Experimental Report

Proposal: **4-01-1219** **Council:** 4/2012

Title: Low-energy dynamic magnetic response in Yb1-xTmxB12 compounds with thermoelectric enhancement

This proposal is a new proposal

Researh Area: Physics

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Samples: Yb_{0.93}Tm_{0.07}^{11}B_{12}
Yb_{0.85}Tm_{0.15}^{11}B_{12}
Lu_{0.93}Tm_{0.07}^{11}B_{12}
Lu_{0.85}Tm_{0.15}^{11}B_{12}
Lu^{11}B_{12}

Instrument	Req. Days	All. Days	From	To
IN4	7	4	01/11/2012	05/11/2012

Abstract:

(Yb,Tm)B12 is a new series of compounds based on the archetype Kondo insulator YbB12. The systems with moderate concentration of Tm demonstrate a considerable enhancement of the thermoelectric power that seems to be related to the magnetic properties of the material. Recently we have studied the high-energy (above 35 meV) dynamical magnetic response in (Yb,Tm)B12. Here we propose to trace the evolution of the spectral response in Tm0.93Tm0.07B12 and Tm0.85Tm0.15B12 in the energy range below ~25 meV, where the spin gap along with cooperative near-gap-edge excitations have been observed in the undoped YbB12. For the precise quantitative subtraction of Tm contribution we further plan to measure the samples Lu0.93Tm0.07B12 and Lu0.85Tm0.15B12, whose magnetic response represents the Tm contribution only.

We estimate that 7 days of beam time in total are needed to collect the data with reasonable statistical accuracy, including the detailed study of the temperature dependence, measurements of the background, of the nonmagnetic reference compound LuB12, and of the vanadium standard.

Low-energy dynamic magnetic response in $Yb_{1-x}Tm_xB_{12}$ compounds with thermoelectric enhancement

Among strongly correlated electron systems Kondo insulators are considered promising for the design of high-performance low-temperature thermoelectric materials because of their rather high Seebeck coefficients (hundreds of μeV/K) [1]. Recently a new series of $Yb_{1-x}Tm_xB_{12}$ compounds has been synthesized [2] based on the archetype Kondo insulator YbB_{12}. It was shown [2] that partial substitution of Yb by Tm ions results in a considerable enhancement of the thermoelectric power. No such effect has been observed [3] on the substitution by Lu ions, which are isovalent to Tm but have no magnetic moment due to their complete *f*-electron shell. Therefore the thermoelectric enhancement has to be somehow related to the magnetic properties of the system.

In the present experiment we have studied the effect of Tm-substitution on the dynamical magnetic response in the series of powder samples $Yb_{1-x}Tm_xB_{12}$ (x = 0, 0.08, 0.15). For the estimation of the phonon contribution to the scattering function we have used the non-magnetic compound LuB_{12}. Tm contribution to the magnetic scattering has been determined from the measurements of the reference sample $Lu_{0.92}Tm_{0.08}B_{12}$. The powder samples (of about 4g for each composition) were prepared using ^{11}B isotope (99.3% or 99.5% enrichment for different samples). The measurements have been performed in the temperature range T = 2 - 80 K with incoming neutron energy E_i=36.32 meV (λ=1.5Å) using PG 004 monochromator.

The magnetic excitation spectra in $Yb_{1-x}Tm_xB_{12}$ (x = 0, 0.08, 0.15) at T = 2 K are shown in Fig. 1. One can see that for x > 0 the spectra are dominated by the strong contribution from crystal field (CF) transitions on Tm^{3+} ions about 7.5 meV and 14 meV. The former is located just in the spin-gap energy range for undoped YbB_{12}, whereas the latter partly overlaps with sharp peak at 15 meV in YbB_{12} spectrum.

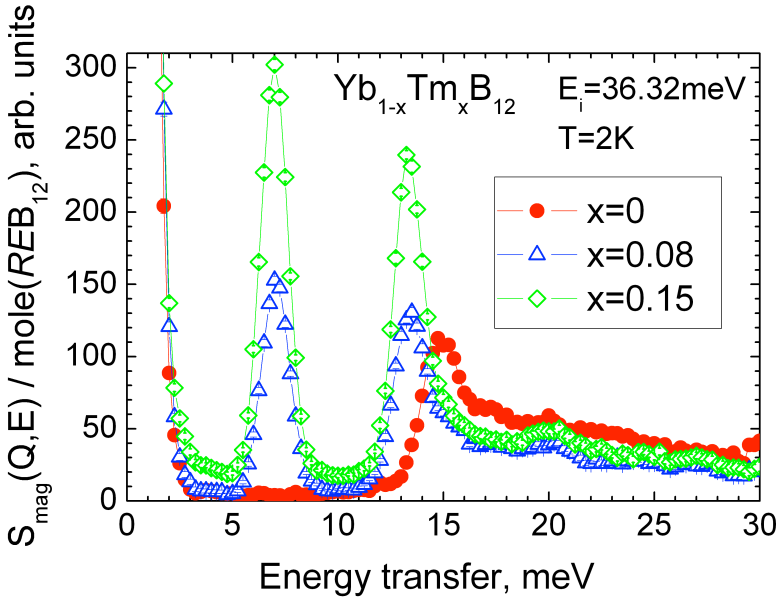

Fig. 1. Magnetic scattering spectra of $Yb_{1-x}Tm_xB_{12}$ (x=0, 0.08, 0.15) at T = 2 K.

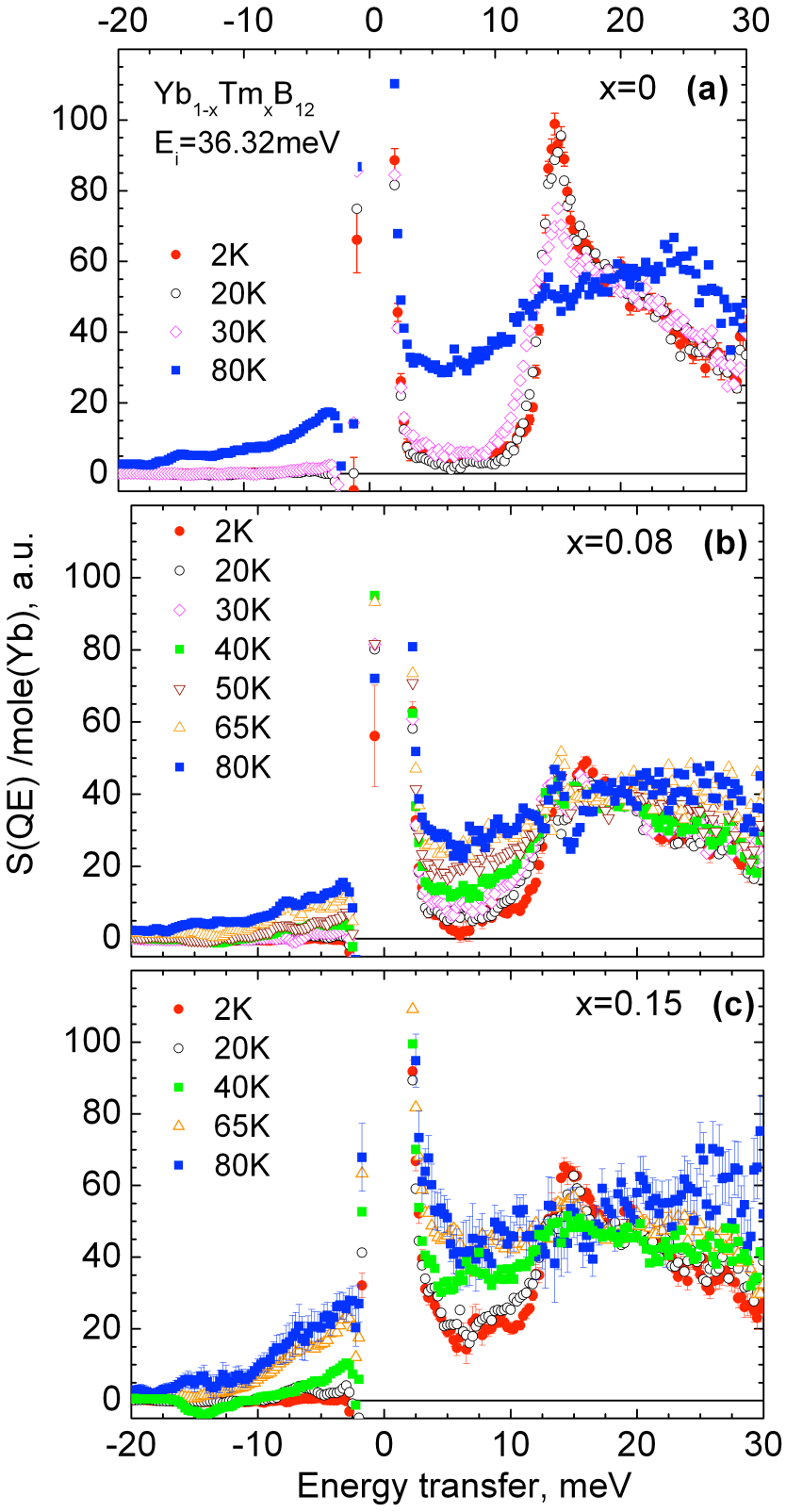

Fig. 2. Yb contribution to the magnetic scattering spectra for $Yb_{1-x}Tm_xB_{12}$, obtained by subtracting the Tm contribution for x=0 (a), 0.08 (b) and 0.15 (c) at different temperature in the range 2 K - 80 K.

For the analysis of the Yb-related magnetic signal, Tm CF contribution has been carefully subtracted using $Lu_{0.92}Tm_{0.08}B_{12}$ reference sample. The spectra after subtraction are shown in Fig. 2. Several observations can be made from these plots. The first one is that, in YbB_{12} (Fig. 2a), the narrow peak at 15 meV associated with the resonant mode (RM) is already strongly suppressed when temperature is raised to no more than 30 K, i.e. just before the spin-gap begins to fill. This is in good agreement with our recent single-crystal data [4], which demonstrated the complete suppression of the RM at q = (1/2, 1/2, 1/2) above 40 K, without a significant shift in its energy. It also confirms a previous study of the temperature evolution of the magnetic spectral response in YbB_{12} powder [5], which concluded to a filling of the gap by a quasielastic signal.

In $Yb_{0.92}Tm_{0.08}B_{12}$ (Fig. 2b) and $Yb_{0.85}Tm_{0.15}B_{12}$ (Fig. 2b) the low-temperature spectra exhibit some residual structure near 15 meV. However, its intensity represents less than 10% of that of the RM peak in undoped YbB_{12}. Furthermore, the effect appears more pronounced in the compound with the larger Tm concentration, which is clearly unphysical. It could result from uncertainties in subtracting out the phonon background, or in separating the Yb magnetic signal from the strong Tm CF excitations. However, if one estimates the magnitude of the latter effect from the residual oscillation it causes in the quasielastic part of the Yb spectrum around E = 7.5 meV (the energy of the lowest Tm^{3+} CF peak), it seems that it cannot account entirely for the observed intensity. Another possible origin could be the existence of mixed excitation modes, in a spectral range where different types of interactions are involved in the excitations probed by neutrons. In any case, the striking result is that the RM peak at 15 meV is essentially suppressed by Tm substitution for such low Tm concentrations as x = 0.15, and even x = 0.08.

Lastly, the spin-gap filling behavior differs depending on Tm concentration. Substituting 8% Tm results in a visible decrease of the temperature at which this process takes places. With 15% Tm, the spectrum changes qualitatively: even at the lowest temperature, T = 2 K, the gap is already partly filled, and the filling is almost completed near 40 K.

References

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