Critical fields and specific heat of $LuNi_2B_2C$

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We have measured the upper critical magnetic field, lower critical magnetic field, and low-temperature specific heat of $LuNi_2B_2C$. Our upper critical-field measurements above ~ 2 K are consistent with earlier measurements by other groups. The upper critical field exhibits a finite slope as $T \rightarrow 0$, and we discuss possible origins of this very unusual behavior. We observe a linear temperature dependence for the lower critical field of our single-crystal sample, a temperature dependence that disagrees with earlier measurements on polycrystals and suggests that the lower critical field may be anisotropic. The Sommerfeld coefficient increases with magnetic field as H^{ϵ} with $\epsilon = 0.63 \pm 0.12$ in qualitative agreement with earlier work. The temperature dependence of the zero-field electronic specific heat is exponential well below T_c. We discuss our results in light of recent proposals of both anisotropic *s*-wave and *d*-wave superconductivity for $LuNi_2B_2C$. Our data seem most consistent with strong-coupling superconductivity and a weakly anisotropic *s*-wave energy gap.

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I. INTRODUCTION

The fascinating interplay between superconductivity and magnetism exhibited by the nickel borocarbides (RNi_2B_2C, where R represents a rare-earth element or Y) continues to attract both experimental and theoretical attention.[1–3] The superconducting transition temperature $T_c \sim 16$ K of the nonmagnetic ($R = $ Y and Lu) materials is quite high for intermetallic compounds. Antiferromagnetism coexists with superconductivity in several of these compounds with a Néel temperature T_N both less than ($R = $ Tm, Er, Ho) and greater than ($R = $ Dy) T_c. In these compounds the rare-earth element appears to act as a magnetic pair breaker that qualitatively follows de Gennes scaling[4,5] although this scaling breaks down when RR' pseudoquaternary samples are examined in detail.[6] Antiferromagnetism also appears in several other members of the series ($R = $ Pr, Nd, Sm, Gd) where superconductivity has not been observed;[1–3] and a ferromagnetic state (with an ordered moment of about $0.3\mu_B$) coexists with the superconducting state below about 2.5 K in the $R = $ Er compound,[7,8] a state that paradoxically coincides with an increase in the critical current density.[9] Lastly, the $R = $ Yb compound has been identified as a heavy fermion system with no long-range order observed to about 50 mK.[10]

The RNi_2B_2C compounds crystallize in a body-centered tetragonal structure where Ni_2 sheets alternate with layers of rare-earth carbides. The R^{3+} ion is in a site of tetragonal point symmetry. This gives rise to considerable crystalline electric-field anisotropy in the magnetic properties of many RNi_2B_2C members.[1,3] However, band-structure calculations[11] predict nearly isotropic electronic behavior for the nonmagnetic compounds, and this prediction is borne out in careful studies of the normal-state magnetoresistance.[12] In the superconducting state, however, both nonmagnetic compounds exhibit an anisotropic equilibrium magnetization in the basal plane.[13–15] In $LuNi_2B_2C$ an anisotropy of about 30% has been reported for measurements of the upper critical field between the [001] and [100] axes by several groups.[16,17] In addition, an upper critical-field anisotropy of about 5% within the basal plane has been observed.[17,18]

Critical-field anisotropies for the Lu compound have been explained by a nonlocal extension of the Ginzburg-Landau equations,[17] a *d*-wave symmetry of the superconducting state itself,[19] a two-band strong-coupling model,[20] and a model involving the suppression of pair breaking due to magnetic correlations.[21] In this paper we report measurements of both upper and lower critical fields and the low-temperature specific heat of $LuNi_2B_2C$. Our results allow us to determine values of the coherence length and penetration depth in the ab plane and along the tetragonal c axis of this material and to address the nature of its superconducting state.

II. EXPERIMENTAL DETAILS

$LuNi_2B_2C$ crystallizes in a body-centered tetragonal $ThCr_2Si_2$-like structure with an additional carbon atom per

formula unit in the rare-earth layer ($a=3.364$ Å and $c=10.631$ Å).[22] The resistance was measured using a standard four terminal technique at a frequency of 16 Hz with excitation currents ranging from 0.1–3 mA depending upon the temperature range. We take the upper critical magnetic field H_{c2} to be the midpoint of a resistive transition with either the magnetic field or the temperature held fixed. Transition widths are taken to be the difference between the 90% and 10% points of the transition. Our polycrystal sample was prepared by arc-melting appropriate quantities of the constituent elements as described elsewhere;[23] it had a T_c of 16.0 K and a transition width of 0.46 K. Our single-crystal sample was grown by a Ni_2B flux method.[24] The single crystals grow as thin plates with the [001] axis normal to the plane. Our single crystal exhibited a T_c of 16.2 K and a transition width of 0.43 K. Typical sample dimensions were $2\times3\times0.5$ mm^3. The specific heat C_p of $LuNi_2B_2C$ with $H\|[001]$ was measured using a standard thermal relaxation technique.[25]

The lower critical magnetic field H_{c1} was determined from isothermal magnetization measurements made with a cantilever magnetometer described elsewhere.[26] To construct the magnetometer the sample was affixed to a thin copper disc that was suspended over a copper plate by fine copper wires forming a capacitor. The capacitance of this device varies with either a linear or angular displacement of the sample. Previous work has shown that the change in capacitance is proportional to both the magnetic force and magnetic torque acting on the sample as long as the change in capacitance is small (a few percent).[27] The magnetometer is placed in the center of a superconducting magnet system consisting of a conventional solenoid and a gradient coil. The conventional coil produces a uniform field (to about one part in 10^4) and the gradient coil generates a uniform field gradient H_z at the position of the sample/cantilever.

We believe that the magnetization is most reliably determined from the magnetic force contribution to the change in capacitance of the cantilever magnetometer because the force contribution is less sensitive to sample shape effects than is the torque contribution.[27] To isolate the magnetic force contribution we first zero-field-cooled the sample to the desired temperature and slowly stepped H through H_{c1}; the resulting change in capacitance of the magnetometer assembly ($C-C_o$, where C_o is the capacitance at $H=0$) is directly proportional to the magnetic torque acting on the sample as long as $C-C_o \ll C_o$. For the measurements reported in this paper $(C-C_o)/C_o \leq 0.01$ with $C_o \simeq 1$ pF. The procedure was then repeated in the presence of a constant field gradient $H_z=0.2$ mT/mm; the resulting change in capacitance contains the above torque contribution plus a second term due to the magnetic force. The difference between these two data sets (ΔC) is directly proportional to the magnetic force and hence the dc magnetization of the sample. [This experimental approach has accurately reproduced the well known high-field magnetization curves of Ni and UBe_{13} as well as the low field magnetization curve of superconducting Pb (Ref. 28).] Typical data on the $LuNi_2B_2C$ single crystal are shown in Fig. 1. The curves labeled ''$\tau+F$'' and ''τ'' in Fig. 1(a) represent $C(H)$ data taken with and without a gradient field, respectively. Figure 1(b) shows the difference between these two data sets ($\Delta C \propto F$) along with a linear fit to the low-H data. We take H_{c1} to be the point at which $\Delta C(H)$ deviates from the linear fit by 1% as shown in Fig. 1(c).

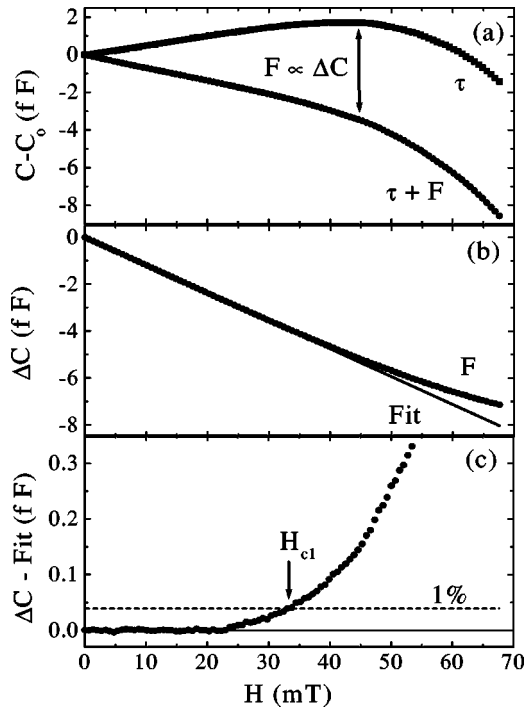

FIG. 1. Typical data used in determining the lower critical magnetic field H_{c1} of $LuNi_2B_2C$. Figure 1(a) shows the change in capacitance of the cantilever magnetometer as a function of uniform magnetic field H without a field gradient (torque only, labeled ''τ'') and with a field gradient (force and torque acting, labeled ''$\tau+F$''). Figure 1(b) shows the difference between the two curves of Fig. 1(a), ΔC as a function of field. ΔC is directly proportional to the magnetic force acting on the sample and hence its magnetization. The solid line is a linear fit to the low-field data. Figure 1(c) shows the fit residuals. The residual at which ΔC differs from the fit by 1% is shown as a dotted line. H_{c1} (uncorrected for shape effects) is identified with an arrow (see text).

The identification of the lower critical field in the dc magnetization is complicated by flux-pinning effects that set in above H_{c1}. In particular, care must be taken to sweep the magnetic field at a sufficiently low rate.[29] In ''stepping'' the external magnetic field through H_{c1} as described above, H is held fixed [for each of the data points in Fig. 1(a)] for intervals of ten seconds before each individual measurement is made. We did not observe any change in our measurements of H_{c1} as this interval was increased.

III. RESULTS

Our single-crystal H_{c2} data above about 1 K are shown in Fig. 2 where the solid and open circles represent data taken with the field parallel to the [100] and [001] axes, respectively. These measurements are consistent with those of other groups,[16,17,20] including the observation of a 30% anisotropy between the two orientations. Our polycrystal H_{c2}

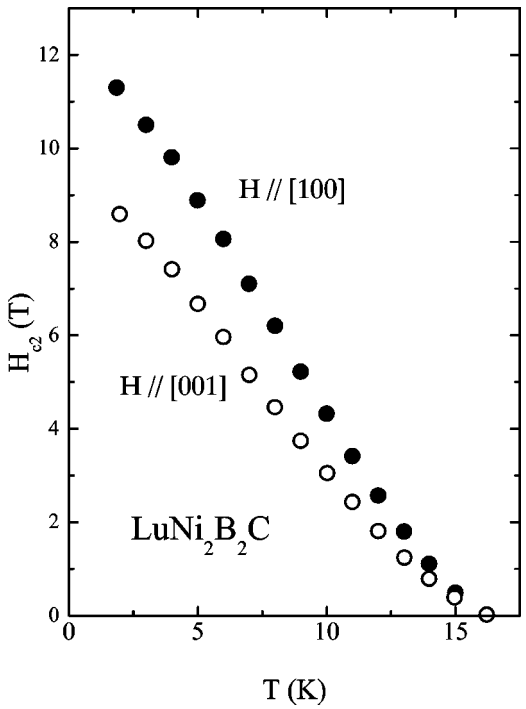

FIG. 2. The upper critical magnetic field of $LuNi_2B_2C$ with the field directed along the *a* or [100] axis (solid circles), and along the *c* or [001] axis (open circles).

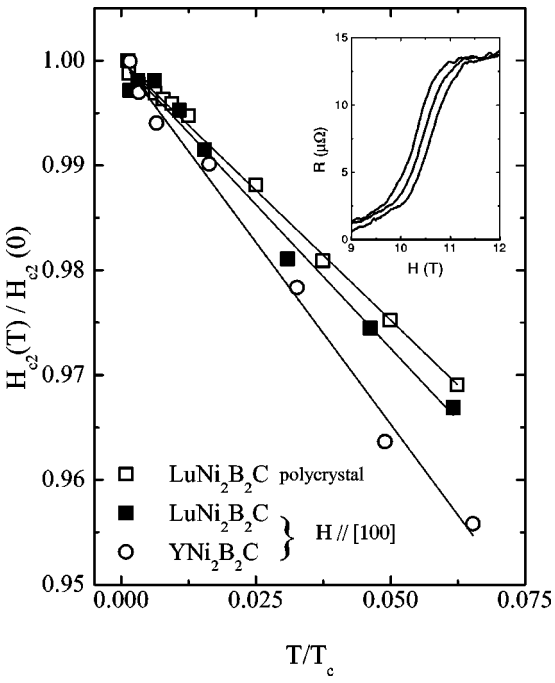

FIG. 3. The low-temperature limit of the upper critical magnetic fields of polycrystal (open squares) and single-crystal ($H\|[100]$, solid squares) $LuNi_2B_2C$ and of a single crystal of YNi_2B_2C with $H\|[100]$ (open circles). The solid lines are guides for the eye. Typical data used in determining the upper critical field of the $LuNi_2B_2C$ single crystal are shown in the inset. The three curves (from left to right) represent data taken at temperatures of 1000 mK, 500 mK, and 50 mK, respectively.

data are similar to the $H\|[001]$ single-crystal data in both shape and magnitude and are not shown.

The low-temperature limit of H_{c2} for three nickel borocarbide samples is shown in Fig. 3. The data are plotted as reduced temperature (T/T_c) versus reduced critical field $[H_{c2}(T)/H_{c2}(0)]$. Data are shown for the $LuNi_2B_2C$ single crystal with $H\|[100]$ and the polycrystal sample. Data are also shown for a YNi_2B_2C single crystal with $H\|[100]$ for comparison. The solid lines represent linear fits to the data. For all three samples H_{c2} decreases linearly with increasing temperature from the lowest temperatures measured (about 25 mK). Typical data used in determining H_{c2}(T) for the $LuNi_2B_2C$ crystal are shown in the inset. The resistive transitions (from left to right) are at temperatures of 1000 mK, 500 mK, and 50 mK, respectively. Reducing the temperature causes these transition curves to shift to higher fields, demonstrating that the observed temperature dependence we report is due to a change in H_{c2} rather than a transition width effect. Such a temperature dependence this far below T_c is unusual but has been observed in some heavy-fermion compounds[30] and in several overdoped cuprates.[31]

Our lower critical-field measurements with $H\|$ [100] are shown in Fig. 4. We have corrected for shape effects using a demagnetization factor $D=0.12$ estimated from the sample dimensions. (We did not measure H_{c1} with $H\|[001]$ owing to the large demagnetization correction that would be required for our platelike crystals.) The solid line in Fig. 4 represents a fit to the functional form $H_{c1}(T)=H_{c1}(0)(1-T/T_c)$, with $H_{c1}(0)=67.2\pm0.6$ mT and $T_c=16.1\pm0.1$ K. H_{c1} measurements on a single crystal of nonmagnetic YNi_2B_2C also exhibit a linear temperature

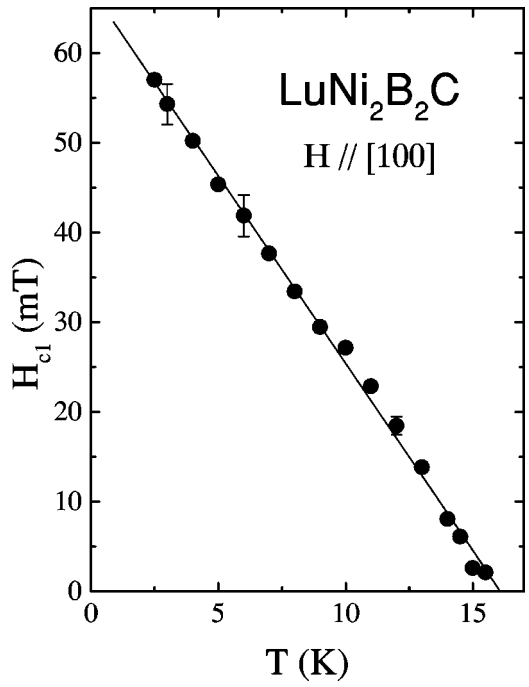

FIG. 4. The lower critical field of $LuNi_2B_2C$ with $H\|[100]$. The solid line is a linear fit to the data (see text).

TABLE I. Measured and derived parameters of $LuNi_2B_2C$ at $T=0$ using anisotropic Ginzburg-Landau theory in the $\kappa \gg 1$ limit.

Quantity	Relation	Value
$H_{c2\|ab}$		12.1 T
$H_{c2\|c}$		9.3 T
$H_{c1\|ab}$		67.2 mT
$H_{c1\|c}$	$(H_{c1\|c}/H_{c1\|ab})=(H_{c2\|ab}/H_{c2\|c})$	87.0 mT
m_c/m_{ab}	$(H_{c2\|ab}/H_{c2\|c})^2$	1.7
ξ_{ab}	$H_{c2\|c}=\Phi_o/2\pi\xi_{ab}^2$	60 Å
ξ_c	$H_{c2\|ab}=\Phi_o/2\pi\xi_{ab}\xi_c$	46 Å
λ_{ab}	$H_{c1\|c}=\Phi_o \ln(\kappa_c)/4\pi\lambda_{ab}^2$	690 Å
λ_c	$H_{c1\|ab}=\Phi_o \ln(\kappa_{ab})/4\pi\lambda_{ab}\lambda_c$	1010 Å
κ_{ab}	$(\lambda_{ab}\lambda_c/\xi_{ab}\xi_c)^{1/2}$	16.0
κ_c	λ_{ab}/ξ_{ab}	11.6
$H_{c\|ab}$	$(H_{c1\|ab}H_{c2\|ab}/\ln\kappa_{ab})^{1/2}$	0.58 T
$H_{c\|c}$	$(H_{c1\|c}H_{c2\|c}/\ln\kappa_c)^{1/2}$	0.54 T

dependence.[32] However, measurements on polycrystals of both $LuNi_2B_2C$ and YNi_2B_2C show a conventional quadratic behavior.[33] (We carried out a limited set of measurements on a polycrystal sample using our technique; the results are consistent with the quadratic temperature dependence reported earlier.)

Calculated values of the coherence lengths (ξ_{ab} and ξ_c), penetrations depths (λ_{ab} and λ_c), Ginzburg-Landau parameters (κ_{ab} and κ_c), thermodynamic critical fields ($H_{c\|ab}$ and $H_{c\|c}$), and effective-mass anisotropy (m_c/m_{ab}) for our single-crystal sample of $LuNi_2B_2C$ are summarized in Table I (where *a*, *b*, and *c*, rather than the Miller indices, denote the crystallographic axes for convenience). These results are generally consistent with those reported by Rathnayaka *et al.*[34] who did not report lower critical-field data for the crystals they studied.

The temperature-dependent specific heat C_p of $LuNi_2B_2C$ with $H\|[001]$ was measured in several fixed magnetic fields. If the Sommerfeld coefficient γ is identified as the $T=0$ limit of C_p/T, we find a $\gamma(H)$ that increases with field as H^ϵ with $\epsilon \simeq 0.5$. This is similar to the results of Nohara *et al.*[35] (who studied a polycrystal) and demonstrates that the observed field dependence of γ is not a consequence of averaging over crystallographic directions. Our raw C_p data well below T_c exhibit a temperature dependence close to T^3, a result that is also in agreement with earlier measurements.[36–38]

To further analyze our data we first assumed that the zero-field specific heat could be written as

$$C_p(T,0)=\gamma_{imp}T+\beta T^3+C_s(T,0), \tag{1}$$

where βT^3 represents the phonon contribution, $\gamma_{imp}T$ represents an impurity contribution (β and γ_{imp} are assumed to be field-independent constants), and C_s is the specific heat of the superconducting state.

We found $\beta=0.17\pm0.01$ mJ/mol K^4 from the slope of $C_p(T,H)/T$ vs T^2 for 1.7 K$\leqslant T \leqslant$5.0 K in the normal state, which was obtained by applying a field of 10 T (i.e., larger than H_{c2} over the temperature range spanned by our data).

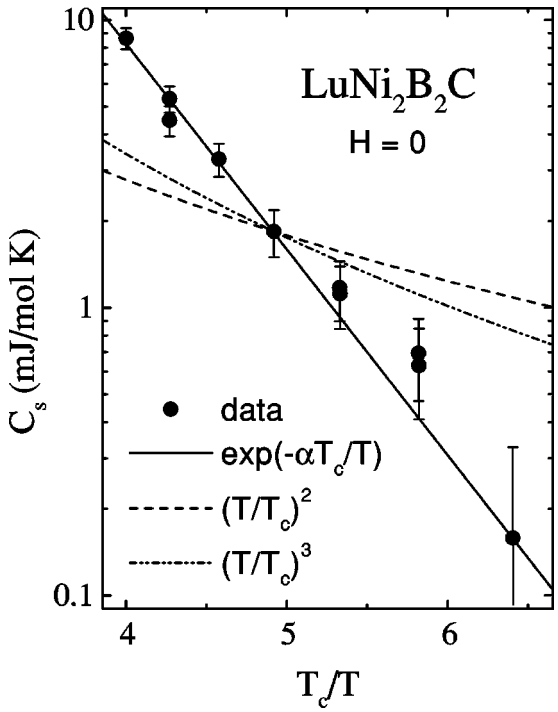

FIG. 5. The superconducting contribution to the specific heat of $LuNi_2B_2C$, C_s in zero magnetic field, plotted on a logarithmic scale against T_c/T. The dashed and dot-dash lines represent quadratic and cubic functions of temperature, respectively, scaled to the center of the data. The solid line is a fit to an exponential temperature dependence (see text).

Despite this temperature range meeting the commonly applied $T<\Theta_D/40$ criteria for characterizing the phonon contribution to the specific heat by a T^3 dependence [where Θ_D is the Debye theta that has been estimated to be in the vicinity of 360 K for $LuNi_2B_2C$ (Refs. 36–38)] one might argue that this is not an appropriate choice since an unusual, temperature-dependent, soft phonon mode has been observed near an energy of 4 meV.[39,40] The T^3 temperature dependence does, however, fit our data very well. With this determination of the phonon contribution we then found that the remaining zero-field specific heat was best fit by $\gamma_{imp}=1.14$ mJ/mol K^2 and an exponentially temperature-dependent superconducting contribution is discussed below. We attribute the impurity contribution to an estimated 5% nonsuperconducting fraction of the sample due to remnants of the flux in which the crystal was grown.

We subtracted the phonon and impurity contributions from our zero-field C_p data to isolate $C_s(T,0)$; the results are plotted versus T_c/T in Fig. 5. $C_s(T,0)$ clearly exhibits the exponential temperature-dependence characteristic of *s*-wave superconductivity. We then fit our $C_s(T,0)$ data to

$$C_s(T,0)=\delta e^{-\alpha(T_c/T)}, \tag{2}$$

where δ and α are constants assumed to be independent of field. This fit, yielding $\delta=6\pm3$ J/mol K and $\alpha=1.65\pm0.12$, is represented by the solid line in Fig. 5. Also shown in Fig. 5 are T^2 and T^3 functions scaled to pass through the center of the data. These power laws are clearly incapable

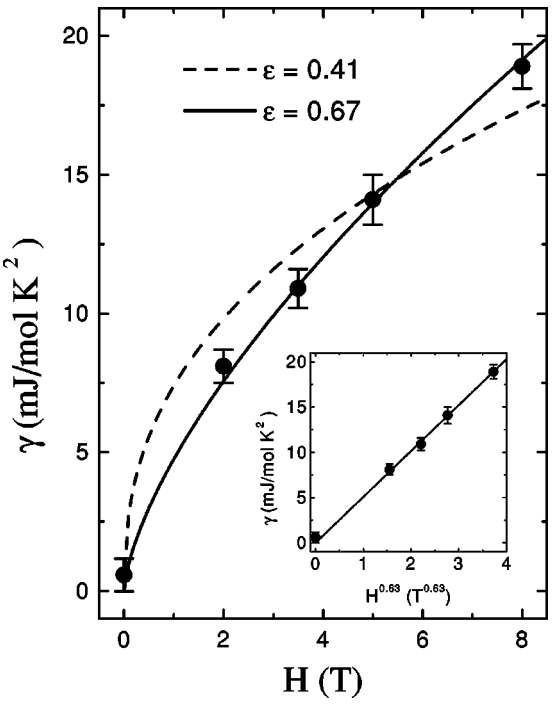

FIG. 6. The Sommerfeld coefficient of $LuNi_2B_2C$, γ, plotted against H. The dotted and solid lines correspond to fits of the data to $\gamma = AH^{\epsilon}$ with $\epsilon = 0.41$ and 0.67, respectively (see text). Shown in the inset is a plot of γ vs $H^{0.63}$ where the solid line represents a fit to the data (see text).

of describing our results. Although not shown, the jump in the specific heat at T_c is $\Delta C_p = 620$ mJ/mol K and the normal-state Sommerfeld coefficient is $\gamma_n = 19$ mJ/mol K^2. Combining these results yields $\Delta C_p/\gamma_n T_c = 2.0$, a result consistent with strong coupling and in general agreement with earlier evaluations.[36–38]

Lastly, after subtracting the phonon and impurity contributions we determined $\gamma(H)$ by linearly extrapolating $C_s(T,H)/T$ to $T=0$ with H fixed. The results were independent of the choice of extrapolation function within experimental uncertainty. We then fit $\gamma(H)$ to

$$\gamma(H) = AH^{\epsilon}, \tag{3}$$

where A is a constant and ϵ is a constant that depends on the symmetry of the order parameter (see below). Our results are summarized in Fig. 6 where the solid and dashed curves represent fits to Eq. (3) with $\epsilon = 0.67$ and 0.41, respectively. Allowing ϵ to be an adjustable parameter yields $\epsilon = 0.63 \pm 0.12$ and $A = 5.1 \pm 1.1$ mJ/mol K^{2}T$^{\epsilon}$. Shown in the inset to Fig. 6 is a plot of γ vs $H^{0.63}$.

IV. DISCUSSION

As mentioned in the introduction the temperature dependence and anisotropy of the upper critical field above about 2 K can be explained with several theoretical approaches. The $T \to 0$ temperature dependence we observe, however, is quite unusual and is inconsistent with conventional descriptions of H_{c2}(T).[41,42] The nonlocal Ginzburg-Landau approach of Metlushko *et al.*[17] predicts a finite slope for H_{c2}(T) as $T \to 0$, but it is not clear that such an approach is valid near $T=0$. Some theories that predict not only positive curvature far below T_c but an infinite H_{c2} near $T=0$ involve Landau-level quantization and this in turn requires both low carrier densities and low effective masses[43] that we believe are incompatible with intermetallic $LuNi_2B_2C$.

A more promising approach to describe our critical-field data involves the high-field suppression of pair breaking due to localized magnetic moments[21] (although the existence of such moments is controversial, see below). Such an approach not only yields a linear temperature dependence near $T=0$ consistent with our data, but is also consistent with the 30% anisotropy in H_{c2} between the [100] and [001] directions and the quasilinear temperature dependence observed for H_{c1} (Ref. 46) (though the low-temperature end of our lower critical-field data diverges from the fit to this theory). To explore the possible existence of local magnetic moments in $LuNi_2B_2C$ and YNi_2B_2C we first note that the agreement between the scaled upper critical-field data of our single and polycrystal samples shown in Fig. 3 suggests that the local moments responsible for the low-temperature linearity of H_{c2}(T) are intrinsic (though this agreement might simply reflect comparable impurity levels between these samples despite being fabricated by different techniques in different laboratories). Indeed, preliminary NMR measurements on both YNi_2B_2C and $LuNi_2B_2C$ suggested the possibility of antiferromagnetic fluctuations of the Ni magnetic moments although more recent ^{11}B NMR measurements on YNi_2B_2C by Suh *et al.* show that this is not the case.[44]

While $LuNi_2B_2C$ and YNi_2B_2C [which also exhibit a linear upper critical field, see Fig. 1(a)] are generally considered nonmagnetic, a small Curie term was reported in the magnetic susceptibility of each compound. If we attribute this to local magnetic moments, either intrinsic or extrinsic, residing in each unit cell, then the estimated magnitudes are $0.06\mu_B$/f.u. and $0.23\mu_B$/f.u. for $LuNi_2B_2C$ (Ref. 45) and YNi_2B_2C (Ref. 44), respectively. Moments of this magnitude are sufficient for the effect on the critical fields to exist.[46,47] However, in addition to the NMR measurements of Suh *et al.*[44] discussed above, one might expect a Schottky contribution to the normal specific heat, a contribution we see no evidence for in our data (see above). Further work is called for to address the existence of local moments in these materials.

We find the temperature dependence of H_{c1} to be linear within our experimental uncertainty. This temperature dependence is very unusual and is consistent with (for example) the presence of line nodes in the energy gap as expected for a *d*-wave superconductor,[48] which contradicts our specific-heat data. However, it may be that this temperature dependence simply reflects the general proportionality between H_{c1} and H_{c2} (which is quasilinear for $LuNi_2B_2C$, see Fig. 2) consistent with a Ginzburg-Landau description.[49] Measurements of H_{c1} on polycrystal samples exhibit a conventional quadratic temperature dependence.[33] This inconsistency may indicate an anisotropy in the temperature dependence of H_{c1}. Further measurements are called for to address this issue.

Although the field dependence of the Sommerfeld coefficient γ is predicted to increase linearly with H for conventional superconductors,[50] a sublinear behavior, similar to a $\sqrt{H}$ dependence, has been observed experimentally in several conventional[51] and unconventional[52] systems. Sonier *et al.* have shown that a $\sqrt{H}$ dependence can result from the expansion of the vortex cores in an *s*-wave superconductor.[53] Volovik has shown that a $\sqrt{H}$ dependence is expected for a superconductor exhibiting *d*-wave symmetry.[54] Ichioka *et al.* have extended Volovik's calculation to include bound states in the vortex core and vortex lattice effects; they predict $\gamma \propto H^{0.41}$ for *d*-wave and $\gamma \propto H^{0.67}$ for *s*-wave superconductivity.[55] The *s*-wave prediction is in good agreement with our fit exponent $\epsilon = 0.63 \pm 0.12$. Lastly, Ichioka *et al.* show that $A = \gamma_n / [H_{c2}(0)]^{\epsilon}$. Using the measured values of γ_n and $H_{c2\|c}$, we find this ratio to be in good agreement with the value of $A = 5.1 \pm 1.1$ mJ/mol $K^2 T^{\epsilon}$ obtained from the fit shown in Fig. 6.

The anisotropy of H_{c2} cannot be used to clearly distinguish between *s*- and *d*-wave superconductivity in $LuNi_2B_2C$ since it can be explained within both scenarios. The field dependence of γ is more consistent with *s*-wave superconductivity. The exponential behavior of $C_s(T)$, however, is incompatible with either point or line nodes in the energy gap. Such nodes will lead to a power-law dependence of $C_s(T)$ that is not observed. Our data therefore suggest that $LuNi_2B_2C$ is a strong-coupling superconductor with a weakly anisotropic *s*-wave gap.

V. CONCLUSIONS

We have measured the upper critical field, lower critical field, and low-temperature specific heat of $LuNi_2B_2C$ and derived several characteristic lengths and thermodynamic quantities using anisotropic Ginzburg-Landau theory. Our upper critical-field measurements above about 2 K are consistent with earlier measurements by other groups. We found a finite slope for H_{c2}(T) as $T \to 0$ and discussed possible origins of this behavior. We observed a linear temperature dependence for the lower critical field (a temperature dependence that has also been observed for a single crystal of YNi_2B_2C) that disagrees with earlier measurements on polycrystals and may indicate an anisotropic temperature dependence. The Sommerfeld coefficient is observed to increase with magnetic field as H^{ϵ} with $\epsilon = 0.63 \pm 0.12$ in qualitative agreement with earlier work. We find the temperature dependence of the zero-field electronic specific heat to be exponential well below T_c. Our data seem most consistent with strong-coupling superconductivity and an anisotropic *s*-wave energy gap.

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