

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In this manuscript, S. Kawasaki et al. report NQR results under pressure in the doped heavy fermion compound $\text{Ce}(\text{Rh}_{0.5}\text{Ir}_{0.5})\text{In}_5$. This family of compound has been heavily investigated in the past two decades and has immensely contributed – and challenged- our understanding of quantum criticality and the interplay between antiferromagnetism and superconductivity in strongly correlated electron systems. While we have accumulated a wealth of data on the pure 115 compounds, like the two end members here, CeRhIn_5 and CeIrIn_5 , there are still gaps in our understanding of how the pressure-temperature phase diagrams evolve from one end compound to the other as a function of doping. In this context the NQR data presented here are extremely valuable and my overall assessment of the paper is that it is well written and the data is clearly presented. My only reservation is with their interpretations, some conclusions drawn here are not as straightforward as they make it sound. They make three strong claims in the paper and I take issue with every single one. Therefore, I recommend the paper for publication with revisions, after the authors satisfactorily address the concerns below.

The first claim is that above 0.8GPa the antiferromagnetism in this compound is of a spin-density wave type. The spin density wave (SDW) is generally understood as a nesting-driven instability of the Fermi Surface and so my question is: what evidence is there in their data (or any other data) to claim that there is a SDW in this doped ($x=0.5$) compound under pressure? In other words, why should we believe that the antiferromagnetism in the doped $\text{Ce}(\text{Rh},\text{Ir})\text{In}_5$ is fundamentally different than that in the parent compound CeRhIn_5 , which we know to be a local type of antiferromagnetism. I find it particularly misleading and confusing to put the X-ray data at room temperature together with NQR data at low temperature. The lattice parameters with pressure decrease monotonically at room temperature, as one would expect, but that doesn't preclude a jump at lower temperature so I am not sure what the x-ray data add to the argument they are trying to make. What we need ideally is a low temperature data of the volume contraction and/or thermal expansion under pressure, which are both rather difficult measurements... Maybe it is best to keep the x-ray data for the supplementary info and show $\ln(2)$ data instead in the main body of the paper?

The related claim they make is that the jump observed in the NQR frequency as a function of pressure is a signature of a small to large Fermi surface change. NQR frequency in fact measures the electric field gradient but since they only focus on $\ln(1)$ nuclei, by symmetry the only component of the EFG gradient they are probing is the one along the c-axis, perpendicular to the CeIn planes. It seems like an over-interpretation to infer the entire Fermi Surface volume is changing based on one component of the EFG gradient alone. Therefore, my question to the authors is have they seen a similar sudden change in NQR frequency for the $\ln(2)$ site? Why or why not? Again, I strongly recommend adding $\ln(2)$ data, if available, in the figure 4 and adjust the discussion accordingly. It is fascinating that the jump in NQR frequency with pressure is observed above and below T_N or T_c . If this effect is truly connected to the Kondo hybridization, one would expect the jump to be visible for all $T < T_{\text{coh}}$, where T_{coh} marks the onset of coherence in the Kondo lattice, and not visible for $T > T_{\text{coh}}$. With the data presented, it is not clear if this is the case so I also ask that the authors clarify up to what temperature they can track the abrupt change in the NQR frequency and add that T^* onto their phase diagram.

-The third big claim of the paper is concerning the residual density of states at the Fermi level below T_c , as evidenced by the $1/T_1T$ plots in fig6. While I agree that this shows there is a significant fraction of unpaired quasiparticles below T_c around the critical pressure for QCP, I take issue with the claim that this is evidence for p-wave pairing. If we have learnt anything about unconventional superconductivity in the past 3 decades, it is that one must be extremely cautious about inferring the pairing symmetry based on "angle averaged" measurements over the whole Fermi surface. There is no evidence that the superconductivity in either CeRhIn_5 (under pressure) or CeIrIn_5 (at ambient pressure) is odd-parity and there is no evidence that superconductivity in $\text{Ce}(\text{Rh},\text{Ir})\text{In}_5$ is of different symmetry than either of its parent compounds. Unless we have the angle-resolved measurements to confirm the symmetry of the doped compound I urge the authors

not to make such a strong statement about it. I think a more fruitful debate one should have on the fig 6 data is to ask ourselves how the antiferromagnetism and superconductivity co-exist on the same (multi-sheet) Fermi surface? Could it be that a nesting –driven SDW gaps most parts of the Fermi surface, decreasing the number of electrons available to form Cooper pairs while increasing the density of quasiparticles below T_c ?

-Also, it is hard for the reader to build his/her own confidence about the validity of the data presented here with so little info about the samples, and I urge the authors to add more detail about sample characterization in the supplementary section. For instance, how confident they are that in the single crystals they studied the distribution of Ir across the sample volume is homogeneous? In other words, we don't have Rh-rich vs Ir-rich parts in the powder? What precautions have they taken to characterize their samples and how they picked what looks like the most homogeneous ones? My own experience with both growing and characterizing doped 115 crystals leads me to believe that the doping distribution we systematically see with SEM is not limited to the surface and that the 115s especially around 50% form inherently inhomogeneous crystals.

Cigdem Capan

Reviewer #2 (Remarks to the Author):

The manuscript "localized-to-itinerant transition, antiferromagnetic quantum critical point, and gapless superconductivity in $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ " presents low temperature NQR experiments on the In(1)-site under hydrostatic pressure. The presented NQR data is exclusively on the first transition, that is $I = \pm 1/2 \rightarrow \pm 3/2$. They found that the In(1) frequency shows a jump upon increasing pressure. The temperature (T) evolution of the jump position extrapolates for $T \rightarrow 0$ to a critical pressure of $p_c = 0.8$ GP, a newly found quantum critical point (QCP) in this compound. The authors interpret the jump as transition from a small Fermi surface (FS) where the 4f-electron is considered to be localized to a large FS where the Ce 4f-electron has itinerant character.

A second QCP appears at higher pressure, $p_c = 1.2$ GPa, evidenced by a maximum in the residual density of states (rDOS), the highest superconducting transition temperature and the position where the θ , a measure for the distance to a 3D antiferromagnetic spin density wave transition (AFM SDW) goes to zero.

The authors further state that the superconducting (SC) state is due to odd-frequency spin-singlet pairing.

To authors, however, impose several constraints to their work which are less straightforward.

(i) The assumption that the hyperfine coupling constant A remains unchanged over the entire pressure range is uncertain. The authors take that conclusion from the evolution of A under pressure in the parent compound CeRhIn_5 . The hyperfine coupling constant strongly depends on the local environment. In CeRhIn_5 , as the authors show, the c/a-ratio increases continuously with pressure, while this is not the case for the 0.5-doped Ir one where the c/a-ratio is almost constant. Hence the local environment will differ and may lead to a change of A with pressure in $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$.

(ii) The author repeatedly stated that Ir acts as chemical pressure and $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ resembles CeRhIn_5 under 1 GPa pressure. Such statement is incorrect and the authors should be more careful. So far, such precise matching of phase diagrams of pressure compared to chemical substitution has been seen in YbRh_2Si_2 with the 5%Ge-doped one, as well as in $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$ -series but as pointed out by Nicklas et al. (Phys. Rev. B 70, 020505(R) (2004)) scaling breaks down for $x > 0.3$, indicating additional effects of Ir substitution. Such is also evident as the T-p phase diagram of $\text{CeRh}_{0.58}\text{Ir}_{0.42}\text{In}_5$ (Luo et al, npj Quant. Mat. 3, 6 (2018)) does not fit into this scaling.

The report $\text{In}(1)$ transition for $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ is slightly smaller than what is found here, yielding 6.24 MHz (Phys. Rev. B 70, 014511 (2004)). The frequency here corresponds more to a value associated with $x = 0.4$. In addition, it is unclear why the entire manuscript presents only the $\text{In}(1)$ $I = \pm 1/2 \rightarrow \pm 3/2$ transition line (not even in the supplemental material anything else is found). The $\text{In}(1)$ line has local tetragonal symmetry and hence the NQR spectra for the $\text{In}(1)$ site consist of four equally spaced transition lines. Why the authors were not using (and presenting) the full $\text{In}(1)$ spectrum in order to determine the $\text{In}(1)$ base frequency and resulting shifts upon applied pressure. The jump itself should be visible in all higher harmonics.

Figure 4(e) actually opposes the authors' claim of a more itinerant behavior for $p > 0.8$ GPa. The $\text{In}(1)$ frequency of CeRhIn_5 is higher than the CeIrIn_5 although in the later the 4f-electron is strongly delocalized. According to this figure one would expect the jump going downwards at $p_c = 0.8$ GPa. The authors should put their calculations in the right context.

More intriguing, the width of the small FS to larger FS transition is constant independent of the temperature. Such points to a real transition line in the T-p phase diagram, not a cross-over like behavior as for instance seen in YbRh_2Si_2 . This is rather unusual. At higher temperatures thermal fluctuations would lead to a broadening of the small FS to large FS transition. The authors should explain such artifact.

This is the first report of a jump in the NQR frequency linked to a Kondo breakdown QCP. The parent compound CeRhIn_5 exhibits a Kondo breakdown QCP at $p_c = 2.3$ GPa, however no group ever presented a frequency jump. While this might have several reasons, in order to point out that such jump is a signature of a Kondo breakdown QCP, the authors should have verified it with CeRhIn_5 to strengthen their claim, despite the additional work it causes.

Moreover, what speaks against a Kondo breakdown QCP at $p_c = 0.8$ GPa is that the rDOS is not enhanced as one would expect in such scenario.

A further problem of the manuscript in this version is that it is unclear whether the authors claim that the first QCP at $p_c = 0.8$ GPa is due to a small FS to large FS transition or a Lifshitz transition.

The subsequent appearance of a Kondo breakdown (or small FS to large FS) transition in this compound is not new and has been reported for $\text{CeRh}_{0.58}\text{Ir}_{0.42}\text{In}_5$ (Luo et al, npj Quant. Mat. 3, 6 (2018)) already. The discussion on the SC state is a marginal contribution to the manuscript and has more speculative character.

In summary, the manuscript contains some interesting observations and ideas. However, so far they seem to be more specific to this material and hence will not enrich the field in a wide context. The presence of two types of QCPs is not new and has been already observed in a similar substituted sample. For this reason, I cannot recommend the manuscript to be published in Communications Physics.

Reviewer #3 (Remarks to the Author):

Referee report on "Localized-to-itinerant transition, and antiferromagnetic quantum critical point, and gapless superconductivity in $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ "
By S. Kawasaki et al.

Summary:

The authors Kawasaki et al. performed NMR and NQR measurements under pressures on a heavy-fermion superconductor $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$. The paper contains interesting experimental results on

an anomalous change of the electronic state at around P_c^* and an enhancement of low-lying magnetic excitations at around P_cSDW . The authors found an anomalous change of the NQR frequency at around P_c^* that is ascribed to the change of Fermi surface (FS) volume from the localized small FS to itinerant large FS.

They also inferred the possible odd-frequency (p-wave) singlet state in $CeRh_{0.5}Ir_{0.5}In_5$ from the anomalously enhanced low-lying excitations in the SC state at around SDW -QCP (P_cSDW). Their data and results are insufficient to support a clear and definite conclusion. Otherwise, this is a good paper and worthy of being published.

Recommendation:

This work is of high quality in general, however some points should be addressed (see below). I recommend publication as a communications physics after revision.

Comments/Questions:

1. The authors discuss the change of the FS volume at P_c^* from the precise pressure dependence of In NQR frequency. They pointed out that the change of the EFG parameter at the In(1) site is due to the change of the on-site electronic state. As the authors mentioned briefly in the DISCUSSION, small amount of Ce-valence change can also be another possible reason. Did the authors calculate the EFG by HiLAPW when the Ce-valence slightly changes? The simulation of the valence-dependence of the EFG strongly supports your claim.

2. The authors found that the coupling constant of $CeRh_{0.5}Ir_{0.5}In_5$ is very close to that of $CeRhIn_5$ under 1GPa, and claimed that Ir substitution with 0.5 is equivalent to the application of a physical pressure “greater than” 1 GPa to $CeRhIn_5$ (Supplementary). Did the authors measure the pressure dependence of the NMR Knight shift? The coupling constant closely connects to a local spin density at In(1) site. For example, Kambe et al. reported that the temperature dependent term of the coupling constant below TK relates to the temperature dependence of the density of state. (PRB 81, 140405R(2010)). Therefore, I guess the coupling constant also changes at around P_c^* .

3. The authors discuss the non-magnetic impurity effect for unconventional superconductors. This material has layered structure and Rh/Ir layer is sandwiched between $CeIn(1)$ layers (Fig.2 (a)). So that, Ce layer is not directly affected by the present substitution. In this case, do the theoretical models for unitary scattering work as it is?

4. The authors mentioned the anomalously high value of $1/T_1T$ (residual DOS) in the SC state at around P_cSDW , comparing with the case for the full gap s-wave and clean d-wave superconductors. However, at just around the SDW QCP, magnetic fluctuations are enhanced, so that one can simply imagine that low-lying magnetic excitations could survive as a background of $1/T_1T$ even in the SC state. How about the case for other QCP materials or Ce_{115} family?

Minor points:

Introduction

The phrase “indirect exchange interaction among weakly screened localized f-electrons,” in line 22 in page 2 should be stated in line 15. In this line, the phrase should be replaced by “the RKKY-type AF order”.

There are two kinds of Γ_7 CEF wave functions ($\Gamma_7(1)$ and $\Gamma_7(2)$) in the tetragonal symmetry. (line 19, page 3) Which state is realized?

Methods

The authors described the single crystals were moderately crushed into grains. For NMR measurements, did the authors use oriented grains for NMR measurements? There are no

statements in Methods.

First of all, we would like to thank all the referees for their time, useful comments and suggestions. We have revised the manuscript by taking these comments/suggestions into account.

Summary of Changes

We have:

1. replaced the words “SDW QCP” and “ P_c^{SDW} ” with “antiferromagnetic QCP” and “ P_c^{AFM} ”.
2. added new references [10], and [36] specified by Reviewer#3.
3. added new figures. The temperature dependence of the In(2) NQR spectrum and its peak position, and In(1) ν_Q at $P = 1.12$ GPa are now shown as Fig. 4d and 4e. The new data point is added in the phase diagram (Fig.8). The previous Figure 4d and 4e now become new Figure 5a and 5b.
4. added the temperature dependence of the 1st and 4th transitions of the In(1) NQR spectrum in Supplementary Fig. 7 of the Supplementary Information.
5. added new descriptions/statements in the main text as shown in red following the reviewers’ suggestions/comments.

Point-to-Point Responses to Reviewers’ comments:

Responses to Reviewer #1:

The first claim is that above 0.8GPa the antiferromagnetism in this compound is of a spin-density wave type. The spin density wave (SDW) is generally understood as a nesting-driven instability of the Fermi Surface and so my question is: what evidence is there in their data (or any other data) to claim that there is a SDW in this doped ($x=0.5$) compound under pressure? In other words, why should we believe that the antiferromagnetism in the doped Ce(Rh,Ir)In5 is fundamentally different than that in the parent compound CeRhIn5, which we know to be a local type of antiferromagnetism.

We are sorry for the confusion. It is not our intension to claim that the antiferromagnetism in this compound is of a spin-density wave type. We merely followed the custom (for example, Q. Si and F. Steglich, Science **329**, 161 (2010).) and used the word SDW to name the type of antiferromagnetic-order QCP.

In the revised manuscript, we replaced the words “SDW QCP” and “ P_c^{SDW} ” with “antiferromagnetic QCP” and “ P_c^{AFM} ” to avoid misunderstanding or confusion. In addition, we

added the above-mentioned review article as new ref. 10 in the introductory paragraph.

I find it particularly misleading and confusing to put the X-ray data at room temperature together with NQR data at low temperature. The lattice parameters with pressure decrease monotonically at room temperature, as one would expect, but that doesn't preclude a jump at lower temperature so I am not sure what the x-ray data add to the argument they are trying to make. What we need ideally is a low temperature data of the volume contraction and/or thermal expansion under pressure, which are both rather difficult measurements... Maybe it is best to keep the x-ray data for the supplementary info and show In(2) data instead in the main body of the paper?

The purpose of the original Fig. 4d-f is to make sure that $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ does not undergo a structural phase transition up to $P = 4$ GPa, at least at room temperature. If a structural phase transition occurs at room temperature, the NQR frequency (ν_Q) measured at low temperature would show a jump.

We agree with the reviewer that it is somehow misleading to show the X-ray data in Fig. 4. In the revised manuscript, we have removed these data to a new figure (Fig. 5).

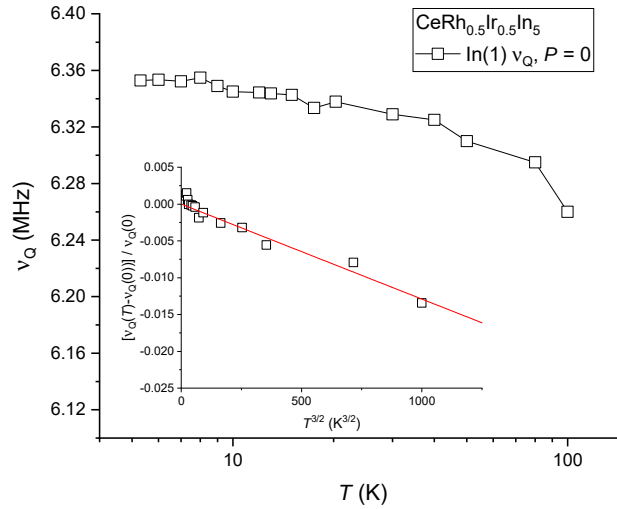
The related claim they make is that the jump observed in the NQR frequency as a function of pressure is a signature of a small to large Fermi surface change. NQR frequency in fact measures the electric field gradient but since they only focus on In(1) nuclei, by symmetry the only component of the EFG gradient they are probing is the one along the c-axis, perpendicular to the CeIn planes. It seems like an over-interpretation to infer the entire Fermi Surface volume is changing based on one component of the EFG gradient alone. Therefore, my question to the authors is have they seen a similar sudden change in NQR frequency for the In(2) site? Why or why not? Again, I strongly recommend adding In(2) data, if available, in the figure 4 and adjust the discussion accordingly.

We measured the temperature dependence of the In(2) In-NQR spectrum at $P = 1.12$ GPa. The results are added as new Fig. 4d and 4e together with the data for the In(1) site at this pressure. The obtained result at the In(2) site agrees well with those of the In(1) site, indicating that the entire Fermi-surface volume changes at (P^*, T^*) . We added a few sentences in the main text along with the results.

It is fascinating that the jump in NQR frequency with pressure is observed above and below T_N or T_c . If this effect is truly connected to the Kondo hybridization, one would expect the jump to be visible for all $T < T_{coh}$, where T_{coh} marks the onset of coherence in the Kondo lattice, and

not visible for $T > T_{\text{coh}}$. With the data presented, it is not clear if this is the case so I also ask that the authors clarify up to what temperature they can track the abrupt change in the NQR frequency and add that T^* onto their phase diagram.

At high temperature, the NQR frequency decreases with increasing temperature regardless of the Kondo hybridization (see attached figure). In general, in conventional non-cubic metals, the temperature dependence of ν_Q due to the lattice contraction can be described by the empirical relation, $\nu_Q(T) = \nu_Q(0)(1 - AT^{3/2})$ (the solid straight line in the inset of the attached graph, see, J. Christiansen *et al.*, Z. Phys. **24**, 177 (1976).). Unfortunately, since the change due to lattice becomes dominant above $T = 4.2$ K, we are unable to know how much ν_Q changes due to Kondo hybridization at T_{coh} .



-The third big claim of the paper is concerning the residual density of states at the Fermi level below T_c , as evidenced by the $1/T1T$ plots in fig6. While I agree that this shows there is a significant fraction of unpaired quasiparticles below T_c around the critical pressure for QCP, I take issue with the claim that this is evidence for p-wave pairing. If we have learnt anything about unconventional superconductivity in the past 3 decades, it is that one must be extremely cautious about inferring the pairing symmetry based on “angle averaged” measurements over the whole Fermi surface. There is no evidence that the superconductivity in either CeRhIn_5 (under pressure) or CeIrIn_5 (at ambient pressure) is odd-parity and there is no evidence that superconductivity in $\text{Ce}(\text{Rh},\text{Ir})\text{In}_5$ is of different symmetry than either of its parent compounds. Unless we have the angle-resolved measurements to confirm the symmetry of the doped compound I urge the authors not to make such a strong statement about it.

It is true that the pairing symmetry in the end members CeRhIn_5 and CeIrIn_5 is most likely of even frequency d -wave. However, $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ possess aspects not shown by CeRhIn_5 nor CeIrIn_5 .

In CeRhIn_5 , although spin fluctuations are strong, the Fermi surface is small. In CeIrIn_5 , on the other hand, the Fermi surface is large but spin fluctuations are weaker. By contrast, in the vicinity of P_c^{AFM} of $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$, Fermi surface is large AND spin fluctuations are strong, which satisfy the conditions for realizing a different pairing symmetry, odd-frequency p -wave spin singlet. We have added one sentence to stress this point in p10 before SUMMARY.

Although we concluded that our observation is a strong indication for such exotic pairing state, we agree with the reviewer that angle-resolved measurements are important to confirm the symmetry. And it is our hope that publication of our work will stimulate such and other measurements, which will expand the research frontier, in our opinion.

I think a more fruitful debate one should have on the fig 6 data is to ask ourselves how the antiferromagnetism and superconductivity co-exist on the same (multi-sheet) Fermi surface? Could it be that a nesting –driven SDW gaps most parts of the Fermi surface, decreasing the number of electrons available to form Cooper pairs while increasing the density of quasiparticles below T_c ?

To our knowledge, it is still unknown how antiferromagnetism and superconductivity coexist on the Fermi surface sheets in the Ce115 system. We speculate that the two orders share the Fermi surface; one part is responsible for magnetism and the other for superconductivity. We hope that our work will also stimulate further studies on this open question. We note, though, that if nesting gapped most parts of the Fermi surface then we might expect a Cr-like anomaly in resistivity. As evident from results of ref. 32, this is not found in the pressure-dependent resistivity of $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$.

-Also, it is hard for the reader to build his/her own confidence about the validity of the data presented here with so little info about the samples, and I urge the authors to add more detail about sample characterization in the supplementary section. For instance, how confident they are that in the single crystals they studied the distribution of Ir across the sample volume is homogeneous? In other words, we don't have Rh-rich vs Ir-rich parts in the powder? What precautions have they taken to characterize their samples and how they picked what looks like the most homogeneous ones? My own experience with both growing and characterizing doped 115 crystals leads me to believe that the doping distribution we systematically see with SEM is not limited to the surface and that the 115s especially around 50% form inherently inhomogeneous crystals.

Our sample is the same used in the previous NQR measurements (ref. 19). Indeed, substitution of Ir for Rh results in randomness of the crystals as evidenced by the broadening of NQR spectrum. However, there is no phase separation of Rh-rich vs Ir-rich parts even in the coarsely crushed powder. This is because no excess peaks in the NMR/NQR spectrum are found and the spectrum can be reproduced by a Gaussian function. Moreover, T_1 is of single component, which also indicates that Ir is randomly distributed. These static and dynamic electronic properties strongly support high homogeneity of the sample used. We should also note that dc susceptibility data of this sample is consistent with the previous report (ref. 18). We have added these statements in Methods.

Responses to Reviewer #2:

(i) The assumption that the hyperfine coupling constant A remains unchanged over the entire pressure range is uncertain. The authors take that conclusion from the evolution of A under pressure in the parent compound CeRhIn_5 . The hyperfine coupling constant strongly depends on the local environment. In CeRhIn_5 , as the authors show, the c/a -ratio increases continuously with pressure, while this is not the case for the 0.5-doped Ir one where the c/a -ratio is almost constant. Hence the local environment will differ and may lead to a change of A with pressure in $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$.

There is NO correlation between the coupling constant A and the c/a -ratio in CeRhIn_5 . The former decreases with increasing pressure but becomes constant above $P = 1$ GPa (see supplementary Fig. 1), in contrast, the latter monotonically increases with increasing pressure up to 4 GPa (see Fig. 7 in P. S. Normile *et al.*, Phys. Rev. B **72**, 184508 (2005)). Our assumption is supported by the experimental fact.

As for the c/a ratio, the data for $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ shows quite a similar trend with CeRhIn_5 except the data point at the highest pressure. As we did not make any claim based on the c/a value, we have removed the data from Fig.5.

(ii) The author repeatedly stated that Ir acts as chemical pressure and $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ resembles CeRhIn_5 under 1 GPa pressure. Such statement is incorrect and the authors should be more careful. So far, such precise matching of phase diagrams of pressure compared to chemical substitution has been seen in YbRh_2Si_2 with the 5%Ge-doped one, as well as in $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$ -series but as pointed out by Nicklas *et al.* (Phys. Rev. B **70**, 020505(R) (2004)) scaling breaks down for $x > 0.3$, indicating additional effects of Ir substitution. Such is also evident as the T-p phase diagram

of CeRh_{0.58}Ir_{0.42}In₅ (Luo et al, npj Quant. Mat. 3, 6 (2018)) does not fit into this scaling.

Although early resistivity measurements (ref. 32) on CeRh_{1-x}Ir_xIn₅ suggested that there might be additional effects of Ir substitution for $x > 0.3$, a more recent systematic study (ref. 44) on $x = 0.42$, which is very close to our composition, has shown that the scaling between pressure and Ir substitution works rather well (see supplemental Fig.S1 of ref.44).

The report In(1) transition for CeRh_{0.5}Ir_{0.5}In₅ is slightly smaller than what is found here, yielding 6.24 MHz (Phys. Rev. B 70, 014511 (2004)). The frequency here corresponds more to a value associated with $x = 0.4$.

We are confused as to where the number 6.24 MHz came from. In this paper, we performed NQR measurements with the same batch of the crystals used in the previous NQR study (ref. 19). We confirmed that the NQR frequency is the same as the previous result within experimental error.

In addition, it is unclear why the entire manuscript presents only the In(1) $I = \pm 1/2 \rightarrow \pm 3/2$ transition line (not even in the supplemental material anything else is found). The In(1) line has local tetragonal symmetry and hence the NQR spectra for the In(1) site consist of four equally spaced transition lines. Why the authors were not using (and presenting) the full In(1) spectrum in order to determine the In(1) base frequency and resulting shifts upon applied pressure. The jump itself should be visible in all higher harmonics.

As the reviewer pointed out correctly, for the In(1) site, four equally spaced NQR transition lines can be observed (see Fig.1 in ref.19). We used the $I = \pm 1/2 \rightarrow \pm 3/2$ transition line to determine ν_Q because it is the most direct measurement to obtain ν_Q . In addition, in the experiment under pressure and low temperature, measurements using the lowest frequency of the In(1) NQR is the best to avoid possible heating effect due to RF pulses.

We speculate that the reviewer might have been concerned about a possible change of the asymmetry parameter η under pressure. In fact, we measured the 1st and 4th transitions ($\pm 7/2 \rightarrow \pm 9/2$) at $P = 1.12$ GPa. The 4th transition peak frequency always equals to 4 times the 1st transition frequency, which assures $\eta = 0$. This indicates that measurements of the In(1) $I = \pm 1/2 \rightarrow \pm 3/2$ transition line are enough to determine $\nu_Q(P, T)$. We added the obtained spectra as Supplementary Fig. 7 in Supplementary Information.

Figure 4(e) actually opposes the authors' claim of a more itinerant behavior for $p > 0.8$ GPa. The

In(1) frequency of CeRhIn₅ is higher than the CeIrIn₅ although in the later the 4f-electron is strongly delocalized. According to this figure one would expect the jump going downwards at $p_c = 0.8$ GPa. The authors should put their calculations in the right context.

Because of lattice contribution (see Fig.2 in ref. 18 and Fig.2 in ref.19), a direct comparison of ν_Q between CeRhIn₅ and CeIrIn₅ is not useful to obtain the information about 4f-electron delocalization. The purpose of the original Fig. 4e (new Fig. 5b) is simply to show that ν_Q of CeRh(Ir)In₅ is larger than LaRh(Ir)In₅, assuring that delocalization of 4f-electron increases ν_Q .

As seen in Fig.4c, the slope of $\nu_Q(P)$ changes below and above P^* as opposed to a monotonically increasing lattice contribution, indicating that a jump of ν_Q is actually due to a sudden increase of the Kondo hybridization at P^* .

More intriguing, the width of the small FS to larger FS transition is constant independent of the temperature. Such points to a real transition line in the T-p phase diagram, not a cross-over like behavior as for instance seen in YbRh₂Si₂. This is rather unusual. At higher temperatures thermal fluctuations would lead to a broadening of the small FS to large FS transition. The authors should explain such artifact.

We agree with the reviewer that, in principle, at higher temperatures thermal fluctuations would lead to a broadening of the small to large FS transition. As we answered to reviewer#1, unfortunately it is not possible to infer how much ν_Q changes due to Kondo hybridization at high temperatures as the lattice contribution to the T -dependence of $\nu_Q(T)$ is large there. Our data are for low temperatures in a narrow temperature range, from which it is difficult to discuss about any thermal broadening effect. Nonetheless, our data do not contradict to the above expectation. Namely, the transition width is about 0.3 GPa for $T = 4.2$ K which is larger than 0.2 GPa for $T = 1.6$ K. We have too few data points for $T = 0.3$ K to infer a transition width.

This is the first report of a jump in the NQR frequency linked to a Kondo breakdown QCP. The parent compound CeRhIn₅ exhibits a Kondo breakdown QCP at $p_c = 2.3$ GPa, however no group ever presented a frequency jump. While this might have several reasons, in order to point out that such jump is a signature of a Kondo breakdown QCP, the authors should have verified it with CeRhIn₅ to strengthen their claim, despite the addition work it causes.

In fact, a previous NMR measurement on CeRhIn₅ under pressure also suggested a ν_Q jump at $P_c^* = 1.75$ GPa (ref.34).

Moreover, what speaks against a Kondo breakdown QCP at $p_c = 0.8$ GPa is that the rDOS is not enhanced as one would expect in such scenario.

To our knowledge, rDOS does not necessarily need to be enhanced at Kondo breakdown QCP.

A further problem of the manuscript in this version is that it is unclear whether the authors claim that the first QCP at $p_c = 0.8$ GPa is due to a small FS to large FS transition or a Lifshitz transition.

We wish to stress that we found a small to large Fermi surface transition experimentally. This is for certain. As to how to interpret the finding, we believe it is most likely due to Kondo breakdown. However, at the moment, one cannot completely exclude the possibility of a more general mechanism for the Fermi surface change. That is why we state in the text that a Lifshitz transition is also consistent with our observations.

The subsequent appearance of a Kondo breakdown (or small FS to large FS) transition in this compound is not new and has been reported for $\text{CeRh}_{0.58}\text{Ir}_{0.42}\text{In}_5$ (Luo et al, npj Quant. Mat. 3, 6 (2018)) already. The discussion on the SC state is a marginal contribution to the manuscript and has more speculative character.

In summary, the manuscript contains some interesting observations and ideas. However, so far they seem to be more specific to this material and hence will not enrich the field in a wide context. The presence of two types of QCPs is not new and has been already observed in a similar substituted sample. For this reason, I cannot recommend the manuscript to be published in Communications Physics.

We respectfully disagree with the assessments.

Ref. 44 showed two quantum critical points at $T = 0$ after suppressing superconductivity by magnetic field. Those measurements, consequently, could not make any conclusion about the nature of superconductivity. Our result is the first observation at zero field and also extending to finite temperatures (T^* line in new Fig.8). In addition, we revealed for the first time the unconventional superconducting properties due to a synergy effect of two QCPs. The physics should also apply to other strongly correlated systems and have a wider impact on condensed matter physics.

Responses to Reviewer #3:

Recommendation:

This work is of high quality in general, however some points should be addressed (see below). I recommend publication as a communications physics after revision.

We appreciate the reviewer for the positive recommendation.

Comments/Questions:

1. The authors discuss the change of the FS volume at P_c^* from the precise pressure dependence of In NQR frequency. They pointed out that the change of the EFG parameter at the In(1) site is due to the change of the on-site electronic state. As the authors mentioned briefly in the DISCUSSION, small amount of Ce-valence change can also be another possible reason. Did the authors calculate the EFG by HiLAPW when the Ce-valence slightly changes? The simulation of the valence-dependence of the EFG strongly supports your claim.

The spirit of first-principles calculations is to determine the spatially distributed electron density in a self-consistent manner, without any input by hand. Any assumption for Ce-valence to determine EFG is physically meaningless. Therefore, we cannot calculate/simulate the EFG by HiLAPW assuming a changed Ce-valence.

2. The authors found that the coupling constant of $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ is very close to that of CeRhIn_5 under 1 GPa, and claimed that Ir substitution with 0.5 is equivalent to the application of a physical pressure “greater than” 1 GPa to CeRhIn_5 (Supplementary). Did the authors measure the pressure dependence of the NMR Knight shift?

The coupling constant closely connects to a local spin density at In(1) site. For example, Kambe et al. reported that the temperature dependent term of the coupling constant below T_K relates to the temperature dependence of the density of state. (PRB 81, 140405R(2010)). Therefore, I guess the coupling constant also changes at around P_c^* .

We did not measure Knight shift under pressure. It would be interesting to perform such measurements in the future, but for the time being, it does not affect our conclusion.

We appreciate the reviewer for pointing out the important paper (Phys. Rev. B **81**, 140405R (2010).) by Kambe *et al* who suggested that the coupling constant at the In(1) site (correctly, their $A_{1,a}$ is related to the In(1) NQR T_1) in CeIrIn_5 does not depend on temperature. Indeed, their result strongly supports our conclusion. We have cited this paper as new ref 36, and

added a corresponding sentence in the first paragraph in page 4.

As we elaborate at length in page 4, line 18-25, the coupling constant at the In(1) site does not change around P_c^* . It was also shown in ref. 34 that the coupling constant does not change across P_c^* in CeRhIn₅.

3. The authors discuss the non-magnetic impurity effect for unconventional superconductors. This material has layered structure and Rh/Ir layer is sandwiched between CeIn(1) layers (Fig.2 (a)). So that, Ce layer is not directly affected by the present substitution. In this case, do the theoretical models for unitary scattering work as it is?

The magnitude of T_1 changes substantially in CeRh_{1-x}Ir_xIn₅ compared to the end members, which indicates that the Rh/Ir substitution directly affects the electronic states to which Ce layer contributes.

The observed anomalously high value of residual DOS (96%) is enough to suppress d -wave completely superconductivity regardless of unitary or Born scattering (ref. 65). Thus, our conclusion does not depend on theoretical models.

4. The authors mentioned the anomalously high value of $1/T_1T$ (residual DOS) in the SC state at around P_cSDW , comparing with the case for the full gap s -wave and clean d -wave superconductors. However, at just around the SDW QCP, magnetic fluctuations are enhanced, so that one can simply imagine that low-lying magnetic excitations could survive as a background of $1/T_1T$ even in the SC state. How about the case for other QCP materials or Ce115 family?

An anomalously high residual DOS (96%) in the SC state has not been found in other QCP materials such as high T_c cuprates (for review see, K. Asayama *et al.*, Progress in Nuclear Magnetic Resonance Spectroscopy **28**, 221 (1996).), the iron-pnictide superconductors: 1111 (T. Oka *et al.*, Phys. Rev. Lett. **108**, 047001 (2012).) and 122 systems (ref.5), although magnetic fluctuations are also strong there.

For the Ce115 family, no significant gapless excitations have been observed so far in the superconducting state, including CeCoIn₅ (Y. Kohori *et al.*, Phys. Rev. B **64**, 134526 (2001).) and CeIrIn₅ (ref. 27), except for the coexistent region of antiferromagnetism and superconductivity in CeRhIn₅ (ref. 47) to which our scenario may also apply.

We have added these statements in P9.

Minor points:

Introduction

The phrase “indirect exchange interaction among weakly screened localized f-electrons,” in line 22 in page 2 should be stated in line 15. In this line, the phrase should be replaced by “the RKKY-type AF order”.

We rephrased the sentences following the reviewer’s suggestion.

There are two kinds of Gamma7 CEF wave functions (Gamma7(1) and Gamma7(2)) in the tetragonal symmetry. (line 19, page 3) Which state is realized?

Gamma7(2) is realized (ref.33 and references therein).

Methods

The authors described the single crystals were moderately crushed into grains. For NMR measurements, did the authors use oriented grains for NMR measurements? There are no statements in Methods.

In Methods, we have added experimental details.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I have reviewed carefully the revised manuscript, supplemental info and the authors reply to all three reviewers. I found that they have addressed all the issues raised by all three reviewers and I do not have any further comment on the manuscript. Therefore, I recommend it for publication as is.

Reviewer #2 (Remarks to the Author):

The authors made several improvements to the manuscript but certain concerns were less well countered.

Throughout the manuscript the authors follow the same line of argument, that is, $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ is equivalent to CeRhIn_5 under applied pressure. The mere influence of Ir-substitution is the decrease of the lattice size. As pointed out in earlier work already, this is acceptable for small substitutions $x < 0.3$ [M. Nicklas et al., Phys. Rev. B 70, 020505(R) (2004)]. For higher substitutions the effect of Ir on the electronic properties cannot be simply neglected as concluded in the above report.

Point (1): The hyperfine constant $115A(1)$ in CeRhIn_5 is constant for $p > 1$ GPa being 7 kOe/ μB . This value is roughly the same for $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ at ambient pressure. The authors used the same value of $115A(1)$ for their pressure study. However, $115A(1)$ of CeIrIn_5 might have a strong pressure dependence, like for CeRhIn_5 for small pressures, i.e. $p < 1$ GPa. Hence, it is rather uncertain that the assumption of the authors regarding the constant value of $115A(1)$ over the entire pressure range is correct. Furthermore, in [C. Lin et al, Phys. Rev. B 92, 155147 (2015)] the pressure dependence of $115A(1)$ for CeRhIn_5 is not really constant for $p > 1$ GPa.

Point(2): No step-like increase of the $\text{In}(1)$ frequencies around the Kondo breakdown QCP in CeRhIn_5 has been reported, which one would expect if such step-like increase of the frequency is a hallmark of a Kondo breakdown scenario and, as the authors point out, $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ is equal to CeRhIn_5 under hydrostatic pressure. The fact that such step-like feature in the $\text{In}(1)$ frequency is missing in CeRhIn_5 is not at all discussed. On the contrary, in their reply the authors point to a manuscript on CeRhIn_5 [C. Lin et al, Phys. Rev. B 92, 155147 (2015)] where the change of electric field gradient (EFG) shows an effect upon pressure which very misleadingly points to a small change of EFG (and thus the frequency) for the $\text{In}(2)$ -site and NOT the $\text{In}(1)$ -site discussed in this manuscript. In addition as shown in Fig. 4 of the reference, the change for the frequency ν_{aa} and ν_{bb} cancels out for ν_{cc} . Moreover, the authors do not explain how this figure relates to their findings, not to mention that there is not even a "step" but a smooth transition.

Point(3): In CeIrIn_5 the frequency ν_2 of $\text{In}(2)$ shows a strong increase at around 1 GPa hinting to a change of Ce-valency [M. Yashima et al., Phys. Rev. Lett. 109, 117001 (2012)]. Note that this is well in agreement with [C. Lin et al, Phys. Rev. B 92, 155147 (2015)]. It is a pity no data is presented on the $\text{In}(1)$ site in this work. The pressure at which this increase happens coincides fairly well with the step-like change of frequency of $\text{In}(1)$ in $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$. What is the correlation between both observations and does this not provide some evidence that Ir-substitution changes more than just the lattice parameters?

The authors also addressed a few other comments only barely.

(a) Comment on reply "We are confused as to where the number 6.24 MHz came from..": there is nothing to be confused of. The number comes straight from Fig.2 from the reference [Phys. Rev. B 70, 014511 (2004)]. Although here the second harmonic (ν_2 -line) is shown (dividing the number by 2 gives the ν_1 frequency yielding 6.24 MHz). So there is a difference in frequency, and moreover, the shape is interesting. The authors show that the ν_1 -line of $\text{In}(1)$ is almost perfectly Gaussian-like, while in the reference [Phys. Rev. B 70, 014511 (2004)] the second harmonic ν_2 is plotted. This line is not at all Gaussian-like. Assuming an internal magnetic field present due to

ordering this can happen. The data are however for $T > T_N$. There seems to be more than just a frequency inconsistency with previous reported data.

(b) Comment on reply "To our knowledge, rDOS does not necessarily need to be enhanced at Kondo breakdown QCP": This requires a particular setting. In general the Fermi Surface (FS) shows an abrupt change accompanied by a tremendous mass enhancement (increase of DOS). This has been seen for several heavy Fermion compounds including CeRhIn5 [H. Shishido et al, J. Phys. Soc. Jpn. 74, 1103 (2005)] and more indirect for CeRh0.58Ir0.42In5 where it is explicitly stated "The maxima of A are about $4 \mu\Omega\cdot\text{cm}/\text{Kn}$, comparable to that in CeRhIn5 at P2, indicating that the effective mass enhancements are similar in these two systems." [Y. Luo . npj Quant. Mat. 3, 6 (2018)]. Thus the lack of an enhancement of rDOS if the authors insist on a description of a Kondo breakdown scenario implies something and the authors should clearly address this issue including what it means for a possible SC order parameter.

The reply of the authors was not satisfying and did not help in increasing confidence in their analysis. As stated in my previous report, the subsequent appearance of a Kondo breakdown (or small FS to large FS) transition in this compound is not new and has been reported for CeRh0.58Ir0.42In5 [Luo et al, npj Quant. Mat. 3, 6 (2018)] already. Here measurements are performed in zero field. Nevertheless, there has been little doubt in the community that the field experiments cannot be extrapolated to zero field. The discussion on the superconducting (SC) state remains speculative without further new input.

The observation of the step in the NQR frequency deserves further attention but so far seems to be specific to CeRh0.5Ir0.5In5. It would be more helpful if experiments on the CeRh0.58Ir0.42In5 would be added which show also such step, as well as some lower Ir concentrations (which if the authors' claim is correct that Ir acts merely as chemical pressure should show a step too). In the current stage the manuscript is just another report on a CeMIn5-compound which shows some feature with some interpretation.

In conclusion, the manuscript contains some interesting observations and ideas. However, two QCPs is not new, the SC state discussion is purely speculation and the step in the NQR frequency so far is sample specific. I cannot recommend the manuscript to be published in Communications Physics.

Reviewer #3 (Remarks to the Author):

Recommendation:

This work is of high quality in general, as mentioned the former review. And the authors respond all criticisms, so that I recommend publication as it is.

Summary:

The authors Kawasaki et al. performed NMR and NQR measurements under pressures on a heavy-fermion superconductor CeRh0.5Ir0.5In5. The paper contains interesting experimental results on an anomalous change of the electronic state at around P_c^* and an enhancement of low-lying magnetic excitations at around P_{cSDW} . The authors found an anomalous change of the NQR frequency at around P_c^* that is ascribed to the change of Fermi surface (FS) volume from the localized small FS to itinerant large FS.

They also inferred the possible odd-frequency (p-wave) singlet state in CeRh0.5Ir0.5In5 from the anomalously enhanced low-lying excitations in the SC state at around SDW-QCP (P_{cSDW}).

Responses to Reviewer #2

Throughout the manuscript the authors follow the same line of argument, that is, $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ is equivalent to CeRhIn_5 under applied pressure. The mere influence of Ir-substitution is the decrease of the lattice size. As pointed out in earlier work already, this is acceptable for small substitutions $x < 0.3$ [M. Nicklas et al., Phys. Rev. B 70, 020505(R) (2004)]. For higher substitutions the effect of Ir on the electronic properties cannot be simply neglected as concluded in the above report.

We have already addressed this issue in our previous response. Just in case, let us point out the most important fact again.

“Although early resistivity measurements (ref. 32) on $\text{CeRh}_{1-x}\text{Ir}_x\text{In}_5$ suggested that there might be additional effects of Ir substitution for $x > 0.3$, a more recent systematic study (ref. 44) on $x = 0.42$, which is very close to our composition, has shown that the scaling between pressure and Ir substitution works rather well (see supplemental Fig.S1 of ref.44).”

Point (1): The hyperfine constant $115A(1)$ in CeRhIn_5 is constant for $p > 1$ GPa being 7 kOe/ μB . This value is roughly the same for $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ at ambient pressure. The authors used the same value of $115A(1)$ for their pressure study. However, $115A(1)$ of CeIrIn_5 might have a strong pressure dependence, like for CeRhIn_5 for small pressures, i.e. $p < 1$ GPa. Hence, it is rather uncertain that the assumption of the authors regarding the constant value of $115A(1)$ over the entire pressure range is correct. Furthermore, in [C. Lin et al, Phys. Rev. B 92, 155147 (2015)] the pressure dependence of $115A(1)$ for CeRhIn_5 is not really constant for $p > 1$ GPa.

First, the issue about the hyperfine coupling constant in our study is whether it changes across the antiferromagnetic quantum critical point (QCP) or not. As one can see clearly in Fig.1, CeIrIn_5 has NO antiferromagnetic QCP under pressure. Therefore, even if the coupling constant of CeIrIn_5 changes under pressure, there is no point to compare with our study.

Second, to be precise, it is the pressure range of $P = 1.2$ GPa or higher in CeRhIn_5 that should be the case of $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$. As can be seen in Supplementary Figure 1, the coupling constant does not change at all there.

Point(2): No step-like increase of the $\text{In}(1)$ frequencies around the Kondo breakdown QCP in CeRhIn_5 has been reported, which one would expect if such step-like increase of the frequency is a hallmark of a Kondo breakdown scenario and, as the authors point out, $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$ is

equal to CeRhIn₅ under hydrostatic pressure. The fact that such step-like feature in the In(1) frequency is missing in CeRhIn₅ is not at all discussed. On the contrary, in their reply the authors point to a manuscript on CeRhIn₅ [C. Lin et al, Phys. Rev. B 92, 155147 (2015)] where the change of electric field gradient (EFG) shows an effect upon pressure which very misleading points to a small change of EFG (and thus the frequency) for the In(2)-site and NOT the In(1)-site discussed in this manuscript. In addition as shown in Fig. 4 of the reference, the change for the frequency ν_{aa} and ν_{bb} cancels out for ν_{cc} . Moreover, the authors do not explain how this figure relates to their findings, not to mention that there is not even a “step” but a smooth transition.

In ref 34, a detailed pressure dependence of In(1) frequency in CeRhIn₅ was not conducted. Also, the In(1) frequency was indirectly determined by NMR, not directly by NQR. In general, it is much more difficult to obtain a precise value of the NQR frequency by NMR technique. This is why it was not clear until we performed the NQR experiment.

However, as we previously responded, the change of the In(2) frequency previously reported at P^* in CeRhIn₅ is consistent with our result.

We have added one sentence in page 5 just before the last paragraph to mention the above statements.

Point(3): In CeIrIn₅ the frequency ν_2 of In(2) shows strong increase at around 1GPa hinting to a change of Ce-valency [M. Yashima et al., Phys. Rev. Lett. 109, 117001 (2012)]. Note that this is well in agreement with [C. Lin et al, Phys. Rev. B 92, 155147 (2015)]. It is a pity no data is presented on the In(1) site in this work. The pressure at which this increase happens coincides fairly well with the step-like change of frequency of In(1) in CeRh_{0.5}Ir_{0.5}In₅. What is the correlation between both observations and does this not provides some evidence that Ir-substitution changes more than just the lattice parameters?

As we responded to Point (1), it does not make much sense to compare CeRh_{0.5}Ir_{0.5}In₅ with CeIrIn₅.

The authors also addressed a few other comments only barely.

(a) Comment on reply “We are confused as to where the number 6.24 MHz came from.”: there is nothing to be confused of. The number comes straight from Fig.2 from the reference [Phys. Rev. B 70, 014511 (2004)]. Although here the second harmonic (ν_2 -line) is shown (dividing the number by 2 gives the ν_1 frequency yielding 6.24 MHz). So there is a difference in frequency, and moreover, the shape is interesting. The authors show that the ν_1 -line of In(1) is almost perfect Gaussian-like, while in reference [Phys. Rev. B 70, 014511 (2004)] the second harmonic

ν_2 is plotted. This line is not at all Gaussian-like. Assuming an internal magnetic field present due to ordering this can happen. The data are however for $T > T_N$. There seems to be more than just a frequency inconsistency with previous reported data.

We believe that this is an error in reading out a number from the figure in Ref.19. Let us assure again that the results are in good agreement.

We agree that the shape of $2\nu_Q$ transition line reported in Ref.19 is not perfect Gaussian-like. That is why the measurement of the $1\nu_Q$ transition line is valuable which is most direct and accurate.

(b) Comment on reply “To our knowledge, rDOS does not necessarily need to be enhanced at Kondo breakdown QCP”: This requires a particular setting. In general the Fermi Surface (FS) shows an abrupt change accompanied by a tremendous mass enhancement (increase of DOS). This has been seen for several heavy Fermion compounds including CeRhIn₅ [H. Shishido et al, J. Phys. Soc. Jpn. 74, 1103 (2005)] and more indirect for CeRh_{0.58}Ir_{0.42}In₅ where it is explicitly stated “The maxima of A are about $4 \mu\Omega\cdot\text{cm}/\text{K}$, comparable to that in CeRhIn₅ at P_2 , indicating that the effective mass enhancements are similar in these two systems.” [Y. Luo . npj Quant. Mat. 3, 6 (2018)]. Thus the lack of an enhancement of rDOS if the authors insist on a description of a Kondo breakdown scenario implies something and the authors should clearly address this issue including what it means for a possible SC order parameter.

An enhancement of the effective mass in CeRhIn₅ (ref.20) and CeRh_{0.58}Ir_{0.42}In₅ (ref.44) around the QCP was observed in the normal state. On the other hand, we are dealing with rDOS in the superconducting state. Therefore, the results from two kinds of completely different measurements should not be compared directly. Namely, an enhancement of the effective mass in the normal state does not necessarily mean that a rDOS needs to appear in the superconducting state.

The reply of the authors was not satisfying and did not help in increasing confidence in their analysis. As stated in my previous report, the subsequent appearance of a Kondo breakdown (or small FS to large FS) transition in this compound is not new and has been reported for CeRh_{0.58}Ir_{0.42}In₅ [Luo et al, npj Quant. Mat. 3, 6 (2018)] already. Here measurements are performed in zero field. Nevertheless, there has been little doubt in the community that the field experiments cannot be extrapolated to zero field. The discussion on the superconducting (SC) state remains speculative without further new input.

The observation of the step in the NQR frequency deserves further attention but so far seems to

be specific to $\text{CeRh}_{0.5}\text{Ir}_{0.5}\text{In}_5$. It would be more helpful if experiments on the $\text{CeRh}_{0.58}\text{Ir}_{0.42}\text{In}_5$ would be added which show also such step, as well as some lower Ir concentrations (which if the authors' claim is correct that Ir acts merely as chemical pressure should show a step too). In the current stage the manuscript is just another report on a CeMIn_5 -compound which shows some feature with some interpretation.

In conclusion, the manuscript contains some interesting observations and ideas. However, two QCPs is not new, the SC state discussion is purely speculation and the step in the NQR frequency so far is sample specific. I cannot recommend the manuscript to be published in Communications Physics.

This is a repeated statement of the previous report. Again, we respectfully disagree. We do not believe that repeating measurements in a sample with very similar Ir-composition will provide useful insights.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I have re-read the reviewer#2's comments and the authors' rebuttal. I remain convinced that the authors have addressed to the best of their ability the points raised by the reviewer#2. What I found new and interesting in this paper, enough to warrant publication in Communications Physics, is the fact that the authors show some clear dissociation between the Kondo breakdown and the antiferromagnetic QCP and that is a big deal! From our knowledge of other heavy fermions systems, the two are expected to be concomitant and in this compound they are simply not. These results will surely motivate more research, both experimental and theoretical, so it should be disseminated widely.