Supplementary Materials for

Voltage-induced ferromagnetism in a diamagnet

Jeff Walter, Bryan Voigt, Ezra Day-Roberts, Kei Heltemes, Rafael M. Fernandes, Turan Birol, Chris Leighton*

*Corresponding author. Email: leighton@umn.edu

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Section A: Structural and Chemical Characterization of FeS$_2$ Single Crystals

For reference, we provide below a summary of structural and chemical characterization data on representative FeS$_2$ crystals. These data demonstrate: (a,b) single-phase X-ray diffraction patterns on powdered crystals, with lattice parameters in accord with prior work; (c) single crystallinity by high-resolution X-ray diffraction; (d) narrow X-ray diffraction rocking curve widths; (e,f) S/Fe ratios from energy dispersive spectroscopy that are consistent with close-to-stoichiometric crystals. With regard to the latter, and as discussed in the main text, prior work has demonstrated fine-tuning of the sulfur vacancy concentration via growth conditions, as detected by electronic transport measurements. The table provided below also provides details on measured impurity concentrations from inductively coupled plasma mass spectrometry.

![Fig. S1. Structural and chemical characterization of FeS$_2$ single crystals.](image)

(a) Wide-angle powder X-ray diffraction (PXRD) from a ground FeS$_2$ single crystal, and (b) a reference pyrite PXRD pattern, resulting in the extracted lattice parameter ($a = 5.417 \pm 0.001$ Å) shown. (c) High-resolution wide-angle XRD from a pristine (111) facet of an FeS$_2$ single crystal, and (d) a rocking curve through the 111 peak of the same crystal, with a full-width-at-half-maximum (FWHM) of 0.009°. (e) Energy-dispersive spectroscopy intensity as a function of energy ($E$), and (f) the reference Fe and S energies. The extracted S:Fe ratio of 2.01 ± 0.09 is shown. All data shown here are from crystals nominally identical (i.e., grown in the same vessel) to Sample 1 from the main text. All results are consistent with prior reports [42,43,47].
Table S1. Quantification of impurity concentrations in FeS$_2$ single crystals. Impurity concentrations of 3$d$ transition metals determined by inductively coupled plasma mass spectrometry in an FeS$_2$ single crystal (reported in [43]) nominally identical to Sample 1 from the main text. Sc and Zn were not quantified due to inconsistent backgrounds/standards. The final column gives the total metals-basis impurity (MBI) concentration, which includes the 3$d$ transition metals listed as well as V, Ga, Rb, Sr, Ag, Sb, Cs, Ba, Pb, Th, U, Mo, and Ge. Of these additional elements, only Mo (1.9 ppm) and Ge (18 ppm) had impurity concentrations above 1 ppm.

|     | Sc  | Ti  | V   | Cr  | Mn  | Co  | Ni  | Cu  | Zn  | Total MBI |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----------|
| ppm | ppm | ppm | ppm | ppm | ppm | ppm | ppm | ppm | ppm | ppm       |
|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----------|
| -   | 0.9 ± 0.1 | < 0.1 | 4.6 ± 0.1 | 1.5 ± 0.1 | 0.8 ± 0.1 | 4.3 ± 0.5 | 2.3 ± 0.1 | -   | 33.1       |
Section B: Surface Conduction and Majority Carrier Type in FeS$_2$ Single Crystals

As noted in the main text, surface conduction is now well established in FeS$_2$ single crystals, leading to the specific form of the temperature ($T$) dependence of the sheet resistance ($R_s$) shown in Figs. 1(b,c). In essence the FeS$_2$ interior exhibits typical carrier freeze-out on cooling, leading to an insulating form of $R_s(T)$. At some point the conductive surface layer then shunts the majority of the current at sufficiently low $T$, leading to the flattening seen in Fig. 1(b) around 150 K, for example. As shown in prior work [42], the FeS$_2$ single crystal interior is $n$-type, due to light doping from S vacancies [43], while the conductive surface layer is heavily $p$-doped [42]. This is thought to occur due to surface states, leading to substantial band bending and a surface Fermi level pinned near the valence band maximum.

Complementary to Fig. 1(b), Fig. S2 below shows Hall measurements as a function of $T$ for a sample that is nominally identical to Sample 1. This sample was grown under the same conditions as Sample 1, and exhibits similar $R_s(T)$. The transverse (Hall) resistance ($R_{xy}$) is shown vs. the perpendicular magnetic field ($H$) at (a) 300 K, (b) 120 K, and (c) 60 K. At 300 K a linear Hall effect with negative Hall coefficient is observed, consistent with the dominance of the $n$-type interior at this $T$. The extracted Hall electron density is $1.2 \times 10^{16}$ cm$^{-3}$, quite typical of such crystals [42]. At 120 K, on the other hand, which is just below the temperature at which an inflection point is observed in Fig. 1(b), distinctly different, non-linear Hall effect emerges. A negative slope at low field is seen to give way to a positive slope at high field, behavior that was previously quantitatively captured by a two-channel conduction model describing an $n$-type interior and a $p$-type surface [42]. The heavily doped $p$-type surface layer was deduced to have a thickness of less than ~1-3 nm in that work [42]. Further cooling to 60 K (Fig. S2(c)) then leads to strong suppression of the Hall effect, resulting, in this particular crystal, in indeterminate majority carrier type. Again consistent with prior work [42], this is a consequence of variable-range hopping conduction in the surface state (see discussion of Fig. 2 in the main text). It is important to note that the nature of the conduction in FeS$_2$ surface states is variable, depending on the exact crystal surface, and in some cases a clear $p$-type Hall effect is observed at low $T$, confirming the typically $p$-doped nature of the surface layer on FeS$_2$ single crystals [42].
Fig. S2. Hall effect measurements elucidating surface conduction and the majority carrier type in FeS$_2$ single crystals. Perpendicular magnetic field ($H$) dependence of the transverse (Hall) resistance ($R_{xy}$) at (a) 300 K, (b) 120 K, and (c) 60 K for an FeS$_2$ single crystal with no ionic liquid, and thus no gate voltage ($V_g$), applied. The crystal used for these measurements was nominally identical to “Sample 1” in the main text, being grown under the same conditions and displaying similar temperature dependence of the sheet resistance.
Section C: Calculation of Accumulation Layer Thickness

In order to calculate the estimated thickness of the accumulation layer in these FeS$_2$ single crystal EDLTs, Thomas-Fermi screening calculations were performed following ref. [16]. Here, the depth ($z$) profile of the gate-induced 3D electron density, $\Delta n(z) = n(z) - n_0$, where $n_0$ is the initial bulk electron density, is related to the electrostatic potential ($\phi$) through the Poisson equation

$$\frac{d^2\phi(z)}{dz^2} = \frac{-4\pi \Delta n(z)}{k},$$

S(1)

where $k$ is the dielectric constant. The 3D electron density, $n(z)$, can be related to $\phi$ using the Thomas-Fermi approach

$$-e\phi(z) = (3\pi^2)^{2/3} e^2 b [n(z)]^{2/3},$$

S(2)

where $b = \hbar^2 (m^* e^2)^{-1}$ and $m^*$ is the electron effective mass. Equations S(1) and S(2) can then be combined, giving

$$\frac{d^2}{dz^2} \left( \frac{\phi}{e/b} \right) = \frac{-2^{7/2}}{3\pi k b^2} \left[ \left( \frac{\phi}{e/b} \right)^{3/2} - \left( \frac{\phi_0}{e/b} \right)^{3/2} \right],$$

S(3)

where the zero bias electrostatic potential is given by $\phi_0 = (3\pi^2)^{2/3} e^2 b n_0^{2/3}$. Using the assumption that the electrostatic potential deep in the FeS$_2$ crystal ($i.e., z \rightarrow \infty$) remains unchanged by the bias application, and using the electric neutrality condition, we then arrive at the boundary conditions

$$\phi(z = \infty) = \phi_0$$

S(4), and,

$$\frac{d\phi(z)}{dz} \bigg|_{z=0} = -\frac{4\pi}{k} \int_0^\infty \Delta n(z) dz = -e\Delta n_{2D},$$

S(5)

where $\Delta n_{2D}$ is the gate-induced surface ($i.e.,$ 2D) charge density. Equation S(3) can then be numerically solved with boundary conditions S(4) and S(5) in order to calculate $n(z)$ depth profiles. To do this we use the following values for FeS$_2$: $k = 10.9$, $n_0 = 10^{15}$ cm$^{-3}$ (typical for these crystals [42,43]) and $m^* = 0.45 m_e$, where $m_e$ is the electron mass. Note here that $z = 0$ corresponds to the ionic liquid/FeS$_2$ interface.

The resulting $n(z)$ and $\phi(z)$ based on this Thomas-Fermi approach are shown in Figs. S3(a,b) below, for various $\Delta n_{2D}$ between $10^{12}$ and $10^{15}$ cm$^{-2}$. In all cases $n(z)$ and $\phi(z)$ exhibit the expected rapid fall-off with increasing $z$, with the $z = 0$ values increasing monotonically with the input value of $\Delta n_{2D}$. For the purposes of simple quantification we define an accumulation layer thickness, $d_\Lambda$, as the thickness of the region in which 90% of the induced electrons reside for each $n(z)$. The resulting $d_\Lambda$ is then plotted vs. $\Delta n_{2D}$ in Fig. S3(d), where it is seen to decrease with increasing $\Delta n_{2D}$ due to the increased screening at higher charge densities. As noted in the main text, the dominance of the anomalous Hall effect over the ordinary Hall effect at high $V_g$ means that it is not possible to extract $\Delta n_{2D}$ from our experimental data. Instead, we note that previous work on bulk Fe$_{1-x}$Co$_x$S$_2$ single crystals found that a ferromagnetic state emerges above $n \approx 1 \times 10^{20}$ cm$^{-3}$ [35]. To compare this to these simulations, the maximum value of $n$ obtained at $z = 0$ ($n_{\text{max}}$) for each of the $n(z,\Delta n_{2D})$ curves in Fig. S3(a) is shown in Fig. S3(c), which also plots the average value of $n(z,\Delta n_{2D})$ within the accumulation thickness $d_\Lambda$. From Fig. S3(c) it can then be seen that the critical threshold for the
onset of ferromagnetism of $1 \times 10^{20}$ cm$^{-3}$ is crossed at $\Delta n_{2D} \approx 1 \times 10^{13}$ and $3 \times 10^{13}$ cm$^{-2}$ for $n_{\text{max}}$ and $n_{\text{ave}}$, respectively. Significantly, and as discussed in the main text, these $\Delta n_{2D}$ values are firmly in the achievable regime of EDLTs, supporting the feasibility of achieving a $V_g$-induced ferromagnetic metallic state in this work. Such calculations also allow us to estimate an upper limit of $d_A \approx 2$ - 2.5 nm for the thickness of the induced ferromagnetic metallic layer from Fig. S3(d); this is the origin of the value of ~2 nm (~4 unit cells) quoted in the main text.

**Fig. S3.** Thomas-Fermi calculations of the accumulation layer thickness in single crystal FeS$_2$ electric double layer transistors. Depth ($z$) dependence of (a) the induced 3D electron density ($n$) and (b) the electrostatic potential ($\phi$), as determined from the Thomas-Fermi method described above. Data are shown for multiple input values of the gate-induced surface electron density ($\Delta n_{2D}$), as indicated in the legend in (b). Note that $z = 0$ is the ionic liquid/FeS$_2$ interface, and that the inset in (a) simply zooms out to show the full range of $n$ for the highest $\Delta n_{2D}$. (c) Extracted $\Delta n_{2D}$ dependence of the maximum 3D carrier density at $z = 0$ ($n_{\text{max}}$) and the average carrier density within the region containing 90% of the induced electrons ($n_{\text{ave}}$). (d) Extracted $\Delta n_{2D}$ dependence of the thickness of the region containing 90% of the induced electrons ($d_A$).
Section D: Sweep-Rate-Dependent $R(V_g)$

As noted in connection with Fig. 6(a) in the main text, the significant hysteresis in $R(V_g)$ at high $T$ implies a $V_g$ sweep-rate dependence. This is confirmed in Fig. S4 below, which shows 240 K $R(V_g)$ data acquired at both 10 mV s$^{-1}$ and 100 mV s$^{-1}$ sweep rates. These data were taken on Sample 4, which is nominally identical to Sample 2 in the main text. As expected, the $R(V_g)$ observed at 10 mV s$^{-1}$ (black line) is rather similar to that seen in Sample 2 in Fig. 6(a), with a pronounced peak occurring at $V_g \approx +1$ V on the forward sweep, followed by a rise around $V_g \approx 0$ on the reverse sweep. We note a “double peak” structure in this particular sample, however, likely related to the conduction inhomogeneity discussed in Section E below. Most importantly, when the sweep rate is increased to 100 mV s$^{-1}$ the overall gate effect (i.e., the magnitude of the resistance changes) is decreased and the peak in $R$ at positive $V_g$ is pushed to higher $V_g$ (~3.5 V). As discussed in the main text, such sweep rate dependence is qualitatively consistent with the hysteresis in Fig. 6(a), and the resulting bistability of $R$ at $V_g = 0$. We ascribe this to sluggish dynamics in the ionic liquid at this temperature in such a macroscopic device.

![Graph](image)

**Fig. S4.** Sweep-rate-dependence of resistance vs. gate voltage sweeps in single crystal FeS$_2$ electric double layer transistors. 240 K gate voltage ($V_g$) dependence of the sample resistance ($R$) (in one van der Pauw orientation) taken at sweep rates of 10 and 100 mV s$^{-1}$ in Sample 4 (nominally identical to Sample 2 in the main text). The ionic liquid used was DEME/TFSI, as in Fig. 6(a).
Section E: Resistance Anisotropy Across Gate Voltage Sweeps

As noted in the main text in connection with Fig. 6, the compensation point resulting in the resistance peak at positive voltages in $R(V_g)$ sweeps is associated with some in-plane transport anisotropy in van der Pauw measurements. This is highlighted in Fig. S5 which plots the $V_g$ dependence of the van der Pauw resistance anisotropy, $A_{\text{vdP}} = R_1/R_2$, where $R_1$ and $R_2$ are the resistances measured in orthogonal orientations. These data are at 180 K, corresponding to Figs. 6(c-j), i.e., they are for Sample 2. The obvious feature in Fig. S5 is that $A_{\text{vdP}}$ is rather constant at a modest value of ~1.5 across the majority of the $V_g$ sweep, except in the vicinity of the compensation point peak at ~1 V. At this voltage $A_{\text{vdP}}$ rises to ~5.5, indicating substantial in-plane resistance anisotropy at the compensation point. As discussed in the main text, we ascribe this to some level of lateral inhomogeneity in the gating process, as might be expected to emerge at a sensitive compensation point.

**Fig. S5.** Resistance anisotropy across gate voltage sweeps in single crystal FeS$_2$ electric double layer transistors. Gate voltage ($V_g$) dependence of the van der Pauw resistance anisotropy ($A_{\text{vdP}}$) for Sample 2. Data were acquired at 180 K, immediately after the Hall sweeps shown in Fig. 6(c-j).