonable agreement is found on the overdoped side, see Fig. 5. This suggests that our SANS data indeed provide a measure of \( \xi \), even though disorder is undoubtedly present. Identifying \( \xi \) with the Pippard coherence length \( \xi_p \sim \hbar v_F(k) / \Delta(k) \) suggests that the relatively short coherence length \( \xi \) around optimally doped LSCO is not only due to the large pairing gap \( \Delta \); the small Fermi velocity is also playing a significant role.

D. Disorder effects and structure factor

On the underdoped side, we find a strong discrepancy between the coherence length estimated from specific heat and the SANS data fitted with the Clem model for the form factor, see Fig. 5. For LSCO \( x = 0.145 \), we find \( \xi \approx 80 \text{ Å} \) corresponding to an unrealistically small upper critical field \( \mu_0 H_2 = \Phi_0 / 2\pi \xi^2 \approx 5 \text{ T} \). A larger coherence length may result from the weakening of superconductivity due to competition with, for example, magnetism. However, this does not explain the discrepancy between our SANS data and the specific heat data.\textsuperscript{50} A more plausible explanation is that the VL disorder potential increases with underdoping. It is possible that VL disorder proliferates as the system is tuned towards the state where magnetism and superconductivity coexist.

The VL displacements throughout the doping range are well-described by elastic theory, namely the Bragg glass (BrG) paradigm.\textsuperscript{16,17} In the presence of disorder, the positional order of an elastic VL decays exponentially with a characteristic length scale \( R_A \). (With increasing disorder \( R_A \rightarrow 0 \).) If such an exponential decay were to persist at all length scales \( R \), a total destruction of long-range order would result.\textsuperscript{55} This proposed destruction, even under weak disorder, presented a long-standing puzzle with respect to experimental observations where Bragg peaks may readily be observed. Theoretically, the puzzle was resolved with the advent of the BrG paradigm, in which an asymptotic regime for \( R > R_A \) enters, where the positional order decays only weakly, leading to algebraically diverging Bragg peaks. At the crossover scale \( R_A \) vortex displacements are comparable to the lattice spacing \( a_0 = \sqrt{\sigma \Phi_0 / \Delta} \). In earlier muon spin rotation (\( \mu \text{SR} \)) work on a LSCO \( x = 0.105 \) sample, an order-disorder transition was observed and associated with a transition out of the quasi-long-range-ordered BrG phase.\textsuperscript{22} VL correlations in the BrG phase have been explored more directly in a study of low-purity niobium.\textsuperscript{22} We point out that the BrG paradigm can explain both the \( H^{-0.5} \) and \( H^{-2} \) field-dependences of intensity (c.f. Fig. 4(b)), as well as the crossover between them. Dependent on whether the instrumental resolution \( s \approx 60a_0 \) is larger or smaller than \( R_A \), a different field dependence of the SANS intensity is predicted.\textsuperscript{22} For \( s > R_A \), the contribution to the structure factor is identical to that of a crystalline VL, hence \( I \sim 1 / \sqrt{H} \). In the other limit \( s < R_A \), an additional factor \( H^{-\mu} \) contributes to the intensity. Elastic theory\textsuperscript{44} yields \( \mu = 3/2 \) and hence \( I \sim H^{-2} \) — as indeed we observed previously in LSCO \( x = 0.105 \) see Fig. 4b. The intensity for \( x = 0.145 \) is also consistent with a \( H^{-0.5} \) to \( H^{-2} \) crossover, see Fig. 3. At the crossover field (\( \sim 0.85 \text{ T} \)), \( R_A \approx s \approx 60a_0 \approx 3 \mu \text{m} \). By contrast, in LSCO \( x = 0.105 \) the crossover field \( \ll 0.05 \text{ T} \), implying that the disorder potential increases dramatically with decreasing doping. This disordering seems to occur as the static magnetic correlation length \( \zeta \) approaches the VL spacing (\( \zeta \rightarrow a_0 \)), suggesting an electronic origin to the VL disorder effectuated by real space competition between \( \zeta \) and \( a_0 \), rather than orthorhombic twin boundaries or impurities as observed in other superconductors.\textsuperscript{22} We noticed that the BrG model also provides an excellent description for the (previously unexplained) field dependence of intensity in \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \) (NCCO) with \( x = 0.15 \) (Fig. 4(b)).\textsuperscript{17} More experimental studies are required on NCCO to establish if the VL disorder therein has origins similar to that in LSCO.

E. Real space picture

We now consider how magnetic and superconducting order parameters might coexist in real space. Around \( x = 0.12 \) doping in LSCO, \( \mu \text{SR} \) measurements revealed magnetic and superconducting fractions that together exceed 100%. It was therefore concluded that the magnetic and superconducting order parameters are not phase separated but rather intertwined on a nanometer scale.\textsuperscript{27,58} This real space picture is not easily reconciled with neutron diffraction studies that report a magnetic correlation length of several hundreds of Angstroms.\textsuperscript{25,31,33,34} One possibility is that the charge of the muon induces magnetism in which case the \( \mu \text{SR} \) technique overestimates the magnetic volume fraction.\textsuperscript{22} Here we showed
that the vortex lattice becomes more disordered as the vortex interspacing approaches the magnetic correlation length. This suggests that the magnetic and superconducting order parameters are coupled. How magnetism, superconductivity, and vortices are arranged in real space when the spin correlation length is larger than the vortex spacing ($\zeta > a_0$) is an interesting question that is difficult to address with the SANS technique since no observable SANS signal is found in that region of the phase diagram, see Fig. 1. Direct imaging techniques are undoubtedly more informative in this regime although it may be experimentally challenging to probe magnetism and vortices simultaneously.

IV. CONCLUSIONS

In summary, our studies of the vortex lattice in LSCO allow us to draw two main conclusions. First, near optimal doping, and far from the SDW instability, the VL structure/orientation and the small superconducting coherence length $\xi$ (and hence large upper critical field $H_{c2}$) may both be rationalized as arising from a vanishing Fermi velocity due to the van-Hove singularity near the zone boundary. Second, we find that the fermiologic picture breaks down as the SDW instability is approached by tuning either the doping or the applied magnetic field. There, the vortex lattice structure factor needs to be accounted for in the field-dependence of the observable SANS intensity. The Bragg glass paradigm, describing vortex lattices in the presence of weak disorder, accounts for the SANS intensity behavior across the entire phase diagram (where a SANS signal is discernible), from the underdoped $x=0.105$, to the optimally doped $x \sim 0.16$ and overdoped $x=0.23$ regimes. In particular, it is able to explain the cross-over as the SDW instability is approached. Evidently, the SDW order acts as a novel electronic provenience of disorder on the vortex lattice in LSCO.

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In the Clem model, the argument $z$ of the modified Bessel function $K_1(z)$ is strictly $z = (G^2 + \lambda^2)^{1/2}$. Here $G \gtrsim 0.003 \AA^{-1}$ and $1/\lambda \lesssim 0.0005 \AA^{-1}$ so $z \approx G\xi$. In the Clem model, $\lambda$ also enters as an $H$-independent prefactor $1/(A K_1(\xi/\lambda))$.

A simple model for VL disorder comprises the static Debye-Waller factor $W = \exp(-4\pi^2(u^2)/a_0^2)$ where $\langle u^2 \rangle$ is the root mean square vortex displacement (along $q$) and $a_0 = \sqrt{\sigma F_0}/H$ is the vortex lattice spacing. Due to the similar field dependence of $F$ and $W$, it is not possible to disentangle contributions arising from the vortex core size $\xi$ and Debye-Waller effects.