

Decrease of upper critical field with underdoping in cuprate superconductors

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It is still unclear why the transition temperature T_c of cuprate superconductors falls with underdoping. The doping dependence of the critical magnetic field H_{c2} is directly relevant to this question, but different estimates of H_{c2} are in sharp contradiction. We resolve this contradiction by tracking the characteristic field scale of superconducting fluctuations as a function of doping, via measurements of the Nernst effect in $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$. H_{c2} is found to fall with underdoping, with a minimum where stripe order is strong. The same non-monotonic behaviour is observed in the archetypal cuprate superconductor $\text{YBa}_2\text{Cu}_3\text{O}_y$. We conclude that competing states such as stripe order weaken superconductivity and cause both H_{c2} and T_c to fall as cuprates become underdoped.

Two paradigms have been proposed to account for the dome-like region of superconductivity in the temperature-doping phase diagram of cuprate superconductors¹. In the first, the amplitude of the superconducting order parameter grows monotonically as the doping p is reduced, but its phase is increasingly disordered², causing T_c to fall at low p . The signature of this scenario is strong phase fluctuations and a superconducting gap above T_c in the underdoped regime. In the second paradigm, the fall of T_c at low p is due to the onset of a state that competes with superconductivity. The signature of this scenario is a small superconducting gap and a small H_{c2} in the underdoped regime.

Whether strong phase fluctuations or a decrease in the pairing gap is causing T_c to fall in underdoped cuprates is currently an open question. Different interpretations of photoemission data disagree on the evolution of the pairing gap^{3–6} and different estimates of the upper critical field H_{c2} are in sharp contradiction^{7,8}. The Nernst signal observed above T_c in underdoped cuprates has been attributed to superconducting fluctuations^{8–10}, and because it persists up to temperatures several times T_c , it was deemed incompatible with the standard Gaussian fluctuations of the superconducting order parameter. It was attributed instead to vortex-like excitations in a phase-fluctuating superconductor^{9,10} with a non-zero pairing amplitude above T_c . The critical field H_{c2} deduced from the Nernst data on cuprates such as $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) was reported to increase with underdoping⁸, even though T_c falls. As shown in Fig. 1a, this is in striking contrast to the rapid drop in H_{c2} deduced from a Gaussian analysis of fluctuations in the magneto-conductivity of $\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO; ref. 7).

Nernst effect in Eu-LSCO

Here we re-examine the Nernst effect in cuprates with a study of $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ (Eu-LSCO), an underdoped cuprate in which the ratio of superconducting (N_{sc}) to quasiparticle (N_{qp}) contributions to the Nernst signal N is exceptionally large—at

least 100 times larger than in previous studies of superconducting fluctuations in cuprates (see Supplementary Table S2). Because of its low T_c , we could determine the quasiparticle background $N_{qp}(T)$ in Eu-LSCO by fully suppressing superconductivity with a magnetic field (Fig. 2a). The large signal-to-background ratio allows us to reliably track N_{sc} up to high temperature, namely up to a reduced temperature $\varepsilon \equiv (T - T_c)/T_c \approx 5$, compared to a typical upper limit of $\varepsilon \approx 0.5$. As we shall see, this gives us access to a regime where the complicating effects of paraconductivity are negligible.

In Fig. 3a, N_{sc} is plotted versus magnetic field H for different temperatures above T_c , for Eu-LSCO at a doping $p = 0.11$. N_{sc} increases linearly at low H , peaks at a field H^* and then decreases monotonically at high H , just as in the conventional superconductor $\text{Nb}_{0.15}\text{Si}_{0.85}$ (refs 11,12; Supplementary Fig. S2). The peak field H^* , also called the ‘ghost critical field’ (ref. 12), is plotted versus ε in Fig. 3b. It obeys $H^* = H_{c2}^* \ln(T/T_c)$ from $\varepsilon \approx 0.5$ to $\varepsilon \approx 5$, where H_{c2}^* is a field scale whose relation to the $T = 0$ upper critical field H_{c2} is discussed below.

Below $\varepsilon \approx 0.5$, H^* deviates from $\ln(T/T_c)$, and remains finite as $\varepsilon \rightarrow 0$. This is because $N_{sc}(\varepsilon) = \alpha_{xy}^{sc}(\varepsilon)/\sigma(\varepsilon)$ is the ratio of two quantities^{13–16}—the off-diagonal Peltier coefficient from superconducting fluctuations α_{xy}^{sc} and the electrical conductivity σ —which both diverge as $\varepsilon \rightarrow 0$ (ref. 13). This causes N_{sc} to saturate at low ε (Fig. 4). The deviation of H^* from $\ln(T/T_c)$ coincides with the onset of paraconductivity below $T \approx 6 \text{ K} \approx 1.5T_c$ (see Supplementary Fig. S3 and Fig. 4c inset). Above $\varepsilon \approx 0.5$, paraconductivity is negligible (Supplementary Fig. S4) and σ reaches its (field-independent) normal-state value, at which point $N_{sc}(H) \sim \alpha_{xy}^{sc}(H)$. We make use of H^* in the latter regime only.

H^* also obeys $H^* = H_{c2}^* \ln(T/T_c)$ in our other Eu-LSCO samples (Supplementary Table S1 and Fig. S1), with $p = 0.08, 0.10$ and 0.125 (Fig. 3c). The value of H_{c2}^* extracted from the fit at each doping is plotted in Fig. 5a. Our first and main finding is this: the field scale for superconductivity, H_{c2}^* , decreases with underdoping, in a non-monotonic way, with a local minimum at $p = 0.11$. This

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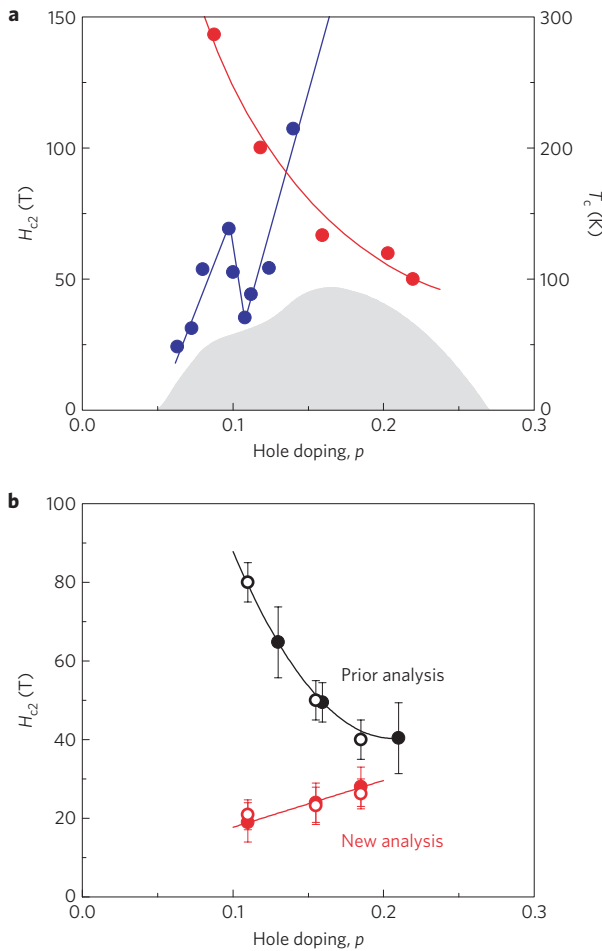


Figure 1 | Doping dependence of the upper critical field H_{c2} in cuprate superconductors. **a**, Upper critical field H_{c2} of cuprate superconductors versus doping p extracted from magneto-conductivity data on YBCO (ref. 7, blue circles; left axis) and from Nernst data on Bi-2212 (ref. 8, red circles; left axis). These two studies of superconducting fluctuations above T_c led to contradictory conclusions on how the superconducting pairing strength in cuprates varies with doping. The superconducting T_c of YBCO is also shown (ref. 34, grey dome; right axis). **b**, Two different definitions of the upper critical field H_{c2} extracted from Nernst data on the same material, Bi-2201, lead to opposite doping dependencies. When H_{c2} is defined as the field H where N versus H at $T = T_c$ extrapolates to zero (filled black circles and error bars, from ref. 8), one finds that H_{c2} increases with underdoping. The same increase is found for the peak field H^* in N versus H (see Fig. 3a) when taken at $T = T_c$ (open black circles; from Fig. 7, $\times 10$). By contrast, a decrease is found if the peak field H^* is taken at $T \approx 1.65T_c$ ($\epsilon \approx 0.65$), outside the region of strong paraconductivity (open red circles; from Fig. 7, $\times 1.9$). The latter result is independent of any theory. Equivalently, H_{c2} may be obtained as H_{c2}^* in a fit of the peak field H^* to $H_{c2}^* \ln(T/T_c)$ (see Fig. 3b), as done in Fig. 7 (filled red circles), again yielding an H_{c2} that decreases with underdoping. This is true whether H^* is obtained from Nernst data (away from the region of strong paraconductivity) or from diamagnetism (Fig. 7). The error bars on the filled red circles reflect the uncertainty in fitting the H^* data points in Fig. 7 to the relation $H_{c2}^* \ln(T/T_c)$. Error bars on open circles (red and black) reflect the uncertainty in determining H^* from the data in refs 10,20 (see Fig. 7).

result comes directly from the Nernst data, free of any model or theory. In fact, the evolution of H_{c2}^* may be directly read off the raw N versus H isotherms: it is simply proportional to the field H^* at which N peaks for a given reduced temperature, say $T = 1.5T_c$ (Supplementary Fig. S5).

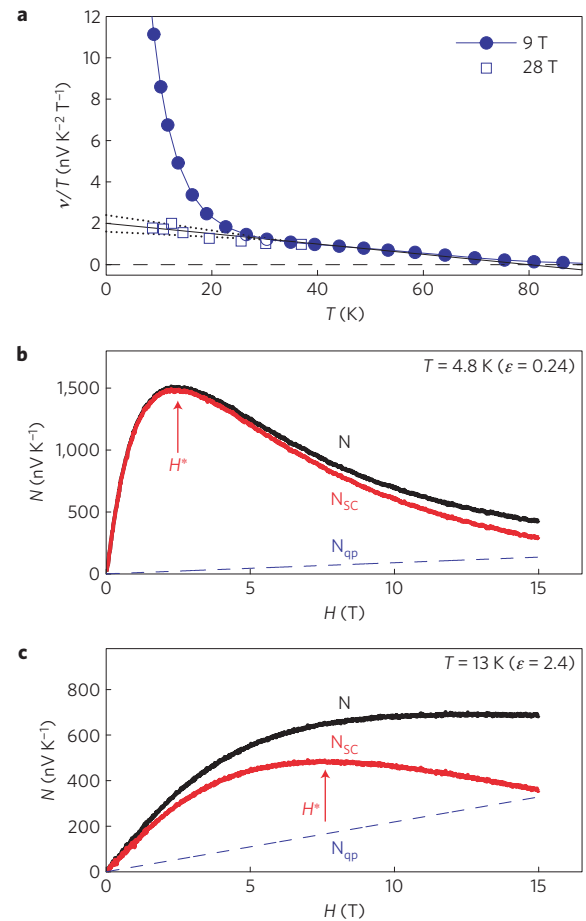


Figure 2 | Quasiparticle and superconducting contributions to the Nernst signal in Eu-LSCO. **a**, Temperature dependence of the Nernst coefficient $\nu \equiv N/H$ of Eu-LSCO at $p = 0.125$, plotted as ν/T versus T , for two magnetic fields as indicated. $\nu(T)$ is independent of H for $T > 35$ K, and hence $\nu(T) \approx \nu_{qp}(T)$ above 35 K (ref. 32). In the range 35–70 K, ν/T is linear, so that $\nu_{qp}/T = a - bT$ (solid black line). Application of a field of 28 T (open squares) to suppress superconductivity such that $\nu_{sc} \ll \nu_{qp}$ shows that the linear T dependence of $\nu_{qp}(T)$ persists as $T \rightarrow 0$. We therefore take $N_{qp} = \nu_{qp}H = (a - bT)TH$ to be our estimate of the quasiparticle contribution used to extract $N_{sc}(T, H) = N(T, H) - N_{qp}(T)$. The same procedure is applied to the data at $p = 0.08, 0.10$ and 0.11 , with parameters a and b that are only slightly different. The dashed lines are the upper and lower bounds on the uncertainty in $\nu_{qp}(T)$. These translate into error bars in the determination of H^* plotted in Fig. 3b,c (see panels below). **b,c**, Nernst signal $N = \nu H$ (black) measured in Eu-LSCO at $p = 0.11$ and quasiparticle contribution $N_{qp} = \nu_{qp}H$ (dashed blue line), for two temperatures as indicated. The difference $N_{sc} = N - N_{qp}$ (red) is the contribution of superconducting fluctuations. In **b**, where $\epsilon < 1$, N_{qp} is small compared with the measured Nernst signal and hence the extraction of the peak field H^* , defined as the field where N_{sc} versus H is maximal, is essentially unaffected by the subtraction of N_{qp} . By contrast for $\epsilon > 1$ (**c**), N_{qp} is large and subtraction is necessary to obtain an accurate determination of H^* .

Nernst data on $\text{Nb}_{0.15}\text{Si}_{0.85}$ (refs 11,12) yield a peak field in agreement with $H^* = H_{c2}^* \ln(T/T_c)$ up to at least $5 T_c$ (Fig. 3c). Pourret *et al.* point out that H^* separates a low- H regime controlled by the temperature-dependent coherence length $\xi(T) = \xi_0 / (\ln(T/T_c))^{1/2}$ and a high- H regime controlled by the magnetic length $l_B = (h/2eH)^{1/2}$ (ref. 17) where ξ_0 is the $T = 0$ superconducting coherence length, h is Planck's constant, and e is the electron charge. They argue that H^* is the field where $\xi(T) = l_B(H^*)$, so that $H^* = (\Phi_0/2\pi\xi_0^2) \ln(T/T_c)$, where $\Phi_0 = h/2e$ is the flux quantum.

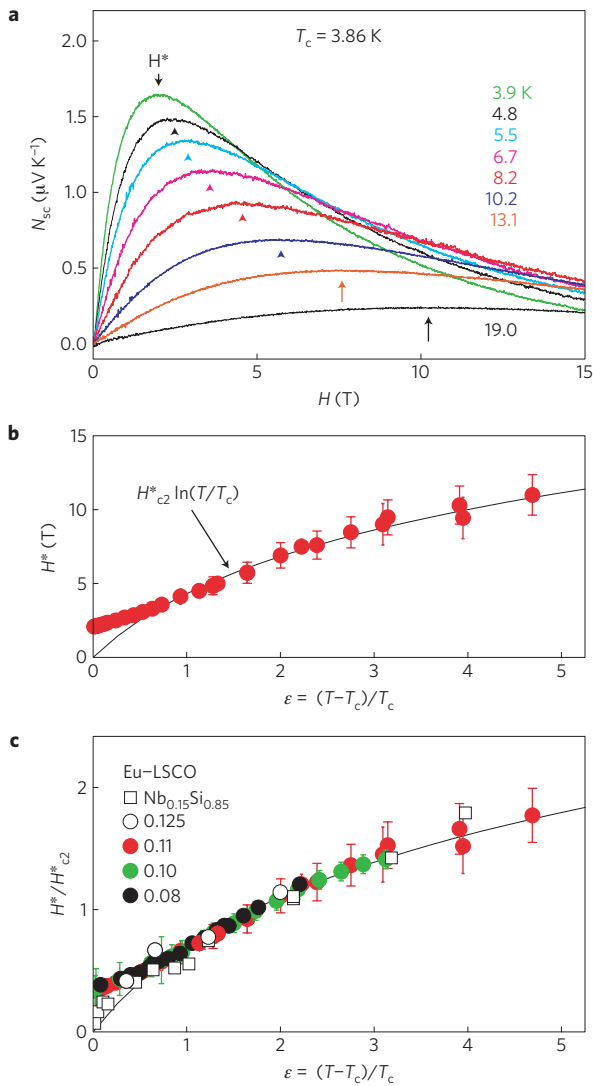


Figure 3 | Peak field H^* in the superconducting Nernst signal above T_c . **a**, Superconducting Nernst signal N_{sc} of Eu-LSCO plotted as a function of magnetic field H at $p = 0.11$, for different values of the temperature T as indicated. A quasiparticle background N_{qp} is subtracted from the raw data of N versus H in Supplementary Fig. S1 (see Fig. 2). The arrows mark the maximum value of $N_{sc} = N - N_{qp}$ for each isotherm, which defines the peak field H^* (also called the ‘ghost critical field’). **b**, Temperature dependence of H^* extracted from the data in **a** plotted versus reduced temperature $\varepsilon \equiv (T - T_c)/T_c$. The solid line is a fit to $H^* = H_{c2}^* \ln(T/T_c)$, which provides a model-free field scale, H_{c2}^* , equal to 6.2 T for this doping. **c**, Peak field H^* in Eu-LSCO at $p = 0.08, 0.10, 0.11$ and 0.125 , and in the conventional superconductor $\text{Nb}_{0.15}\text{Si}_{0.85}$ (from ref. 11), plotted as H^*/H_{c2} versus ε , with H_{c2}^* obtained as in **b** for each sample. The values of H_{c2}^* are given in Supplementary Table S1 and plotted in Fig. 5a. Error bars on H^* in **b** and **c** come from the uncertainty in N_{qp} (Fig. 2).

This makes the field scale H_{c2}^* equal to the $T = 0$ upper critical field $H_{c2} \equiv \Phi_0/2\pi\xi_0^2$. In Supplementary Fig. S6, we use resistivity data on Nd-LSCO, a material very similar to Eu-LSCO, to show that the field H_{c2}^{res} needed to suppress superconductivity in Nd-LSCO at $p = 0.12$ and $T = T_c/20$ is roughly equal to H_{c2}^* in Eu-LSCO at $p = 0.125$ (Fig. 5a). This confirms experimentally that $H_{c2}^* \approx H_{c2}$.

Comparison to Gaussian theory

We now compare our data against the theory of Gaussian fluctuations^{13–15}. The calculated curve of α_{xy}^{sc} versus H (ref. 15) is

in excellent agreement with the measured curve of N_{sc} versus H (Fig. 6). The peak field in α_{xy}^{sc} versus H increases with temperature roughly as $H^* \sim \ln(T/T_c)$ (Supplementary Fig. S7), with a prefactor that is proportional to $1/\xi_0^2$ (Supplementary Fig. S7). In the $H = 0$ limit, theory predicts^{14,15}: $\nu_{sc} \sigma = \alpha_{xy}^{\text{sc}}/H \sim \xi_0^2/T \ln(T/T_c)$. In Fig. 4a, the Nernst coefficient $\nu (\equiv N/H)$ of Eu-LSCO at $p = 0.11$ is plotted versus H for different temperatures above T_c . Its value in the $H = 0$ limit, $\nu_0 \equiv \nu(H \rightarrow 0)$, is plotted in Fig. 4b as a function of ε . In Fig. 4d, the data are seen to follow the theoretical temperature dependence precisely, from $1.02 T_c$ up to at least $5 T_c$, as previously found^{14,15} in $\text{Nb}_{0.15}\text{Si}_{0.85}$ (refs 11,12). As shown in Fig. 6 (and explained in the Supplementary Information), the data also follow the theoretical dependence on magnetic field, both at $T < T_c$ and at $T > T_c$. We conclude that our Nernst data on Eu-LSCO are consistent with several non-trivial features of Gaussian theory. This validates the earlier use of Gaussian theory to analyse conductivity data⁷ in YBCO (in a context of much smaller signal-to-background ratio; Supplementary Table S2), discussed below. Note that the quantum oscillations observed in YBCO at $p \sim 0.1$ (ref. 18) are consistent with Fermi-liquid theory¹⁹, the framework on which Gaussian theory is based. Agreement with Gaussian theory and consistency of the different measures of H_{c2} indicate that the superconducting fluctuations in this cuprate are controlled entirely by the coherence length, and there is only one temperature scale, T_c , and one field scale, H_{c2} , for superconductivity. The fluctuations seem to be no different from those of conventional superconductors, and there is no need to invoke unusual vortex-like excitations^{8–10,16,20,21}, at least for $p = 0.08$ and above.

Nernst effect and diamagnetism in Bi-2201

Wang *et al.*⁸ extracted a field scale from their raw Nernst data on the cuprates Bi-2212 and $\text{Bi}_2\text{Sr}_{2-y}\text{La}_y\text{Cu}_2\text{O}_6$ (Bi-2201), and found it to increase with underdoping (Fig. 1). However, because they used Nernst data at $T = T_c$, their analysis was contaminated by paraconductivity. Our analysis of their data⁸ away from T_c yields a field scale that decreases with underdoping, in agreement with diamagnetism data²⁰ on Bi-2201 (Fig. 7). The H dependence of M_d , the diamagnetic component of magnetization, is very similar to that of N_{sc} (ref. 21), as expected theoretically^{16,22}. Magnetization data on an underdoped sample of Bi-2201 with $T_c = 12$ K (ref. 20) yield a peak value H_d^* that obeys $H_{c2}^* \ln(T/T_c)$ all the way from $T \approx T_c$ to $T \approx 4T_c$ (Fig. 7), with $H_{c2}^* \approx 19$ T. Applying the same fit to published Nernst data on a Bi-2201 sample of the same doping⁸ yields the same value of H_{c2}^* (Figs 1b and 7). In summary, both diamagnetic and Nernst signals in the cuprate Bi-2201 obey the relation $H^* = H_{c2}^* \ln(T/T_c)$ (Fig. 7), from which the same field scale can be reliably extracted, and this field scale (proportional to H_{c2}) decreases with underdoping (Fig. 1b). This resolves the apparent contradiction highlighted in Fig. 1a.

Critical field H_{c2} in YBCO

In YBCO, a cuprate with $T_c \approx 60$ K at $p = 0.11$, the effect of superconducting fluctuations on the in-plane electrical conductivity σ was analysed up to $\varepsilon \approx 1$ for a range of dopings⁷, using the Aslamazov–Larkin theory of Gaussian fluctuations. The only fit parameter in the theory is ξ_0 , plotted in Fig. 5a as $H_{c2} = \Phi_0/2\pi\xi_0^2$ versus p . H_{c2} is seen to have a minimum at $p = 0.11$, just as in Eu-LSCO. In Fig. 5b, we show that this value of H_{c2} (obtained from fluctuations above T_c) is in good agreement with the value of H_{c2}^{res} measured at $T \ll T_c$ directly by high-field transport^{23,24}. H_{c2}^{res} is defined as the field where ρ or the Hall coefficient R_H or the Seebeck coefficient S has reached its normal-state value^{23,24}. For example, in YBCO at $p = 0.11$, S versus H at $T = 2$ K yields $H_{c2}^{\text{res}} \approx 25$ T (ref. 24). A third, independent estimate of the coherence length ξ_0 is the vortex core radius measured by muon spin relaxation deep in the vortex state (at low H and $T \ll T_c$). In YBCO at

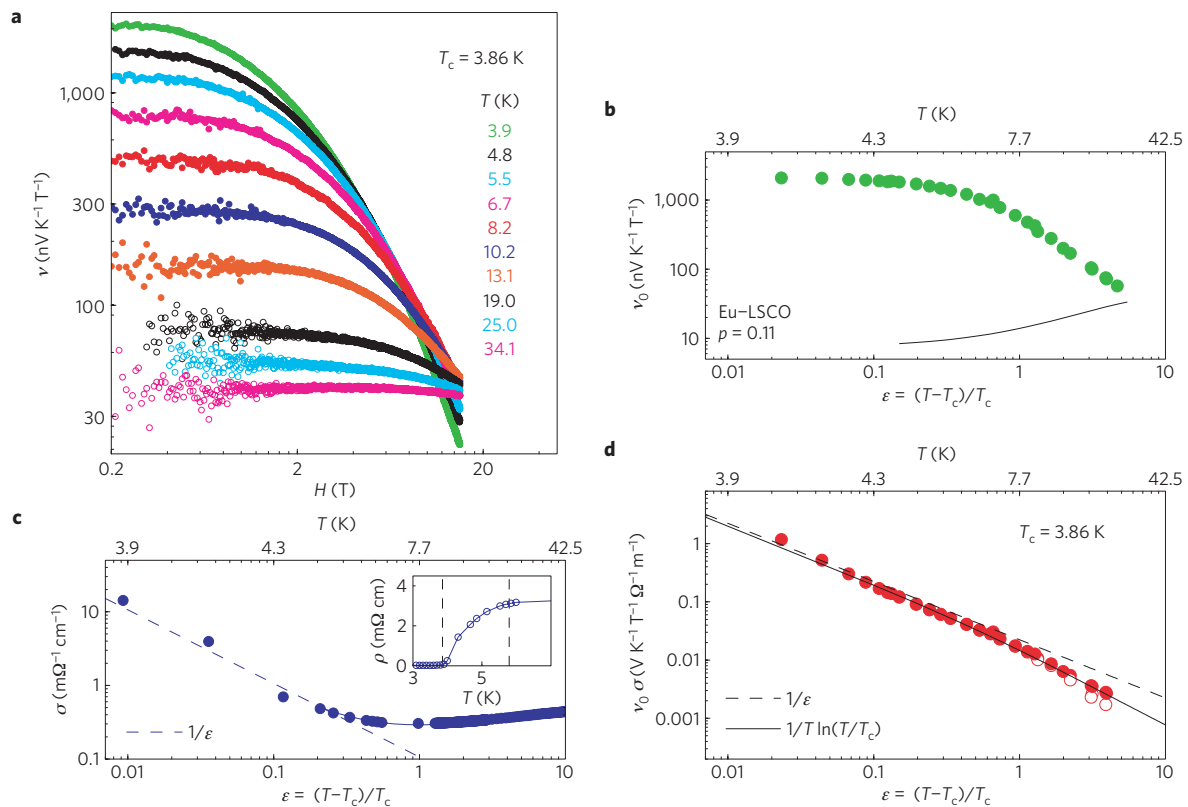


Figure 4 | Temperature dependence: comparison to Gaussian theory. **a**, Nernst coefficient ν of Eu-LSCO at $p = 0.11$ as a function of magnetic field H , plotted on a log–log scale for various values of the temperature T as indicated. **b**, Temperature dependence of the Nernst coefficient in the $H = 0$ limit, ν_0 , plotted versus reduced temperature ϵ on a log–log scale. The solid line is the quasiparticle background ν_{qp} obtained in Fig. 2. **c**, Zero-field conductivity $\sigma (=1/\rho)$ from Supplementary Fig. S3. The dashed line shows the temperature dependence expected from Gaussian fluctuation theory close to T_c (ref. 13). Inset: zoom on the resistivity near the transition. The lower vertical dashed line marks $T_c = 3.86$ K; the upper vertical dashed line marks $T = 1.5T_c$. **d**, Product of ν_0 in **b** and σ in **c**. Open circles show the effect of subtracting ν_{qp} from ν_0 , to obtain $\nu_{sc}(B \rightarrow 0) = \nu_0 - \nu_{qp}$. The solid line is the theoretical expectation for the Peltier coefficient in the $B \rightarrow 0$ limit, $\alpha_{xy}^{sc} \sim \nu_{sc}\sigma$, from Gaussian fluctuations of the order parameter in a 2D superconductor^{14,15}. The dashed line is the expected limiting behaviour close to T_c (ref. 13).

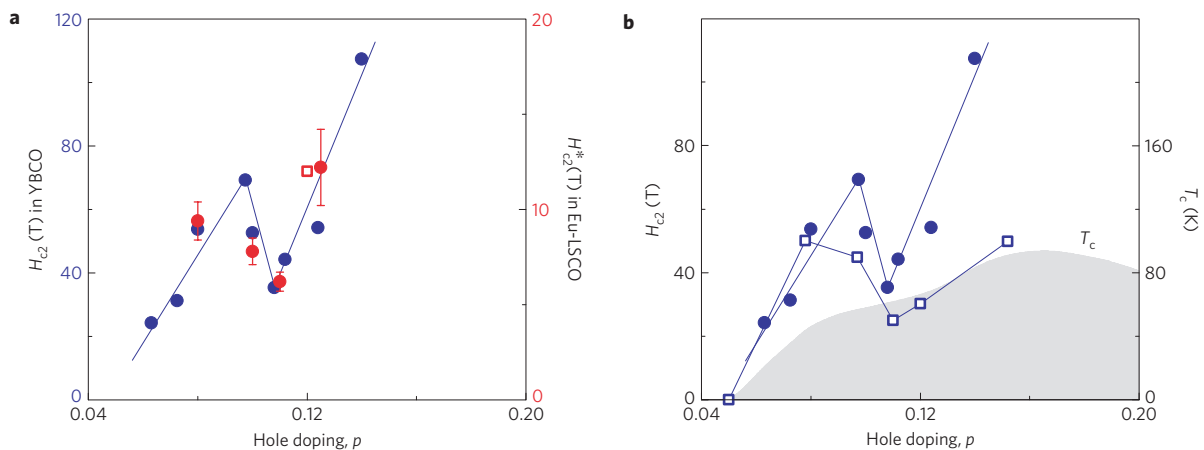


Figure 5 | Doping dependence of H_{c2} in YBCO and Eu-LSCO. **a**, Upper critical field H_{c2} obtained from the superconducting fluctuations above T_c , plotted as a function of hole doping p . For YBCO (blue symbols; left axis), H_{c2} is defined via the relation $H_{c2} \equiv \Phi_0/2\pi\xi_0^2$, in terms of the zero-temperature coherence length ξ_0 obtained from a Gaussian analysis of the fluctuation magneto-conductivity (ref. 7). For Eu-LSCO (red symbols; right axis), H_{c2} is taken to be H_{c2}^* in the fit of the peak field H^* to $H^* = H_{c2}^* \ln(T/T_c)$ (Fig. 3). Error bars on H_{c2}^* correspond to the uncertainty in fitting to the H^* data points in Fig. 3c. The red square marks the value of H_{c2} obtained directly from resistivity measurements at $T \rightarrow 0$ on Nd-LSCO, a material very similar to Eu-LSCO (Supplementary Fig. S6). **b**, Comparison of H_{c2} in YBCO determined in two different ways: from ξ_0 above T_c (circles), as in **a**, and from high-field transport measurements^{23,24} (squares) that suppress superconductivity at low temperature ($T \ll T_c$). The two measures of H_{c2} are in reasonable agreement; in particular, they both have a minimum at $p = 0.11$, where $H_{c2} \approx 30$ T. The doping dependence of the zero-field T_c is shown for comparison (grey dome, right axis; ref. 34).

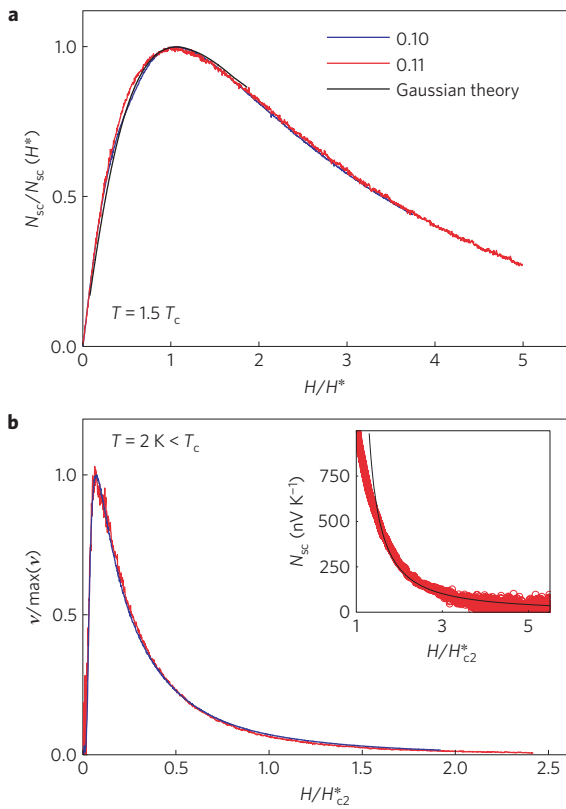


Figure 6 | Field dependence: comparison to Gaussian theory. **a**, Nernst signal N of Eu-LSCO at $p = 0.10$ (blue; $T_c = 5.0$ K) and $p = 0.11$ (red; $T_c = 3.86$ K) as a function of H/H^* , for a temperature $T = 1.5T_c$. At this temperature, the peak field H^* , at which N_{sc} versus H is maximal, is $H^* = 4.0$ T and 3.0 T, respectively. The solid line is a theoretical calculation of $\alpha_{xy}^{sc} (=N_{sc}\sigma)$, as in ref. 15, for a diffusion constant $D = 0.178$ cm² s⁻¹ (K. Michaeli, personal communication). Note that the experimental σ at $T = 1.5T_c$ is essentially independent of H so that the H dependences of N_{sc} and α_{xy}^{sc} are the same. The theoretical curve is independent of D when normalized at H^* , for a wide range of D values (K. Michaeli, personal communication; Supplementary Fig. S7). **b**, Nernst coefficient of Eu-LSCO at $p = 0.10$ (blue) and $p = 0.11$ (red), for a temperature $T = 2$ K $< T_c$, plotted versus H/H_c2^* , where H_c2^* is obtained from the peak field $H^* = H_c2^* \ln(T/T_c)$ (Fig. 3c). The fact that the data for different dopings collapse on the same curve when normalized by H_c2^* for both $T < T_c$ and $T > T_c$ (see also Supplementary Fig. S8) shows that H_c2^* is the relevant field scale throughout the H - T phase diagram. Inset: N_{sc} versus H/H_c2^* for $p = 0.11$ at the high-field end (up to 34 T), compared against the limiting behaviour expected theoretically^{14,15} for $\alpha_{xy} = N_{sc}\sigma$ at $H \gg H_c2$, namely $\alpha_{xy} \sim 1/H \ln(H/H_c2)$ (solid line).

$p \approx 0.11$, the vortex core radius is ≈ 3 nm, so that $H_c2 \approx 30$ T (ref. 25). The fact that three very different measures of H_c2 agree is compelling evidence that the correct value of H_c2 has been reached in underdoped YBCO, with a minimum of $H_c2 \approx 30$ T at $p = 0.11$. The agreement also confirms the validity of Gaussian theory. We conclude that the upper critical field H_c2 of cuprates decreases with underdoping, in the same non-monotonic fashion in Eu-LSCO and YBCO.

We attribute this non-monotonic weakening of superconductivity to the competing effect of stripe order. Stripe order is present in Eu-LSCO above $p = 0.08$ (ref. 26). In YBCO, stripe order was recently inferred from Seebeck measurements of Fermi-surface reconstruction²⁴ and confirmed by high-field NMR measurements²⁷. This scenario of phase competition is akin to that found in iron-based, heavy-fermion and organic superconductors,

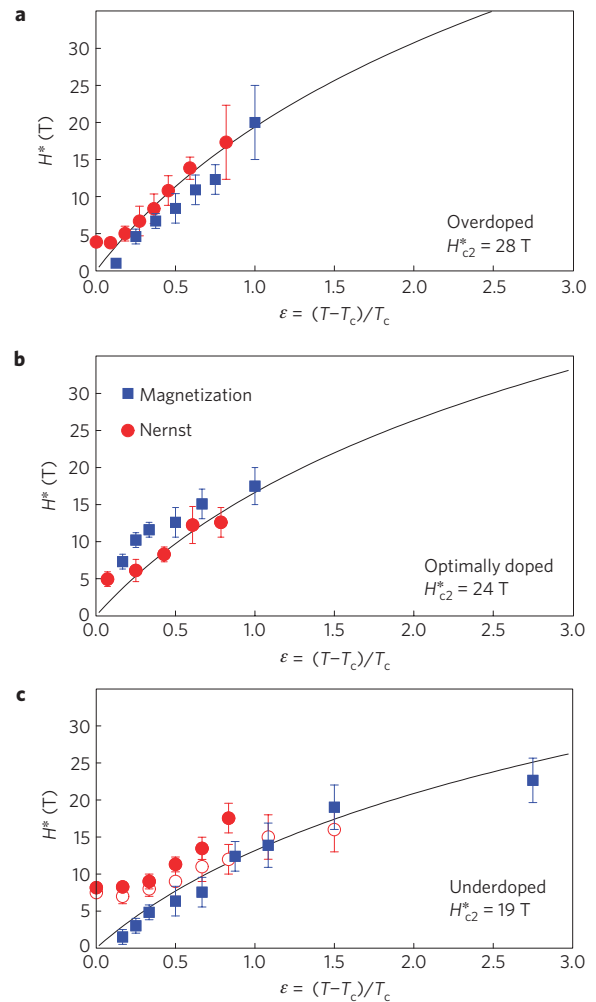


Figure 7 | Peak field H^* in Bi-2201. Peak field H^* , plotted versus $\epsilon = (T - T_c)/T_c$, obtained from published data^{10,20} on the cuprate Bi-2201, for three different dopings: **a**, overdoped ($T_c = 22$ K for Nernst¹⁰ and $T_c = 20$ K for magnetization²⁰), **b**, optimally doped ($T_c = 28$ K for Nernst¹⁰ and $T_c = 30$ K for magnetization²⁰), and **c** underdoped ($T_c = 12$ K for both Nernst¹⁰ and magnetization²⁰). H^* is extracted from Nernst data¹⁰, as the peak in N versus H (filled red circles), and H_d^* is extracted from magnetization data²⁰, as the peak in the diamagnetic signal $-|M_d|$ versus H (blue squares). In the overdoped and optimally doped samples, where the quasiparticle term N_{qp} is very small¹⁰, H^* agrees well with H_d^* (above $\epsilon \approx 0.2$). In the underdoped sample ($T_c = 12$ K), however, a large positive N_{qp} must be subtracted to get a meaningful H^* , as in underdoped Eu-LSCO (Fig. 2). The open red circles are obtained assuming the same N_{qp} as in underdoped Eu-LSCO (Fig. 2a); they agree well with the magnetization data (blue squares). The solid lines are fits of the Nernst data to the expression $H^* = H_c2^* \ln(T/T_c)$, giving values of H_c2^* as indicated, with error bars of ± 5 T. The fits reveal that H_c2^* decreases with decreasing p . The opposite conclusion would be reached if, as done previously⁸, the focus were placed on the Nernst data at $T \approx T_c$ (Fig. 1b), where contamination by field-dependent paraconductivity is maximal (Supplementary Fig. S4). The error bars reflect the uncertainty in determining H^* from the data of refs 10 and 20.

where the competing phase in these cases is spin-density-wave order. In YBCO at lower doping ($p < 0.08$), the rapid drop in T_c and H_c2 (Fig. 5b) may be due to other phases, such as spin-density-wave order below $p \approx 0.08$ (ref. 28) and antiferromagnetism below $p \approx 0.05$. At low doping, the approach to the Mott insulator may also play a role.

Methods

Nernst effect. The Nernst effect²⁹ is the development of a transverse electric field E_y across the width (y axis) of a metallic sample when a temperature gradient $\partial T/\partial x$ is applied along its length (x axis) in the presence of a perpendicular magnetic field H (along the z axis). The Nernst signal is $N = E_y/(\partial T/\partial x)$ and the Nernst coefficient $\nu = N/H$. Two mechanisms can give rise to a Nernst signal: superconducting fluctuations, which give a positive signal N_{sc} , and charge carriers (quasiparticles), which give a signal N_{qp} that may be of either sign. The measured signal is their sum: $N = N_{sc} + N_{qp}$ (see Supplementary Information). The superconducting term N_{sc} is strongly dependent on the field strength H , whereas the quasiparticle term N_{qp} is essentially linear in field (that is, ν_{qp} is constant).

Samples. Single crystals of $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ (Eu-LSCO) were grown by the travelling floating zone method in Tokyo. The hole doping p is taken to be the nominal Sr concentration x . The characteristics of our four samples are listed in Supplementary Table S1. The superconducting transition temperature T_c was determined as the temperature below which the resistivity $\rho = 0$. For each sample, three pairs of silver epoxy contacts were diffused into the surface. Contacts used to measure the temperature gradient were separated by a distance L and the transverse contacts used to measure the Nernst voltage were separated by a distance w . The ratio L/w was typically in the range 0.5–3.

Single crystals of $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ (Nd-LSCO) used for determining H_{c2} from the resistivity (Supplementary Fig. S6) were described in ref. 30; their T_c and H_{c2} values are given in Supplementary Table S1.

Electrical resistivity. A standard four-probe method was used to measure the resistivity ρ in zero field. In Supplementary Fig. S3, we show ρ versus T for our four Eu-LSCO samples. Both temperature and doping dependences of the resistivity are consistent with published data³¹, as is the magnitude just above T_c ($\rho \approx 1 \text{ m}\Omega \text{ cm}$). The resistivity of Nd-LSCO samples is shown in Supplementary Fig. S6.

Nernst measurements. The Nernst effect was measured using a one-heater two-thermometer setup, as described in refs 32 and 33. The magnetic field, applied along the c axis, was swept between -15 and $+15 \text{ T}$ at a rate of 0.4 T min^{-1} or slower. Also, for Eu-LSCO samples with $p = 0.10$ and 0.125 ($p = 0.11$), the Nernst coefficient was measured up to 28 T (34 T) at the high-field laboratory in Grenoble Laboratoire National des Champs Magnétiques Intenses (LNCMI). Heat was applied horizontally in the ab plane and the temperature gradient $dT = \Delta T/L$ was obtained by measuring the temperature difference ΔT between two contacts separated by the length L , using two uncalibrated Cernox chips referenced to a third calibrated chip. The Nernst voltage $V_y = E_y/w$ across two transverse contacts separated by the length w was measured using a nanovolt preamplifier and a nanovoltmeter.

The Nernst signal N is, as the Hall signal, anti-symmetric in field, so that by taking the difference

$$N = \{(V_y(H) - V_y(-H))/dT\}(L/2w)$$

any symmetric component is eliminated. For example, contamination from the Seebeck effect due to slightly misaligned contacts is removed in this fashion. A constant background coming from the measurement circuit is also eliminated.

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References

1. Norman, M. R. *et al.* The pseudogap: Friend or foe of high T_c ? *Adv. Phys.* **54**, 715–733 (2005).
2. Emery, V. J. & Kivelson, S. A. Importance of phase fluctuations in superconductors with small superfluid density. *Nature* **374**, 434–437 (1995).
3. Kanigel, A. *et al.* Evidence for pairing above the transition temperature of cuprate superconductors from the electronic dispersion in the pseudogap phase. *Phys. Rev. Lett.* **101**, 137002 (2008).
4. Chatterjee, U. *et al.* Observation of a d -wave nodal liquid in highly underdoped Bi2212 . *Nature Phys.* **6**, 99–103 (2010).
5. Tanaka, K. *et al.* Distinct Fermi-momentum-dependent energy gaps in deeply underdoped Bi2212 . *Science* **314**, 1910–1913 (2006).
6. Kondo, T. *et al.* Competition between the pseudogap and superconductivity in the high- T_c copper oxides. *Nature* **457**, 296–300 (2008).
7. Ando, Y. & Segawa, K. Magnetoresistance of untwinned $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystals in a wide range of doping: Anomalous hole-doping dependence of the coherence length. *Phys. Rev. Lett.* **88**, 167005 (2002).
8. Wang, Y. *et al.* Dependence of upper critical field and pairing strength on doping in cuprates. *Science* **299**, 86–89 (2003).
9. Xu, Z. A. *et al.* Vortex-like excitations and the onset of superconducting phase fluctuations in underdoped LSCO. *Nature* **406**, 486–488 (2000).
10. Wang, Y. *et al.* Nernst effect in high- T_c superconductors. *Phys. Rev. B* **73**, 024510 (2006).
11. Pourret, A. *et al.* Observation of the Nernst signal generated by fluctuating Cooper pairs. *Nature Phys.* **2**, 683–686 (2006).
12. Pourret, A. *et al.* Nernst effect as a probe of superconducting fluctuations in disordered thin films. *New J. Phys.* **11**, 055071 (2009).

13. Ussishkin, I., Sondhi, S. L. & Huse, D. A. Gaussian superconducting fluctuations, thermal transport, and the Nernst effect. *Phys. Rev. Lett.* **89**, 287001 (2002).
14. Serbyn, M. N. *et al.* Giant Nernst effect due to fluctuating Cooper pairs in superconductors. *Phys. Rev. Lett.* **102**, 067001 (2009).
15. Michaeli, K. & Finkel'stein, A. M. Fluctuations of the superconducting order parameter as an origin of the Nernst effect. *Europhys. Lett.* **86**, 27007 (2009).
16. Podolsky, D., Raghun, S. & Vishwanath, A. Nernst effect and diamagnetism in phase-fluctuating superconductors. *Phys. Rev. Lett.* **99**, 117004 (2007).
17. Pourret, A. *et al.* Length scale for the superconducting Nernst signal above T_c in $\text{Nb}_{0.15}\text{Si}_{0.85}$. *Phys. Rev. B* **76**, 214504 (2007).
18. Doiron-Leyraud, N. *et al.* Quantum oscillations and the Fermi surface in an underdoped high- T_c superconductor. *Nature* **447**, 565–568 (2007).
19. Sebastian, S. E. *et al.* Fermi-liquid theory in an underdoped high- T_c superconductor. *Phys. Rev. B* **81**, 140505 (2010).
20. Li, L. *et al.* Diamagnetism and Cooper pairing above T_c in cuprates. *Phys. Rev. B* **81**, 054510 (2010).
21. Wang, Y. *et al.* Field-enhanced diamagnetism in the pseudogap state of the cuprate superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ in an intense magnetic field. *Phys. Rev. Lett.* **95**, 247002 (2005).
22. Schneider, T. & Weyeneth, S. Diamagnetism, Nernst signal, and finite-size effects in superconductors above the transition temperature T_c . *Phys. Rev. B* **83**, 144527 (2011).
23. LeBoeuf, D. *et al.* Lifshitz critical point in the cuprate superconductor $\text{YBa}_2\text{Cu}_3\text{O}_y$ from high-field Hall effect measurements. *Phys. Rev. B* **83**, 054056 (2011).
24. Laliberté, F. *et al.* Fermi-surface reconstruction by stripe order in cuprate superconductors. *Nature Commun.* **2**, 432 (2011).
25. Sonier, J. E. *et al.* Hole-doping dependence of the magnetic penetration depth and vortex core size in $\text{YBa}_2\text{Cu}_3\text{O}_y$: Evidence for stripe correlations near $1/8$ hole doping. *Phys. Rev. B* **76**, 134518 (2007).
26. Fink, J. *et al.* Phase diagram of charge order in $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$ from resonant soft x-ray diffraction. *Phys. Rev. B* **83**, 092503 (2011).
27. Wu, T. *et al.* Magnetic-field-induced charge-stripe order in the high-temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_y$. *Nature* **477**, 191–194 (2011).
28. Haug, D. *et al.* Neutron scattering study of the magnetic phase diagram of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$. *New J. Phys.* **12**, 105006 (2010).
29. Behnia, K. The Nernst effect and the boundaries of the Fermi-liquid picture. *J. Phys.: Condens. Matter* **21**, 113101 (2009).
30. Daou, R. *et al.* Linear temperature dependence of the resistivity and change in the Fermi surface at the pseudogap critical point of a high- T_c superconductor. *Nature Phys.* **5**, 31–34 (2009).
31. Hess, C. *et al.* Nernst effect of stripe ordering $\text{La}_{1.8-x}\text{Eu}_{0.2}\text{Sr}_x\text{CuO}_4$. *Eur. Phys. J. Spec. Top.* **188**, 103–112 (2010).
32. Cyr-Choinière, O. *et al.* Enhancement of the Nernst effect by stripe order in a high- T_c superconductor. *Nature* **458**, 743–745 (2009).
33. Daou, R. *et al.* Broken rotational symmetry in the pseudogap phase of a high- T_c superconductor. *Nature* **463**, 519–522 (2010).
34. Liang, R. *et al.* Evaluation of CuO_2 plane hole doping in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ single crystals. *Phys. Rev. B* **73**, 180505 (2006).

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Author contributions

J.C. initiated the project; J.C., N.D.-L., O.C.-C., F.L., E.H., J.-Ph.R. and R.D. performed the Nernst measurements in Sherbrooke; J.C., N.D.-L., F.L., O.C.-C. and G.G. performed the Nernst measurements at the LNCMI in Grenoble; S.P., T.T. and H.T. prepared the Eu-LSCO samples and measured their resistivity; J.C., N.D.-L. and L.T. analysed the data; J.C., N.D.-L. and L.T. wrote the manuscript; L.T. supervised the project.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to L.T.

Competing financial interests

The authors declare no competing financial interests.

Decrease of upper critical field with underdoping in cuprate superconductors

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In the version of this Article originally published online, the unit on the y axis of Fig. 4c should have read ($\text{m}\Omega^{-1} \text{cm}^{-1}$). This error has been corrected in the HTML and PDF versions of the Article.