Field-induced nematic-like magnetic transition in an iron pnictide superconductor, 
\( \text{Ca}_{10}(\text{Pt}_3\text{As}_8)((\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2)_5 \)

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We report a high magnetic field study up to 55 T of the nearly optimally doped iron-pnictide superconductor \( \text{Ca}_{10}(\text{Pt}_3\text{As}_8)((\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2)_5 \) \((x=0.078(6))\) with a \( T_c \approx 10 \) K using magnetic torque, tunnel diode oscillator technique and transport measurements. We determine the superconducting phase diagram, revealing an anisotropy of the irreversibility field up to a factor of 10 near \( T_c \) and signatures of multiband superconductivity. Unexpectedly, we find a spin-flop like anomaly in magnetic torque at 22 T, when the magnetic field is applied perpendicular to the \((ab)\) planes, which becomes significantly more pronounced as the temperature is lowered to 0.33 K. As our superconducting sample lies well outside the antiferromagnetic region of the phase diagram, the observed field-induced transition in torque indicates a spin-flop transition not of long-range ordered moments, but of nematic-like antiferromagnetic fluctuations.

It is widely believed that superconductivity in iron pnictides and selenides is intimately related to their magnetic properties. The unusual stripe order [1] which breaks the four-fold symmetry (or, more precisely, fluctuations associated with this order) are often considered to play a key role in superconducting pairing. According to this point of view, short-range antiferromagnetic fluctuations survive well above the Néel temperature and gain additional stability by acquiring a nematic component, that is, the simultaneous excitation of antiferromagnetic fluctuations with the same wave vector but different phases leads to orbital fluctuations and additional energy gain. Detecting such nematic fluctuations above the structural transition is extremely challenging, although there is some experimental evidence to their existence [2]. In this letter we present a magnetic-field tuned experiment that suggests the existence of nematic-like antiferromagnetic fluctuations with preferential spin directions in an nearly optimally doped Fe-based superconductor in the absence of a true long-range magnetic order.

Recently, \( \text{Ca}_{10}(\text{Pt}_3\text{As}_8)(\text{Fe}_2\text{As}_2)_5 \) was found to be the parent compound of a new class of Fe-based superconductors, commonly called 10-3-8 [6]. As shown in Fig. 1(b) and (c), the characteristic Fe-As layers are separated by Ca atoms and a Pt3As8 plane and the phase diagram for this system shows similar features to those found in other families of iron-based superconductors. Recently, optical imaging [8], NMR [4], powder x-ray diffraction and \( \mu \)SR measurements [10] have indicated the structural / magnetic phase transitions occur in the parent 10-3-8 compound around 100 K, where the NMR measurement probing the \( ^{75}\text{As} \) environment suggests that the Fe-As planes have a striped antiferromagnetic order similar to \( \text{BaFe}_2\text{As}_2 \). Superconductivity occurs under applied pressure [11] or with electron doping, either by La doping onto the Ca site [12], or Pt doping onto the Fe site (Fig. 1(a)). As the symmetries of the Fe-As and Pt-As planes are incompatible in the 10-3-8 phase, the system crystallizes in the triclinic \( P1 \) group [6]. However, given the large separation of the two subsystems the interaction of Fe-As with the Pt-As layer is weak and thus the electronic properties of the Fe-As layer are expected to follow the tetragonal symmetry as found in other iron-based superconductors. This has been confirmed.

![Fig. 1](image-url)

**Fig. 1.** a) Schematic phase diagram of \( \text{Ca}_{10}(\text{Pt}_3\text{As}_8)((\text{Fe}_{1-x}\text{Pt}_x)_2\text{As}_2)_5 \) as a function of electron doping \( x \) based on Refs. [6, 7]. Our compound lies at the right of this phase diagram as indicated by the arrow. (b) The structure of the 10-3-8 phase in which the Fe-As layers, Ca atoms and Pt-As layers (in (c)) form a layered structure that crystallizes in the low-symmetry \( P1 \) triclinic space group.
FIG. 2. a) Magnetic torque in an applied magnetic field at constant temperatures between 0.33 K and 3.23 K for a 10-3-8 sample S1 when \( H \perp ab \) (within \( \theta = 4.3^\circ \), where \( \theta \) is the angle between the magnetic field and the normal to the \( ab \) planes). Solid arrows indicate the positions of the irreversibility field \( \mu_0H_{irr} \), and the peak of the magnetic anomaly at \( \mu_0H_M \), fitted to a Lorentzian. c) Field dependence of torque as function of angle \( \theta \) at 0.33 K. d) A simulation of torque based on a spin-flop and a paramagnetic contribution described below, assuming the magnetic moments are aligned perpendicular to the \( ab \) plane in zero field.

by angle-resolved photo-emission spectroscopy [13, 14].

In this letter we report high magnetic field studies up to 55 T of the nearly optimally-doped superconducting Ca_{10}(Pt_3As_8)(Fe_{0.922}Pt_{0.078})_2As_2, with \( T_c \approx 10 \) K, using magnetic torque, tunnel diode oscillator technique and transport measurements. We determine the superconducting phase diagram, finding a high anisotropy up to \( \gamma_H = 10 \) near \( T_c \) and an irreversibility field up to 32 T. Furthermore, we find an anomaly at low temperatures at 22 T that is consistent with a spin-flop transition, when the magnetic field is perpendicular to the Fe-As planes and in close proximity to the superconducting phase. We suggest that this transition - which occurs in the absence of any long-range magnetic order in a quasi-two-dimensional system - could originate from strong nematic-like antiferromagnetic fluctuations between two configurations with different preferential orientations of the Fe spins.

Sample growth details are presented elsewhere [6]. X-Ray measurements confirmed the \( P1 \) triclinic symmetry group [15]. Wavelength-dispersive X-ray measurements give a doping level of \( x = 0.078(6) \) by constraining Fe+Pt=13 in the chemical formula. [16] Transport measurements were performed in a Quantum Design PPMS in magnetic fields up to 14 T and also in pulsed fields up to 55 T at LNCMI, Toulouse with low-resistance electrical contacts made using Sn solder. Torque measurements were performed at low temperatures (down to 0.33 K) in magnetic fields up to 33 T at the HMFL in Nijmegen and in pulsed fields up to 55 T at LNCMI in Toulouse. Single crystals with typical size \( \approx 150 \times 150 \times 30 \) \( \mu \)m were used for torque measurements using highly sensitive piezo-resistive microcantilevers. All samples measured from the batch were found to be superconducting.

Fig. 2 shows magnetic torque data on two crystals (S1 and S2) when the magnetic field is approximately perpendicular to the \( ab \) planes. The typical hysteresis that occurs in the superconducting state due to the pinning of the flux vortices allows the determination of the superconducting phase diagram from the position of the irreversibility field \( \mu_0H_{irr} \), as indicated in Fig. 2(a) and (b). Furthermore, at magnetic fields above the superconducting hysteresis, we detect a clear anomaly at 22 T in the magnetic torque data that becomes sharper as the temperature is lowered to 0.33 K. This effect was reproduced in all the crystals investigated and no further anomalies were detected up to 55 T. This anomaly is well-defined only when the applied field is nearly perpendicular to the \( ab \) plane, and it is symmetric around this direction, as shown in Fig. 2(c).

The width of this peak at \( \mu_0H_M \) (the maximum in the torque signal after subtracting the featureless 9.6 K sweep), quantified by the half-width of Lorentzian fit to the subtracted data (inset of Fig. 2(b)), shows a linear decrease with temperature suggestive of slowing down of magnetic fluctuations. The observed behavior of magnetic torque in 10-3-8 is rather unusual and is in contrast to the paramagnetic \( \sim H^2 \) background torque typically observed in other iron-based superconductors above the irreversibility field [17, 18]. The observed anomaly at 22 T is likely to be of magnetic origin and the large magnitude of the anomaly compared to the superconducting hysteresis suggests that a significant proportion of the magnetic ions are involved, so an explanation involving impurities is unlikely.

In order to understand this behavior we consider first the Pt ions. Heavy Pt ions have strong spin-orbit coupling, and may experience large magnetic anisotropy. In the doped material, there are two distinct types of Pt: one in the Fe plane (Pt\(^{2+}\)) and another one in the Pt-As layer (Pt\(^{2+}\)) [19]. We do not expect the latter to be magnetic, given the d\(^{10}\) configuration of Pt\(^{2+}\), but the former, on the first glance, might spin polarize. Even with a relatively weak polarization it would have provided for a measurable anisotropy of the magnetic response. For that reason, we performed density functional theory (DFT) calculations on a \( \sqrt{5} \times \sqrt{5} \) 10-3-8 supercell where one Fe was substituted by Pt (see Supplementary Material, SM) and looked for spin polarization on Pt. We performed full lattice relaxation to avoid applying artificial internal pressure to the in-plane Pt. However, for both Pt positions (in-plane and intermediate layer) our calculations render completely polarization-free Pt, even though the surrounding Fe atoms acquire full polarization. Adding Hubbard \( U=2.5 \) eV and \( J=0.5 \) eV does not create spin polarization on Pt either.

Another way to rationalize this result is to look for spin-flop-like physics in the Fe subsystem. Although there is no reported long-range antiferromagnetic order at this doping...
(see Fig. 1), there are certainly antiferromagnetic fluctuations, which could become sufficiently long-lived at low temperatures to contribute to the magnetic torque. At a temperature $T \lesssim K$, where $K$ is the single-ion magnetic anisotropy energy, the fluctuating spins at zero field will lie predominantly along the easy axis and may be driven into a spin-flop transition by an external field; this will manifest in magnetic torque as a peak at $H_{SF}$, when the magnetic field is aligned along the zero-field easy axis. In experiments (Fig. 2(a,b)) the sharpness of the anomaly at $H_M$, which we now associate with a spin-flop field $H_{SF}$, varies substantially between 0.33 K and 3.23 K, and is completely featureless by $\sim 10$ K. Therefore a qualitative association of the temperature and magnetic anisotropy energy scales would suggest that $K$ is of the order of $\sim 1$ K (86 $\mu$eV). A direct calculation of $K$ from DFT for the Ca-10-3-8 parent compound ($\tau=0$) gives a value of $K \approx 106$ $\mu$eV which is of the same order of magnitude as the measured magnetic energy scale but predicts $b$ as the easy axis. In our experimental geometry the spin-flop explanation requires the $c^*$ axis, perpendicular to the $ab$ planes, to be the easy axis. However, analysis of the magnetic anisotropy in other pnictide compounds shows that DFT easy axis predictions do not always agree with experimental observations (see SM). A spin-flop field $H_{SF}$ could be linked to $ab$-initio calculations by the exchange energy that is calculated using the difference, $\Delta$, between the ferro- and antiferromagnetic ordered states, derived as $\mu_0 H_{SF} = 2\sqrt{K\Delta}/M$, where $M$ is the Fe moment (see SM). However, in Ca-10-3-8 the ferromagnetic state converges only in a fixed moment calculation which results in a large overestimation of $\Delta$ and correspondingly, $H_{SF}$. Comparison of $H_{SF}$ calculated for Ca-10-3-8 and other iron-based superconductors shows that DFT overestimates this value (see SM).

We can simulate the angular dependence of torque in a spin-flop scenario, including a Lorentzian broadening term ($\Gamma \approx 2.1$ T) and adding an additional $\tau \sim H^2 \times \sin 2\theta$ term to account for a paramagnetic contribution, as described in the SM. While a magnetization measurement of a spin-flop transition usually shows a magnetization step or slope change at the spin-flop field, magnetic torque as a function of applied field gives rise to a peak if the field is applied parallel to the easy axis [20]. Despite the simplicity of the model, the correspondence with the experimental data above $\mu_0 H_{tr}$ is remarkable, as shown in Fig. 3(d). The model assumes that the (fluctuating) magnetic moments are aligned perpendicular to the $ab$ plane in contrast to the in-plane collinear antiferromagnetic order typically found in the parent compounds of the iron-based superconductors. However, the anisotropy energies are not large (see SM) and given the more unusual crystal structure of 10-3-8, it is possible that the easy axis is perpendicular to the $ab$ planes. One may ask why similar spin-flop transitions have not been observed in other Fe pnictides. On this point, it is important to keep in mind that the magnetic anisotropy in these materials is not universal: neutron scattering studies suggest that in LaFeAsO, SrFe$_2$As$_2$ and BaFe$_2$As$_2$ the spins are aligned in-plane [11], but in NdFeAsO [21] along $c$. In the latter case the spin flop may be masked by spin-reorientation transitions in the Nd subsystem. Additionally, there is evidence in polarized neutron scattering measurements that the anisotropy of low-energy spin fluctuations [22] in electron-doped Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ does not reflect the easy axis of the ordered phase.

We have also performed in-plane and out-of-plane transport measurements to characterize the superconducting phase and to determine whether the transition in applied field can be detected in transport. Fig. 3(a) shows the temperature dependence of the in-plane resistance that has a crossover from a metallic-like to semiconductor-like regime [21] at a temperature $T^*$ around 80 K before it becomes superconducting at $T_c \approx 9.7$ K. The application of a magnetic field perpendicular to the planes (Fig. 3(c)) causes a substantial broadening of the superconducting transition. However, the superconducting properties are highly anisotropic, and the application of a field parallel to the $ab$ planes in Fig. 3(d) does not suppress the superconducting transition to the same extent. This broaden-
FIG. 4. (a) The superconducting field–temperature phase diagram of nearly optimally doped Ca$_{10}$(Pt$_3$As$_8$)((Fe$_{1-x}$Pt$_x$)$_2$As$_2$)$_2$, for two different orientations with respect to the (ab plane), as obtained from magnetic torque (circles) and transport measurements (squares). Open symbols indicate the pulsed fields measurements. Dashed line is the WHH fit to $H || \text{ab}$ data; the coloured phase boundaries are a guide to the eye. (b) The magnetic field–temperature phase diagram for $H \perp \text{ab}$ showing the position of the magnetic anomaly at $H_M$ (stars - taken from Fig. 2(a)) and the irreversibility field, $H_{irr}$, in the superconducting (SC) state. Black arrows represent the energetically favorable orientations of the fluctuating antiferromagnetic spins.

...ing indicates strong thermal fluctuations of the vortex lattice, which can be quantified by the Ginzburg number $G_i \approx 0.16$ [23], significantly higher than typical pnictides (using a large penetration depth of $\lambda_{ab}(0) \approx 1000$ nm [23] and combined with the measured upper critical field). In contrast with the in-plane measurements, the inter-plane anisotropy (Fig. 3(b)) is found to increase with decreasing temperature by a factor $\sim 6$ suggesting strongly incoherent transport between the planes. In-plane resistivity measurements (Fig. 3(e)) and penetration depth measurements [24] (TDI in Fig. 3(f)) in very high magnetic fields up to 55 T show no anomalies that could be associated with the magnetic transition (when $H \perp \text{ab}$), beside the expected transition from superconducting to the normal state. By suppressing the superconducting state by the application of a magnetic field of 55 T, the normal state resistance tends to increase exponentially at low temperatures suggesting field-induced charge localization in 10-3-8, as shown in the inset of Fig. 3(e).

Based on the transport and torque data we have constructed the phase diagram of the superconducting Ca$_{10}$(Pt$_3$As$_8$)((Fe$_{1-x}$Pt$_x$)$_2$As$_2$)$_2$, as shown in Fig. 4(a). Near $T_c$, the anisotropy parameter $\gamma_H = H_{c2}^{|| \text{ab}}(T)/H_{c2}^{\perp \text{ab}}(T)$ has a value of 10, similar to the 1111 iron pnictides [25] but significantly larger than the value of $\sim 2$ in the 122s [26]. When the magnetic field is parallel to the conducting plane, we use the single-band Werthamer-Helfand-Hohenberg (WHH) model [27] and fit the zero–resistance data points near $T_c$ to predict $H_{c2}(0) \approx 30$ T, which is close to the irreversibility field of 32 T measured in torque at 1.5 K. When the magnetic field is perpendicular to the Fe-As layers ($H \perp \text{ab}$), the superconducting phase boundary shows a strong concave curvature that cannot be captured by the one-gap WHH model and is suggestive of multi-band superconductivity in this system, as seen in the closely related 10-4-8 superconducting phase [23] or other anisotropic systems such as the 1111 pnictides [29] and cuprates.

While the superconducting phase diagram of the 10-3-8 has features found in other anisotropic superconductors, the magnetic region induced by the high magnetic fields at low temperatures is rather unusual. The phase diagram under pressure [11], which is similar overall to that as function of doping shown in Fig. 1(a), tracks the minimum in the resistivity at a temperature $T^*$ well above the structural transition, suggesting that this temperature could be linked to the gradual appearance of nematic fluctuations (and the corresponding loss of the carrier density). Due to the more strongly two-dimensional character of the 10-3-8 compared to other Fe-pnictides (for instance, the long interlayer stacking distance of 10.27 Å and the lower N´eel temperature in the parent compound suggest weaker interlayer coupling), nematic antiferromagnetic spin fluctuations in the Fe-As planes without 3D long-range order are likely to exist over a large area of the phase diagram, roughly characterized by the resistivity upturn at $T^*$ (Fig. 1). It is these antiferromagnetic fluctuations along the c-axis that could be responsible for the observed spin-flop transition in torque in an applied magnetic field.

In conclusion, we have used high magnetic fields to map out the superconducting phase diagram of Ca$_{10}$(Pt$_3$As$_8$)((Fe$_{0.922}$Pt$_{0.078}$)$_2$As$_2$)$_2$, finding a high anisotropy up to $\gamma_H = 10$, and an irreversibility field up to $\approx 32$ T for $H || \text{ab}$. Most importantly, we reveal a field-induced magnetic transition in torque measurements at 22 T at low temperature not previously observed in Fe-based superconductors, which we attribute to a spin-flop of antiferromagnetic fluctuations in the Fe-As plane. This thermodynamic measurement at low temperature demonstrates that the magnetic field is an effective tuning parameter of these antiferromagnetic fluctuations which may be responsible for the pairing in the 10-3-8 iron-based superconductors. These fluctuations are likely to be nematic fluctuations that reflect the breaking of the local tetragonal symmetry in the Fe-As plane in 10-3-8 over a large range of temperature and doping. Further work to quantify the nature of these magnetic fluctuations needs to be provided by other experimental techniques, such as polarized neutron scattering experiments.

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[1] M. D. Lumsden and A. D. Christianson, J. Phys. Cond. Mat. 22, 203203 (2010)

[2] S. Kasahara, H. J. Shi, K. Hashimoto, S. Tonegawa, Y. Mizukami, T. Shibue, K. Sugimoto, T. Fukuda, T. Terashima, A. H. Nevidomskyy, and Y. Matsuda, Nature 486, 382 (2012)

[3] T. Shimojima, T. Sonobe, and W. Malaeb, arXiv: 1305.3875 (2013)

[4] J.-H. Chu, H.-H. Kuo, J. G. Analytis, and I. R. Fisher, Science 337, 710 (2012)

[5] R. M. Fernandes, L. H. VanBebber, S. Bhattacharya, P. Chandra, V. Keppens, D. Mandrus, M. A. McGuire, B. C. Sales, A. S. Sefat, and J. Schmalian, Phys. Rev. Lett. 105, 157003 (2010)

[6] N. Ni, J. M. Allred, B. C. Chan, and R. J. Cava, PNAS 108, E1019 (2011)

[7] Z. J. Xiang, X. G. Luo, J. J. Ying, X. F. Wang, Y. J. Yan, A. F. Wang, P. Cheng, G. J. Ye, and X. H. Chen, arXiv:1203.1486v1 (2012)

[8] K. Cho, M. a. Tanatar, H. Kim, W. E. Straszheim, N. Ni, R. J. Cava, and R. Prozorov, Phys. Rev. B 85, 020504 (2012)

[9] T. Zhou, G. Koutroulakis, J. Lodico, N. Ni, J. D. Thompson, R. J. Cava, and S. E. Brown, J. Phys. Cond. Mat. 25, 122201 (2013)

[10] T. Stürzer, G. M. Friederichs, H. Luetkens, A. Amato, H.-H. Klauss, and D. Johrendt, J. Phys. Cond. Mat. 25, 122203 (2013)

[11] P. Gao, L. Sun, N. Ni, J. Guo, and Q. Wu, arXiv: 1301.2863 (2013)

[12] N. Ni, W. E. Straszheim, D. J. Williams, M. A. Tanatar, R. Prozorov, E. D. Bauer, F. Ronning, J. D. Thompson, and R. J. Cava, Phys. Rev. B 87, 060507 (2013)

[13] M. Neupane, C. Liu, S.-Y. Xu, Y.-J. Wang, N. Ni, J. M. Allred, L. A. Wray, N. Alidoust, H. Lin, R. S. Markiewicz, A. Bansil, R. J. Cava, and M. Z. Hasan, Phys. Rev. B 85, 094510 (2012)

[14] S. Thirupathaiah, T. Stürzer, V. B. Zabolotnyy, D. Johrendt, B. Büchner, and S. V. Bortsenko, arXiv 1307.1608v2 (2013)

[15] The lattice parameters are $a=8.8751(13)\AA$, $b=8.7852(12)\AA$, $c=10.6745(17)\AA$, $\alpha=94.66(1)^{\circ}$, $\beta=104.23(1)^{\circ}$, $\gamma=89.91(1)^{\circ}$, slightly different from those found for the parent compound reported in Ref. [3].

[16] WDX was performed on several areas of a mm-sized crystal from the same batch as the samples in which the anomaly was observed. Our value of $x = 0.078(6)$ is found by constraining $N(\text{Fe}^+\text{Pt}^2) = 13$. The point-to-point variation was on the order of the measurement uncertainty.

[17] A. I. Coldea, J. D. Fletcher, A. Carrington, J. G. Analytis, A. F. Bangura, J.-H. Chu, A. S. Erickson, I. R. Fisher, N. E. Hussey, and R. D. McDonald, Phys. Rev. Lett. 101, 216402 (2008)

[18] C. Putzke, A. I. Coldea, I. Guillamón, D. Vignolles, A. Mc-Collam, D. LeBoeuf, M. D. Watson, I. I. Mazin, S. Kasahara, T. Terashima, T. Shibue, Y. Matsuda, and A. Carrington, Phys. Rev. Lett. 108, 047002 (2012)

[19] The square-planar coordination of Pt in the Pt-As layer creates a strong crystal field, with a high antibonding $d_{x^2-y^2}$ band, and all other $d$-states occupied, so the valency of the interlayer Pt is $2^+$, as opposed to the Pt$^{4+}$ substituting for Fe. The arsenics in the $Pt_3As_2$ plane form covalently bound dimers with an effective valency of $4^+$ per dimer so that the ionic electron count is $Ca_{10}Pt_5^{2+}(As_2)_{4+}Fe_{10}As_{10}$.