Interplay Between Kondo and Magnetic Interactions in $Pr_{0.75}Gd_{0.25}ScGeH$

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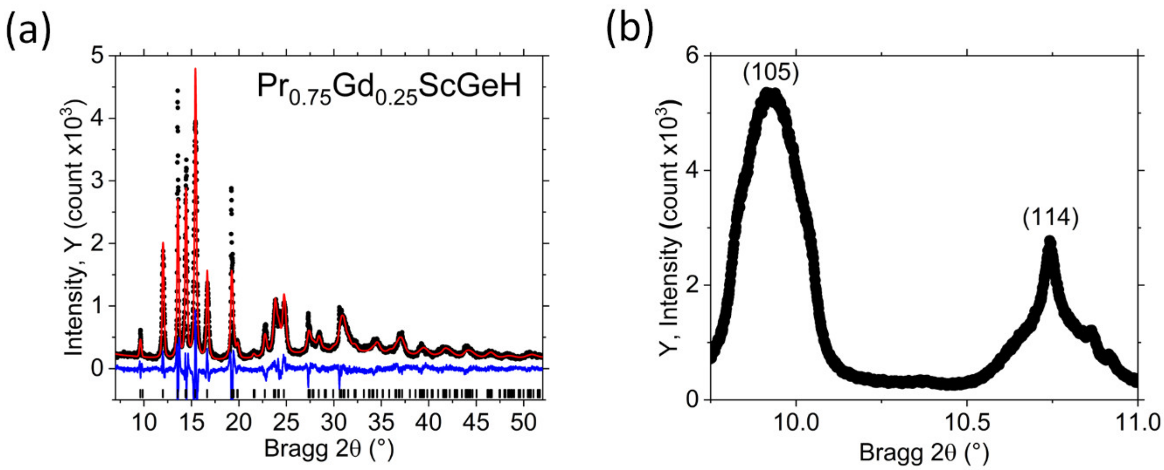

Figure S1. (a) Rietveld refinement and (b) the shapes of (105) and (114) Bragg peaks of $Pr_{0.75}Gd_{0.25}ScGeH$ hydrogenated at 150 bar. The data presented in (a) are from a laboratory powder diffractometer using Mo-Kα radiation, while the data in (b) are from APS, ANL using $\lambda = 0.457897$ Å. The noticeable mismatch of intensities in (a) is due to deteriorated crystallinity and non-analytical peak shapes after hydrogenation, exemplified in (b).

Figure S2 depicts the heat capacity of $Pr_{0.75}Gd_{0.25}ScGeH$ and LaScGeH. Contributions from magnetic and nuclear effects (exemplified by the low-temperature differences between LaScGeH and $Pr_{0.75}Gd_{0.25}ScGeH$) makes low-temperature fitting to determine the electronic specific heat, γ, impractical. Assuming that isostructural LaScGeH has the same Debye temperature, θ_D, as $Pr_{0.75}Gd_{0.25}ScGeH$, it is still possible to estimate γ for the latter as the constant difference between their C_P/T in the region (~30 – 80 K) where both the nuclear and magnetic contributions become negligible, see Eq. S1 and Figure S2.

$$\gamma_{Pr_{0.75}Gd_{0.25}ScGeH} = \gamma_{LaScGeH} + \left(\left(\frac{C_P}{T}\right)_{Pr_{0.75}Gd_{0.25}ScGeH} - \left(\frac{C_P}{T}\right)_{LaScGeH}\right) \qquad \text{(Eq. S1)}$$

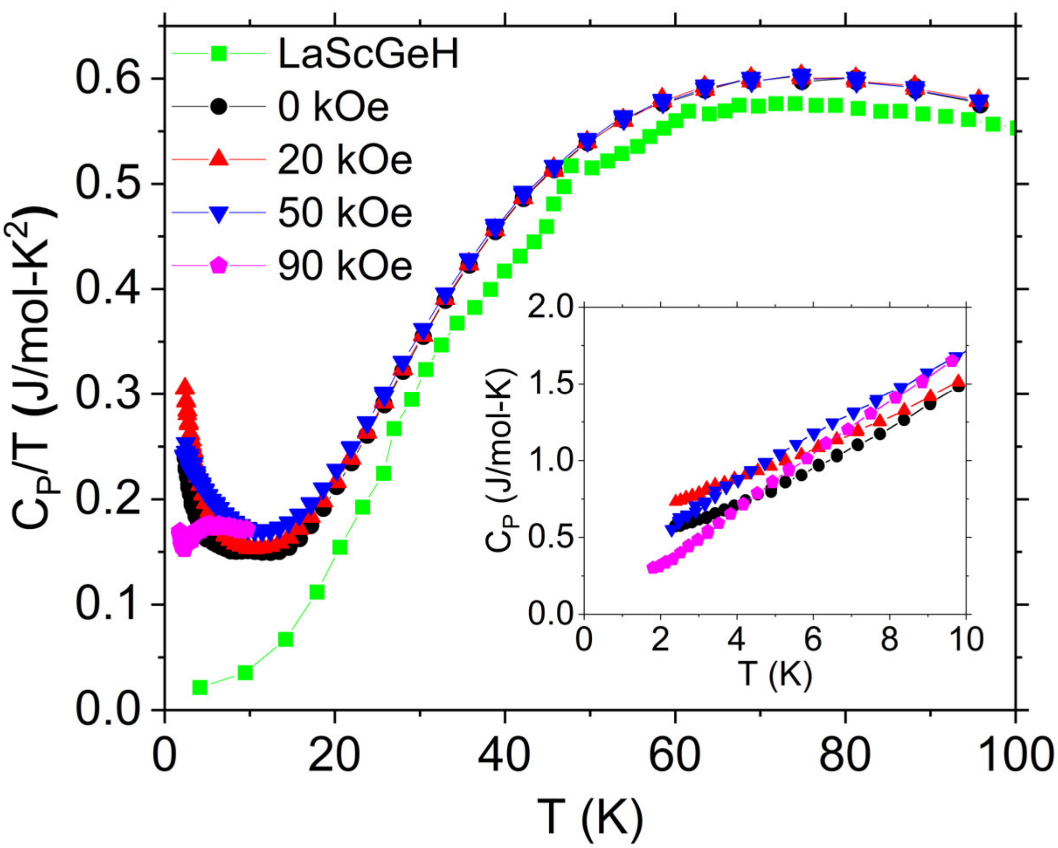

Figure S2. Heat capacity (C_p) of $Pr_{0.75}Gd_{0.25}ScGeH$ measured in zero and applied magnetic fields of 20, 50, and 90 kOe shown as C_p/T vs. temperature (T). LaScGeH data are taken from Ref [1]. Inset shows low-temperature details as C_p vs T.

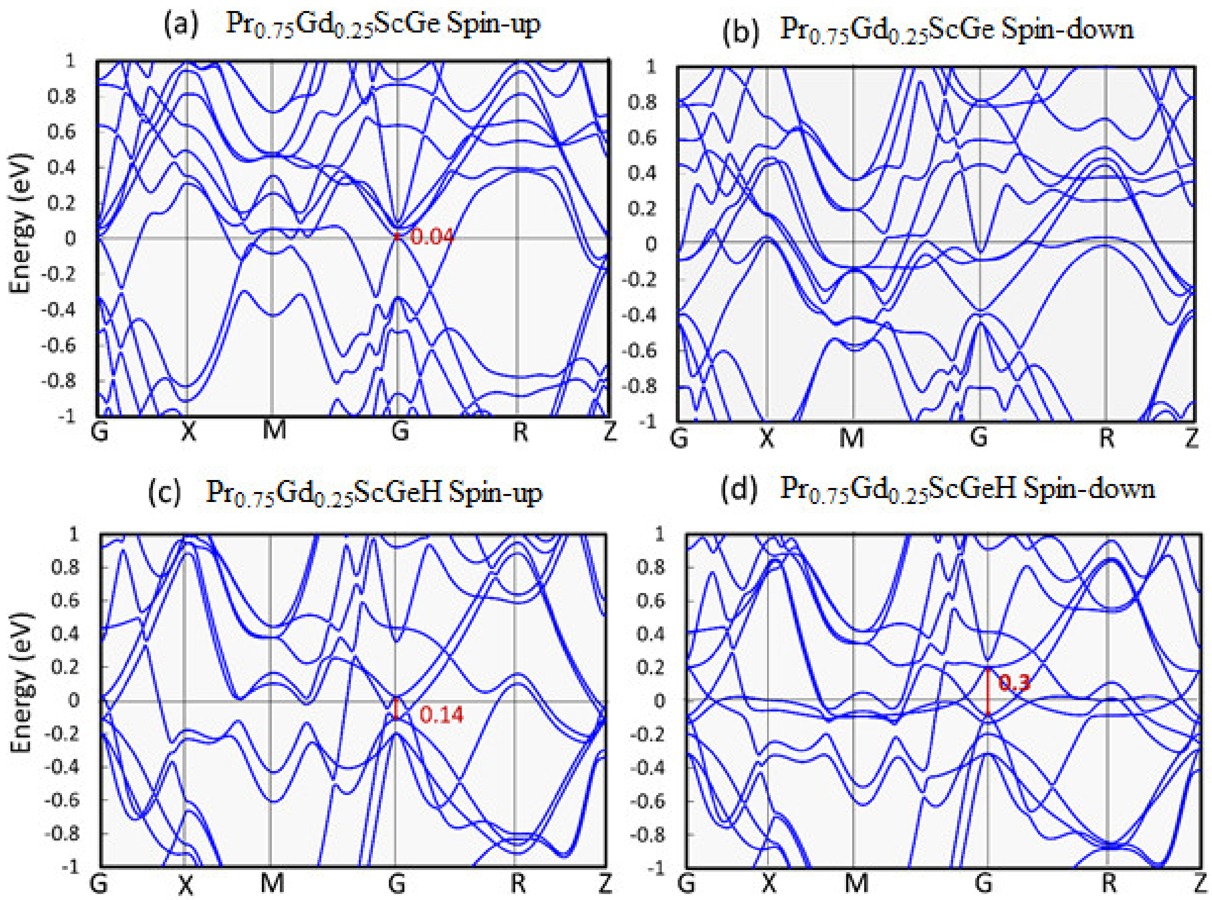

Figure S3: Spin-polarized DFT band structure of (a, b) $Pr_{0.75}Gd_{0.25}ScGe$ and (c, d) its hydride, showing both spin-up (a, c) and spin-down (b, d) electron manifolds.

The band structures, plotted in Figure S3, indicate a sharp peak of the 4*f* states close to the E_F. Additionally, the Gamma point has a very narrow gap at E_F in the majority-spin channel (Figure S3a), whereas the minority-spin channel is metallic in $Pr_{0.75}Gd_{0.25}ScGe$ (Figure S3b). As the density of states for a system is inversely proportional to the derivative of energy dispersion E_k, the sharp minority-spin 4*f*-peak in PDOS (see Figure 4b) of hydrogenated $Pr_{0.75}Gd_{0.25}ScGe$ around E_F is flat in the band structure (Figure S3b). Flat bands mean larger effective electron mass or possible heavy-fermion behavior however, they are not exactly at the E_F. Therefore, the flat 4*f* band leads to an enhanced effective electron mass without necessarily reflecting heavy-fermionlike state. To confirm this, electronic specific heat (γ) has been calculated for both $Pr_{0.75}Gd_{0.25}ScGe$ and its hydride, which are 9.35 and 13.22 mJ/mol. K^2, respectively. The theoretical value of γ for the hydride is smaller than experimental values (28.7 mJ/mol. K^2) by a factor of nearly two, whereas both theory and experiment agree for the $Pr_{0.75}Gd_{0.25}ScGe$ system (Table 1). Electron mass enhancement in rare-earth metals and the effect on specific heat were discussed by Fulde et al. [2]. Experimental electronic specific heat of Pr metal determined from heat capacity data between 1 and 6 K is larger than the DFT value by a factor of ~4, and the difference is reduced by applying a magnetic field. The reason for the mass enhancement of conduction electrons in Pr-metal (and other lanthanides) at low temperature and low magnetic field is the interaction of conduction electrons with localized 4*f* moments, which is neglected in calculating γ by DFT methods. Hence, theoretically calculated values are commonly different from experimental ones. Here, the predicted interactions of conduction and 4*f*-electrons of Pr1 atoms around E_F leads to electron mass enhancements in hydrogenated $Pr_{0.75}Gd_{0.25}ScGe$ compared to the non-hydrogenated parent. Although γ is enhanced with H-insertion, the increase is far from sufficient to classify the hydride as a heavy-fermion system.

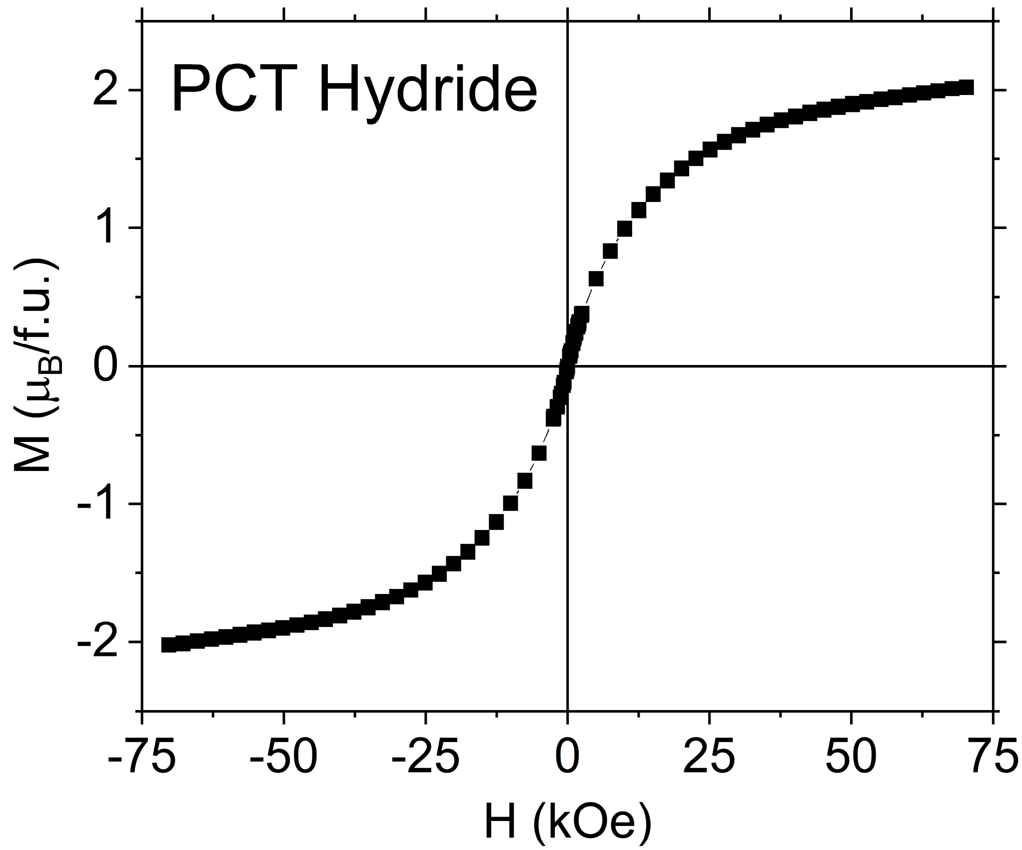

Figure S4: Magnetization of the PCTPro hydride measured as a function of applied magnetic field at T = 2 K.

References

[1] Mahon, T. *et al.* "Hydrogen insertion in the intermetallic GdScGe: A drastic reduction of the dimensionality of the magnetic and transport properties." *Inorganic Chemistry* 57, 14230 (2018).

[2] Fulde, P. & Jensen, J. "Electronic heat capacity of the rare-earth metals." *Physical Review B* 27, 4085 (1983).