Dimensionality Driven Enhancement of Ferromagnetic Superconductivity in URhGe

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In most unconventional superconductors, like the high-T_c cuprates, iron pnictides, or heavy-fermion systems, superconductivity emerges in the proximity of an electronic instability. Identifying unambiguously the pairing mechanism remains nevertheless an enormous challenge. Among these systems, the orthorhombic uranium ferromagnetic superconductors have a unique position, notably because magnetic fields couple directly to ferromagnetic order, leading to the fascinating discovery of the reemergence of superconductivity in URhGe at a high field. Here we show that uniaxial stress is a remarkable tool allowing the fine-tuning of the pairing strength. With a relatively small stress, the superconducting phase diagram is spectacularly modified, with a merging of the low- and high-field superconducting states and a significant enhancement of the superconductivity. The superconducting critical temperature increases both at zero field and under a field, reaching 1 K, more than twice higher than at ambient pressure. This enhancement of superconductivity is shown to be directly related to a change of the magnetic dimensionality detected from an increase of the transverse magnetic susceptibility: In addition to the Ising-type longitudinal ferromagnetic fluctuations, transverse magnetic fluctuations also play an important role in the superconducting pairing.

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For usual s-wave superconductors, superconductivity and ferromagnetism are antagonist states, as the ferromagnetic exchange field easily destroys the superconducting pairs. Therefore, the discovery of the microscopic coexistence of superconductivity and ferromagnetism in three orthorhombic uranium-based heavy-fermion systems (UGe_2, URhGe, and UCoGe) [1–6] was one of the most exciting events in recent condensed matter physics. This coexistence strongly suggests a superconducting state with triplet pairing, where the Pauli limiting mechanism is not active and the Cooper pairs can survive in the strong exchange field. The direct coupling between a static uniform field and ferromagnetism also leads to fascinating behavior of the superconducting state under a magnetic field. In these systems, the pairing mechanism can actually be tuned by a magnetic field, either increased [7,8] or suppressed [9], respectively, when the magnetic field is applied perpendicular to (transverse field configuration) or along (longitudinal configuration) the easy magnetization axis. The most striking case is URhGe. When the magnetic field is applied along the b axis of the orthorhombic crystal (perpendicular to the easy magnetization c axis), superconductivity is initially suppressed for a field of about 2 T due to the usual orbital effect. However, at a higher field, superconductivity reappears in the field region from 9 to 13 T [8]. The superconducting critical temperature (T_{SC}) of the reentrant phase reaches a value of 0.4 K, almost twice higher than T_{SC} in zero field, at a field $H_R \approx 12$ T, where a rotation of the direction of the magnetic moments occurs, from the c to the b axis [8].

Hydrostatic pressure is a powerful method to reveal unconventional superconductivity in many systems, mainly by driving them to the threshold of an electronic instability [10,11]. Previous studies [12,13] show, however, that in URhGe hydrostatic pressure increases the Curie temperature T_{Curie}, driving the system further away from its instability. Simultaneously, H_R was found to increase, accompanied by a collapse of zero-field superconductivity at about 4 GPa and of the reentrant superconductivity at an even lower pressure of 2 GPa [13]. Uniaxial stress has proved often a much more efficient tuning tool than hydrostatic pressure in many strongly correlated systems such as URu_2Si_2 [14], UCoAl [15], and Sr_2RuO_4 [16]. Here we demonstrate that, in URhGe, uniaxial stress applied along the b axis boosts the magnetic fluctuations and drastically decreases H_R, rapidly leading to a strong increase of the superconducting critical temperatures at zero field and at H_R. This increase is accompanied by a merging of the low- and high-field superconducting phases even before a significant effect of stress has been detected on T_{Curie}. The driving force for the enhancement of the pairing mechanism, even in zero magnetic field, seems to be the increase of the b-axis susceptibility, moving the system away from the Ising-type limit. This mechanism is predicted by microscopic theories of anisotropic ferromagnetic superconductors, as arising from an

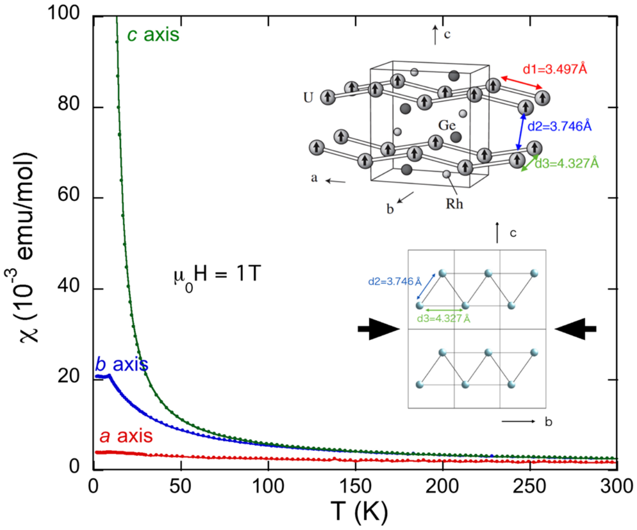

FIG. 1. Magnetic susceptibility and orthorhombic crystal structure of URhGe showing the position of the U atoms in the *cb* plane: Applied stress (black arrows) along the *b* axis reduces the U-U distance in the *b* direction, while it increases in the *c* direction.

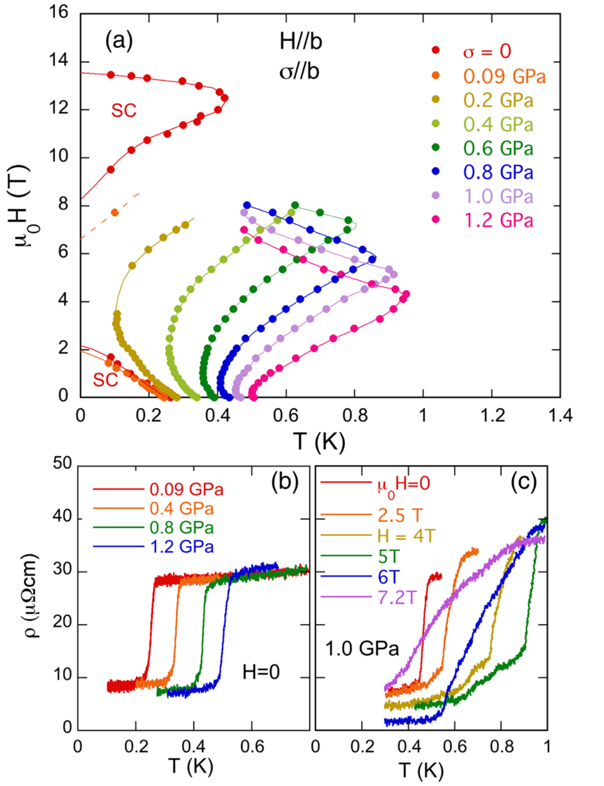

FIG. 2. (T, H) phase diagram of superconductivity for the transverse ($H\|b$) configuration. (a) shows the phase diagram for different values of stress, together with a previous zero pressure measurement [7]. It is obtained with a criterion of 50% of the total resistance drop. Lines are guides to the eye. (b) and (c) show the superconducting transitions at different values of stress and field, respectively. See Supplemental Material [19] for a discussion on the shape of the transitions.

enhanced coupling of the spin-polarized bands by transverse fluctuations [17].

A single crystal of URhGe was grown by the Czochralski technique. A bar-shaped sample was cut with the long direction along the *b* axis. Quite a large sample was needed, so the sample quality was less than that of the best samples, as indicated by the residual resistivity ratio: RRR = 10. Also, at ambient pressure the sample showed a sharp but incomplete superconducting transition. Nevertheless, we can reasonably assume that the transition measured here reflects the behavior of the bulk superconducting phase found in high-quality samples [18] (see Supplemental Material for a further discussion of this point [19]). The sample resistance was measured with the current along the *b* axis, in a dilution cryostat. A magnetic field of maximum 8 T was applied along the *b* axis. The sample was compressed along the *b* axis between two sapphire anvils. The stress was applied and changed *in situ* using helium-filled bellows. This device is based on a previously reported system for tuning the pressure in a diamond anvil cell [20]. Stress was increased up to a maximum value of 1.2 GPa. Magnetization was measured in a clamp cell adapted to a SQUID magnetometer (Quantum Design MPMS) up to 0.6 GPa and a maximum field of 5 T. The force applied at room temperature determined the stress value. More details are given in Supplemental Material [19].

Figure 1 shows the temperature dependence of the magnetic susceptibility for the three crystallographic directions and the orthorhombic crystal structure of URhGe. The U atoms form zigzag chains along the *a* direction with the magnetic moments aligned in the *c* direction. The U atoms form distorted hexagons in the *bc* plane, slightly elongated along the *b* axis. Hence, the structural anisotropy in the *bc* plane is rather small. While the *a* axis is the hardest magnetic axis at all temperatures, the magnetic susceptibilities along the *b* and *c* axes are similar above 50 K. Thus, URhGe selects the easy magnetization *c* axis only when a coherent state of renormalized heavy quasiparticles develops. The low energy scale governing the emergence of the *bc* anisotropy is certainly a key element driving the response of URhGe to a magnetic field $H\|b$.

When stress is applied along one crystallographic axis, the unit cell is compressed in this direction but expands in the other directions. From thermal expansion measurements [18] according to the Ehrenfest relation, applying uniaxial stress along the *b* axis of URhGe would tend to reduce T_{Curie} with an expected slope of −1.6 K/GPa. We also expect that, as the characteristic magnetic energy scales are lowered, the rotation of the moments from the *c* to the *b* axis will occur at a lower field. The first goal is therefore to determine the relations between T_{Curie}, H_R, and T_{SC}, the latter both in zero field and at H_R.

Figure 2 shows the (T,H) superconducting phase diagram in the transverse field configuration ($H\|b$) for

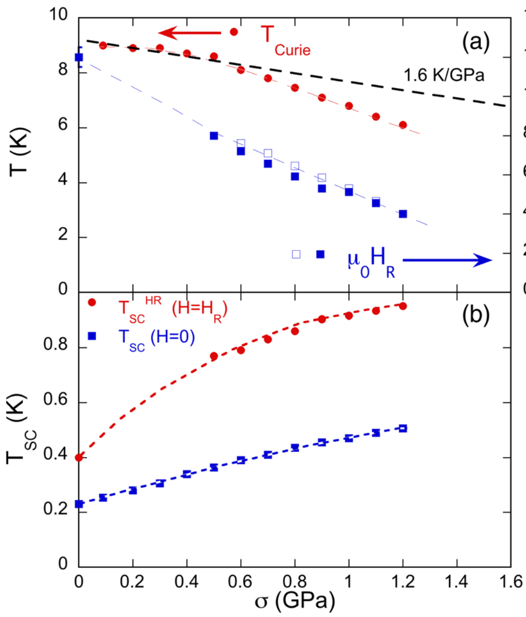

FIG. 3. (a) Stress dependence of the parameters T_{Curie} and field H_R where the rotation of the moments occurs. H_R was determined by the maximum of T_{SC} (full symbols) and by the peak in the normal state magnetoresistance (open symbols; see Supplemental Material [19]). H_R for $\sigma = 0$ was not measured, so a typical value of 12 T was taken; the error bar shows the possible uncertainty, mainly due to the precision of the alignment with the field. The dashed black line shows the expected initial slope for T_{Curie} of 1.6 K/GPa from the thermal expansion measurements. (b) Stress dependence of the superconducting critical temperature $T_{\mathrm{SC}}^{(H=0)}$ at zero field (red circles) and at the maximum value $T_{\mathrm{SC}}^{\mathrm{HR}}$ obtained for $H = H_R$. Lines are guides for the eyes.

different values of uniaxial stress applied along the b axis. We have defined H_R as the field of the maximum T_{SC} of the high-field phase; it corresponds also to the maximum of the normal state magnetoresistance (see Supplemental Material [19]). We find two main effects. First of all, H_R decreases strongly: The high-field superconducting pocket moves to lower fields and merges with the low-field superconducting phase already at 0.2 GPa. Second, the superconductivity is significantly enhanced, both at zero field and even more strongly at H_R.

In Fig. 3, we show the stress dependence of the different parameters T_{Curie}, H_R, $T_{\mathrm{SC}}^{(H=0)}$, and the maximum $T_{\mathrm{SC}}^{\mathrm{HR}}$. As expected from the thermal expansion results, T_{Curie} decreases with stress. This decrease is very weak below 0.5 GPa, compatible with a slope of ~ -1.6 K/GPa, deduced from thermal expansion. This decrease is more pronounced above 0.6 GPa. In contrast, H_R is extremely sensitive to uniaxial stress. H_R could not be determined for stress below 0.6 GPa, as H_R is larger than 8 T, the maximum field we could apply for these measurements; however, the stress dependence of H_R is close to linear over the whole measured range. At 0.6 GPa, H_R has decreased to less than 8 T and is reduced to 4 T at 1.2 GPa, the maximum

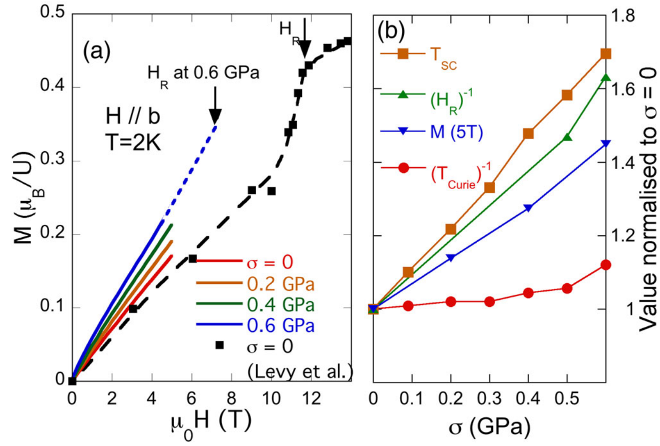

FIG. 4. Magnetization (a) of URhGe under uniaxial stress for $H \| b$ and $\sigma \| b$, compared to ambient pressure data (Levy *et al.* [8]). Arrows show the value of H_R at ambient pressure and $\sigma = 0.6$ GPa. (b) shows that the normalized changes of χ (taken as M at 5 T), $1/H_R$, and T_{SC} are very similar and seem uncorrelated with those of $1/T_{\mathrm{Curie}}$. Lines are guides for the eyes.

stress achieved. This rapid decrease of H_R is accompanied by a significant enhancement of the superconducting critical temperatures: $T_{\mathrm{SC}}^{(H=0)}$ and $T_{\mathrm{SC}}^{\mathrm{HR}}$ both increase continuously, $T_{\mathrm{SC}}^{(H=0)}$ reaching 0.5 K and $T_{\mathrm{SC}}^{\mathrm{HR}}$ almost attaining 1 K at 1.2 GPa. Like for H_R, their strong initial variations under stress contrast with the weak decrease of T_{Curie}. And this variation is not altered above 0.6 GPa, when T_{Curie} is decreasing faster. So qualitatively, the evolution of the critical field H_R and of the superconducting transitions seems *not* driven by the mere evolution of T_{Curie} under stress.

A hint for the factor which may be most influenced by stress comes from the strong decrease of H_R: The relation between the high-field superconducting state and the rotation of the moments at H_R was established early on [8]. So qualitatively, the lowering of H_R means that, under stress, a smaller field is required to align the moments along the b axis. In other words, the anisotropy between the c axis (the easy magnetization axis) and the b axis is getting smaller. Quantitatively, it was noticed that the rotation occurs when the magnetization in the b direction approaches the spontaneous zero-field magnetization $M_c = 0.4\mu_B$ in the easy c direction [7,18]. This means that, if H_R occurs at lower fields, the susceptibility along the b axis, $\chi_b = \partial M/\partial H$, should increase, with a proportionality of $1/\chi_b \approx H_R$.

Figure 4(a) shows our magnetization measurements at 2 K under uniaxial stress: A significant and rapid increase of the susceptibility χ_b with increasing stress is found. In Fig. 4(b), we show the relative change of the different parameters versus stress. The variations of $1/\chi_b$ and H_R are quite similar, as expected, whereas the relative change of T_{Curie} is much weaker. It is also clear that T_{SC} is correlated to $1/\chi_b$ and H_R rather than to T_{Curie}. This extreme sensitivity of H_R to stress, and the fact that T_{Curie} is much less so, points to strongly anisotropic magnetocrystalline effects. The situation is quite different with hydrostatic pressure, where, up to 0.84 GPa, T_{Curie} and H_R were found to increase with similar relative changes [13]. This can be qualitatively understood by considering the crystal structure of URhGe (see Fig. 1). Applying stress along the b axis will reduce the nearest-neighbor distance of U atoms in the b direction, while the nearest-neighbor distance along the c axis will increase, leading through the variation of the exchange integrals to an increase of the susceptibility along the b axis and a decrease of T_{Curie}. It will also reduce the distortion of the hexagons leading to a more isotropic bc plane.

The other spectacular result is that, with increasing stress, superconductivity is strongly enhanced, both at zero field and at H_R, with the maximum $T_{\mathrm{SC}}^{\mathrm{HR}}$ more than doubling between ambient pressure and 1.2 GPa. Of course, as H_R decreases, the maximum $T_{\mathrm{SC}}^{\mathrm{HR}}$ occurs at a lower field, so the orbital pair-breaking effect of the field is weaker and will naturally lead to a higher T_{SC}.

To eliminate the influence of the reduced orbital effect when H_R occurs at a lower field and better quantify the reinforcement of superconductivity, we have analyzed the superconducting upper critical field (H_{C2}) curves using a strong-coupling model [21], following the approach developed by Wu *et al.* [9]. The hypothesis is that the changes of T_{SC} and of the orbital limit are controlled by that of the electronic correlations responsible for the pairing: T_{SC} and the effective mass m^* are controlled by a unique (field- and stress-dependent) strong-coupling parameter λ (see Supplemental Material for more details [19]). Note that, in Sr_2RuO_4, where a similar increase of T_{SC} under stress is observed, the mechanism is different, involving mainly the occurrence of a van Hove singularity on the 2D-band structure [16]. The results are shown in Fig. 5 together with those obtained from the analysis of the ambient pressure data. As the stress is increased, the values of λ at zero field and at H_R clearly increase [Fig. 5(a), inset], showing that the pairing strength is significantly enhanced as H_R moves to lower values. However, as shown in Fig. 5(b), the enhancement of λ between zero field and H_R, plotted versus H/H_R, appears to be independent of the stress. In other words, the value of λ (and therefore T_{SC}) at H_R under stress scales with its zero-field value.

This increase of T_{SC} with uniaxial stress at zero field, while T_{Curie} is almost constant, is quite remarkable. We have shown that the increase of T_{SC} matches the increase of the susceptibility along the b axis, which can be seen as a weakening of the uniaxial anisotropy. This is at odds with initial theoretical studies predicting that Ising-type ferromagnets would be more favorable to equal spin pairing (ESP) p-wave superconductors due to the suppression of the "pair-breaking" transverse magnetic fluctuations [22–24].

Recent work has shown that this is not the case in anisotropic (orthorhombic) systems [17]. Taking into account the coupling between the two components ($\Delta_{\uparrow\uparrow}$ and $\Delta_{\downarrow\downarrow}$) of the ESP p-wave order parameter, a weak-coupling expression for T_{SC} very similar to that for a multiband superconductor is derived [17]. Intraband couplings $g_{1\uparrow,\downarrow}$ are proportional to the susceptibility χ_c at the Fermi wave vector along the (easy) c axis and to the respective averaged density of states for spin $\uparrow$, $\downarrow$ bands. Interband couplings $g_{2\uparrow,\downarrow}$ depend mainly on the same density of states and on $(\chi_b - \chi_a)$. In the isotropic systems considered in the early theories, $g_{2\uparrow,\downarrow} = 0$. For an orthorhombic anisotropy, $g_{2\uparrow,\downarrow} \neq 0$, and T_{SC} is expressed as [17]

$$T_{\mathrm{SC}} \propto \exp\left(-\frac{1}{g}\right),$$

$$g = \frac{g_{1\uparrow} + g_{1\downarrow}}{2} + \sqrt{\frac{(g_{1\uparrow} - g_{1\downarrow})^2}{4} + g_{2\uparrow}g_{2\downarrow}}.$$

$g_{2\uparrow}g_{2\downarrow} \propto (\chi_b - \chi_a)^2$ is a positive term, so the anisotropy of the transverse susceptibilities increases T_{SC}. We claim that this is the reason why stress is so effective in boosting

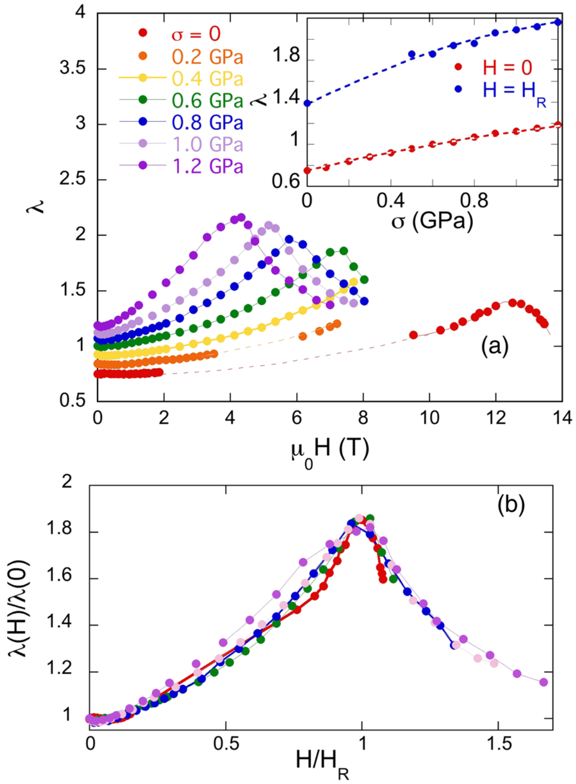

FIG. 5. Analysis of H_{C2}. (a) Field dependence of the strong coupling parameter λ determined from the fits of the upper critical field H_{C2} for different applied stresses. See Supplemental Material [19] for a description of the procedure. The red points are taken from Levy *et al.* [8], where the zero pressure analysis was performed. The insert shows the pressure dependence of λ at $H = 0$ and $H = H_R$. (b) shows the normalized values; the enhancement of λ at H_R is constant at all values of stress. Lines are guides for the eye.

T_{SC}: Enhancing χ_b drives the system from a "1D" towards a "2D" magnetic anisotropy, increasing the coupling between the superconducting order parameters of the opposite spin bands, which enhances T_{SC} as in any two-band superconductor. This mechanism seems much more efficient than the simultaneous weak decrease of T_{Curie}.

The situation for the reentrant phase at H_R is more complex. Close to this field, NMR studies have shown that both the longitudinal and transverse ferromagnetic fluctuations are strongly enhanced [25,26]. Our results strongly suggest that both types of fluctuations might contribute to reinforce the superconductivity at H_R.

Therefore, the zero-field results yield the most solid experimental argument demonstrating that, in ferromagnetic systems, a large transverse susceptibility allowing the presence of both longitudinal and transverse ferromagnetic fluctuations is more effective for superconductivity than a strong Ising character. If this is well understood as arising from "multiband superconductivity," a detailed microscopic theory of the magnetic properties of URhGe is still missing, and the details of the electronic structure are a difficult and open question.

We hope that this work will motivate theoretical efforts towards a more quantitative understanding of the interplay between ferromagnetism and superconductivity in uranium systems. Experimentally, these results should stimulate new investigations on URhGe and might also guide explorations to find other ferromagnetic superconductors. A challenge is to perform experiments at higher stress. Extrapolating our results, H_R should be tuned to zero at a critical value of stress of about 1.7 GPa. At this point, URhGe might switch from an easy c-axis to a b-axis ferromagnetic state. What will happen then to superconductivity at this point is a completely open question.

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