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Maxime Leroux, Vivek Mishra, Christine Opagiste, Pierre Rodière, Asghar Kayani, et al.. Charge density wave and superconductivity competition in Lu 5 Ir 4 Si 10: A proton irradiation study. Physical Review B, 2020, 102 (9), pp.094519. 10.1103/PhysRevB.102.094519 . hal-02952064

HAL Id: hal-02952064

<https://hal.science/hal-02952064>

Submitted on 12 Nov 2020

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Charge density wave and superconductivity competition in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$: A proton irradiation studyMaxime Leroux ^{1,*}, Vivek Mishra,^{1,‡} Christine Opagiste ², Pierre Rodière,² Asghar Kayani,³
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(Received 11 June 2020; revised 10 September 2020; accepted 10 September 2020; published 28 September 2020)

Real-space modulated charge density waves (CDW) are a ubiquitous feature in many families of superconductors. In particular, how CDW relates to superconductivity is an active and open question that has recently gathered much interest since CDWs have been discovered in many cuprates superconductors. Here we show that disorder induced by proton irradiation is a full-fledged tuning parameter that can bring essential information to answer this question as it affects CDW and superconductivity with different and unequivocal mechanisms. Specifically, in the model CDW superconductor $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ that develops a 1D CDW below 77 K and *s*-wave superconductivity below 4 K, we show that disorder enhances the superconducting critical temperature T_c and H_{c2} while it suppresses the CDW. Discussing how disorder affects both superconductivity and the CDW, we make a compelling case that superconductivity and CDW are competing for electronic density of states at the Fermi level in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, and we reconcile the results obtained via the more common tuning parameters of pressure and doping. Owing to its prototypical, 1D, Peierls type CDW and the *s*-wave, weak-coupling nature of its superconductivity, this irradiation study of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ provides the basis to understand and extend such studies to the more complex cases of density waves and superconductivity coexistence in heavy fermions, Fe-based, or cuprates superconductors.

DOI: [10.1103/PhysRevB.102.094519](https://doi.org/10.1103/PhysRevB.102.094519)**I. INTRODUCTION**

A charge density wave (CDW) is a spatial modulation of the electronic density of states which opens a gap at the Fermi level. CDW can arise from electronic instabilities such as Fermi surface nesting in low-dimension metals [1,2] or a peak in electron-phonon coupling [3]. This charge modulation is usually accompanied by a periodic lattice distortion, via the electron-phonon coupling. An analog modulation known as a spin density wave (SDW) also exists for the electronic spin density [1,2]. The presence of real-space modulated CDW or SDW is a feature of many families of superconductors [2,4].

Recently, CDWs have been found to be ubiquitous in many cuprates superconductors, whether in hole-doped $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ [5], $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ [6–12], $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ [13], $\text{HgBa}_2\text{CuO}_{4+\delta}$ [14], or in electron doped $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ [15]. Other examples include: Fe-based superconductors in which superconductivity seems to compete with spin density waves [16,17], heavy fermion compounds where the SDW appears linked to *d*-wave superconductivity [18], transition metal

dichalcogenides where the CDW is well known to compete with superconductivity in 2H-TaS₂ and 2H-TaSe₂ [19] but 1T-TiSe₂ has been proposed as an excitonic superconductor enhanced by the CDW [20], and finally organic superconductors also exhibit superconductivity in coexistence with density waves [e.g. (TMTSF)₂PF₆] [21]. Thus, whether CDW compete with [9,10,22] or on the contrary are a key ingredient in explaining the origin of cuprates' high temperature superconductivity [23,24], the relation between density waves and superconductivity is an active and open question [25].

Among superconductors with CDW, $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ is a well established case of *s*-wave superconductivity coexisting with a standard Peierls-type CDW [26–29]. This compound possesses a first order CDW transition below $T_{\text{CDW}} = 77$ K [30,31] and it also becomes superconducting below $T_c = 4$ K [32]. The CDW develops on 1D Lutetium atom chains along the *c* axis, following the nesting mechanism [29], with clear signatures in electrical transport [26,28], x-ray [33], specific heat [26,33,34], or elastic constants [30,31]. The CDW is commensurate with a periodicity of seven unit cells as evidenced by x-ray diffraction [33]. The CDW gaps an estimated 36% of the density of states at the Fermi level as determined from resistivity and specific heat measurements [26], with more recent optical estimates ranging from 16% [35] to 30% [29]. The effect of pressure points to a competition scenario: From 0 to 2 GPa T_{CDW} decreases continuously and T_c is constant, but above 2 GPa the CDW suddenly vanishes and T_c jumps from 4 to 9 K [26]. Chemical doping also points

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to a competition scenario: The CDW state is suppressed and T_c continuously increases up to at least 6 K for increasing doping [27,32,36]. We note that the increase of T_c is binary in the former case but progressive in the latter. To explain this differing behavior of T_c between pressure and doping, it has been proposed that $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ presents a sharp feature in the electronic density of states just above the Fermi level [37].

In this paper, we establish disorder induced by irradiation as a full-fledged axis in the phase diagrams of superconductors, via an extensive study of this model compound $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$. In particular, we evidence the mechanism through which proton irradiation acts as a tuning parameter suppressing the CDW in favor of superconductivity and we show how this tuning parameter brings its own set of unique information on superconductivity-CDW competition. In sharp contrast with the expected effect of disorder on superconductivity, we observe an *increase* of T_c after irradiation. Proton irradiation produces cascade-type clusters of defects, which are typically a few nanometer in diameter, along with a small fraction of point defects. This irradiation induced disorder strongly suppresses the CDW and broadens its transition, thus revealing the precise mechanism of CDW suppression. Moreover the increase of $H_{c,2}$ with disorder reveals that the channel for competition between CDW and superconductivity is the electronic density of states at the Fermi level. These results make a compelling case that reconciles how CDW and superconductivity competes in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ with pressure, doping, and disorder. This extensive set of results in a BCS s-wave compound with a prototypical 1D CDW of the Peierls type provides the basis to pursue such irradiation studies in the more complex cases of density wave coexistence in heavy fermions, Fe-based, or cuprates superconductors [22].

The paper is organized as follows: Section II presents the materials and methods. Section III presents how irradiation induced disorder raises T_c and reduces T_{CDW} . Section IV presents the evolution of $H_{c,2}$ as a function of disorder. Section V reconciles the different evolutions of T_c with pressure, doping, and disorder by discussing how they relate to the CDW suppression mechanisms.

II. MATERIALS AND METHODS

$\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ has a tetragonal unit cell with lattice parameters $a = 12.484(1)$ and $c = 4.190(2)$ Å [38] and space group symmetry $P4/mbm$. The CDW forms along the c axis on quasi-1D chains of lutetium atoms. The samples are high quality single crystals that grow in needle shape along the c axis and have been characterized previously [32,38]. We used the tandem van de Graaf accelerator at Western Michigan University to irradiate a sample several times with 4 MeV protons. This sample has dimensions $10\ \mu\text{m} \times 65\ \mu\text{m} \times 500\ \mu\text{m}$ ($a \times b \times c$). The $10\ \mu\text{m}$ thickness of the sample ensures uniform irradiation damage and negligible proton implantation, as SRIM calculations [39] show the projected range of protons is $67\ \mu\text{m}$ in these conditions.

For the irradiations, the sample is mounted onto an aluminum sample holder that allows for linear and rotational motion. In order to avoid heat damage of the sample we use a relatively low beam current of 500 nA and a cooling stage that maintains the sample at -10°C during irradiation. The

incident proton beam of 4.7 mm diameter is homogenized by passing through a $1\ \mu\text{m}$ gold foil placed at 240 mm upstream from the sample. The beam is defined through a 7.8 mm aperture placed at 40 mm upstream. This setup is calibrated with the help of a Faraday cup placed downstream from the sample which captures all protons passing the aperture while the sample is moved out of the beam path. The sample is electrically connected to the sample holder and the irradiation chamber which, in turn, is isolated from all other electronics and from the beam pipe through plastic rings. This approach allows us to accurately determine the irradiation dose by integrating the current from the chamber, not affected by spurious effects due to the emission of secondary electrons. The sample was irradiated in four sessions at Western Michigan University to a rather high cumulative dose of 12×10^{16} p/cm² (protons per cm²). Such a high dose is known to start to degrade some superconducting properties in several families of superconductors [22]. After each irradiation we measured the resistivity using a Keithley 2182 voltmeter and 6221 current source, with currents ranging from 50 μA to 1 mA, in a helium 4 cryostat with a 7 T magnet. Contacts were made with sputtered platinum and silver epoxy Epotek H20E. Typical contact values are $\lesssim 5\ \Omega$. The voltage contacts were spaced 200 μm apart along the c axis.

III. DISORDER INCREASES T_c AND DECREASES T_{CDW}

Figure 1 shows the evolution of the superconducting [panel (a)] and of the CDW transition [panel (b)] with increasing irradiation dose. We define T_c as the point where the resistivity dropped to 50% (midpoint). Figure 1(a) reveals a clear increase of T_c with irradiation, from 4.15 K in the pristine state to 5.25 K at the highest irradiation dose. Even though there appear shoulders of unknown origin near the top/bottom of the superconducting transitions at high doses, the width of the main part of the transition (15–85%) surprisingly decreases upon irradiation.

Figure 1(b) shows an overview of the c -axis resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ in semilog scale. The transition to the CDW phase at low temperature appears as a large increase of resistivity below $T_{\text{CDW}} \approx 77$ K, as previously observed [26]. We define T_{CDW} as the midpoint (50%) of this increase in resistivity. As the irradiation dose increases, T_{CDW} shifts toward lower temperature and the amplitude of the increase in resistivity is reduced. Contrary to the superconducting transition, the width of the CDW transition strongly increases with irradiation dose. The CDW transition also has an hysteresis of approximately 1 K, as was previously observed [32,38] and recently studied in details [31]. We find that this hysteresis survives up to the highest irradiation dose. However, after irradiation the hysteresis occurs only below T_{CDW} (midpoint), whereas, in the pristine state, the hysteresis extends up to the onset of the transition (83.5 K).

We note that Matthiessen's rule seems to fail both above and below T_{CDW} in Fig. 1(b). But one needs to be very careful when applying Matthiessen's rule in a CDW system. In the CDW phase, the density of states (DOS) at the Fermi level decreases (gapped charge carriers) but there are also fewer electrons to scatter off, meanwhile the CDW will also contribute to electronic scattering. So, the resistivity can

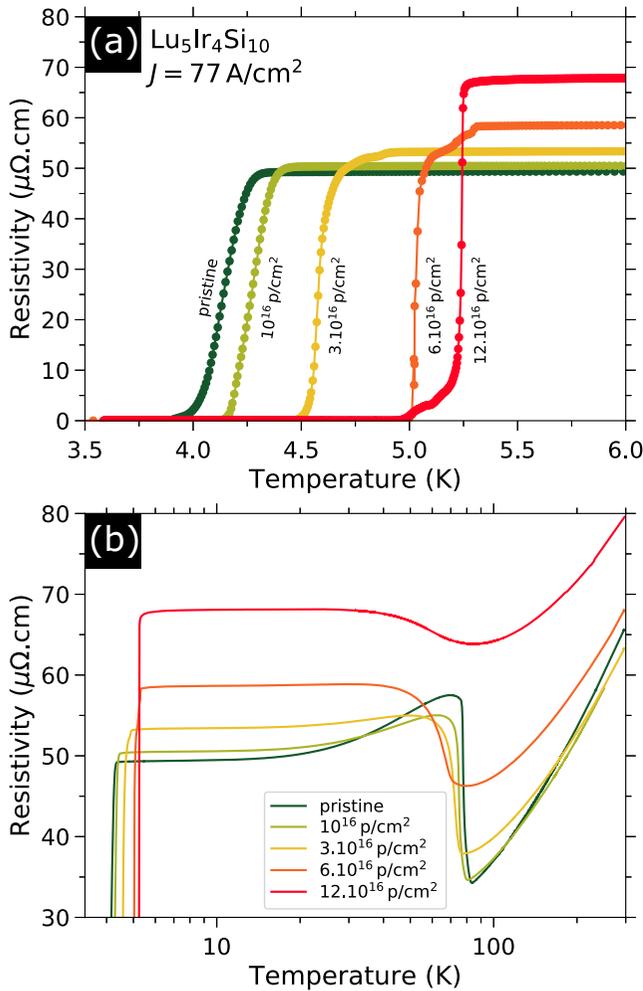


FIG. 1. Temperature dependence of the c -axis resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ for increasing irradiation doses. (a) The superconducting transition shifts to *higher* temperature after irradiation, in contrast with expected behavior. The transition width (15–85%) surprisingly *decreases* with irradiation, evidencing uniform irradiation damage (see text), even though there appear shoulders of unknown origin near the top/bottom of the superconducting transitions at high doses. (b) The large increase in resistivity below 80 K is caused by a CDW which gaps density of states at the Fermi level and increases electronic scattering. Contrary to T_c , the CDW transition temperature shifts to *lower* temperature, the transition width *increases*, and the amplitude of the increase is reduced for increasing irradiation dose.

go either way at the CDW transition. A blatant example of this mercurial behavior are the 1T and 2H polytypes of the dichalcogenide compound TaS_2 : The resistivity increases in the CDW phase of 1T- TaS_2 (dominated by DOS effects), while the resistivity decreases in the CDW phase of 2H- TaS_2 (dominated by electron scattering effects). Note that a similar effect of increase/decrease in resistivity can also occur at a SDW transition as observed in $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$: Irradiating the sample with electrons makes the transition split into an upturn and subsequent downturn upon cooling [40]. Besides, the CDW is commensurate in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, so that there is also a Fermi surface reconstruction, and it is a system with tens of electronic bands, eleven of which cross the Fermi level

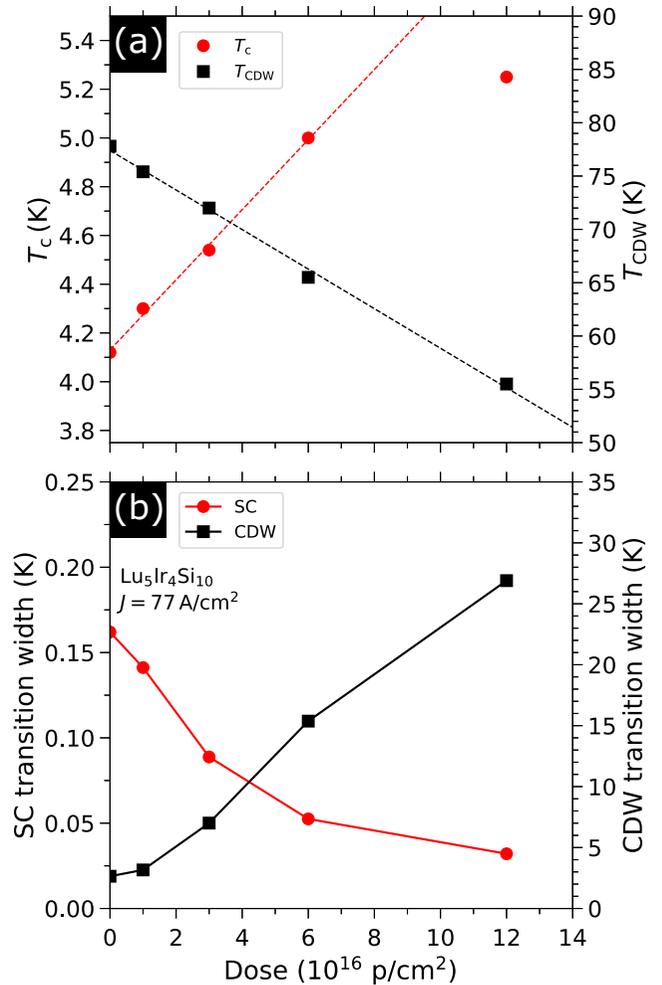


FIG. 2. Superconducting and CDW transitions as a function of irradiation dose. (a) T_c and T_{CDW} vary linearly up to high irradiation doses. T_{CDW} decreases at a constant rate of $-1.85 \text{ K}/10^{16} \text{ p}/\text{cm}^2$, whereas T_c increases at a rate of $+0.14 \text{ K}/10^{16} \text{ p}/\text{cm}^2$, which appears to saturate at the highest irradiation dose. (b) For both transitions we define the width using a 15%–85% criterion (for the CDW: between the min/max resistivity above/below T_{CDW} , respectively). As irradiation dose increases, the superconducting transition width is reduced whereas the CDW transition width increases, evidencing the different mechanisms through which disorder affects them (see text).

[29]. Finally, a short range CDW order can persist above the damage level erasing the trace of transition in the resistivity versus temperature curve, as evidenced in NbSe_2 in Ref. [41], and short range CDW correlations can also persist above T_{CDW} (e.g., CDW satellite peaks already appear in x ray below 85 K in Ref. [33]). Such a short range CDW order will not lead to Fermi surface reconstruction but will still contribute to transport properties.

The simultaneous variations of T_{CDW} and T_c are summarized in Fig. 2 as a function of irradiation dose. T_c increases almost linearly at a rate of $+0.14 \text{ K}/10^{16} \text{ p}/\text{cm}^2$ (or $0.093 \text{ K}/\mu\Omega \text{ cm}$) and starts saturating after the last irradiation. T_{CDW} decreases linearly in the whole range of irradiation doses at a rate of $-1.85 \text{ K}/10^{16} \text{ p}/\text{cm}^2$ (or $-1.18 \text{ K}/\mu\Omega \text{ cm}$).

According to Anderson's theorem [42], in an isotropic s-wave superconductor small concentration of nonmagnetic defects should not affect T_c while magnetic defects should be pair breaking and reduce T_c . Generally, the effect of pair-breaking scattering is described by Abrikosov-Gorkov theory [43,44]. In this theory, T_c is found to always decrease [45–47]. We thus conclude that the increase of T_c we observed cannot be explained by the standard effects of disorder on a superconductor.

Rather, such an increase of T_c with irradiation dose arises naturally from a competition scenario between the CDW and superconductivity, if irradiation suppresses the density wave more than superconductivity [16,17,48,49]. We recently demonstrated such an increase of T_c via competition with CDW using irradiations in the d-wave cuprate superconductor $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ [22]. This has also been evidenced in the dichalcogenides superconductors using irradiation induced disorder [19,41,50] and substitution disorder [51]. Both types of disorder strongly suppress CDW, either via real-space phase fluctuations [52,53] (domains) or by pair breaking [54]. A competition scenario was also proposed for $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ based on pressure [26] and doping [32,36] studies. The increase of T_c that we observe upon irradiation is therefore definitive evidence that the CDW is competing with superconductivity in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$.

Quantitatively, $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ is a weak coupling limit s-wave superconductor ($\Delta C/\gamma T_c = 1.41$ [26], close to 1.43), so that the electronic density of states released by the CDW should yield an exponential increase of T_c following the standard formula for a BCS superconductor: $T_c = \alpha \theta_D \exp(-\frac{1}{N(E_F)V})$ where $\alpha \approx 1.14$ in the weak coupling limit, $\theta_D = [315\text{--}366]$ K is the Debye temperature [26,32], $N(E_F)$ is the density of states at the Fermi level involved in Cooper pairs, and V is the attractive potential between the electrons of the pair.

The evolution of the residual resistivity as a function of irradiation dose is presented in Fig. 3. Before irradiation, the residual resistivity is $\rho_0 = 49.3 \mu\Omega \text{ cm}$ and the residual resistivity ratio (RRR) is $\rho_{295\text{K}}/\rho_0 = 1.3$, in line with previous studies [26,36,38]. After irradiation, we find that the residual resistivity increases linearly at a rate of $1.57 \mu\Omega \text{ cm}/10^{16} \text{ p/cm}^2$ without saturation up to $12 \times 10^{16} \text{ p/cm}^2$. Such a linear increase is what is typically expected in metals following the ‘‘unitary limit’’ [55], but this was not *a priori* obvious in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ because of the CDW. Indeed, on one hand, irradiation suppresses the CDW, which increases the density of states at the Fermi level and should reduce the residual resistivity. But on the other hand, irradiation increases the number of defects and reduces the size of CDW domains, both of which should raise electronic scattering and increase the residual resistivity. Here, we can at least conclude that the increased scattering more than compensates the increased density of states, as the residual resistivity increases overall. We also find that $\rho^{\text{max}} - \rho^{\text{min}}$, the amplitude of the jump in resistivity at T_{CDW} , is reduced after irradiation (see Fig. 3). Again, a natural explanation for this reduction would be that after the CDW is suppressed by disorder, it does not gap as much electronic density of states.

We also note that there appear to be slight discrepancies in the literature values of T_c and T_{CDW} in pristine single crystals

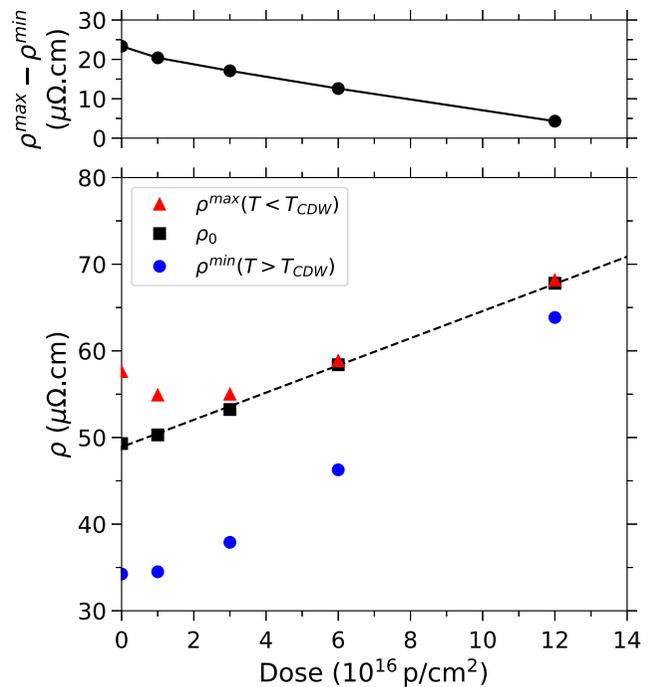


FIG. 3. Irradiation dose dependence of the residual resistivity ρ_0 and the resistivity jump at T_{CDW} . We define the amplitude of the jump as the difference $\rho^{\text{max}} - \rho^{\text{min}}$ between the maximum and minimum resistivity below and above T_{CDW} , respectively [see Fig. 1(b)]. ρ_0 increases linearly with dose at a rate of $1.57 \mu\Omega \text{ cm}/10^{16} \text{ p/cm}^2$. No saturation of defects creation is observed up to $12 \times 10^{16} \text{ p/cm}^2$.

[26–28,33,36]: ranging from $T_c = 3.8$ K and $T_{\text{CDW}} = 79$ K, to $T_c = 3.9$ K and $T_{\text{CDW}} = 83$ K. This could be due to different criteria to determine the transition temperatures, for instance using the onset of the jump in resistivity versus the midpoint to define T_{CDW} . This onset is also at 83 K in our work and we expect it to be less sensitive to disorder as it relates to short range CDW fluctuations. But based on our present results, the spread in T_c and T_{CDW} values in the literature could simply reflect the slightly different levels of disorder in as-grown crystals.

IV. INCREASE OF $H_{c,2}$ WITH DISORDER

As expected from the increase of T_c , we find an increase of the in-plane upper critical field ($H_{c,2}$) for increasing irradiation dose. Our measurements of $H_{c,2}$ are reported in Fig. 4(a), as a function of temperature for several irradiation doses, and where we define $H_{c,2}$ as the point where the resistivity drops 10% below the residual resistivity value ρ_0 . Figure 4(c) shows the detailed magnetic field dependence of the R(T) curves at the superconducting transition. We note that, notwithstanding the peak effect related to vortex lattice softening, the R(T) curves are shifted downward as the field increases, with the same shape and without broadening, even after repeated irradiations and in large magnetic fields. Our data is in good agreement with published $H_{c,2}(T)$ data in non-irradiated $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ [26,57]. We find that $H_{c,2}(T)$ follows the Werthamer-Helfand-Hohenberg-Maki (WHHM) scaling [56] at all irradiation doses. Using the WHHM scaling, Ref. [26]

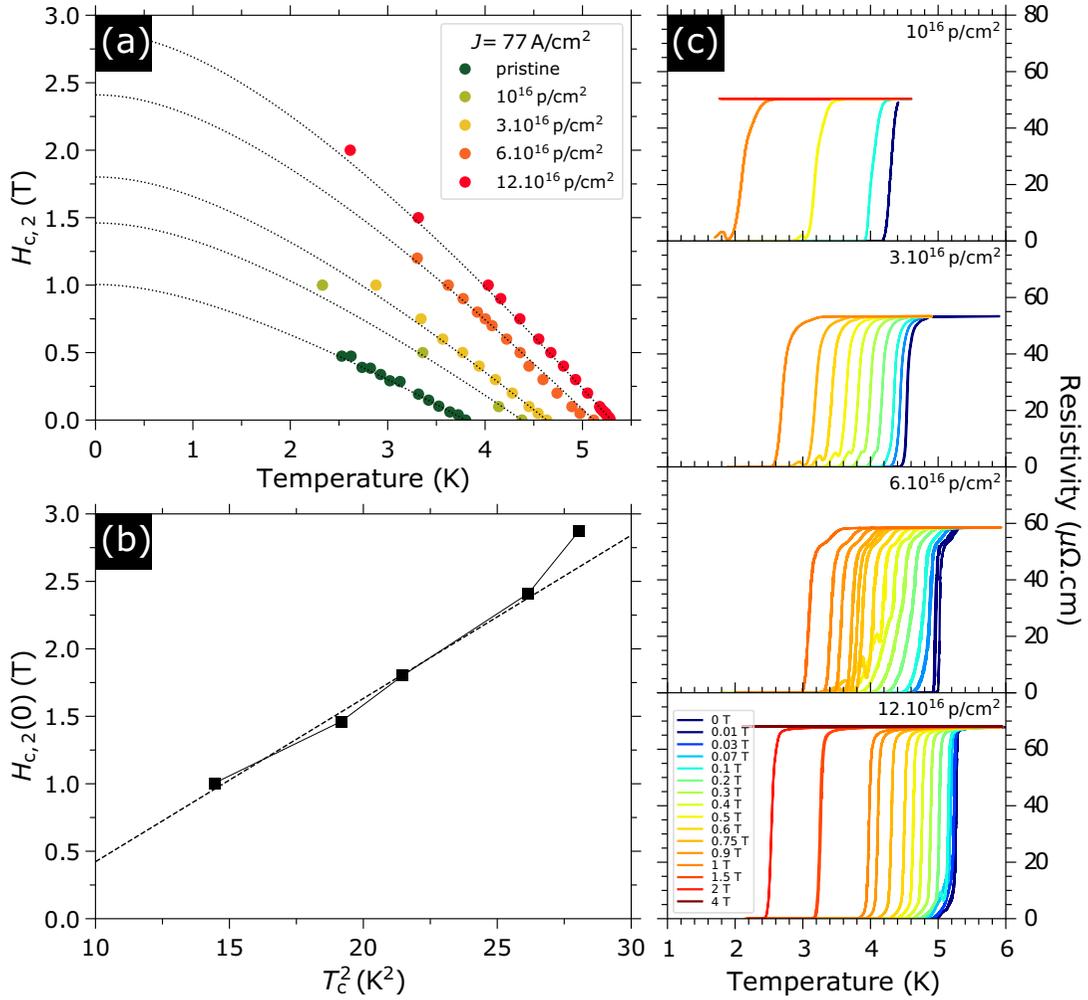


FIG. 4. $H_{c,2}$ vs irradiation dose. (a) Superconductivity second critical field $H_{c,2}$ for H in plane as a function of temperature and irradiation dose. Lines are theoretical curves from Werthamer-Helfand-Hohenberg-Maki (WHHM) theory [56] with $\alpha = 0.21$ and $\lambda_{SO} = 0$. Data in the pristine state is taken from Ref. [26]. (b) Superconductivity second critical field at zero temperature $H_{c,2}(0)$, extrapolated from the WHHM curves in (a), appears to increase linearly with T_c^2 at a rate of 0.12 T/K² (see text). (c) Detailed magnetic field dependence ($H//ab$) of the c -axis resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ at the superconducting transition, using a current density of 77 A/cm² and for increasing irradiation doses (top to bottom). The superconducting transition does not broaden, even after repeated irradiations and in a magnetic field. A vortex lattice peak effect is also visible in the superconducting transition (see also Fig. 7 in Appendix).

found best fit parameters values $\alpha = 0.21$ for the Maki parameter and $\lambda_{SO} = 9.0$ for the spin-orbit coupling. However, due to the small value $\alpha = 0.21$, fits are essentially insensitive to the choice of the spin-orbit coupling λ_{SO} , so that we can adopt in the following $\lambda_{SO} = 0$ and extrapolate the value of $H_{c,2}(0)$ with little uncertainties (see Appendix A for details).

In Fig. 4(b) we find that $H_{c,2}(0)$ is in good agreement with a T_c^2 dependence. In a usual s-wave isotropic superconductor in the dirty limit, $H_{c,2}(0)$ should scale with T_c . Indeed, the upper critical field is equal to $\frac{\phi_0}{2\pi\xi_0^2}$ and in the dirty limit the coherence length ξ_0 is renormalized to $\xi_{0,d} = \sqrt{\xi_0\bar{l}_0}$, where $\bar{l}_0 = v_F\tau_0$ and $\xi_0 = \frac{\hbar v_F}{1.76\pi k_B T_c}$ in the weak-coupling BCS theory, yielding:

$$H_{c,2}(0) = \frac{\phi_0}{2\pi\xi_{0,d}^2} \approx \frac{\phi_0 1.76 k_B}{2\hbar v_F^2} \frac{1}{\tau_0} T_c. \quad (1)$$

So usually, in the dirty limit $H_{c,2}(0) \propto T_c$, however here $H_{c,2}(0) \propto T_c^2$. This unusual scaling can be easily explained by the fact that $1/\tau_0$ varies linearly with T_c due to the competition with the CDW, namely: The prefactor $\frac{\phi_0 1.76 k_B}{2\hbar v_F^2}$ is independent of irradiation dose, whereas $1/\tau_0$ is usually proportional to the irradiation dose for uniform nonoverlapping defects in metals [55], and in this compound, empirically, T_c increases linearly with the irradiation dose (see Fig. 2).

V. PRESSURE, DOPING AND IRRADIATION: PROGRESSIVE VERSUS BINARY INCREASE OF T_c

Interestingly, the competition scenario between CDW and superconductivity still requires clarification in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$. Indeed, pressure studies [26,28] show a *binary* effect on T_c : Below 2 GPa, T_c is 4 K and constant, whereas above 2 GPa, T_c is 9 K and constant; in contrast doping studies [27,32,36] show a *progressive* increase of T_c from 4 K to 6 K. This is

all the more surprising as in both cases the CDW is progressively suppressed. Hence, naively, shouldn't one expect that T_c also increases progressively with pressure? We argue that this can be explained by the different mechanisms through which the CDW is suppressed when using pressure, doping, and irradiation.

The variations as a function of irradiation dose of the width (15–85%) of the superconducting and CDW transitions are summarized in Fig. 2(b). As can be seen, the width of the superconducting transition decreases with irradiation dose. In general, for the superconducting pairs in a s-wave superconductor, disorder only acts via the pair-breaking mechanism from magnetic defects. The defects introduced by proton irradiation are nonmagnetic in nature. Therefore, they are not pair breaking for a s-wave superconductor like $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$. The Anderson theorem remains valid even in the presence of cascade defects, as they are not magnetic defects [42]. In addition, the suppression mechanism by real-space phase fluctuations from disorder is usually irrelevant for superconductivity unless the superconducting condensate is modulated in real space, that is Cooper pairs have a nonzero center of mass momentum as in the FFLO, stripes, or pair-density wave superconducting states [25,58]. The question remains open in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ whether the superconducting condensate is modulated in the CDW phase (beyond a trivial spatial segregation), but we do not think this is highly likely as it is a rather standard BCS s-wave compound. STM studies as in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ [59] could shed light on this interesting open question. So, except for the elusive non-zero-momentum Cooper pairs states, in a standard s-wave superconductor no change of the superconducting transition width is expected with disorder at first. However, there could still be a “trivial” broadening of the superconducting transition via the competition with the CDW, that is: A spatially inhomogeneous CDW will yield a spatially inhomogeneous superconducting order parameter (stronger where the CDW is weaker), hence a broader T_c . Thus, here the decreased superconducting-transition width after irradiation shows that the irradiation damage caused by proton irradiation is very uniform, to the point that disorder in the sample is actually more uniform after irradiation.

Let us now turn to how disorder influences the CDW. By contrast to the superconducting transition, as shown in Fig. 2, the width of the CDW transition significantly increases with irradiation dose, and both T_{CDW} and the jump in resistivity decreases with increasing disorder. These are strong indications that the CDW is suppressed by real-space phase fluctuations.

While the almost linear variation of residual resistivity with irradiation dose points to the main contribution arising from scattering in the unitary limit, 4-MeV proton irradiation produces a complex morphology of defects: Beyond vacancy-interstitial pairs (atomic point defects), cascade-type clusters of defects are also generated. These clusters are typically a few nanometers in diameter, and, because of this larger size, they are expected to be less pair breakers for superconductivity than point defects as bigger defects should scatter in a smaller volume of reciprocal space and be closer to or larger than the coherence length. There is also experimental evidence for this lesser pair breaking effect: An extreme example is commercial superconducting tapes based on cuprates. Despite the huge density of tiny nanoparticles in these tapes, T_c

is almost the same as in the clean bulk cuprate and proton irradiation also does not affect T_c much in these tapes [60]. Also, in iron-based superconductors, it was found that the rate of depression of T_c per unit of residual resistivity increase is twice higher when using electron irradiation [61] (producing solely point defects) rather than proton irradiation [62], suggesting that cascade defects could have less pair breaking effect on superconductivity too. We note that in the case of proton irradiation the scattering process from combined point defects and cascade-type clusters of defects has an undetermined parameter: the ratio between those two channels. In superconductors where nonmagnetic defects are pair breakers, such as those with s + - or d-wave gap, this complex damage morphology can impede conclusive statements on pairing mechanism. For instance, a decrease of T_c followed by a saturation of T_c for increasing proton irradiation dose has been attributed to a crossover in pairing mechanism from s + - to s + + [63], but this effect could not be reproduced in electron irradiation studies [64].

In $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, the cascade defects are expected to not be pair breaking at all as we know that nonmagnetic defects do not decrease T_c in an isotropic s-wave superconductor (which is the case here until proven otherwise). However, nonmagnetic defects irrespective of their sizes do suppress the CDW order, which is consistent with the Imry-Ma theorem [65] and some earlier studies [66,67]. Therefore, the cascade defects have pair-breaking effect only on the CDW phase. In addition, we expect the cascade-type defects produce strong pinning of the CDW and contribute to the broadening of the CDW transition. Therefore, for the study of the coexistence of superconductivity and CDW, it could be seen as a benefit of proton irradiation that it provides these few nanometers wide cascade-type defects. These cascades should indeed mostly suppress the CDW while not affecting much superconductivity (even if of the d-wave type [22,60]), thus enabling to evidence the competition or synergy between the two more clearly.

Two different mechanisms have been proposed for CDW suppression by disorder: (i) a pair-breaking mechanism [50,68] where disorder increases the scattering rate, which induces a broadening of the Fermi function and reduces the peak in electronic susceptibility. This process reduces the jump in resistivity at the CDW transition, but it does not affect the macroscopic coherence of the CDW and produces a uniform global reduction of the CDW. Thus this mechanism cannot account for the broadening of the CDW transition. (ii) a real-space phase fluctuations mechanism where disorder pins the phase of the periodic spatial modulation of the electronic density and associated lattice distortion. This breaks up the CDW into small domains, which broadens the transition (Ref. [69] §2.7.2). Thus, our data indicate that real-space fluctuations are clearly contributing to the suppression of the CDW by irradiation in this study. In doping studies [27,36] in which Sc (Co) were introduced on the Lu (Ir) sites, respectively, pronounced broadening and suppression of the CDW was observed, in analogy to the results presented here, suggesting that real-space phase fluctuations are suppressing the CDW in doping studies as well. However, in doping studies, additional effects may arise from doping-induced changes of the Fermi surface.

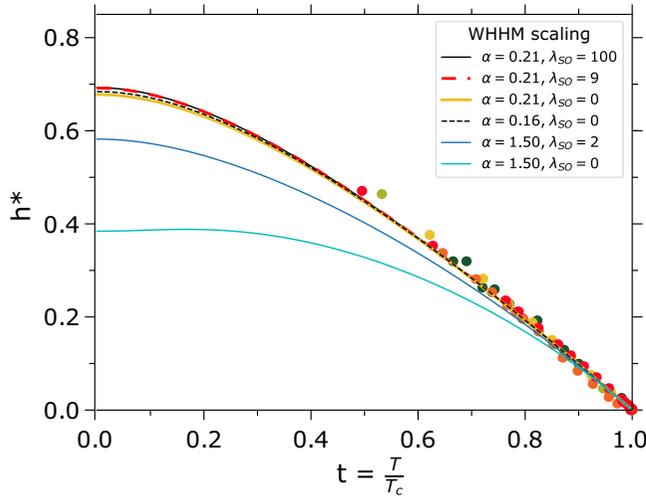


FIG. 5. $H_{c,2}$ and WHHM scaling. Superconductivity reduced second critical field $h^* = H_{c,2}/(-dH_{c,2}/dt)_{t=1}$ as a function of the reduced temperature $t = T/T_c$, for all irradiation doses. No significant changes occur with irradiation in this reduced plot. The lines are theoretical curves from WHHM theory. Data in the pristine state was taken from Ref. [26].

Conversely, in pressure studies, the mechanism by which the CDW is suppressed must be different as there is no change in the number of defects, hence no pair breaking nor phase fluctuations. This is also quite strikingly evidenced experimentally [26,28]: Even though T_{CDW} decreases by up to a factor of 10, there is no significant change in the CDW transition width under pressure. A natural explanation for this is that pressure stiffens the elastic constants of the crystal, which makes the periodic lattice distortion less favorable energetically and reduces T_{CDW} . This suppression-by-elastic-stiffening mechanism follows from the standard CDW stability criterion of Chan and Heine [70]. As this mechanism is global in essence, it explains why the CDW transition width remains sharp and constant even though T_{CDW} diminishes. Finally, as T_c remains essentially constant up to 2 GPa, it also means that the density of states at the Fermi level is essentially constant with pressure in the CDW phase. To first order, the elastic stiffening modifies only the temperature at which the Chan and Heine criterion is met, without affecting the density of electronic states involved.

Thus, we can now reconcile the effects of pressure, doping, and irradiation on the CDW and superconductivity, by considering the differences between the three mechanisms of CDW suppression: pair breaking, real-space phase fluctuations, and elastic constants stiffening. The main effect of pressure on the CDW is to reduce T_{CDW} via elastic constants stiffening, but both the CDW-transition width and the amount of density of states gapped by the CDW remains constant. Therefore T_c increases in a binary way: As pressure increases T_c is essentially constant and T_{CDW} decreases, until $T_{CDW} < T_c$, at which point T_c jumps to another constant value. By contrast, with doping or irradiation there are both phase fluctuations and pair breaking which not only reduces T_{CDW} but also reduces the amount of density of states gapped by the CDW. Thus T_c increases in a progressive way: Doping/irradiation frees

density of states that was gapped by the CDW, which then raises T_c [71] (as superconductivity is not subject to the same strong pair-breaking or phase fluctuations effects of disorder).

We note here that CDW and superconductivity cannot live *independently* as the CDW gap can develop only on some parts of the Fermi surface, but the superconducting gap always opens over the entire Fermi surface. So, in this sense, CDW and superconductivity are always competing for electrons at the Fermi level, albeit somewhat trivially. An even stronger evidence for competition would be a reduction of the CDW diffraction peak upon entering the superconducting state, as in x-ray studies of $YBa_2Cu_3O_{6.67}$ [10]. To our knowledge, this is an open question that has not been settled experimentally in $Lu_5Ir_4Si_{10}$ and would greatly benefit from low temperature x-ray studies. Likewise, if the CDW modulation survives in the superconducting state, we would expect some comodulation of the superconducting and CDW order parameters, if only around defects and close to vortex cores, which an STM study might be able to evidence, if proper surfaces of $Lu_5Ir_4Si_{10}$ can be produced.

VI. CONCLUSION

We showed that irradiation induced disorder enhances the superconducting critical temperature T_c and H_{c2} while it suppresses the CDW in $Lu_5Ir_4Si_{10}$. We showed how this increase of T_c cannot be accounted for by the expected effect of disorder and instead stems from the increase of density of states at the Fermi level. Our results thus make a very compelling case that superconductivity and CDW are competing for electronic density of states at the Fermi level in $Lu_5Ir_4Si_{10}$. Owing to its prototypical, 1D, Peierls type CDW and the s-wave, weak-coupling nature of its superconductivity, $Lu_5Ir_4Si_{10}$ thus provides a platform from which to understand the more complex cases of density waves and superconductivity coexistence in heavy fermions, Fe-based, or cuprates superconductors. Recently [72] it was shown that, in an unconventional superconductor mediated by spin fluctuation, very inhomogeneous conditions may result in an increase of T_c . However we do not think this latter case is relevant to the standard s-wave superconductor studied here. Also very recently, it was found that, while the effect of disorder on the dichalcogenide $NbSe_2$ in bulk form is explained in terms of CDW-superconductivity competition and synergy [41], in monolayer $NbSe_2$ a much larger T_c dome was discovered as a function of disorder and this has been proposed to be due to the wave-function multifractality in a 2D monolayer system [73,74]. Thus, disorder as a tuning parameter is finding relevance not only for the study of bulk superconductors with density waves but also for 2D materials such as monolayer transition metal dichalcogenides.

ACKNOWLEDGMENTS

M.L. acknowledges fruitful discussions with S. Eley, F. Ronning, C. Proust, and P. Monceau. The experimental study at Argonne National Laboratory was supported by the US Department of Energy (DOE), Office of Science, Materials Sciences and Engineering Division. V.M. was supported by the Center for Emergent Superconductivity, an Energy Frontier Research Center, funded by the US DOE, Office of

Science, Office of Basic Energy Sciences. Proton irradiation was performed at Western Michigan University. C.O. and P.R.'s work is supported by the ANR-DFG Grant No. ANR-18-CE92-0014-03 "Aperiodic." The samples used in this study were grown at the Néel Institute by C. Opagiste.

APPENDIX A: WHHM SCALING

To support the relevance of the WHHM theory in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$, despite its complex Fermi surface [29], we verify that the α values estimated in two independent ways are close, following the recommendation of Ref. [56]. First, using the normal state values just above T_c : $\alpha = \frac{3e^2 \hbar \gamma \rho_0}{2m\pi^2 k_B^2} \approx 0.16$ (SI units), with $\gamma = 119.1 \text{ J/m}^3/\text{K}^2$ (the volumic Sommerfeld coefficient from $\gamma = 23.42 \text{ mJ/mol/K}^2$ in Ref. [26]) and $\rho_0 = 56.2 \mu\Omega \text{ cm}$ (also from Ref. [26]). This result is close to the value deduced from the slope

of $H_{c,2}$ near T_c (in T/K): $\alpha = 0.52758 \times -\left(\frac{dH_{c,2}}{dT}\right)_{T_c} \approx 0.21$ (SI units). In Fig. 5, we show that because of the small value $\alpha = 0.21$, the WHHM scaling is essentially insensitive to the choice of the spin-orbit coupling λ_{SO} in this compound.

APPENDIX B: DETAILED CDW AND SC TRANSITION CURVES AND PEAK EFFECT

In Fig. 6, we show that the point where resistivity drops below the resolution of the instruments ($\approx 10^{-3} \mu\Omega \text{ cm}$) also shifts to higher temperature with increasing irradiation dose, in the same manner as the midpoint of the transition. We also note that in the pristine (nonirradiated) state, higher current densities shift the transition to lower temperature, whereas after irradiation the curves become almost identical at all current densities. We found no effect of the current density

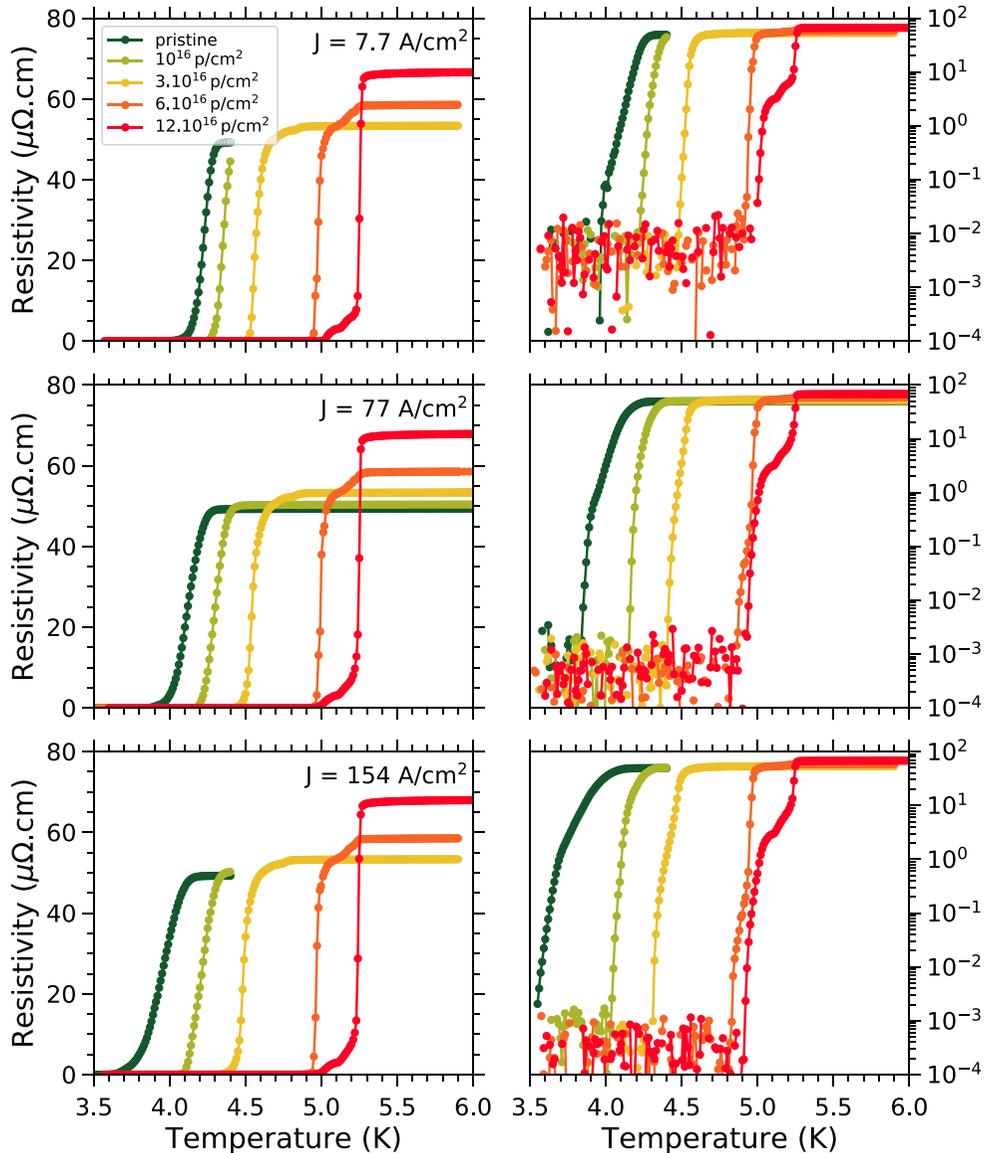


FIG. 6. Detailed temperature dependence of the c -axis resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ at the superconducting transition: for increasing current densities (top to bottom) and in linear (left column) and semilog scale (right column).

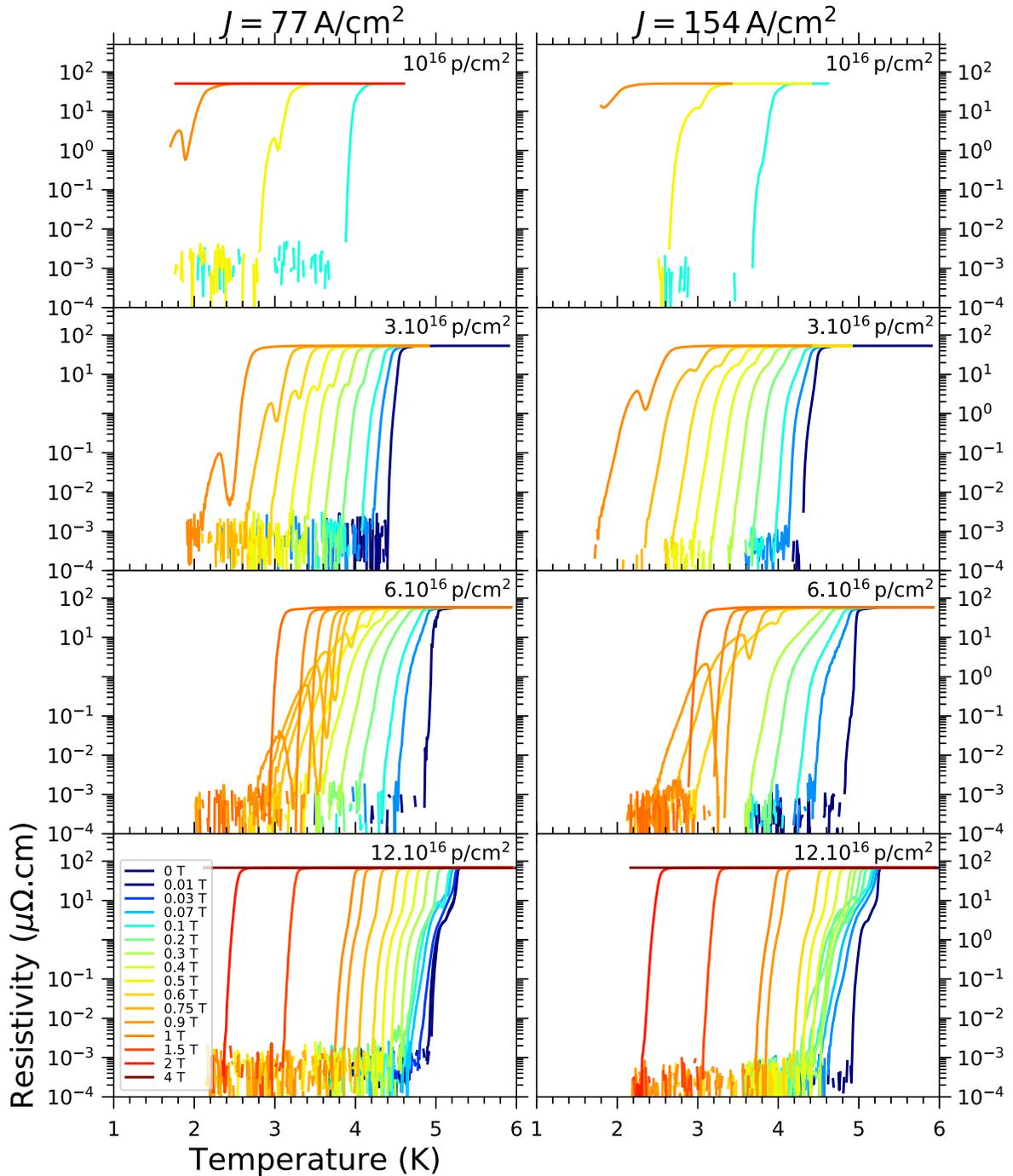


FIG. 7. Detailed magnetic field dependence ($H//ab$) of the c -axis resistivity of $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$ at the superconducting transition: for increasing irradiation doses (top to bottom) and using a current density of 77 (left column) and 154 A/cm^2 (right column). A peak effect is visible in the superconducting transition.

on the CDW transition in the range of current density that we explored ($\leq 154 \text{ A}/\text{cm}^2$).

In Fig. 7, we show that we observe a clear peak effect in the middle of the superconducting transition, at both current densities and for all irradiation doses, which shows this peak effect is robust to disorder. Such a peak effect [75–79] is usually caused by the softening of the vortex lattice near T_c

which enables it to better adapt to the distribution of defects, hence the drop in resistivity. To our knowledge, such a peak effect had never been reported in $\text{Lu}_5\text{Ir}_4\text{Si}_{10}$. We also find that the irreversibility field (H_{irr}) defined as the point where the resistivity drops below the resolution of our instruments (1 $\text{n}\Omega\text{ cm}$) increases with irradiation dose, in line with $H_{c,2}$.

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