Magnetic ground state at the ytterbium site in $YbNiAl_4$

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Magnetic, specific heat, and ^{170}Yb-Mössbauer spectroscopy measurements are presented for polycrystalline $YbNiAl_4$. Although the low temperature specific heat data are consistent with the early stages of a magnetic transition, there is no clear evidence for magnetic order down to the lowest experimental temperature of 1.5 K. It is concluded that this is due to an even stronger quenching effect than is predicted by the current crystal field theory. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3067526]

I. INTRODUCTON

The orthorhombic intermetallic series $RNiAl_4$ (R =rare earth) is currently of interest because of its intriguing magnetic properties. The Ni and Al sublattices take no part in the magnetic order but the R sublattice orders antiferromagnetically and exhibits metamagnetism and multiple magnetic phase transitions as a function of both temperature and applied magnetic field.[1] Based on the broad dependence of the Néel temperature on the de Gennes factor that has been observed for other members of the series, $YbNiAl_4$ is expected to order at about 5 K. The "easy" magnetization axis depends on the rare earth and, because of this, the crystal field (CF) interaction at the R site has recently been investigated using a combination of ^{155}Gd-Mössbauer spectroscopy for $GdNiAl_4$ (Ref. 2) and inelastic neutron scattering for $ErNiAl_4$.[3] When the preliminary CF characterization is converted for application to the Yb^{3+} ion, the predicted ground state is a Kramers doublet which is well isolated from the next excited doublet. This is likely to suppress the ordering temperature. As a test of the CF predictions, we report here on new magnetic, specific heat, and ^{170}Yb-Mössbauer spectroscopy measurements for polycrystalline $YbNiAl_4$.

II. EXPERIMENTAL DETAILS

The polycrystalline $YbNiAl_4$ specimen was prepared by repeated argon arc melting of stoichiometric proportions of Yb (99.9%) and Ni and Al (99.99+%). X-ray powder diffraction revealed traces of less than 8 wt % Yb_2O_3. The magnetization measurements (B=1 T) and specific heat measurements (zero applied field) were carried out on a Quantum Design Physical Property Measurement System (PPMS) with a base temperature of 2 K. ^{170}Yb-Mössbauer spectra were recorded with both the source and the absorber ($\approx$440 mg cm^{-2} of specimen material) mounted vertically inside a helium-flow cryostat. The 20 mCi ^{170}Tm source was prepared by neutron activation of $\approx$25 mg of Tm(10 wt %)Al and the Mössbauer drive was calibrated using an optical interferometer.

III. RESULTS AND DISCUSSION

The temperature dependence of the magnetization for $YbNiAl_4$ powder is shown in the inset of Fig. 1. At high temperatures (100–300 K), the magnetization falls to a constant value that is some 10–20 times smaller than what is observed at room temperature for other members of the $RNiAl_4$ series and is suggestive of Pauli paramagnetism. There is no evidence for magnetic order down to the base temperature of 2 K. However, the low temperature inverse susceptibility (Fig. 1) has a negative temperature intercept that is consistent with antiferromagnetic coupling. The intercept corresponds to $\theta_N \approx 2.1$ K and the slope yields an ef-

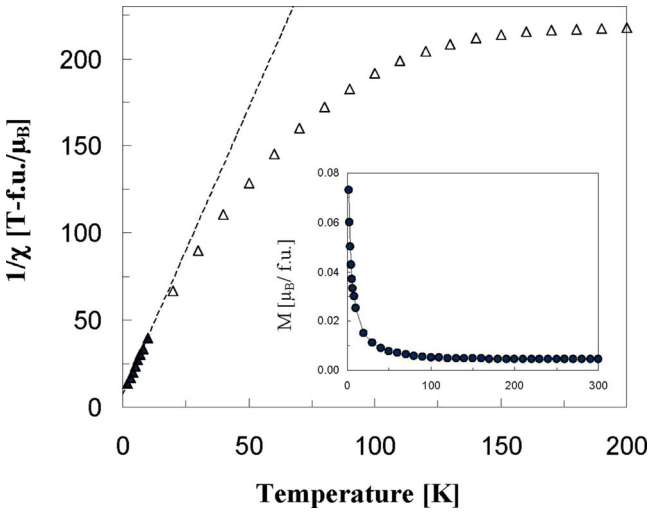

FIG. 1. (Color online) Magnetization data recorded for $YbNiAl_4$ as a function of temperature with B=1 T. The low temperature inverse susceptibility approaches Curie–Weiss behavior with $p_{eff} \approx 1.17\mu_B$. This reduces to $p_{eff} \approx 1.03\mu_B$ if the high temperature Pauli susceptibility contribution (inset) is stripped away.

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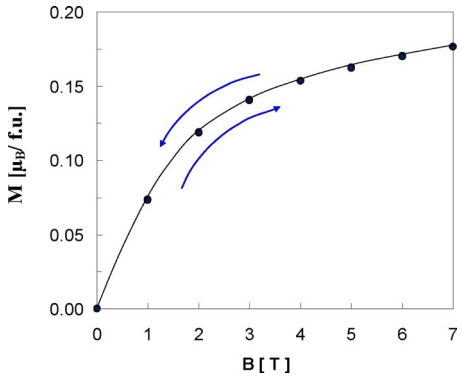

FIG. 2. (Color online) Magnetization curve recorded for polycrystalline $YbNiAl_4$ as a function of applied magnetic field with $T=2$ K.

fective moment of $p_{\text{eff}} \approx 1.17\mu_B$ (these values reduce to 1.3 K and $1.03\mu_B$, respectively, if the constant Pauli susceptibility contribution is first stripped away). For the local orthorhombic (C_{2v}) Yb^{3+}-site symmetry, the preliminary CF theory predicts a ground state Kramers doublet of the form

$$|\pm\rangle = 0.083\left|\pm\tfrac{7}{2}\right\rangle + 0.658\left|\pm\tfrac{5}{2}\right\rangle + 0.711\left|\pm\tfrac{3}{2}\right\rangle - 0.233\left|\pm\tfrac{1}{2}\right\rangle,$$

with anisotropic g-factor components $g_x \approx 2.82$, $g_y \approx 5.09$, and $g_z \approx 0.75$ yielding ${g_{\text{powder}}}^2 = \frac{1}{3}({g_x}^2 + {g_y}^2 + {g_z}^2)$ and an effective moment of p_{eff} (powder) $\approx 2.9\mu_B$. At $1.17\mu_B$, the experimental effective moment is therefore substantially smaller than both the $2.9\mu_B$ derived from the CF analysis and the Yb^{3+} free ion effective moment of $g_J\sqrt{J(J+1)} = 4.54\mu_B$. In Fig. 2, the magnetization at 2 K is observed to approach an intermediate state with $\mu \approx 0.2\mu_B$/f.u. (compared with the free ion moment of $g_J J = 4\mu_B$) as the external field is increased to $B=7$ T. The magnetization data are identical when the applied field is reduced.

Given that no magnetic hysteresis is observed at 2 K, it is interesting that the low temperature specific heat (Fig. 3) starts to increase below 3 K, consistent with the early stages of a magnetic transition. For temperatures above 7 K, the low temperature C/T data exhibit a linear dependence on T^2

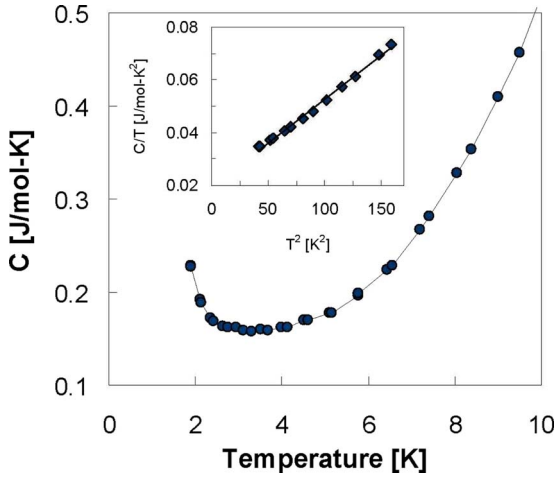

FIG. 3. (Color online) Low temperature specific heat data for $YbNiAl_4$. Above 7 K, the data are consistent with electronic and Debye contributions according to $C = \gamma T + \beta T^3$.

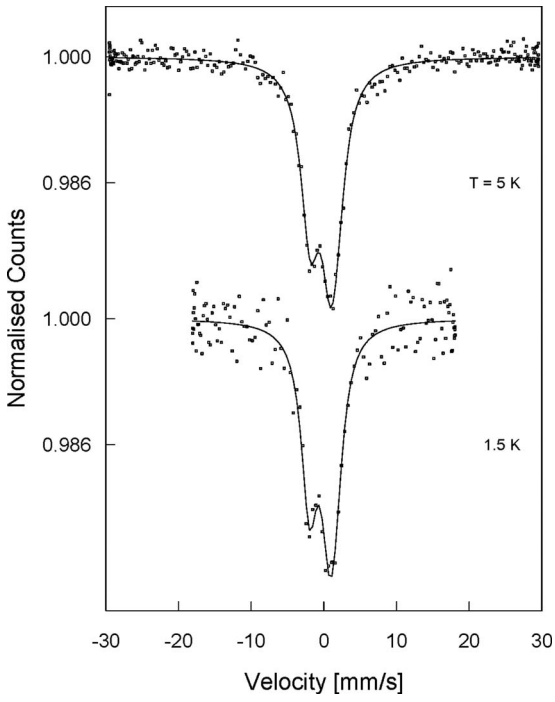

FIG. 4. ^{170}Yb-Mössbauer spectra recorded for $YbNiAl_4$ at 1.5 and 5 K. The solid curves represent the fitted quadrupole interaction theory.

(inset of Fig. 3) that corresponds to an electronic coefficient of $\gamma \approx 19.61$ mJ mol^{-1}, a phonon coefficient of $\beta \approx 0.330$ mJ mol^{-1} K^{-3}, and a Debye temperature of $\Theta \approx 181$ K.

The ^{170}Yb-Mössbauer spectra (Fig. 4) provide no evidence for magnetic hyperfine splitting or magnetic relaxation effects down to 1.5 K, despite the specific heat data's indication of the onset of a magnetic transition. Both spectra (Fig. 4) were able to be fitted to a single quadrupole-split subspectrum with a total electric field gradient of $V_{zz} = 11.8 \times 10^{21}$ V m^{-2} and asymmetry parameter of $\eta = 0$. Again this is substantially less than (and of opposite sign to) the CF theory prediction of $V_{zz} \approx -34 \times 10^{21}$ V m^{-2} with $\eta = 0.3$. In fact, the experimental result is closer in value to the lattice contribution alone of $V_{zz}(\text{latt}) \approx +8.0 \times 10^{21}$ V m^{-2} as determined by ^{155}Gd-Mössbauer spectroscopy for isostructural $GdNiAl_4$ and suggests that the Kramers ground state contribution to the electric field gradient at the nucleus should be negligible.

IV. CONCLUSION

Despite indications that antiferromagnetic order is imminent, no such order has been observed for polycrystalline $YbNiAl_4$ down to the lowest experimental temperature of 1.5 K. It is concluded that this is due to an even stronger CF quenching effect than is predicted by the current theory. This information will assist with the further refinement of the CF characterization for the $RNiAl_4$ intermetallic series.

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