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**LETTER TO THE EDITOR**

# Irreversible magnetization deep in the vortex-liquid state of a 2D superconductor at high magnetic fields

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**Abstract**
The remarkable phenomenon of weak magnetization hysteresis loops, observed recently deep in the vortex-liquid state of a nearly two-dimensional (2D) superconductor at low temperatures and high magnetic fields, is shown to reflect the existence of an unusual vortex-liquid state, consisting of collectively pinned crystallites of easily sliding vortex chains.

(Some figures in this article are in colour only in the electronic version)

Many potentially important superconductors, such as some of the high-$T_c$ cuprates as well as the ET-based organic conductors, are extremely type II superconductors with small in-plane coherence lengths and nearly 2D electronic structure. ET (or BEDT-TTF) stands for bisethylenedithio-tetrathiafulvalene. For these materials, drastic deviations from mean-field-theory predictions due to strong fluctuations in the superconducting (SC) order parameter are therefore expected, in particular under strong perpendicular magnetic field [1]. The great fundamental interest in the latter class of materials stems from their moderately low upper critical fields, which enable us to investigate the virtually unexplored phase diagram and vortex dynamics of strongly type II superconductors at low temperatures and high magnetic fields.

Of special interest is a recent striking observation in $\beta''$-$(ET)_2SF_5CH_2CF_2SO_3$ of small, but very clear magnetization hysteresis loops appearing well above the 'major' irreversibility field at low temperature, where significant de Haas–van Alphen (dHvA) oscillations are observable as well [2] (figure 1). It should be stressed that the occurrence of such high-field hysteresis tails is not peculiar to this particular material as it can be observed in e.g. $\kappa$-$(ET)_2Cu(NCS)_2$ as well [3].

The unusual feature of this irreversibility effect is associated with its appearance deep in the vortex-liquid phase, where one usually expects unrestricted motion of flux lines through the

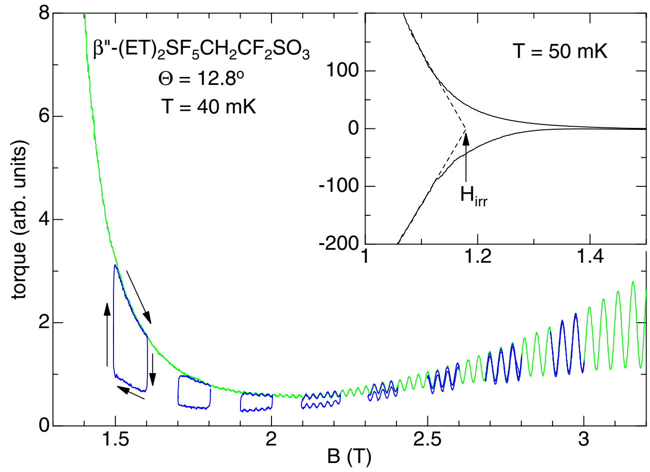


**Figure 1.** Torque signal with several small hysteresis loops. Arrows indicate the field-sweep directions. The determination of the major irreversibility field, $H_{irr}$, is shown in the inset.

entire SC sample, ensuring the establishment of thermodynamic equilibrium. In the present letter we argue that the observed hysteresis effect is a general feature which reflects the unusual nature (namely the nematic liquid-crystalline structure) of the low-temperature vortex-liquid state in 2D superconductors well above the vortex-lattice melting point, which was recently predicted theoretically [4].

According to this theory the vortex state above the major irreversibility field, $H_{irr}$ (found to be close to the vortex-lattice melting field, see below), is not an isotropic liquid but some mixed phase containing SC domains of easily sliding parallel vortex chains which are stabilized by a small number of strong pinning centres. This model resembles the sluggish vortex-fluid picture proposed by Worthington *et al* [5] to describe the intermediate vortex-liquid phase in defect-enhanced high-$T_c$ crystals and applied more recently by Sasaki *et al* [6] to account for a similar behaviour in the organic superconductor $\kappa$-$(ET)_2Cu(SCN)_2$. As shown below, a simple Bean-like model for the magnetic-induction profiles, associated with the vortex chains injected into the SC sample, yields very good quantitative agreement with the measured field dependence of the magnetization hysteresis.

Our model consists of independent 2D SC layers in the $x$–$y$ plane under the influence of an external uniform magnetic field $\vec{H} = H\hat{z}$, $H > 0$, perpendicular to the layers (figure 2). The SC sample is confined between two boundary planes at $x = 0$, and $L_x$. $H$ is varied, at low temperatures, from above $H_{c2}$ to the irreversibility field $H_{irr} < H_{c2}$, with clearly observable magnetic quantum oscillations. The dHvA effect is measured by means of the torque method, in which the signal is detected during steady (upward and downward) sweeps of the external magnetic field. Note that, for the sake of simplicity, the small in-plane component of the external magnetic field, required for this high-resolution measurement, is neglected in our analysis.

The influence of a steady increase (or decrease) of the magnetic field on the distribution of magnetic flux lines within the SC region is determined by the pinning forces acting on quantized SC vortex lines near the normal–superconducting (N–S) boundary planes. The pinning-force resistance against flux injection leads to the establishment of a flux-density gradient perpendicular to the magnetic-field direction along the normal of the N–S boundary plane (along $x$). Assuming the flux injection to be uniform along $y$, the current density $\vec{j} = j\hat{y}$

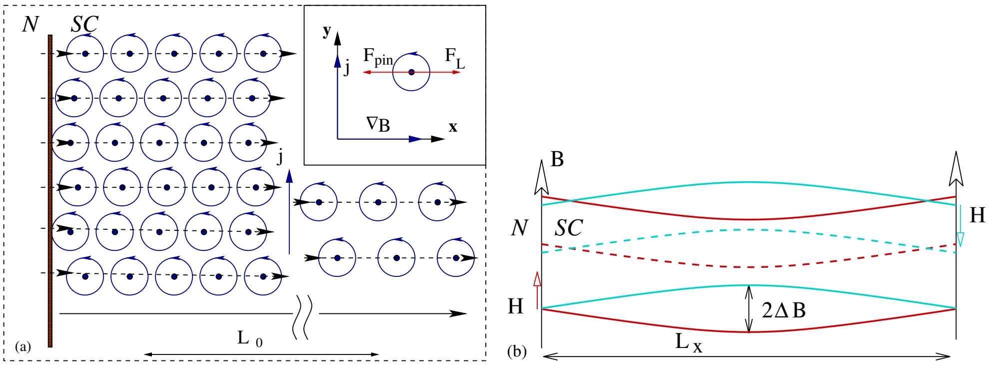


**Figure 2.** (a) Sketch of a vortex crystallite, oriented with its easily sliding Bragg chains perpendicular to the N–S boundary, under injection of flux lines. Note the scaling of the magnetic length over a macroscopic distance $L_0$. (b) Magnetization profiles during upward and downward field sweeps.

and the field gradient, $\frac{\partial B}{\partial x}$, are connected by Ampere's law:

$$\frac{\partial B}{\partial x} = -[\vec{\nabla} \times \vec{B}] \cdot \hat{y} = -\frac{4\pi}{c} j, \tag{1}$$

where the Lorentz-force density (per unit volume), exerted on the vortex current by the magnetic field, is $\vec{F}_{\mathrm{L}} = \frac{1}{c}[\vec{j} \times \vec{B}] = F_{\mathrm{L}}\hat{x}$.

We focus here on the field range $H > H_{\mathrm{irr}}$ where the reduced pinning force cannot balance the driving Lorentz force and the motion of vortices in the entire SC region becomes a continuous flow. This vortex movement with velocity $\vec{v}_\phi$ is opposed by a dynamic friction force, $\vec{F}_\eta = -\eta\vec{v}_\phi = F_\eta\hat{x}$, which balances the action of the Lorentz force at the critical velocity, $v_\phi^{\mathrm{c}} = j_{\mathrm{c}}B/c\eta$. In the corresponding steady state the motion of flux lines generates an electric field [7], $E = \frac{1}{c}Bv_\phi^{\mathrm{c}}$, parallel to the current density $\vec{j}$. Consequently, the system develops a finite electrical resistivity to the critical-current flow, $\rho(B) = E/j = B^2/c^2\eta$, so that the Bardeen–Stephen (BS) relation [8, 9], $\eta \approx BH_{\mathrm{c2}}/c^2\rho_{\mathrm{n}}$, with $\rho_{\mathrm{n}} = \rho(H_{\mathrm{c2}})$, together with the flux-line conservation law, lead to the nonlinear 'diffusion' equation for the magnetic-flux density:

$$\frac{\partial B}{\partial t} = \frac{D_\phi}{H_{\mathrm{c2}}}\left[B\left(\frac{\partial^2 B}{\partial x^2}\right) + \left(\frac{\partial B}{\partial x}\right)^2\right], \tag{2}$$

with the diffusion coefficient $D_\phi \equiv \frac{c^2\rho_{\mathrm{n}}}{4\pi}$. In the high-field limit of interest here, when the range $\Delta H$ of a single field-sweep cycle is much smaller than the initial field of the hysteresis loop, $H_0$, the nonuniform part of the magnetic induction associated with the flux flow is very small, i.e., $|b(x,t)| \equiv |B(x,t) - H(t)| \ll H(t) \simeq H_0$. Under this condition the second term on the RHS in equation (2) can be neglected, and the equation may be linearized to $\frac{\partial B}{\partial t} \approx \left(\frac{H_0}{H_{\mathrm{c2}}}\right)D_\phi\frac{\partial^2 B}{\partial x^2}$. The solution of this equation, satisfying steady upward and downward field-sweep boundary conditions, $\frac{\partial B}{\partial t}(x = 0, t) = \frac{\partial B}{\partial t}(x = L_x, t) = \frac{\Delta H}{\tau}$ for $0 \leqslant t \leqslant \tau$, and $-\frac{\Delta H}{\tau}$ for $\tau < t \leqslant 2\tau$, takes the forms $B_+(x,t) = H_0 + \frac{\Delta H t}{\tau} + \Delta B\left[\left(\frac{2x}{L_x} - 1\right)^2 - 1\right]$ and $B_-(x,t) = H_0 + \Delta H(2 - \frac{t}{\tau}) - \Delta B\left[\left(\frac{2x}{L_x} - 1\right)^2 - 1\right]$, respectively, with $\tau$ being half the period of the field-sweep cycle, and $\Delta B$ a constant.

Note the spatial rigidity of the induction profiles $B_\pm(x,t)$, which is assured, in the high-field limit $|b(x,t)| \ll H$, by the nearly instantaneous propagation of any local fluctuation of the magnetic-flux density. Indeed, the propagation velocity of such a fluctuation, $v_{\mathrm{f}} = \frac{\partial B}{\partial t}/\frac{\partial B}{\partial x}$, is found by equation (2) to be $v_{\mathrm{f}} = \frac{c^2\rho_{\mathrm{n}}}{4\pi H_{\mathrm{c}2}}\frac{\partial}{\partial x}\left(B\frac{\partial B}{\partial x}\right)/\frac{\partial B}{\partial x}$, so that the flux-line velocity $v_\phi$ satisfies $|v_\phi| \simeq \left|v_{\mathrm{f}}\right|\left(\frac{\partial B}{\partial x}\right)^2/B\left|\frac{\partial^2 B}{\partial x^2}\right| \ll |v_{\mathrm{f}}|$.

The resulting parabolic induction profile can be readily exploited to evaluate, for each small hysteresis loop, the jump, $\Delta M_{\uparrow\downarrow}$, of the magnetization occurring at a point where the field sweep is reversed (figure 2). The corresponding jump, $\Delta M_{\uparrow\downarrow} = \frac{1}{3\pi}\Delta B$, is determined by the spatial average of the induction difference, $[B_-(x,\tau) - B_+(x,\tau)]$.

In what follows we present a simple model of the flux flow, which is based on the main features of the vortex-liquid state in a 2D superconductor at high magnetic fields and which can account for the observed experimental data. In the field range of interest here the vortex system is in a liquid-crystal-like state, with long-range orientational order [4], which tends to respond to flux injection by a smectic flow of vortices. We thus imagine an ensemble of vortex crystallites, having their principal lattice vector aligned perpendicular to the N–S boundary, i.e., along $x$, where a weak driving force can inject vortex chains into the SC region. We will assume that these crystallites are stabilized by a dilute network of very strong pinning centres. The building block of this system is a cluster of vortex chains, pinned to the underlying metallic lattice through its two boundary chains. Thus, all the vortex chains within the cluster are assumed to move freely, subject only to the intrinsic vortex–vortex interactions, while the two edge chains, which are considered fixed in the laboratory frame. In this model the frictional force, which opposes the action of the driving Lorentz force, arises solely from the vortex–vortex interactions activated in shear distortions along the principal chain axis.

A microscopic description of the critical state established in the smectic flow outlined above will enable us now to estimate the characteristic energy scale corresponding to the friction of moving vortices. Comparison of this estimate with the experimental data will provide support for the proposed picture. Suppose that the macroscopic driving Lorentz force is generated by a continuous injection of vortex chains along the easy crystallographic axis into the SC sample. In the critical state this injection supplies to a vortex chain just the minimal kinetic energy $m_{\mathrm{v}}v_{\phi 0}^2/2$ required to overcome the energy barrier (between adjacent minima and maxima) of the vortex–vortex potential. Here $m_{\mathrm{v}}$ is the dynamic mass per vortex and $v_{\phi 0}$ its initial velocity at the bottom of a potential well. Note that all extensive chain parameters are expressed here per single vortex. The energy barrier is of the order of the phase-dependent minimal coupling, $\varepsilon_{\mathrm{ph}} \approx 4\lambda^2\varepsilon_0$, which is of the order of the characteristic vortex-lattice melting energy, with $\varepsilon_0$ being the SC condensation energy per vortex and $\lambda \approx 0.066$ [4]. Suppose now that the entire energy supplied to the vortex chain in a jump over a single barrier is dissipated through a frictional force with the other chains. We shall now show that this leads to a friction coefficient $\eta$ identical to that derived from Caldeira–Leggett theory [10] for moving vortex chains.

Since the average velocity, $v_{\phi 0}/2 \approx \sqrt{\varepsilon_{\mathrm{ph}}/m_{\mathrm{v}}}$, of such a vortex chain can be identified in the considered model with the critical-state velocity $v_\phi^{\mathrm{c}}$, the energy dissipated in a single jump is $\varepsilon_{\mathrm{dis}} \sim \eta v_\phi^{\mathrm{c}}\Delta x$, where $\Delta x \sim a_H$ is the distance between neighbouring vortex sites along the chain. Using the above expression for $v_\phi^{\mathrm{c}}$ together with the condition $\varepsilon_{\mathrm{dis}} = \varepsilon_{\mathrm{ph}}$, we find that $\eta \approx \sqrt{m_{\mathrm{v}}\varepsilon_{\mathrm{ph}}}/a_H$, which is identical to the result obtained [11] from the Caldeira–Leggett theory [10]. In the above expressions $a_H = \sqrt{c\hbar/eH}$ is the magnetic length.

Let us now relate the dissipation energy $\varepsilon_{\mathrm{dis}}$ to $\Delta M_{\uparrow\downarrow}$. In the critical-state model described above the work (per unit volume) done by the Lorentz force in moving vortex chains within the entire cluster over a distance $\Delta x$ between two neighbouring sites (i.e. $\Delta x \sim a_H$) is $W_{\mathrm{L}} = \int_0^{L_x}\frac{1}{c}\left|j_{\mathrm{c}}(x)\,B(x)\right|\,\mathrm{d}x \approx \left(\frac{H_0}{4\pi}\right) \times 2\int_0^{L_x/2}\left|\frac{\partial B}{\partial x}\right|\,\mathrm{d}x = \left(\frac{3}{2}\right)H_0\Delta M_{\uparrow\downarrow}$. This may be rewritten in units of the maximal SC condensation-energy density, $E_{\mathrm{cond}} = \frac{H_{\mathrm{c}2}^2}{16\pi\kappa^2}$, so that

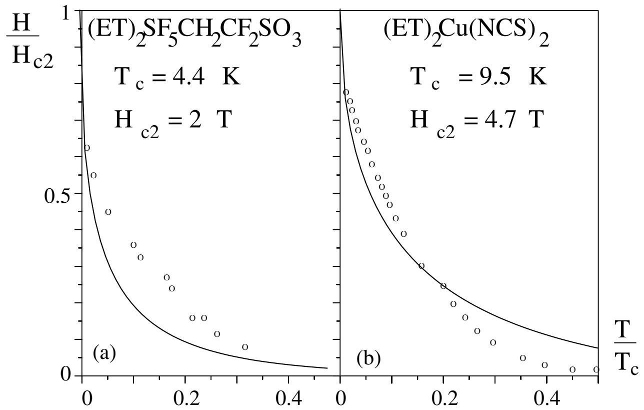


**Figure 3.** Experimental major irreversibility fields (open circles) and theoretical melting fields (solid curves) as functions of temperature for two organic superconductors. Data for $\kappa$-$(ET)_2Cu(NCS)_2$ from [3].

for $B \approx H_{c2}$, $\tilde{\varepsilon}_L \equiv W_L/E_{cond} \approx 24\kappa^2\Delta M_{\uparrow\downarrow}/H_{c2}$. This dimensionless energy scale should be compared to the dissipation energy per vortex, estimated above from the condition $\varepsilon_{dis} = \varepsilon_{ph} \approx 4\lambda^2\varepsilon_0$, after normalizing with respect to the SC condensation energy per vortex, $\varepsilon_0$.

Using the experimental value of the magnetization jump at $B = 2$ T, that is $\Delta M_{\uparrow\downarrow} \approx 4 \times 10^{-8}$ T, we find that $\tilde{\varepsilon}_L \approx 4 \times 10^{-3}$, where $\kappa \approx 46$ is used [12][4]. This result is of the same order of magnitude as $\tilde{\varepsilon}_{dis} = \varepsilon_{ph}/\varepsilon_0 \approx 4\lambda^2 \sim 10^{-2}$, which is the characteristic energy scale associated with the melting of the vortex lattice in 2D superconductors. This agreement lends support to the proposed model that most of the vortex crystallites are aligned with their principal lattice vector along the direction of the flux-line motion [13], so that the flux flow is dominated by easily sliding vortex chains, which are subject only to the residual shear resistance characterizing the vortex-liquid state above the melting transition [1]. This picture is very similar to the moving smectic state found recently in many numerical simulations of 2D vortex lattices in the presence of random pinning centres [14].

The similar values of $\tilde{\varepsilon}_{ph}$ and $\tilde{\varepsilon}_L$, found above, may also reflect the correlation between the onset of irreversibility and the vortex-liquid freezing transition. This may be verified by calculating the temperature-dependent melting field, $H_m(T)$, and comparing it to the irreversible field, $H_{irr}(T)$, extracted from the experimental data. Similar to [3], $H_{irr}(T)$ is determined here by the onset of the major hysteresis in the magnetization curve (see the inset in figure 1). Within the Ginzburg–Landau (GL) approach, $H_m(T)$ has been derived by several authors [4, 15]. It can be obtained from $\xi^2(H_m, T) = g_m^2$, where $\xi^2(H, T) \equiv \varepsilon_0/k_BT$, with $\varepsilon_0 = \alpha^2/2\beta$, and $\alpha$ and $\beta$ are the GL parameters. Here we estimate $|\Delta_0|^2 = \frac{\alpha}{\beta} = (1.76k_BT_c)^2 \ln\left(\frac{H_{c2}}{B}\right)$, and $\beta = \frac{1.38}{E_F(\hbar\omega_c)^2}$, where $\omega_c = eB/m_cc$ is the cyclotron frequency (see [1]). Using our estimate, $g_m^2 \approx 1/4\lambda^2$, and neglecting the weak temperature dependence of $H_{c2}$ at low $T$, we obtain the simple equation for $h_m(t) = H_m(T)/H_{c2}(0)$: $\ln(h_m) = -k_0\sqrt{t}h_m$, where $t = T/T_c(0)$ and $k_0$ is a single dimensionless parameter depending on the properties of the superconductor through $k_0^2 \simeq 0.15g_m^2E_F(\hbar\omega_{c2})^2/(k_BT_c)^3$. The function $h_m(t)$ for $\beta''$-$(ET)_2SF_5CH_2CF_2SO_3$, with $T_c(0) = 4.4$ K and $H_{c2} = 2$ T is shown in figure 3(a). The experimental irreversibility line agrees rather well with the calculated melting curve. A similar

[4] The value $\kappa \approx 46$ is recalculated using the reduced value of $H_{c2}$ found in the present paper.

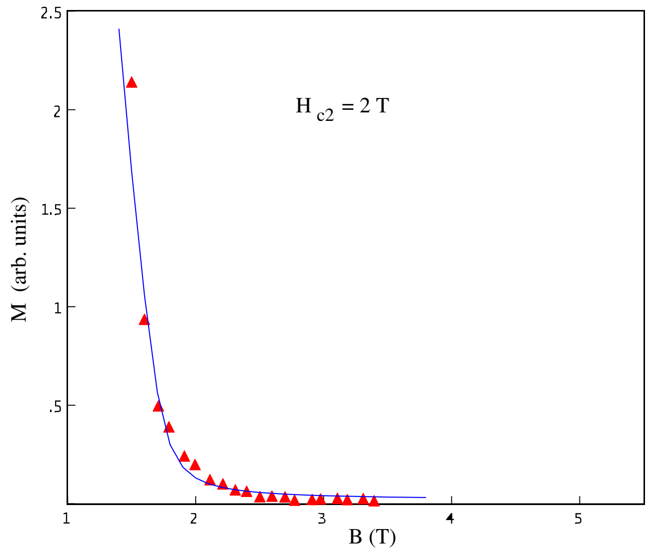


**Figure 4.** Field dependences of the experimental magnetization jumps (triangles) and the fit (solid curve) according to equation (3).

procedure is applied to $\kappa$-(ET)$_2$Cu(NCS)$_2$. The parameter $H_{c2} \approx 4.7$ T was determined by fitting the additional damping amplitude of the dHvA oscillation in the mixed SC state, as calculated from our SC fluctuation theory [1], to the corresponding experimental data [3]. Again, the calculated melting curve is found to be close to the irreversible line obtained in [3] (see figure 3(b)). It is interesting to note that at $T = 0$ the resulting melting field coincides with the mean field $H_{c2}$. The sharp increase toward $H_{c2}$, characterizing $H_m$ at $T \to 0$, is similar to that of $H_{irr}$ observed experimentally in both materials.

The proximity of the melting and the irreversibility lines, $H_{irr} \approx H_m$, justifies the use of our fluctuating-vortex-chain model above $H_m$ [1] in attempting to account for the magnetic-field dependence of the jump $\Delta M_{\uparrow\downarrow}$ above $H_{irr}$, extracted from the experimental result shown in figure 1. Since the generators of the flux-density gradient, i.e., the pinning force and the vortex–vortex interaction, all originate in the existence of local SC order at the vortex position, it should be proportional to the mean-square SC order parameter, $\langle|\Delta_0(H)|^2\rangle$ [16], and so the magnetization jump can be written as $\Delta M_{\uparrow\downarrow} = \frac{1}{3\pi}\Delta B \approx \frac{L_x}{3\pi}\overline{\left|\frac{\partial B}{\partial x}\right|} \propto \frac{1}{B}\langle|\Delta_0(B)|^2\rangle$, where the bar means spatial averaging. Using the limit of independently fluctuating vortex chains in the GL theory [1] to describe the vortex-liquid state above $H_{irr}$, i.e., writing $\langle|\Delta_0(B)|^2\rangle = \sqrt{\frac{2k_B T}{\pi^2\beta}}\Phi(\xi)$, where $\Phi(\xi) = \sqrt{\pi}\left[\xi + \frac{\exp(-\xi^2)}{2\int_{-\infty}^{\xi} d\varsigma\, \exp(-\varsigma^2)}\right]$, we find that

$$\Delta M_{\uparrow\downarrow} \propto \frac{1}{B}\langle|\Delta_0(B)|^2\rangle \propto \Phi(\xi), \qquad (3)$$

which is a universal function of the dimensionless parameter $\xi(B, T) = \sqrt{\varepsilon_0/k_B T}$. Employing the material parameters, $T_c = 4.4$ K, $\frac{m_c}{m_e} = 2.0$, where $m_e$, $m_c$ are the free electron and cyclotron mass respectively, and $E_F/k_B = 133$ K, at the temperature of the experiment, $T = 40$ mK, so that $\xi \approx (31[T]/B[T])\,(1 - B/H_{c2})$, and treating $H_{c2}$ and the proportionality factor in equation (3) as adjustable parameters, the best fit is obtained for $H_{c2} = 2$ T (figure 4). The resulting curve reflects the crossover between the vortex state below $H_{c2}$, which is well described by mean-field theory, and the normal state far above $H_{c2}$, with the long tail of the field-dependent magnetization hysteresis corresponding to the enhanced influence of 2D SC fluctuations.

In conclusion, we have shown that the striking phenomenon of the weak magnetization hysteresis loops, observed deep in the vortex-liquid state of a nearly 2D superconductor, can be reasonably explained as arising from shear viscous flow of easily sliding vortex chains, which are clustered around a small number of strong pinning centres.

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