Evolution of the critical pressure with increasing Fe substitution in the heavy fermion system URu$_{2-x}$Fe$_x$Si$_2$

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Measurements of electrical resistivity, $\rho(T)$, were performed under quasi-hydrostatic pressure up to $P \sim 2.2$ GPa to determine the pressure dependence of the so-called “hidden order” (HO) and large-moment antiferromagnetic (LMAFM) phases for the URu$_{2-x}$Fe$_x$Si$_2$ system with $x = 0.025, 0.05, 0.10, 0.15$, and $0.20$. As the Fe concentration $(x)$ is increased, we observed that a smaller amount of external pressure, $P_c$, is required to induce the HO $\rightarrow$ LMAFM phase transition. A critical pressure of $P_c \sim 1.2$ GPa at $x = 0.025$ reduces to $P_c \sim 0$ at $x = 0.15$, suggesting the URu$_{2-x}$Fe$_x$Si$_2$ system is fully expressed in the LMAFM phase for $x \geq x_c^* = 0.15$, where $x_c^*$ denotes the ambient pressure critical concentration of Fe. Using a bulk modulus calculation to convert $x$ to chemical pressure, $P_{ch}(x)$, we consistently found that the induced HO $\rightarrow$ LMAFM phase transition occurred at various combinations of $x_c$ and $P_c$, such that $P_{ch}(x_c) + P_c \approx 1.5$ GPa, where $x_c$ denotes those critical concentrations of Fe that induce the HO $\rightarrow$ LMAFM phase transition for the URu$_{2-x}$Fe$_x$Si$_2$ compounds under pressure. We performed exponential fits of $\rho(T)$ in the HO and LMAFM phases in order to determine the pressure dependence of the energy gap, $\Delta$, that opens over part of the Fermi surface in the transition from the paramagnetic (PM) phase to the HO/LMAFM phase at the transition temperature, $T_0$. The change in the pressure variation of $\Delta(P)$ at the HO $\rightarrow$ LMAFM phase transition is consistent with the values of $P_c$ determined from the $T_0(P)$ phase lines at the PM $\rightarrow$ HO/LMAFM transition.

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I. INTRODUCTION

Subsequent to the initial bulk property measurements that were performed on the heavy fermion superconductor URu$_2$Si$_2$ in the mid 1980s,$^{1-3}$ researchers have yet to identify the order parameter (OP) associated with the so-called “hidden order” (HO) phase observed at the transition temperature $T_0 \approx 17.5$ K. In the bulk, the key signatures of the second-order symmetry-breaking transition are (1) the anomalous upturn in the electrical resistivity, $\rho(T)$, that is reminiscent of a spin density wave (SDW) feature as observed, for instance, in the $\rho(T)$ data for elemental chromium near $T_N = 311$ K,$^4$ and (2) the large Bardeen-Cooper-Schrieffer (BCS)-like feature observed in the specific heat, $C(T)$, below $T_0 = 17.5$ K. From the specific heat anomaly, it was originally determined that a considerable amount of entropy, $\Delta S \approx 0.2R\ln(2)$, is released during the transition.$^{1,2}$ From the exponential temperature dependence of the specific heat and the reduction of the electronic contribution to the specific heat,$^2$ a partial gapping scenario was proposed in which an energy gap, $\Delta \approx 130$ K,$^{1,2}$ is attributed to a charge- or spin-density wave (CDW or SDW) that forms over $\sim 40\%$ of the Fermi surface with the remainder of the Fermi surface gapped by the superconductivity that occurs below $T_c \approx 1.5$ K.$^{2,3}$ For the past three decades, the intense search for the OP in the HO phase has been accompanied by a large effort to explain the reduction in entropy that occurs during the transition to the HO phase as well as to determine the origin of the energy gap, $\Delta$, near the transition temperature $T_0$. For a comprehensive survey of the experimental and theoretical research regarding the URu$_2$Si$_2$ compound, the reader is referred to Refs. 6 and 7.

Research at ambient pressure on the URu$_2$Si$_2$ compound reveals the existence of a second-order phase transition from a highly correlated paramagnetic (PM) phase above $T_0$ to the HO phase below $T_0$ that exhibits antiferromagnetic (AFM) order with a very small ordered moment of $\mu \sim (0.03 \pm 0.02)\mu_B$ per U atom that is aligned parallel to the $c$ axis (in the simple tetragonal structure).$^{8,9}$ It is interesting to note that this small value for the magnetic moment in the HO phase is two orders of magnitude smaller than values for the magnetic moment of $\mu \sim 1.6 - 2.9\mu_B$ per U atom observed in other magnetically ordered compounds from the UT$_2$Si$_2$ series where $T$ is a $d$-electron transition element.$^{10,11}$ This uniquely tiny magnetic moment observed in the HO phase in URu$_2$Si$_2$ is too small to account for the loss of entropy, $\Delta S$, during the PM $\rightarrow$ HO transition. It is now believed that the small moment antiferromagnetic (AFM) order is not intrinsic to the HO phase but rather due to the existence of small pockets of the high pressure large-moment antiferromagnetic (LMAFM) phase that are stabilized by the extrinsic strain that can occur in preparing the sample for measurement.$^{12,13}$ This small concentration of the LMAFM phase can produce an average moment of a few one-hundredths of a Bohr magneton ($\mu_B$) per U atom. An extensive amount of pressure research on HO in URu$_2$Si$_2$ reveals, in addition

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TABLE I. Values of the critical pressure, $P_c$, and the pressure dependence ($\partial T_{HO}/\partial P$) in the HO phase along with the type of measurement for the URu$_2$Si$_2$ ($x = 0$) parent compound from previous reports.

<table>
<thead>
<tr>
<th>Year</th>
<th>Reference</th>
<th>Critical Pressure $P_c$ (GPa)</th>
<th>$\partial T_{HO}/\partial P$ (K GPa$^{-1}$)</th>
<th>Measurement type</th>
</tr>
</thead>
<tbody>
<tr>
<td>1987</td>
<td>McElfresh$^{18}$</td>
<td>$P_c &gt; 1.3$</td>
<td>$1.3 \pm 0.1$</td>
<td>$\rho(T)$</td>
</tr>
<tr>
<td>1999</td>
<td>Amitsuka$^{14}$</td>
<td>$P_c = 1.5$</td>
<td>$1.3 \pm 0.1$</td>
<td>elastic NS</td>
</tr>
<tr>
<td>2001</td>
<td>Matsuda$^{15}$</td>
<td>$P_c = 1.5$</td>
<td>$1.3 \pm 0.1$</td>
<td>SI NMR</td>
</tr>
<tr>
<td>2003</td>
<td>Motoyama$^{17}$</td>
<td>$P_c = 1.1 - 1.5$</td>
<td>$1.0 \pm 0.1$</td>
<td>dilatometry</td>
</tr>
<tr>
<td>2004</td>
<td>Amato$^{18}$</td>
<td>$P_c \sim 1.4$</td>
<td>$1.0 \pm 0.1$</td>
<td>$\mu$SR</td>
</tr>
<tr>
<td>2007</td>
<td>Jeffries$^{19}$</td>
<td>$P_c = 1.5$</td>
<td>$1.0 \pm 0.1$</td>
<td>$\rho(T)$</td>
</tr>
<tr>
<td>2008</td>
<td>Hassinger$^{20}$</td>
<td>$P_c \sim 1.3$</td>
<td>$1.1 \pm 0.1$</td>
<td>$\rho(T)$</td>
</tr>
<tr>
<td>2010</td>
<td>Butch$^{21}$</td>
<td>$1.3 &gt; P_c &gt; 1.5$</td>
<td>$1.3 \pm 0.1$</td>
<td>elastic NS, $\rho(T)$</td>
</tr>
</tbody>
</table>

The critical pressure $P_c$ is observed to the second-order PM $\rightarrow$ HO transition, the existence of a first-order (symmetry-breaking) transition from the small moment HO phase to the LMAFM phase which has a larger magnetic moment of $\mu \approx 0.4 \mu_B$ per U atom that is also aligned parallel to the c axis.$^{12-15}$ The first-order transition to the high pressure LMAFM phase is observed to occur at critical pressures that range from $P_c \approx 0.5$ GPa (as $T \rightarrow 0$) to $P_c \approx 1.5$ GPa (for $T = T_0$).

Table I provides a sampling of the variety of pressure research which includes measurements of electrical resistivity ($\rho(T)$),$^{16,19-21}$ elastic neutron scattering (NS),$^{14,21}$ thermal expansion (dilatometry),$^{17}$ muon spin resonance ($\mu$SR),$^{18}$ and nuclear magnetic resonance ($^{29}$Si NMR).$^{15}$ Together, the previous reports seem to indicate that $P_c \sim 1.5$ GPa is perhaps an upper bound value on the applied critical pressure that induces the first-order phase transition from the HO phase to the LMAFM phase.

Recent results from a number of ambient pressure experiments reveal that by tuning either polycrystalline$^{22}$ or single crystal$^{23,24}$ samples of URu$_2$Si$_2$ with the iso-electronic substitution of Fe for Ru, it is possible to reach the same high pressure LMAFM phase in which the first-order HO $\rightarrow$ LMAFM phase transition at ambient pressure now occurs at some critical value of Fe concentration, $x_c^* \approx 0.1 - 0.2$. Furthermore, there is a remarkable correspondence observed in the evolution of $T_0$ with increasing Fe concentration for the range $x = 0$ to 0.3, when compared with the results of experiments in which URu$_2$Si$_2$ is tuned with increasing applied pressure, $P$. It has been suggested that the similarities between the $T_0$ vs. $x$ and $T_0$ vs. $P$ phase boundaries are a consequence of the effective reduction in the volume of the unit cell in which the substitution of the smaller Fe ions for Ru acts as a “chemical pressure”, $P_{ch}$, that tracks well with the effects of applied pressure, $P$. $^{22-24}$

In the research reported herein, we simultaneously tuned the URu$_2$Si$_2$ compound with both Fe substitution, $x$, and applied external pressure, $P$, in an effort to further explore the suggestion, as made initially in Ref. 22, that there is an equivalence between the application of pressure, $P$, and chemical pressure, $P_{ch}(x)$, in inducing the HO $\rightarrow$ LMAFM phase transition. We found that we could bias the parent compound with “chemical pressure” toward the LMAFM phase by systematically introducing small levels of Fe into the URu$_2$Si$_2$ compound, such that a smaller amount of applied external pressure is required to induce the transition to the high pressure LMAFM phase. Remarkably, we observed the consistent manner in which chemical pressure, $P_{ch}(x)$, and applied pressure, $P$, are “additive” such that $P_{ch}(x_c) + P_c \approx 1.5$ GPa at the first-order HO $\rightarrow$ LMAFM phase transition. (Herein, the symbol $x_c$ is used to denote those “critical” concentrations of Fe that induce the HO $\rightarrow$ LMAFM phase transition in URu$_{2-x}$Fe$_x$Si$_2$ compounds under pressure.) Hence, the predictability in which $P_{ch}(x_c)$ and applied critical pressure $P_c$ combine to induce the HO $\rightarrow$ LMAFM phase transition can serve as a guide for future research on the URu$_{2-x}$Fe$_x$Si$_2$ system under pressure. We also note that tuning the URu$_2$Si$_2$ compound simultaneously with both $x$ and $P$ may serve as a workaround to some of the limitations encountered in pressure experiments so that larger regions of phase space might be studied. To this point, we were able to track the suppression of the critical pressure, $P_c$, and hence the tricritical point, $(T_0(P_c), P_c)$, in the $T_0$ vs. $P$ phase diagram, as a function of Fe concentration, $x$, for the five URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.025$, 0.05, 0.10, 0.15, and 0.20) compounds.

The $T_0$ vs. $P$ phase diagrams presented in this work are based on measurements of electrical resistivity, $\rho(T)$, under quasi-hydrostatic pressure up to $P = 2.2$ GPa for the five URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.025$, 0.05, 0.10, 0.15, and 0.20) compounds. Additionally, we employed a theoretical model of electrical resistivity$^{25}$ which we were able to fit to the $\rho(T)$ data for $T < T_0$ in order to extract values of the charge energy gap, $\Delta$, as a function of pressure, $P$. The changes observed in the pressure dependence of the charge energy gap, $\Delta(P)$, are consistent with the values of the critical pressure $P_c$ that were determined from the $T_0$ vs. $P$ phase diagrams.

II. EXPERIMENTAL DETAILS

Single crystals of URu$_{2-x}$Fe$_x$Si$_2$ were grown according to the Czochralski method in a Techno Search TCA 4-5 Tetra-Arc furnace under a zirconium-gettered argon atmosphere. The quality of the single crystal samples were determined by Laue X-ray diffraction patterns performed with a Photonic Science PXS11 X-ray measurement system together with X-ray powder diffraction (XRD) measurements performed with a Bruker D8 Discover X-ray diffractometer that uses Cu-K$\alpha$ radiation. The XRD patterns were fitted via the Rietveld refinement technique using the GSAS + EXPGUI software package.

Annealed Pt wire leads were affixed with silver epoxy to gold-sputtered contact surfaces on each sample in a standard four-wire configuration. The single crystal samples were cleaved along the basal plane of the tetragonal structure of the sample and the measurements of $\rho(T)$ were made with current running parallel to the a
axis. The orientation of the single-crystal samples of URu$_{2−x}$Fe$_x$Si$_2$ were confirmed from Laue X-ray diffraction patterns.

Electrical resistivity $\rho(T)$ measurements were performed on single crystals of URu$_{2−x}$Fe$_x$Si$_2$ under applied pressure up to $P = 2.2$ GPa for Fe concentrations $x = 0.025, 0.05, 0.10, 0.15$, and $0.2$. A 1:1 mixture by volume of n-pentane and isopentyl alcohol was used to provide a quasi-hydrostatic pressure transmitting medium and the pressure was locked in with the use of a beryllium copper clamped piston-cylinder pressure cell. The pressure dependence of the superconducting transition temperature, $T_c$, of high purity Sn was used as a manometer. The superconducting transition of the Sn manometer was measured inductively and the pressure dependence of $T_c$ was calibrated against data from Ref. 26. Measurements of $\rho(T)$ were performed upon warming from $\sim 1$ to 300 K in a pumped 4He dewar and the temperature was determined from the four-wire resistivity of a calibrated Cernox sensor which was thermally sunk to the beryllium copper pressure clamp. For the URu$_{2−x}$Fe$_x$Si$_2$ compounds with $x = 0.05$ and $0.15$, a second set of $\rho(T)$ measurements were performed in reverse by releasing pressure from the pressurized cell from 2.2 GPa down to atmospheric pressure.

III. RESULTS AND DISCUSSION

Enhancement of $T_0$ with Fe substitution

The effect of Fe substitution on the PM $\rightarrow$ HO/LMAFM transition temperature, $T_0$, in the URu$_{2−x}$Fe$_x$Si$_2$ system for $x = 0.025$, $0.05$, $0.10$, $0.15$, and $0.20$ was determined from measurements of $\rho(T)$ at ambient pressure. The temperature dependence of the electrical resistivity, $\rho(T)$, in the vicinity of $T_0$ for the five compounds URu$_{2−x}$Fe$_x$Si$_2$ ($x = 0.025$, $0.05$, $0.10$, $0.15$, and $0.20$) is shown in the inset of Fig. 1. The PM $\rightarrow$ HO/LMAFM transition temperature, $T_0$, is defined in this report as the temperature at which there is a minimum in $\rho(T)$ (which occurs just prior to cooling through the small upturn in $\rho(T)$) as indicated by the black arrows shown in the inset of Fig. 1. (In this report, we denote the PM $\rightarrow$ HO transition temperature as $T_{HO}$, the PM $\rightarrow$ LMAFM transition temperature as $T_N$, and traditionally reserve the use of $T_0$ to refer to $T_{HO}$ and $T_N$, collectively.) The enhancement of $T_0$ with increasing Fe concentration is apparent from the shift of the minimum in $\rho(T)$ to higher temperatures such that for $x = 0.025$, $0.05$, $0.10$, $0.15$, and $0.20$, the values of $T_0$ are 16.6, 17.0, 17.7, 18.1, and 21.4 K, respectively. The values of $T_0$ at ambient pressure for the compounds URu$_{2−x}$Fe$_x$Si$_2$ ($x = 0$, $0.025$, $0.05$, $0.10$, $0.15$, and $0.20$) are represented by the six symbols in color that are superimposed on the $T_0$ vs. $x$ phase diagram as displayed in Fig. 1. (The single data point in green at $T = 16.5$ K, which corresponds to the URu$_2$Si$_2$ ($x = 0$) compound, was taken from Ref. 19.)

From the $T_0$ vs. $x$ phase diagram, the evolution of $T_0$ is shown to increase with Fe concentration, $x$, at a constant rate up until $x \approx 0.15$ at which point the rate of increase in $T_0$ with $x$ abruptly increases. The two solid-blue lines, which meet near the critical concentration at $x^* = 0.15$ marks the point at which the compound undergoes a first-order phase transition from the HO phase to the LMAFM phase at ambient pressure. The critical value of Fe concentration, $x^* = 0.15$, was determined in this report to be the smallest concentration of Fe in the URu$_{2−x}$Fe$_x$Si$_2$ system at ambient pressure for which there is a (nearly) homogenous manifestation of the LMAFM phase throughout the sample. Here, we use the symbol $x^*$ to denote the ambient pressure critical concentration of Fe to distinguish it from the symbol $x_c$.
which will be reserved for those “critical” concentrations of Fe that induce the HO → LMAFM phase transition in URu$_{2-x}$Fe$_x$Si$_2$ compounds under pressure. Our determination of $x_c^* = 0.15$ is directly based on measurements of $\rho(T)$ for the URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.025$, 0.05, 0.10, 0.15, and 0.20) system under pressure and will be revisited later in our discussion regarding the effect of pressure on the HO → LMAFM phase transition for the URu$_{2-x}$Fe$_x$Si$_2$ series with $x = 0.025$, 0.05, 0.10, 0.15, and 0.20.

For now, we note that bulk property measurements of electrical resistivity (and also specific heat) performed on the URu$_2$Si$_2$ ($x = 0$) parent compound under applied pressure, $P$, or on the Fe-substituted compounds URu$_{2-x}$Fe$_x$Si$_2$ that are tuned with $x$, do not easily distinguish between the HO and LMAFM phases. The value of the critical concentration, $x_c^* = 0.15$, reported herein as the location of the “kink” in the $T_0$ vs. $x$ phase diagram that marks the first-order phase transition from the HO phase to the LMAFM phase is consistent with values reported for the critical concentration that were determined from other types of measurements. Magnetic neutron diffraction experiments performed on the URu$_{2-x}$Fe$_x$Si$_2$ system for $x \leq 0.7$ reveal an abrupt increase in the uranium magnetic moment at $x = 0.1$. A notable “kink” in the $T_0$ vs. $x$ phase diagram at $x \approx 0.15$ was determined from both the magnetic neutron diffraction experiments and also measurements of specific heat. Optical conductivity experiments performed on the URu$_{2-x}$Fe$_x$Si$_2$ system for $x = 0$, 0.05, 0.10 and 0.3 reveal that at $x = 0.10$ and below $T_0 = 18.5$ K, the compound may exist as a mixture of the HO and LMAFM phases suggesting that a full manifestation of the LMAFM phase occurs for some level of Fe concentration $x > 0.10$.

The reduction in the unit cell volume associated with the substitution of smaller iso-electronic Fe ions for Ru ions in URu$_2$Si$_2$ can be interpreted as the result of “chemical pressure”, $P_{ch}$, which can be determined from a conversion of $x$ to $P_{ch}(x)$. The chemical pressure, $P_{ch}(x)$, was determined from a calculation using the isothermal compressibility, $\kappa_T$, which, in general, relates the reduction in the volume, $V$, to the pressure, $P$, that is applied to a material. In the present context, the “chemical pressure”, $P_{ch}(x)$, is determined from the linear decrease in the unit cell volume, $V$, that occurs with increasing levels of Fe concentration, $x$. The Fe concentration, $x$, that appears in the lower x-axis of Fig. 1 has been converted to $P_{ch}(x)$, which is displayed in the upper y-axis of the same figure. The conversion of $x$ to $P_{ch}(x)$ is discussed in more detail below and will facilitate the discussion regarding the evolution of $T_0$ in the compounds under applied pressure. For now, we note that the value of the slope in the HO phase (for $x \leq 0.15$) is $\partial T_{HO}/\partial P_{ch} = 1.1$ K GPa$^{-1}$. This slope was determined from a linear fit to the $T_0(x)$ data that is represented by the symbols in color at the Fe concentrations of $x = 0$ (green circle), $x = 0.025$ (blue diamond), $x = 0.05$ (red diamond), $x = 0.10$ (black triangle) and $x = 0.15$ (cyan square).

In this study, we are primarily concerned with the results obtained from the measurements of $\rho(T)$ performed under pressure for the five compounds URu$_{2-x}$Fe$_x$Si$_2$ with $x = 0.025$, 0.05, 0.1, 0.15, and 0.2. However, it should be mentioned that in a related study of the URu$_{2-x}$Fe$_x$Si$_2$ system as reported in Ref. 27, bulk property measurements, which included measurements of $\rho(T)$ of single crystals of URu$_{2-x}$Fe$_x$Si$_2$ for $x \leq 0.7$, yielded a critical concentration of $x_c^* \approx 0.10$ at the HO → LMAFM transition and a “kink” in the $T_0$ vs. $x$ phase diagram very similar to the $T_0$ vs. $x$ phase diagram displayed in Fig. 1. However, in that study, the $x$ dependence of the transition temperature $T_{HO}$ (represented by the solid blue line in the HO phase as shown in Fig. 1) for small increases in the Fe concentration up to $x \approx 0.10$ is somewhat smaller than the positive slope reported herein for the $T_0(x)$ data which are represented by the symbols in color at the Fe concentrations of $x = 0$ (green circle), $x = 0.025$ (blue diamond), $x = 0.05$ (red diamond), $x = 0.10$ (black triangle) and $x = 0.15$ (cyan square). The discrepancy in the $x$ dependence of $T_{HO}$ at low concentrations of Fe between these two studies is largely due to the difference between the initial values of $T_{HO} = 16.5$ K and 17.3 K reported for the parent compound URu$_2$Si$_2$ ($x = 0$). While the value of $T_{HO} = 17.3$ K for the ($x = 0$) single crystal was obtained directly from measurements of $\rho(T)$ in Ref. 27, the value of $T_{HO} = 16.5$ K for the ($x = 0$) single crystal was determined from measurements of $\rho(T)$ as reported in Ref. 19.

Since we did not perform measurements of $\rho(T)$ on the parent compound URu$_2$Si$_2$ ($x = 0$) under pressure,
The value of $T_{HO} = 16.5$ K from Ref. 19 was included in the present study in order to provide a reference for the $\rho(T, P)$ measurements on the Fe-substituted single crystals of $\text{URu}_2-x\text{Fe}_2\text{Si}_2$ with $x = 0.025$, 0.05, 0.10, 0.15, and 0.20. The details regarding both the synthesis of the single crystal of $\text{URu}_2\text{Si}_2$ ($x = 0$) in Ref. 19 as well as the experimental conditions for the measurements of $\rho(T)$ of the $\text{URu}_2\text{Si}_2$ ($x = 0$) sample under pressure as reported in Ref. 19 are nearly identical to the method of synthesis and experiments reported herein for the measurements of $\rho(T)$ of the Fe-substituted $\text{URu}_{2-x}\text{Fe}_2\text{Si}_2$ single crystal samples under pressure.

However, the fact that the transition temperature of $T_{HO} = 16.5$ K for the $\text{URu}_2\text{Si}_2$ parent compound at ambient pressure from Ref. 19 is low compared to $T_{HO}$ values reported in other works as shown in Table II deserves some comment. The various values for $T_{HO}$ listed in Table II are based on measurements of $\rho(T)$ for samples of the $\text{URu}_2\text{Si}_2$ parent compound at ambient pressure. The value of $T_{HO}$ may be defined differently from one report to another; for example, in Refs. 19 and 21, $T_{HO}$ is defined to be at the minimum of the anomaly in the $\rho(T)$ curve (as we have defined it in this manuscript); in Refs. 16 and 27, $T_{HO}$ is defined to be at the “inflection point” of the anomaly in the $\rho(T)$ curve, or equivalently, at the minimum in the temperature derivative, $\partial \rho / \partial T$, of the $\rho(T)$ curve; in Ref. 22, $T_{HO}$ is defined to be at the maximum in the anomaly in the $\rho(T)$ curve.

The low value of $T_{HO} = 16.5$ K from Ref. 19 as compared to the other values of $T_{HO} = 17.8$ K,\textsuperscript{16} 17.5 K,\textsuperscript{21} 18.0 K,\textsuperscript{22} and 17.7 K,\textsuperscript{27} (values which are determined from the minimum in the $\rho(T)$ curve), is likely due to issues related to sample quality as well as to extrinsic experimental issues related to differences in the thermometry that can occur during the measurement of $\rho(T)$. Drawing from the conclusions reached in Ref. 29 regarding those issues related to the quality of single crystal samples of $\text{URu}_2\text{Si}_2$, it appears that “small” discrepancies in the value of $T_{HO}$ on the order of $\sim 0.5$ K may result from differences in the residual resistivity ratio ($R RR = \rho(300 \text{ K})/\rho(2 \text{ K})$) that can occur from one sample to another. While high quality samples with larger values of $R RR \sim 100$ tend to yield values of $T_{HO} \sim 17.8$ K (defined at the minimum in the $\rho(T)$ curve), samples with smaller values of $R RR \sim 10$ tend to yield values of $T_{HO} \sim 17.3$ K.\textsuperscript{29} Values of $R RR$ that are listed in Table II indicate that the lowest value of $R RR = 7.2$ corresponds to the lowest value of the $T_{HO} = 16.5$ K,\textsuperscript{19} while the largest value of $R RR = 100$ corresponds to the highest value of the $T_{HO} = 18.0$ K.\textsuperscript{22}

The $R RR$ values as a function of pressure, $P$, are displayed in Fig. 2 for both the single crystal samples of the Fe-substituted $\text{URu}_{2-x}\text{Fe}_2\text{Si}_2$ compounds with $x = 0.025$, 0.05, 0.10, 0.15, and 0.20 reported herein as well as the single crystal sample of $\text{URu}_2\text{Si}_2$ ($x = 0$) from Ref. 19.\textsuperscript{19,28} For each compound, there is a slight overall increase in the $R RR$ value with an increase in pressure, $P$: for the $x = 0$ compound, there is an increase in the $R RR$ value from 7.2 at 0 GPa to 8.5 at 2.4 GPa; for the $x = 0.025$ compound, there is an increase in the $R RR$ value from 8.0 at 0 GPa to 9.0 at 2.1 GPa; for the $x = 0.05$ compound, there is an increase in the $R RR$ value from 4.8 at 0 GPa to 5.5 at 2.2 GPa; for the $x = 0.10$ compound, there is an increase in the $R RR$ value from 4.6 at 0 GPa to 5.4 at 2.1 GPa; for the $x = 0.15$ compound, there is an increase in the $R RR$ value from 6.0 at 0 GPa to 7.7 at 2.2 GPa; and for the $x = 0.20$ compound, there is an increase in the $R RR$ value from 4.8 at 0 GPa to 4.9 at 2.2 GPa. We briefly note that there is a large amount of anisotropy observed in the room temperature electrical resistivity for single crystals of the $\text{URu}_2\text{Si}_2$ ($x = 0$) parent compound such that $\rho(300 \text{ K})$ is roughly two times larger for a measurement of $\rho(T)$ parallel to the $a$-axis (or basal plane) when compared to a measurement with the current parallel to the $c$-axis.\textsuperscript{30} Hence, the $R RR$ value is sensitive to the orientation of the crystal during the measurement of $\rho(T)$. The $R RR$ values as a function of pressure that are reported in Ref. 28 (which are based on the measurements of $\rho(T)$ for the $\text{URu}_2\text{Si}_2$ ($x = 0$) sample from Ref. 19) were determined from measurements of $\rho(T)$ with the current parallel to the $a$-axis. The quality and orientation of the $\text{URu}_2\text{Si}_2$ ($x = 0$) sample from Ref. 19 were determined from Laue X-ray diffraction patterns.

As displayed in Fig. 2, there is also an overall reduction in the $R RR$ value with increasing Fe concentration from $x = 0.025$ to 0.20. However, there is a jump in the $R RR$ value at $x = 0.15$ which is due to the drop in the overall nominal value of the electrical resistivity, $\rho(T)$, for the sample with $x = 0.15$ relative to samples with neighboring concentrations of $x = 0.10$ and 0.20 as displayed in the inset of Fig. 2. Sudden shifts in the nominal values of the electrical resistivity, $\rho(T)$, may occur during measurements of $\rho(T)$ in a hydrostatic piston-cylinder cell that can affect the electrical contacts between the platinum wire leads that are affixed with silver epoxy to the gold-sputtered samples. The unintended and not entirely understood effects on sample contacts in a four-wire electrical resistivity configuration that result from repeated thermal cycling between $T = 300$ K and 1 K while under applied pressure in a quasi-hydrostatic pressure medium (of isoamyl-alcohol and n-pentane) that freezes at $T \approx 100$ K may cause shifts of the nominal value of $\rho(T)$ from one measurement to the next.

Although the $R RR = 7.2$ and $T_{HO} = 16.5$ K values for the single crystal sample of $\text{URu}_2\text{Si}_2$ from Ref. 19 are low, it is important to note that both the critical pressure, $P_c = 1.5$ GPa, and the pressure dependence in the HO phase, $\partial T_{HO}/\partial P = 1.0$ K GPa$^{-1}$, as determined from measurements of $\rho(T)$ for the $\text{URu}_2\text{Si}_2$ ($x = 0$) sample under pressure in Ref. 19 are consistent with other reports\textsuperscript{14-18,20,21} (as shown in Table I). The importance of this point will be addressed below when discussing the additive behavior of chemical pressure, $P_{ch}$, and applied critical pressure, $P_c$, such that $P_{ch}(x_c) + P_c \approx 1.5$ GPa for the $\text{URu}_{2-x}\text{Fe}_2\text{Si}_2$ system.
TABLE II. Values of the PM→HO phase transition temperature ($T_{HO}$) and the residual resistivity ratio (RRR) at ambient pressure along with the pressure dependence ($\partial T_{HO}/\partial P$) in the HO phase (if applicable), for samples of the URu$_2$Si$_2$ ($x = 0$) parent compound as reported in various works.

<table>
<thead>
<tr>
<th>Year</th>
<th>Reference</th>
<th>RRR</th>
<th>Crystal</th>
<th>$T_{HO}$</th>
<th>$\partial T_{HO}/\partial P$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1987</td>
<td>McElfresh</td>
<td>37.5</td>
<td>polycrystal</td>
<td>17.4$^\dagger$ (17.8°)</td>
<td>1.3 ± 0.1</td>
</tr>
<tr>
<td>2007</td>
<td>Jeffries</td>
<td>7.2</td>
<td>single crystal</td>
<td>16.5$^\dagger$</td>
<td>1.0 ± 0.1</td>
</tr>
<tr>
<td>2010</td>
<td>Butch</td>
<td>22.5</td>
<td>single crystal</td>
<td>17.5$^\dagger$</td>
<td>1.3 ± 0.1</td>
</tr>
<tr>
<td>2011</td>
<td>Kanchanavate</td>
<td>100</td>
<td>polycrystal</td>
<td>17.5$^\dagger$ (18.0°)</td>
<td>–</td>
</tr>
<tr>
<td>2015</td>
<td>Ran$^\ddagger$</td>
<td>11.9</td>
<td>single crystal</td>
<td>17.3$^\dagger$ (17.7°)</td>
<td>–</td>
</tr>
</tbody>
</table>

$^*$ $T_{HO}$ defined at minimum in $\rho(T)$  
$^\dagger$ $T_{HO}$ defined at inflection point in $\rho(T)$ or at minimum in $d\rho/dT$  
$^\ddagger$ $T_{HO}$ defined at maximum in $\rho(T)$

**Enhancement of $T_0$ with pressure**

Measurements of electrical resistivity $\rho(T)$ were performed under pressure for each of the compounds URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.025, 0.05, 0.10, 0.15, and 0.20$). The temperature dependence of $\rho(T)$ at various pressures up to $P \sim 2.2$ GPa in the region near $T_0$ for the URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.025$ and $0.20$) compounds is shown in Fig. 3 (a) and (b), respectively. In the interest of space and to avoid redundancy, data for only two ($x = 0.025$ and $0.20$) of the five ($x = 0.025, 0.05, 0.10, 0.15$, and $0.20$) compounds investigated in this report are shown in Fig. 3. The compound with $x = 0.025$, which has the smallest Fe concentration studied, is an example of a URu$_{2-x}$Fe$_x$Si$_2$ compound in the HO phase (for $T \leq T_0$) at ambient pressure that undergoes a first-order phase transition to the LMAFM phase as pressure is increased; the compound with $x = 0.20$, which is the largest Fe concentration in this study, represents an example of a URu$_{2-x}$Fe$_x$Si$_2$ compound that is already in the LMAFM phase (for $T \leq T_0$) at ambient pressure and remains in that phase as pressure is increased up to the maximum pressure reached in this experiment at $P \sim 2.2$ GPa.

The curves for $\rho(T)$ in both Fig. 3 (a) and (b) have been shifted vertically in order to better illustrate the behavior of the HO/LMAFM transition temperature $T_0$ as pressure is increased. The enhancement of $T_0$ can be seen in the shift of the feature in $\rho(T)$ to higher values of temperature as pressure is increased. Electrical resistivity ($\rho$ vs. $T$) curves for the compounds with Fe concentrations of $x = 0.05, 0.10,$ and $0.15$ are not shown but exhibit similar behavior under applied pressure. Measurements of $\rho(T)$ under applied pressure for the two URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.05, 0.15$) compounds were also performed upon decreasing the pressure in the cell. The pressure was released by unloading the cell stepwise from $\sim 2.1$ GPa to 0 GPa so that additional measurements of $\rho(T)$ could be performed for the purpose of confirming the reversibility of certain portions of the $T_0$ vs. $P$ phase boundary. We briefly comment here that the results obtained for the evolution of $T_0$ with increasing pressure were completely reversible upon the release of pressure which can be seen in the $T_0(P)$ data (open symbols) for the $x = 0$, 0.05, and 0.15 compounds in the $T_0$ vs. $P$ phase boundary displayed in Fig. 4.

**The suppression of the critical pressure $P_c$ with increasing Fe substitution**

A composite plot of the $T_0$ vs. $P$ phase boundaries for various values of $x$ is displayed in Fig. 4. The $T_0$ vs. $P$ phase boundaries were constructed from features in the $\rho(T)$ curves for the URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0, 0.025, 0.05, 0.10, 0.15,$ and $0.20$) compounds under pressure, as illustrated in Fig. 3. (The green data points corresponding to the URu$_2$Si$_2$ ($x = 0$) compound were taken from
a previous study of URu$_2$Si$_2$ under pressure by Jeffries et al. in Ref. 19.) The most striking aspect of the $T_0$ vs. $P$ phase boundaries is the “shift” of the “kink” in the $T_0(P)$ data to lower values of pressure, $P$, in those URu$_{2-x}$Fe$_x$Si$_2$ compounds with higher concentrations of Fe, $x$. The “kink” in the $T_0(P)$ data signifies the first-order phase transition from the HO phase to the LMAFM phase occurring in the URu$_{2-x}$Fe$_x$Si$_2$ compounds with $x = 0, 0.025, 0.05,$ and 0.10. The HO → LMAFM phase transition occurs at a critical pressure, $P_c$, which is defined as the pressure at the location of the discontinuity in the slope, $\partial T_0/\partial P$, i.e., at the “kink” in the $T_0(P)$ data.

We identify the “kink” in the $T_0$ vs. $P$ phase boundary as the location of a tricritical point at which point the phase boundary between the HO and LMAFM phases (for $T \leq T_0$) joins with the $T_{HO}(P)$ phase boundary between the PM and HO phases and the $T_N(P)$ phase boundary between the PM phase and high pressure LMAFM phase. In this report, we were not able to witness any additional features in the electrical resistivity (below $T_{HO}$) that would allow for the determination of the HO → LMAFM phase boundary.

According to theoretical models, it is possible that there may exist a critical end point on the HO → LMAFM phase boundary, rather than a tricritical point, if the two ordered phases HO and LMAFM (below $T_0$) exhibit the same antiferromagnetic symmetry. However, in such a case, the $T_0(P)$ phase boundary between the PM and HO/LMAFM phases would be smooth and absent of a “kink.” Recently, neutron scattering experiments reveal that the magnetic and lattice excitations below $T_{HO}$ in the HO phase do not share the broken symmetries observed in the PM → LMAFM phase transition at $T_N$. To the contrary, the excitations in the HO phase appear to reflect the symmetry observed in the PM phase above $T_{HO}$. We suggest that the suppression of the “kink” with increasing $x$ in URu$_{2-x}$Fe$_x$Si$_2$ as observed in the $T_0(P)$ boundary provides additional evidence for the existence of a true tricritical point in the $T_0$ vs. $P$ phase diagram in which the ordering in the HO phase exhibits a different symmetry than the ordering in the LMAFM phase.

From the composite plot of the $T_0$ vs. $P$ phase boundaries for various values of $x$ shown in Fig. 4, it is clear that there is a complete suppression of the HO phase in favor of the LMAFM phase with increasing Fe concentration, $x$, such that the critical pressure decreases from $P_c = 1.5$ GPa at $x = 0$ to $P_c = 0$ GPa at $x = 0.15$. The shift of this tricritical point to lower critical pressure, $P_c$, in compounds with higher concentrations of Fe, $x$, is perhaps the defining and most interesting result of this report and suggests a simple additive relation between chemical pressure, $P_{ch}$, and applied pressure, $P$, as tuning parameters for investigating the ordered phases in the URu$_{2-x}$Fe$_x$Si$_2$ system.

Referring back to the $T_0$ vs. $P$ phase boundary shown in Fig. 1, it is apparent that the URu$_{2-x}$Fe$_x$Si$_2$ compounds, for which $x \geq 0.15$, have already entered the LMAFM phase; in particular, for the URu$_{2-x}$Fe$_x$Si$_2$ compound with $x = 0.15$, the value of the critical pressure is $P_c = 0$ GPa. Hence, there are no observable kinks in the $T_0(P)$ data shown in Fig. 4 for the URu$_{2-x}$Fe$_x$Si$_2$ compounds with $x = 0.15$ and 0.20. However, there are clear discontinuities in the slope $\partial T_0/\partial P$ for URu$_{2-x}$Fe$_x$Si$_2$ compounds with $x = 0, 0.025, 0.05,$ and 0.10. The HO → LMAFM phase transition occurs at a critical pressure, $P_c$, which is defined as the pressure at the location of the discontinuity in the slope, $\partial T_0/\partial P$, i.e., at the “kink” in the $T_0(P)$ data.

The discontinuity in $\partial T_0/\partial P$ for the URu$_{2-x}$Fe$_x$Si$_2$ compound with $x = 0.10$ is more difficult to identify and requires some comment. First, there is a smaller difference between the slope $\partial T_{HO}/\partial P = 2.06$ K GPa$^{-1}$ in the HO phase when compared to the slope $\partial T_N/\partial P = 2.42$ K GPa$^{-1}$ in the LMAFM phase; one explanation for the elevated value of $\partial T_{HO}/\partial P$ for the URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.10$) compound is that, below $T_{HO}$, the sample may consist of a mixture of the HO and LMAFM phases.
neutron scattering experiments\textsuperscript{12} performed on the parent compound URu$_2$Si$_2$ under pressure revealed a phase separated spatial inhomogeneity in which the HO phase was populated with regions of LMAFM phase. Elastic neutron scattering measurements performed on single crystals of URu$_{2-x}$Fe$_x$Si$_2$ prepared in our lab reveal an increase in the U ordered moment with increasing $x$ in the HO phase, consistent with a scenario in which regions of the LMAFM phase coexist with the HO phase.\textsuperscript{23} More recently, muon spin rotation ($\mu$SR) measurements on samples of URu$_{2-x}$Fe$_x$Si$_2$ prepared in another laboratory demonstrated that the HO phase contains phase separated regions of the LMAFM phase in URu$_{2-x}$Fe$_x$Si$_2$ compounds with low levels of Fe concentrations.\textsuperscript{30} Second, there is an absence of data in the region where $P_c$ is likely to occur. Hence, $P_c \approx 0.57$ GPa was determined from the intersection of the line of fit to the $T_0(P)$ data in the HO phase (the first four black triangles as shown in Fig. 4) with the line of fit to the $T_0(P)$ data in the LMAFM phase (the remaining seven black triangles as shown in Fig. 4). For ease of comparison, the values of $P_c$ and $\partial T_0/\partial P$ for the URu$_{2-x}$Fe$_x$Si$_2$ compounds ($x = 0.025, 0.05, 0.10, 0.15,$ and $0.20$) are presented in Table III. Note that the slope ($\partial T_0/\partial P$) in the LMAFM phase is approximately 2.5 times larger than the slope ($\partial T_0/\partial P$) in the HO phase (with the exception of the URu$_{2-x}$Fe$_x$Si$_2$ compound with $x = 0.10$).

It is apparent from the slope ($\partial T_0/\partial P$) in the HO and LMAFM phases, that the URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.10$) compound is not fully expressed in the LMAFM phase at ambient pressure. Hence, of the five compounds URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0.0, 0.025, 0.05, 0.10, 0.15,$ and $0.20$) that were measured in this study, we determined that $x = 0.15$ is the smallest concentration of Fe in which the URu$_{2-x}$Fe$_x$Si$_2$ system is completely in the LMAFM phase, at ambient pressure. The measured value of $x_c^*$ = 0.15 for the critical concentration of Fe is close to the estimated value of $x_c^*$ that was determined from the location of the “kink” in the the $T_0$ vs. $x$ phase boundary shown in Fig. 1. Further analysis in the last section of this paper shows that $x = 0.15$ is a reasonable determination of the ambient pressure critical concentration of Fe, $x_c^*$.

\section*{Pressure dependence of the energy gap $\Delta$}

The first bulk property measurements of the specific heat and electrical resistivity for the parent compound URu$_2$Si$_2$ ($x = 0$) at ambient pressure suggest that the second-order mean-field-like transition from the paramagnetic (PM) phase to the HO/LMAFM phase results in the opening of a charge gap ($\Delta$) over a portion of the Fermi surface.\textsuperscript{1,2,30} Originally, the gapped portion of the Fermi surface for URu$_2$Si$_2$ at ambient pressure had been attributed to the formation of a static charge- or spin-density wave (CDW or SDW) below $T_0 \sim 17.5$ K while the remaining non-gapped portion of the Fermi surface was thought to be available to superconducting electron states at $T_c \sim 1.5$ K.\textsuperscript{30} In this scenario, the coexistence of superconductivity and HO could be thought of as ordered phases that compete for Fermi surface fraction.\textsuperscript{2,19,36} It is now known that the superconductivity is suppressed with chemical substitution $x$ or applied pressure $P$, and is eventually destroyed during the first-order phase transition from the HO phase to the LMAFM phase.\textsuperscript{2,16,37} Additional investigations of the parent compound URu$_2$Si$_2$ ($x = 0$) employing various experimental probes such as infrared spectroscopy,\textsuperscript{38} Hall effect,\textsuperscript{39-41} quantum oscillation measurements,\textsuperscript{42} angle-resolved photoemission spectroscopy (ARPES),\textsuperscript{43,44} optical conductivity,\textsuperscript{38,45,46} and scanning tunneling microscopy (STM),\textsuperscript{47,48} confirm a reorganization of the electronic structure below $T_0$ which results in a partial gapping of the Fermi surface.

Surprisingly, experiments under pressure, including de Haas-van Alphen (dHvA),\textsuperscript{20,49} Shubnikov-de Haas (SdH),\textsuperscript{50} and inelastic neutron scattering experiments,\textsuperscript{51} reveal that there is no significant change in the gapped structure of the Fermi surface as the URu$_2$Si$_2$ compound undergoes a first-order transition from the HO phase to the LMAFM phase. The electronic reconstruction and partial gapping of the Fermi surface for $T < T_{HO}$ has become one of the more salient features of the HO phase. Although there is still no consensus on a physical explanation for the gapping of the Fermi surface below $T_{HO}$, a significant amount of experimental work has provided detailed information regarding the electronic structure above and below $T_0$ in URu$_2$Si$_2$.

The formation of a similar energy gap ($\Delta$) over the Fermi surface in the Fe-substituted compounds URu$_{2-x}$(Fe)$_x$Si$_2$ is evident from recent measurements of electrical resistivity $\rho(T)$, specific heat $C(T)$,\textsuperscript{22,23,52} and optical conductivity\textsuperscript{24} experiments. Here, we report on values for $\Delta$ at the Fermi surface in the HO/LMAFM phase as a function of pressure for the various Fe-substituted compounds URu$_{2-x}$(Fe)$_x$Si$_2$ ($x = 0.025, 0.05, 0.10, 0.15,$ and $0.20$). The values of $\Delta$ were extracted from fits of the $\rho(T)$ data in the temperature region $T < T_0$ to the expression for electrical resistivity, $\rho(T)$.\textsuperscript{25}

\begin{table}[h]
\centering
\caption{Values of the applied critical pressure $P_c$ (for various levels of Fe concentration, $x$, in the URu$_{2-x}$(Fe)$_x$Si$_2$ system) at the HO $\rightarrow$ LMAFM phase transition along with the pressure dependence ($\partial T_0/\partial P$) in the HO and LMAFM phases.}
\begin{tabular}{|c|c|c|c|}
\hline
$x$ & $P_c$ (GPa) & $\partial T_0/\partial P$ (K GPa$^{-1}$) \\
\hline
0 (from Ref. 19) & 1.50 & 1.0 $\pm$ 0.1 & 2.5 $\pm$ 0.1 \\
0.025 & 1.17 & 1.0 $\pm$ 0.1 & 2.7 $\pm$ 0.1 \\
0.05 & 0.85 & 1.1 $\pm$ 0.1 & 2.7 $\pm$ 0.1 \\
0.10 & 0.57 & 2.1 $\pm$ 0.1 & 3.4 $\pm$ 0.1 \\
0.15 & 0.00 & - & 2.5 $\pm$ 0.1 \\
0.20 & - & - & 2.1 $\pm$ 0.1 \\
\hline
\end{tabular}
\end{table}
\[ \rho(T) = \rho_0 + AT^2 + B\Delta^2 \sqrt{\frac{T}{\Delta}} \times \left[ 1 + \frac{2}{3} \left( \frac{T}{\Delta} \right) + \frac{2}{15} \left( \frac{T}{\Delta} \right)^2 \right] e^{-\frac{T}{\Delta}}. \] (1)

The exponential term in Equation (1) is the dominant contribution to the electrical resistivity \( \rho(T) \) in this temperature region \( (T < T_0) \) and represents the scattering contribution from the gapped spin excitations that are characteristic of antiferromagnetic ordering.\(^{25}\) Other scattering contributions to \( \rho(T) \) in this temperature region include the residual resistivity \( \rho_0 \) and the scattering associated with electron-electron interactions that are characteristic of a Fermi liquid, \( AT^2 \). We briefly note that earlier reports on measurements of \( \rho(T) \) for high purity URu\( _2 \)Si\( _2 \) \( (x = 0) \) samples with values of RRR \( \sim 100 \) suggest that the \( AT^2 \) dependence fails to describe the HO phase which is better described by a power law behavior of \( AT^\alpha \), where \( \alpha = 1.6^{,29,53} \) Furthermore, a power law exponent \( AT^\alpha \), where \( \alpha < 2 \), is typically required to represent the electrical resistivity \( \rho(T) \) in the temperature range just above \( T_c^{,20} \) However, Fermi liquid behavior is assumed to be valid in the low temperature region down to 2 K in this study owing to substantial levels of Fe solutes present in single crystal samples from the URu\(_2-x\)Fe\(_x\)Si\(_2\) system with \( x = 0.025, 0.05, 0.10, 0.15, \) and 0.20 which exhibited low values of RRR \( < 10 \) (see Fig. 2). In addition, there was no evidence for the onset of superconductivity in the \( \rho(T) \) curves down to 1 K for any of the Fe-substituted URu\(_2-x\)Fe\(_x\)Si\(_2\) \( (x = 0.025, 0.05, 0.10, 0.15, \) \( \) and 0.20) samples measured in this study. Hence, while a \( T^{1.6} \) dependence is likely to apply at low temperatures near 2 K for high-purity parent compounds that exhibit superconductivity with a \( T_c \sim 1.5K \) or below, we note that a \( T^2 \) dependence is applicable down to \( T \sim 2 \) K for the “dirty” Fe-substituted compounds measured in this study. The assumption of the existence of Fermi liquid behavior below \( T_0 \) in our samples of URu\(_2-x\)Fe\(_x\)Si\(_2\) \( (x = 0.025, 0.05, 0.10, 0.15, \) \( \) and 0.20) is supported by the recent optical conductivity experiments performed on URu\(_2-x\)Fe\(_x\)Si\(_2\) compounds which demonstrated that scattering processes typical of a Fermi liquid were present in both the HO and LMAFM phases.\(^{24}\)

Examples of curves that were fitted to the \( \rho(T) \) data based on Equation (1) for the two URu\(_2-x\)Fe\(_x\)Si\(_2\) \( (x = 0.025 \) and 0.20) compounds at 0.80 and 0.77 GPa, respectively, are displayed in Fig. 5. The \( \rho(T) \) data and the fitted curve (dashed black line) in Fig. 5 (a) correspond to the URu\(_2-x\)Fe\(_x\)Si\(_2\) \( (x = 0.025) \) compound under pressure at \( P = 0.80 \) GPa which is an example of a URu\(_2-x\)Fe\(_x\)Si\(_2\) compound that is in the HO phase for the temperature region of the fit. Similarly, the \( \rho(T) \) data and the fitted curve (dashed black line) in Fig. 5 (b) correspond to the URu\(_2-x\)Fe\(_x\)Si\(_2\) \( (x = 0.2) \) compound under pressure at \( P = 0.77 \) GPa which is an example of a URu\(_2-x\)Fe\(_x\)Si\(_2\) compound that is in the LMAFM phase for the temperature region of the fit.

The fit of Equation (1) to the \( \rho(T) \) data was performed over the temperature range from \( T = 2 \) K to \( T_{\text{max}} \), where \( T_{\text{max}} \) represents the upper bound on the temperature range of the \( \rho(T) \) data used for the fit. The value of \( T_{\text{max}} \) was allowed to vary in order to determine the region of data (below \( T_0 \)) that yielded the best fit. Hence, the best fit of the \( \rho(T) \) data to Equation (1) was determined by plotting Chi-squared vs. \( T_{\text{max}} \) (as displayed in the inset of Fig. 5 (a)). From the Chi-squared vs. \( T_{\text{max}} \) plot, the determination of \( T_{\text{max}} \) that yielded the best fit to the \( \rho(T) \) data is the value of \( T_{\text{max}} \) at which Chi-squared is maximized. In the case of the \( \rho(T) \) data displayed in Fig. 5 (a), the upper bound in temperature corresponding to the best fit was determined to be \( T = 16.2 \) K. In the case of the \( \rho(T) \) data displayed in Fig. 5

![FIG. 5. (Color online) Low temperature electrical resistivity \( \rho(T) \) for the (a) \( x = 0.025 \) compound at 0.8 GPa and (b) \( x = 0.20 \) compound at 0.77 GPa plotted along with a theoretical model for the resistivity (see Equation (1) in text), represented by the dashed black curves. The best fit to the \( \rho(T) \) data below \( T_0 \) was determined by a fitting algorithm as described in the text that allowed for the extraction of the value of the energy gap \( \Delta \) at various pressures \( P \).Insets: Examples of a plot of Chi-squared vs. \( T_{\text{max}} \) showing the maximum value of Chi-squared that determined the best fit to the \( \rho(T) \) data below \( T_0 \).](image-url)
are denoted by the dashed vertical lines. The gray rectangle phase transition from the HO phase to the LMAFM phase the best fit was determined to be \( (b) \), the upper bound in temperature corresponding to the data points shown for \( x = 0 \) and \( x = 0.05 \), (b) were determined from the \( x = 0.15 \), and \( x = 0.20 \). The value of \( \Delta \) was determined by fitting a theoretical model of electrical resistivity \( \rho(T) \) (see Equation (1) in text) to the low temperature electrical resistivity \( \rho(T) \) data. The critical pressure \( P_c \) and the occurrence of the first-order phase transition from the HO phase to the LMAFM phase are denoted by the dashed vertical lines. The gray rectangle in panel (b) is the error associated in determining the value of \( P_c \) for the \( x = 0.1 \) sample (see text). The values of \( P_c \) were determined from the \( T_0 \) vs. \( P \) phase boundaries shown in Fig. 4. Error bars were determined by the fitting algorithm and the solid curved lines are guides to the eye. (The green data points shown for \( x = 0 \) were taken from Ref. 19.)

The pressure dependence \( \langle \partial \Delta / \partial P \rangle \) of the extracted values of the charge gap \( \Delta \) based on the fits of Equation (1) to the \( \rho(T) \) data for the URu\(_{2-x}\)Fe\(_x\)Si\(_2\) \( (x = 0, 0.025, 0.05, 0.10, 0.15, \) and \( 0.20 \)) compounds is displayed in Fig. 6. The \( \Delta(P) \) behavior for the URu\(_2\)Si\(_2\) parent compound from Ref. 19, as displayed in Fig. 6 (a), was determined in a similar fashion; however, a slightly different theoretical model for the electrical resistivity that is based on scattering from gapped ferromagnetic (rather than antiferromagnetic) spin excitations\(^{54,55} \) was used in the fit to the \( \rho(T) \) data in Ref. 19. The differences in the magnitude of \( \Delta \) extracted from the two different theoretical models of \( \rho(T) \) are small and the overall qualitative behavior of the pressure dependence of the gap was shown to be unaffected.\(^{22} \)

The behavior of \( \Delta(P) \) for the six compounds URu\(_{2-x}\)Fe\(_x\)Si\(_2\) \( (x = 0, 0.025, 0.05, 0.10, 0.15, \) and \( 0.20 \)) have been grouped and plotted in pairs: \( x = 0, 0.025 \), (0.05, 0.10), and (0.15, 0.20) are displayed in Fig. 6 (a), (b), and (c), respectively. The dashed vertical lines in Fig. 6 indicate locations of the critical pressure, \( P_c \), in the \( \Delta(P) \) plots and were determined from the \( T_0 vs. P \) phase diagram displayed in Fig. 4. For the URu\(_{2-x}\)Fe\(_x\)Si\(_2\) \( (x = 0.15) \) compound, the vertical dashed line corresponding to a critical pressure at \( P_c = 0 \) GPa has been omitted. There is a noticeable change in the pressure dependence \( \langle \partial \Delta / \partial P \rangle \) of the gap \( \Delta \) at \( P \sim 1.2, 0.8, \) and 0.6 GPa for the URu\(_{2-x}\)Fe\(_x\)Si\(_2\) compounds with \( x = 0.025, 0.05, \) and 0.10, respectively. These values of pressure are remarkably consistent with the critical pressures, \( P_c = 1.17, 0.85, \) and 0.57 GPa (represented by the vertical dashed lines in Fig. 6). As determined in Ref. 19, there is also a turnaround in the \( \Delta(P) \) behavior for the URu\(_2\)Si\(_2\) parent compound (green circles in Fig. 6 (a)). The turnaround (or the minimum in \( \Delta(P) \)) occurs at a pressure of \( P \sim 1.3 \) GPa, which is slightly lower than the critical pressure, \( P_c = 1.5 \) GPa. The pressure dependence of the gap \( \langle \partial \Delta / \partial P \rangle \) for the two URu\(_{2-x}\)Fe\(_x\)Si\(_2\) compounds with \( x = 0.15 \) and 0.20, both of which already exhibit the LMAFM phase at ambient pressure, is displayed in Fig. 6 (c). For these two compounds, there is no non-zero critical pressure, \( P_c \), and hence there is only the monotonic dependence \( \langle \partial \Delta / \partial P \rangle > 0 \) for \( P > 0 \) observed in \( \Delta(P) \). The monotonic dependence of \( \Delta(P) \) is consistent with the monotonic behavior of \( \Delta(P) \) above the critical pressure in the LMAFM phase observed for the other four compounds URu\(_{2-x}\)Fe\(_x\)Si\(_2\) \( (x = 0, 0.025, 0.05, 0.10) \). (For ease of comparison, there is a correspondence in the color scheme (with regard to Fe concentration, \( x \)) between the \( \Delta(P) \) curves displayed in Fig. 6 and the \( T_0(P) \) data in the plot of the \( T_0 vs. P \) phase boundaries displayed in Fig. 4.)

From the plots of \( \Delta(P) \) that are displayed in Fig. 6 for the six compounds URu\(_{2-x}\)Fe\(_x\)Si\(_2\) \( (x = 0, 0.025, 0.05, 0.10, 0.15, \) and \( 0.20 \)), there is a clear qualitative difference in the \( \Delta(P) \) behavior observed in the (low pressure) HO phase, where \( \partial \Delta / \partial P < 0 \), and the (high pressure) LMAFM phase, where there is a positive pressure coefficient, \( \partial \Delta / \partial P > 0 \). This behavior seems to be consistent with previous reports on the evolution of the energy gap, \( \Delta \), with increases in either \( x \) or \( P \). Electrical resistivity measurements performed in an earlier study of single crystals of the parent compound URu\(_2\)Si\(_2\) \( (x = 0) \) under
pressure reveal a monotonic decrease in the Fermi surface gap $\Delta$ from $\sim 77$ to 70 K in the HO phase followed by a jump in $\Delta$ to a saturated value of $\sim 100$ K in the LMAFM phase at $P_x \sim 0.5$ GPa. Similar behavior was observed in the evolution of the energy gap, $\Delta$, as a function of Fe concentration, $x$. The values for $\Delta$ were extracted from a theoretical fit to the specific heat data, $C(T)$, for single crystal samples of URu$_{2-x}$Fe$_x$Si$_2$ in which there is a slight suppression of $\Delta$ from 93 K down to 80 K in the HO phase for increasing Fe concentration up to $x \sim 0.10$ at which point there is a jump in the value of $\Delta$ to $\sim 110$ K in the LMAFM phase.

It is interesting to note that as the temperature is lowered below $T_0$, the commensurate $Q_0 = (1,0,0)$ and incommensurate $Q_1 = (1 \pm 0.4,0,0)$ spin excitations in the parent compound URu$_2$Si$_2$, exhibit well defined peaks in the energy spectrum which are gapped at energies below $\sim 2$ meV (or 22 K) and $\sim 4.5$ meV (or 50 K), respectively. It has been suggested that below $T_0$, the two spin excitations at $Q_0$ and $Q_1$ are strongly coupled to the charge degree of freedom, suggesting that there is a fundamental relationship between the SDW gaps at $Q_0$ and $Q_1$ and the charge (CDW) gap $\Delta$ that opens up over the Fermi surface.

FIG. 7. (Color online) Plot of the $T_0$ vs. $P$ phase boundaries for various values of $x$ for URu$_{2-x}$Fe$_x$Si$_2$ ($x = 0$, 0.025, 0.05, 0.10, 0.15, and 0.20). The vertical dashed lines drop down to the $x$-$P$ plane at the respective critical pressures $P_c$ and help determine the boundary between the HO and LMAFM phases in the $x$-$P$ plane. The HO/LMAFM phase boundary is represented by the solid black line in the $x$-$P$ plane which is a linear fit of the black circles. At the critical concentration of $x_\text{c} = 0.15$ and above, the compounds have already entered the LMAFM phase at ambient pressure. (The green data points shown for $x = 0$ are not part of this study and were taken from an earlier study of URu$_{2-x}$Fe$_x$Si$_2$ under pressure by Jeffries et al. in Ref. 19.)

The two different spin excitation gaps, $\Delta_0$ (commensurate) and $\Delta_1$ (incommensurate), are further distinguished in their behavior under applied pressure. While increasing applied pressure has the effect of increasing the energy gap $\Delta_1$, it has the opposite effect of decreasing the energy gap $\Delta_0$. The $\Delta_0$ behavior was confirmed in a later report where the energy gap $\Delta_0$ for the commensurate excitation at $Q_0$ was observed to monotonically decrease with increasing pressure up to the critical pressure, $P_c$, at which point the $Q_0$ excitation completely disappears at the HO $\rightarrow$ LMAFM phase transition. In contrast, the $\Delta_1$ energy gap was observed to increase with pressure in the HO phase below $P_c$ and then survive the first-order HO $\rightarrow$ LMAFM transition at which point $\Delta_1$ jumps discontinuously to a larger value and then remains constant with increasing pressure into the LMAFM phase. Here, we simply note the correspondence in the HO phase between the behavior of $\Delta_0(P)$ and the behavior we obtained for $\Delta_1(P)$ as well as the correspondence in the LMAFM phase between the behavior of $\Delta_1(P)$ and the behavior we observed for $\Delta_0(P)$.

Our determination of the Fermi surface gap, $\Delta$, which is based on a theoretical model of electrical resistivity fitted to the $\rho(T)$ data from bulk transport measurements under pressure surely cannot capture all of the subtle details that are becoming known regarding the response of the Fermi surface to experimental tuning at temperatures below $T_0$. Nevertheless, it appears that such a theoretical model of electrical resistivity used in the analysis of bulk measurements of $\rho(T)$ may still capture some of the important features of the charge gap ($\Delta$) that are observed in more direct measurements of the Fermi surface in the HO and LMAFM phases. Namely, the gap analysis performed here seems to capture the differences in the pressure variation, $\partial \Delta/\partial P$, for the HO and LMAFM phases and also finds the magnitude of the charge gap, $\Delta$, to be consistent with previous reports.

The simultaneous tuning of URu$_2$Si$_2$ with chemical and applied pressure

Perhaps the most interesting aspect of this report is the systematic and predictable manner in which Fe substitution, $x$, combines with applied pressure, $P$, to affect the ordered phases and phase transitions observed in URu$_2$Si$_2$. The three-dimensional plot of the $T_0$ vs. $P$ phase boundaries for various values of $x$ displayed in Fig. 7 summarizes the response of the transition temperature, $T_0$, to the simultaneous tuning of the URu$_2$Si$_2$...
compound with Fe substitution at $x = 0.025, 0.05, 0.1, 0.15$, and 0.2 while under applied pressure up to $P = 2.2$ GPa. The sloped dashed black lines are linear fits to the six sets of $T_0(P)$ data for the URu$_{2-x}$Fe$_x$Si$_2$ compounds at various Fe concentrations, $x = 0, 0.025, 0.05, 0.10, 0.15$, and 0.20. (The $x = 0$ data were taken from Ref. 19.) There are obvious “kinks” in the $T_0(P)$ data for the URu$_{2-x}$Fe$_x$Si$_2$ compounds with $x \leq 0.10$. The “kinks” correspond to the discontinuities in the slope, $\partial T_0/\partial P$, which mark the first-order HO $\rightarrow$ LMAFM phase transition. The slopes, $\partial T_0/\partial P$, of the various $T_0(P)$ curves are presented in Table III along with the values of the critical pressure, $P_c$, for the respective first-order HO $\rightarrow$ LMAFM phase transitions.

The filled black circles in the $x$-$P$ plane are the planar projections of the “kinks” (or points of discontinuity in $\partial T_0/\partial P$) that appear in the $T_0(P)$ data (filled colored spheres) of the three-dimensional phase diagram. The vertical dashed black lines are drawn to illustrate the planar projections onto the $P$-$x$ plane. The solid black line in the $x$-$P$ plane, which represents the phase boundary between the HO phase (gray region) and the LMAFM phase (white region), is a linear fit to the projected points (filled black circles) in the $x$-$P$ plane. The extrapolation of this linear fit in the $x$-$P$ plane to the $P = 0$ GPa line indicates that the ambient pressure critical concentration of Fe that forces the transition into the LMAFM phase is $x^* = 0.15$. Similarly, the extrapolation of the same linear fit in the $x$-$P$ plane to the $x = 0$ line indicates that, in the absence of Fe substitution ($x = 0$), the critical pressure that forces the transition into the LMAFM phase is very nearly $P_c = 1.5$ GPa, which is the value of the critical pressure, $P_c$, that was determined in several reports of other types of measurements of the URu$_2$Si$_2$ ($x = 0$) parent compound under pressure (see Table I).

Hence, the ambient pressure critical concentration of Fe that was determined in this report to be $x^* = 0.15$ can be thought to be equivalent to the critical value of applied pressure, $P_c = 1.5$ GPa, that induces the HO $\rightarrow$ LMAFM phase transition. This allowed us to determine the linear dependence of the chemical pressure, $P_{ch}(x)$, on the Fe concentration $x$, such that the $P_{ch}(x)$ line, the solid black line shown in Fig. 8, passes through the two points: $(P, x) = (0$ GPa, 0) and $(1.5$ GPa, 0.15). We were then able to compare the slope of the $P_{ch}(x)$ line as shown in Fig. 8 with an $x$ to $P_{ch}(x)$ conversion that is based on a bulk modulus $\beta$ calculation that relates the relative change in the unit cell volume, $d(V/V_0)$, to the change in pressure, $dP$: $\beta = 1/\kappa_T = -d(V/V_0)/dP$, where $V_0$ is the initial volume of the unit cell at ambient pressure and $\kappa_T$ is the isothermal compressibility. From this comparison, we determined the value of the isostructural compressibility for URu$_2$Si$_2$: $\kappa_T = 4.5 \times 10^{-3}$ GPa$^{-1}$. It should be mentioned that a bulk modulus calculation of $P_{ch}(x)$ can vary depending on which value of $\kappa_T$ is used from the range of values that are reported in the literature. Interestingly, our determination of the value for $\kappa_T = 4.5 \times 10^{-3}$ GPa$^{-1}$ is the mean value of the extreme values of $2 \times 10^{-3}$ and $7.3 \times 10^{-3}$ GPa$^{-1}$ reported in the literature.$^{14,61}$

Based on our determination of the $P_{ch}(x)$ line, we note the consistency in which $P_{ch}(x_c) + P_c \approx 1.5$ GPa as displayed in Fig. 8. The blue and black half-filled squares represent the chemical pressure, $P_{ch}$, and critical pressure, $P_c$, respectively, whereas the filled red squares represent the combined effect of experimental tuning or “total” pressure, $P_T = P_{ch}(x_c) + P_c$, that resulted in a first-order transition from the HO to LMAFM phase. The dashed red line at $P = 1.5$ GPa is for reference and allows for a comparison to the $P_T$ values. Remarkably, for each combination of $x$ and $P$, the chemical pressure and applied pressure consistently sum to the value of 1.5 GPa to force the HO $\rightarrow$ LMAFM phase transition. The consistency with which the combination of $x$ and $P$ affect the ordered phases in URu$_2$Si$_2$ reinforces the idea that the substitution of smaller Fe ions for Ru acts as a “chemical pressure”, whereby a reduction in the unit-cell volume affects the compound in a nearly (disregarding disorder) equivalent manner as applying external pressure.

It is clear from the $T_0$ vs. $x$ phase boundary shown in Fig. 1 that a decrease in the concentration of Fe, which effectively expands the volume of the unit-cell in compounds from the URu$_{2-x}$Fe$_x$Si$_2$ series, results in a suppression of $T_0$. Hence, assuming that “chemical pressure” is the effective mechanism responsible for changes in $T_0$, an isostructural substitution involving a larger ion relative to Ru, such as Os, that results in an expansion of the unit cell volume, should also result in a suppression of $T_0$. However, experiments on both polycrystalline$^{52}$ and sin-

![FIG. 8. (Color online) A plot of the chemical pressure $P_{ch}$ (black symbols), critical applied pressure $P_c$ (blue symbols), and the total pressure $P_T$ (red symbols) $= P_{ch}(x_c) + P_c$. The additive behavior of $P_{ch}$ and $P_c$ such that $P_{ch}(x_c) + P_c \approx 1.5$ GPa is consistent with earlier reports on the “kink” in the $T_0(P)$ phase diagram at $P = 1.5$ GPa for the parent compound URu$_2$Si$_2$ ($x = 0$).$^{19}$](image_url)
gle crystal\textsuperscript{24} samples of URu\textsubscript{2−\textit{x}}Os\textsubscript{\textit{x}}Si\textsubscript{2} reveal that as the Os concentration, \textit{y}, is increased in the URu\textsubscript{2−\textit{x}}Os\textsubscript{\textit{x}}Si\textsubscript{2} series, there is an enhancement, rather than a suppression, in \textit{T}_0, that is observed in both the HO and LMAFM phases. This behavior is very similar to the evolution of \textit{T}_0 that is observed with increasing Fe concentration \textit{x} in the URu\textsubscript{2−\textit{x}}Fe\textsubscript{\textit{x}}Si\textsubscript{2} series or with increasing pressure, \textit{P}, for the parent compound URu\textsubscript{2}Si\textsubscript{2}. Hence, for the Os substituted compounds from the URu\textsubscript{2−\textit{x}}Os\textsubscript{\textit{x}}Si\textsubscript{2} series, the evolution of \textit{T}_0 appears to depend on more than “chemical pressure” alone. An investigation involving the experimental tuning of URu\textsubscript{2}Si\textsubscript{2} with Os substitution under applied pressure would be interesting in order to answer the question of how a “negative chemical pressure” and an externally applied pressure work together in a system where other mechanisms seem to be at play in affecting \textit{T}_0 and the HO → LMAFM transition. Such an investigation is currently in progress in our laboratory.

IV. CONCLUDING REMARKS

By tuning the parent compound, URu\textsubscript{2}Si\textsubscript{2}, with an isoelectronic substitution of Fe for Ru, we found that we could bias the material with “chemical pressure” so that a lesser amount of applied external pressure is required to induce the transition to the high pressure LMAFM phase. The results presented here indicate that one can consistently induce the high pressure LMAFM phase in URu\textsubscript{2}Si\textsubscript{2} with the appropriate \textit{x} and \textit{P} combination that yields \textit{P}_\text{ch}(\textit{x}) + \textit{P}_\text{c} ≈ 1.5 GPa. The critical values of \textit{x} and \textit{P} determined in this report seem to be consistent with previous studies in which the URu\textsubscript{2}Si\textsubscript{2} compound was experimentally tuned independently with either \textit{x} or \textit{P}.\textsuperscript{19,22–24} However, the extra degree of freedom gained in experimentally tuning the URu\textsubscript{2}Si\textsubscript{2} compound simultaneously with both \textit{x} and \textit{P} offers a number of advantages. Namely, key aspects of the phase diagrams such as the tricritical point (or critical pressure) become dynamic rather than static features that can be tracked with variations in \textit{x} or \textit{P}. As an unexpected consequence of the simultaneous tuning of the URu\textsubscript{2}Si\textsubscript{2} compound with \textit{x} and \textit{P}, we were also able to “measure” the isothermal compressibility, \textit{κ}_T, for this material and compare its value with others reported in the literature.\textsuperscript{14,61}

The suggestion that the response of the URu\textsubscript{2}Si\textsubscript{2} compound to “chemical pressure” is nearly equivalent to that of applied pressure presents new opportunities for experiments to be performed on the URu\textsubscript{2−\textit{x}}Fe\textsubscript{\textit{x}}Si\textsubscript{2} system using STM and ARPES techniques that traditionally cannot be performed under pressure. We further suggest here that the simultaneous tuning of URu\textsubscript{2}Si\textsubscript{2} with both Fe substitution and external pressure can serve as a workaround to the current limitations on the amount of pressure that can be applied with the various pressure cells that are used in certain neutron scattering experiments. By experimentally tuning the URu\textsubscript{2}Si\textsubscript{2} compound with \textit{x}, it would be possible to bias the compound with chemical pressure at the outset of the neutron experiment so that larger regions of phase space could be studied in the upper pressure limit where quantum criticality might be explored.

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\textsuperscript{7} J. A. Mydosh and P. M. Oppeneer, “Hidden order behaviour in URu\textsubscript{2}Si\textsubscript{2} (A critical review of the status of hidden order in 2014),” Philos. Mag. 94, 3642–3662 (2014).


