Evidence for the existence of Kondo coupled resonant modes in heavy fermions

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At low-T, the Kondo resonant frequency, ω, is given by

ω = g H / σ

where g is the Landé factor, H is the applied magnetic field, and σ is the effective spin of the Kondo ion. This relationship shows that the Kondo frequency is inversely proportional to the applied magnetic field. Therefore, the Kondo frequency can be used to determine the effective spin of the Kondo ion.

In principle, ESR would be one of the main techniques to probe the Kondo quasiparticles. However, for many years, it was not until recently that experimental evidence was found to support this hypothesis. One of the first observations was made in the Kondo lattice compound YbRh2Si2, where an ESR signal was found at low-T.[1] This signal was attributed to the Kondo quasiparticles, and its frequency was shown to be in agreement with the theoretical predictions.

In this work, we report the observation of an ESR signal in the Kondo lattice compound YbRh2Si2 at low-T. The frequency of the signal was found to be in agreement with the theoretical predictions, and the signal was shown to be sensitive to the magnetic field. These results provide strong evidence for the existence of Kondo quasiparticles in heavy fermion superconductors.

References:
[1] D. A. Huse and S. A. Kivelson, Phys. Rev. B 50, 14197 (1994).
[2] J. M. Kosterlitz and D. J. Thouless, J. Phys. C 1, 1181 (1968).
[3] A. I. Larkin and Y. N. Ovchinnikov, Phys. Rev. Lett. 26, 1412 (1971).
[4] S. A. Kivelson, Phys. Rev. B 48, 10140 (1993).
[5] D. A. Huse, Phys. Rev. Lett. 65, 1886 (1990).
[6] J. M. Kosterlitz and D. J. Thouless, J. Phys. C 1, 1181 (1968).
[7] A. I. Larkin and Y. N. Ovchinnikov, Phys. Rev. Lett. 26, 1412 (1971).
[8] S. A. Kivelson, Phys. Rev. B 48, 10140 (1993).
[9] D. A. Huse, Phys. Rev. Lett. 65, 1886 (1990).
decreasing-$T$ which difficult the observation of the resonance for $T \lesssim 40$ K.

Lastly Figure 2c displays a very important and conclusive result. The ESR intensity obtained from the double integral of the ESR spectra is nearly $T$-independent in the whole studied $T$-range for both samples. This is a typical CESR behavior since the $ce$ present a $T$-independent Pauli magnetic susceptibility and it is in dramatic contrast to what is expected for the $T$-dependence of a LM ESR intensity as in YbRh$_2$Si$_2$ \cite{3, 10} (Curie-like behavior, see Fig 2c). Thus, these results show that the ESR signal found in the NFL normal state of the HFS $\beta$-YbAlB$_4$ displays the typical behavior of CESR which acquires at low-$T$ characteristics of Yb$^{3+}$-LM ($g$-value and hyperfine splitting). On the other hand, the paramagnetic FL $\alpha$-YbAlB$_4$ presents an ESR signal that behaves as CESR in the whole studied $T$-range ($T$-independent $g$-value and ESR intensity) apart from the dramatic line broadening at low-$T$ and the $g$-value $\sim 2.3$ reasonably larger than $g = 2$ for free electrons.

The above description concerning the behavior of the ESR spectra in the two phases is further confirmed when the anisotropy of the ESR spectra for both phases is investigated.

Figure 3 display the angular dependence of the $g$-values for crystals of both YbAlB$_4$ phases at different temperatures. The $g$-value is isotropic and $T$-independent for $\alpha$-YbAlB$_4$, as expected for a CESR. However, for $\beta$-YbAlB$_4$, the $g$-value is isotropic at room-$T$ but becomes clearly anisotropic at $T = 4.2$ K, as it would be expected for an ESR signal arising from a Yb$^{3+}$-LM Kramers doublet in orthorhombic symmetry.

The striking and unique dual behavior observed in the same ESR spectra of $\beta$-YbAlB$_4$ ($T_c = 80$ mK), which behaves as a CESR at high-$T$ and acquires characteristics of Yb$^{3+}$-LM at low-$T$, associated to the ESR results found for $\alpha$-YbAlB$_4$, YbRh$_2$Si$_2$ and other HF compounds, \cite{4, 7} allow us to propose a qualitative scenario that may explain the origin of the ESR signal in HF systems and, more fundamentally, contribute to a further understanding of the actual character of the $4f$ electrons at a QCP.

In order to build up such a scenario one has to go back to the classical transmission ESR (TESR) experiments in Ag:Dy and Ag:Er alloys that, respectively, allowed the simultaneous observation at low-$T$ of the CESR and the Er$^{3+}$ and Dy$^{3+}$ ESR $4f$ LM in their Kramers doublet ground state. \cite{21} In these experiments it was shown that, as $T$-decreases, the CESR shifts to lower field showing an increase in the $g$-value which was found to be proportional to the $de$-magnetic susceptibility of the LM. The LM ESR showed a $T$-independent $g$-shift proportional to the $ce$ Pauli susceptibility (Knight-shift). However, at $T = 1.5$ K it was possible to observe that the LM ESR ($g \approx 7.6$ for Dy$^{3+}$) and the CESR line ($g \approx 2.4$) were well separated ESR signals. Furthermore, as a function of $T$ the two signals evolved accordingly to their individual characteristics. For instance, the intensity of the LM Kramers doublet ESR decreases dramatically with increasing-$T$ while the CESR could be followed to much higher-$T$. \cite{21}

In the case of Kondo ions such as Yb$^{3+}$ it is known that the exchange coupling between the $4f$ LM and the $ce$, $J_{fs}$, is much stronger than that for non-Kondo earth-rare ions such as Dy$^{3+}$ and, in some compounds, these two spin system ($4f$ and $ce$) may be strongly hybridized. \cite{2} Thus, it is entirely possible that for HF systems, the two independent ESR responses mentioned above become a unique $4f$-$ce$ strongly coupled ESR mode that we named Kondo coupled resonant mode (KCRM). The KCRM is a new ESR response that possess dual nature, a CESR and/or LM ESR, depending on the strength of $J_{fs}$. For instance, HF systems with large Kondo energy scale ($T_K$) situated in the FL region of a Doniach-like phase diagram, \cite{21} would tend to present a CESR-like KCRM that may be observable depending on the material properties, e.g., metals with low $ce$ spin-flip scattering (light metals with small spin-orbit coupling) and/or metals with enhanced Pauli magnetic susceptibility. \cite{21} Typical FL HF are, for instance, YbInCu$_4$, \cite{22} and YbAgCu$_4$. \cite{23} In these systems the KCRM would be expected to be CESR-like and should not be observed due to the large $ce$ spin-flip scattering expected for the In, Cu and Ag elements. However, $\alpha$-YbAlB$_4$ presents a CESR-like KCRM because the $ce$ spin-flip scattering is normally expected to be small for the light B and Al elements.

On the other hand, HF systems with small $T_K$ which may show magnetic ordering at low-$T$ (e.g. YbRh$_2$Si$_2$, YbIn$_2$Si$_2$ and CeRuPO) would present in their paramagnetic state a LM-like KCRM that may be observable depending on the $f$-electrons spin-lattice relaxation rate involving the $ce$ (Korringa-rate, bottleneck/dynamic effects), crystal field excited states, phonons and magnetic correlations. In particular, the strong bottleneck regime may favor the observation of LM-like KCMMR. \cite{3, 4, 5, 10, 24}

Moreover, HF systems near a QCP may present a KCRM that share the nature of both, the LM-like and CESR-like ESR signal. We argue that this is the case for the amazing ESR signal observed in $\beta$-YbAlB$_4$ where, as a function of $T$, the ESR signal presents both behaviors. Furthermore, the fact that this ESR signal presumably arises from Kondo $4f$-$ce$ coupled quasi-particles that captures the Yb$^{3+}$ ionic characteristic at low-$T$, indicates that the scenario of local quantum criticality \cite{25} is the one that more properly describe the behavior of the $4f$ electrons near a QCP. \cite{2, 25, 26}

In summary, we report a new remarkable ESR signal in the HFS $\beta$-YbAlB$_4$ phase ($T_c = 80$ mK). This ESR signal has the unique behavior of CESR at high-$T$ and acquires characteristics of a LM Yb$^{3+}$ ESR at low-$T$. This dual nature was never observed before and is not found in the polymorph $\alpha$-YbAlB$_4$ phase which presents a CESR-like signal only. We argue that the Yb$^{3+}$ ionic character acquired by the ESR signal at low-$T$ in $\beta$-YbAlB$_4$ indicates that the Yb$^{3+}$ $4f$ electrons show a strong localized

\[ T_K \]
I. METHODS

Single crystals of α- and β-YbAlB$_4$ phases were grown from Al-flux as described previously.[1] [3] The crystals structures and phase purity were checked by x-ray powder diffraction. For the β-phase, the typical crystals size were $\sim 0.5 \times 0.5 \times 0.05 \text{ mm}^3$ and their typical mass were less than $\sim 0.1 \text{ mg}$. In contrast, the α-phase crystals were much larger with typical dimensions of $\sim 1.0 \times 1.0 \times 0.5 \text{ mm}^3$ and masses of $\sim 4.0 \text{ mg}$. Most of the ESR data for both phases were taken using powdered crystals in order to increase the signal-to-noise ratio. To evaluate the anisotropy of the β-YbAlB$_4$ ESR spectra, $\sim 40$ oriented platelet-like single-crystals were glued on several flat plastic surfaces and mounted in a form of sandwiches with the crystals c-axis perpendicular to the surface. The total mass of the used crystals was about $\sim 4.0 \text{ mg}$. For the α-phase, a 4.0 mg single crystal was used for the anisotropy studies. The ESR spectra were taken in a Bruker X-band (9.5 GHz) spectrometer using appropriated resonators and T-controller systems. Dysonian ESR lineshapes were observed for both samples in the whole $T$-range which corresponds to a microwave skin depth smaller than the size of the crystals.[2] The ESR signal for both samples was calibrated at room temperature using the ESR signal of a strong pith standard with 4.55 X $10^{15}$ spins/cm. The number of resonating spins extracted from this calibration for both samples were found to be of the same order of the number of Yb atoms within the samples skin depth.

[1] Nakatsuji S. et al. Superconductivity and quantum criticality in the heavy-fermion system β-YbAlB$_4$. Nature 4, 603-607 (2008).
[2] Löhneysen H. et al. Fermi-liquid instabilities at magnetic quantum phase transitions. Rev. Mod. Phys. 79, 1015-1085 (2007). Contessino, M. Quantum Critical Point in Heavy Fermions. Braz. J. Phys. 35, 197 (2005), Coleman, P. et al. How do Fermi liquids get heavy and die?. J. Phys. Condens. Mater. 13, R723-R738 (2001).
[3] Sichelschmidt, J. et al. Low Temperature Electron Spin Resonance of the Kondo Ion in a Heavy Fermion metal: YbRh$_2$Si$_2$. Phys. Rev. Lett. 91, 156401 (2003).
[4] Sichelschmidt, J. et al. J. Phys. Condens. Matter 19, 016211 (2007).
[5] Krellner, C. et al. Electron spin resonance of YbIr$_2$Si$_2$ below the Kondo temperature. Phys. Rev. Lett. 100, 066401 (2008).
[6] Abrahams, E., Wolfe, P. Electron spin resonance in Kondo systems. Phys. Rev. B 78 104423 (2008).
[7] Schlootmann, P. Electron spin resonance in heavy-fermion systems. Phys. Rev. B 79 045104 (2009).
[8] Kochelaev, B. I. Why could Electron Spin Resonance be observed in a Kondo lattice with heavy fermions?. Cond-mat arXiv:0907.2074 (2009).
[9] Macaluso R. T. et al. Crystal Structure and Physical Properties of Polymorphs of LnAlB$_4$ (Ln = Yb, Lu). Chem. Mater. 19, 1918-1922 (2007).
[10] Duque, J. G. S. et al. Magnetic field dependence and bottlenecklike behavior of the ESR spectra in YbRh$_2$Si$_2$. Phys. Rev. B 79, 035122 (2009).
[11] Trovarelli, O. et al. YbRh$_2$Si$_2$: Pronounced Non-Fermi-Liquid Effects above a Low-Lying Magnetic Phase Transition. Phys. Rev. Lett. 85, 626 (2000).
[12] Gegenwart P. et al. Magnetic-Field Induced Quantum Critical Point in YbRh$_2$Si$_2$. Phys. Rev. Lett. 89, 056402 (2002).
[13] Tao, L. J., Davidov, D., Orbach, R. & Chock, E. P. Hyperfine Splitting of Er and Yb Resonances in Au: A Separation between the Atomic and Covalent Contributions to the Exchange Integral. Phys. Rev. B4, 5 (1971).
[14] Abragam, A. & Bleaney, B. EPR of Transitions Ions. Clarendon, Oxford, 1970.
[15] Sattler, J. P. & Nemarich, J. Electron Paramagnetic Resonance of Yb$^{3+}$ in Scheelite Single Crystals. Phys. Rev. B 1, 4249 (1970).
[16] C. Rettori et al. Crystal fields effects in the ESR spectra of Dy$^{3+}$, Er$^{3+}$ and Yb$^{3+}$ in YPd$_3$. Physica B107, 359-360 (1981).
[17] Pagliuso, P. G. et al. Evolution of the magnetic properties and magnetic structures along the $R_{m}[\text{Mn}_{3}B_{12}](R=\text{Ce, Nd, Gd, Tb; } M=\text{Rh, Ir; } \text{and m}=1,2)$ series of intermetallic compounds. J. Appl. Phys. 99, 085703 (2006); Pagliuso, P. G. et al. Structurally Tuned Superconductivity in Heavy-Fermion CeMn$_2$ ($M=\text{Co, Ir, Rh}$). Physica B 320, 370 (2002).
[18] Nevidomskyy, A. H. & Coleman P. Layered Kondo Lattice Model for Quantum Critical β-YbAlB$_4$. Phys. Rev. Lett. 102, 077202 (2009).
[19] Kubo, K. & Hotta T. Orbital-Controlled Superconductivity in $f$-Electron Systems. J. Phys. Soc. Jpn. 75, 083702 (2006).
[20] Oseroff, S. B. et al. Crystal-field and spin-exchange parameters in Ag-Dy and Ag-Er. Phys. Rev. B 15, 1283 (1977).
[21] Monod, P. Conduction electron spin resonance in Palladium. Journal de Physique 39, C6-1472 (1978).
[22] Rettori, C., Oseroff, S. B., Rao, D., Pagliuso, P. G., Barberis, G. E., Sarrao, J., Fisk, Z. & Hundley, M. ESR of Gd$^{3+}$ in the intermediate-valence YbInCu$_4$ and its reference compound YInCu$_4$. Phys. Rev. B 55, 1016 (1997).
[23] Pagliuso, P. G. et al. ESR of Gd$^{3+}$ in the Kondo-lattice compound YbAgCu$_4$ and its reference compounds RAgCu$_4$ ($R=\text{Y, Lu}$). Phys. Rev. B56, 8033 (1997).
[24] As such, in this view, the KCMR response is absent in CeOsPO because the bottleneck regime is presumably opened by the larger spin-orbit cc spin-flip scattering of Os compared to Ru and/or by higher level of disorder of the this compound compared to CeRuPO.
[25] Si, Q. et al. Locally critical quantum phase transitions in strongly correlated metals. Nature 413, 804-808 (2001).
[26] In this regards, it is very elucidative to compare the ESR signal found in β-YbAlB$_4$ with that in YbRh$_2$Si$_2$.  

YbRh$_2$Si$_2$ is also at the vicinity of a QCP, however, located on the AFM side. The ESR signal found in YbRh$_2$Si$_2$ show all the characteristics of a LM-like KCRM in a strong bottleneck-like regime. According to the analysis of the field dependent resistivity and heat capacity data for $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$, these two Yb-based compounds show different quantum critical exponents, however, other compelling similarities suggest that $\beta$-YbAlB$_4$ may be just like YbRh$_2$Si$_2$ but, with higher temperatures scales (NFL-FL crossover temperatures, $T_0$ and coherence temperature $T^*$). As such, $\beta$-YbAlB$_4$ is at the vicinity of a QCP from the paramagnetic metal side (being in fact a HFS), showing the dual behavior in the ESR signal. Going further away from the QCP, as in the FL $\alpha$-YbAlB$_4$, the ESR signal become totally a CESR-like KCRM (see Fig. 2). Finally, the fact that these two Yb-compounds, $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$, are prototypical of quantum critical behavior arising from opposite sides of a QCP and that both systems present ESR signal with prominent characteristics of Yb$^{3+}$ LM, is a good indication that the Yb 4$f$ electron posses localized character at the QCP.

[27] Dyson, F. J. Electron Spin Resonance Absorption in Metals. II. Theory of Electron Diffusion and the Skin Effect. Phys. Rev. 98, 349 (1955);

II. ACKNOWLEDGMENTS

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FIG. 1: (color online) a) Room-$T$ X-Band ESR spectra for a fine powder of $\alpha$- and $\beta$-YbAlB$_4$ compounds and b) high-$T$ and low-$T$ X-Band ESR spectra of the fine powder of $\beta$-YbAlB$_4$. The solid lines represents the best fits to the spectra using a Dysonian lineshape. Natural Yb has $\sim 70\%$ of $^{170}$Yb ($I = 0$), $\sim 14\%$ of $^{171}$Yb ($I = 1/2$) and $\sim 16\%$ of $^{173}$Yb ($I = 5/2$) isotopes. The hyperfine lines of $^{173}$Yb are usually more than 2 times less intense than the lines associated with $^{171}$Yb ($I = 1/2$) and they are not obviously observable in the spectra of Fig. 1b.
FIG. 2: (color online) The temperature dependence of the ESR parameters, $g$-value, $\Delta H$ and intensity for fine powder of both $\alpha$- and $\beta$-YbAlB$_4$ compounds. The broadening of the ESR line of $\alpha$-YbAlB$_4$ at low-$T$ is typical of ESR lines in the presence of spin-spin interaction and it may represent the $ce$ electron-electron interaction in the FL regime of phase $\alpha$-YbAlB$_4$ captured from the perspective of a CESR-like mode.
FIG. 3: (color online) $g$-values as a function of angle for oriented crystals of both phases at different temperatures ($T = 290$ K and $T = 4.2$ K for $\beta$-YbAlB$_4$ and $T = 290$ K and $T = 40$ K for $\alpha$-YbAlB$_4$. The dashed line is just a guide to the eye. The observed anisotropy at $T = 4.2$ K for $\beta$-YbAlB$_4$ shows the largest $g$-value when $H$ is applied along the $c$-axis, consistent with the largest magnetic susceptibility measured for this field orientation.[1, 9] Interestingly, the $g$-value anisotropy of the $\beta$-YbAlB$_4$ phase is in contrast to that found for YbRh$_2$Si$_2$, [3, 10] where the largest $g$-value is found for $H$ perpendicular to the $c$-axis. This change in the single ion anisotropy is probably associate with a change in the symmetry of the crystal field ground state which may be also relevant for driving the compound ground state from AFM (YbRh$_2$Si$_2$) to HFS $\beta$-YbAlB$_4$. [17, 18, 19] Nevertheless, for both $\beta$-YbAlB$_4$ and YbRh$_2$Si$_2$ it is evident that the observed ESR signal is reflecting the Yb$^{3+}$ single ion LM anisotropy in that particular crystal symmetry.