Supplemental Material: Native defects and impurity band behavior in half-Heusler thermoelectric NbFeSb

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(Dated: June 14, 2018)

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I. STRETCHED-EXPONENTIAL SPIN-LATTICE RELAXATION ANALYSIS

This work includes an analysis of the stretched-exponential NMR T_1 behavior due to paramagnetic impurities combined with a single-exponential relaxation process. A full description of this analysis procedure is as follows:

Tse and Hartmann [1] originally demonstrated that dilute paramagnetic impurities could produce a relaxation function of the form $S(t) = S_0 \exp[-(t/\tau_1)^{1/2}]$, a result which is valid in the long-time limit. This behavior is due to an inhomogeneous distribution of fluctuating fields due to random positioning of paramagnetic defects. A given nucleus thus undergoes statistical behavior corresponding locally to ordinary exponential relaxation. This superposition of nuclear responses can be written,

$$S(t) = S_0 \exp[-(t/\tau_1)^{1/2}] = S_0 \sum_i c_i \exp[-t/(T_{1s})_i], \tag{1}$$

where the c_i are a set of weighting factors corresponding to the distribution of exponential relaxation times $(T_{1s})_i$, and the summation in Eqn. (1). represents a discrete approximation to the continuous inhomogeneous distribution.

We examine the situation where this is combined with a second uniform relaxation process due to metallic electrons, which alone would produce a relaxation function $S(t) = S_0 \exp[-(t/T_{1,\mathrm{exp}})]$. Since for locally exponential processes the rates will add, we obtain the combined relaxation function,

$$S(t) = S_0 \exp[-(t/T_{1,\mathrm{exp}})] \sum_i c_i \exp[-t/(T_{1s})_i]. \tag{2}$$

To obtain the parameters for the summation in Eqn. (1), we chose seven $(T_{1s})_i$ values which were initially logarithmically distributed over 4 orders of magnitude, and fitted to the stretched exponential $\exp[-(t/\tau_1)^{1/2}]$ by performed a least squares minimization of the parameters $(T_{1s})_i$ and c_i, the result being a sum of exponentials with single effective parameter τ_1 which could be varied by scaling all $(T_{1s})_i$ values by an identical factor.

Results of the fitting process are shown in Fig. 1, for the three temperatures near the observed relaxation peak. As explained in the main text, we obtained τ_1 from the paramagnetic impurity density and moment determined in this work, using Eqn. (3) in the main text, which contains the function $(\omega\tau_c/(1+\omega^2\tau_c^2))^{1/2} \equiv F(\omega, \tau_c)$, containing τ_c which is the spin-lattice relaxation time for the local moment. Since the temperature dependence of τ_c is not

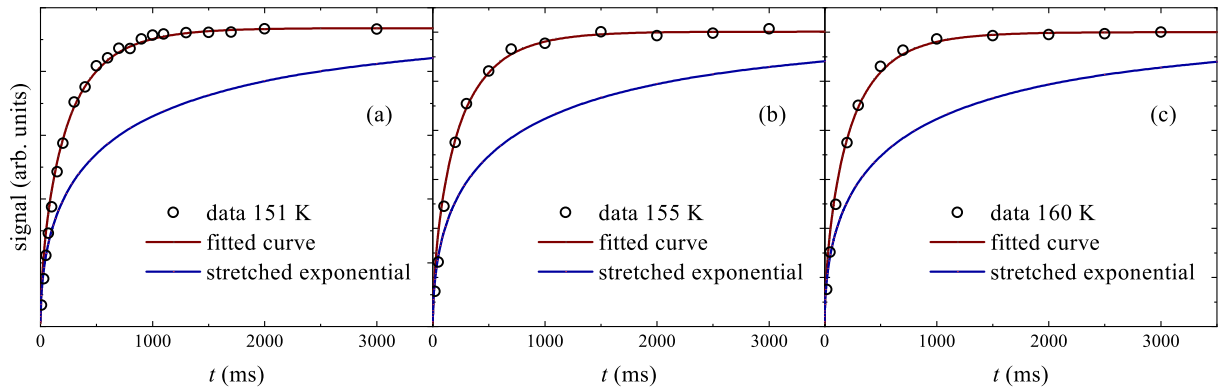

FIG. 1. Saturation-recovery plots for three temperatures, (a) 151 K, (b) 155 K, and (c) 160 K, with fitted curves described in text. Stretched exponentials alone (fitted curves with $\exp[-(t/T_{1,\mathrm{exp}})]$ removed) are also shown for comparison.

known, $F(\omega, \tau_c)$ cannot be in general be evaluated except at its maximum point ($\omega = \tau_c^{-1}$) where $F(\omega, \tau_c) = 2^{-1/2}$. Therefore, the fitting process was carried out for temperatures only at the apparent peak of the exponential-fitted NMR relaxation rate vs temperature. With τ_1 fixed the fitting to $(S_0 - S(t))$ involved two parameters, S_0 and $T_{1,\mathrm{exp}}$, for each temperature. Since the stretched-exponential recovery function is not valid at very short times during the initial recovery process, the fitting was limited for these curves to $t \geq 200$ ms, for which $S(t)/S_0 < 0.5$.

[1] D. Tse and S. Hartmann, Phys. Rev. Lett. **21**, 511 (1968).