Electrical resistivity of UBe_{13} in high magnetic fields

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We have measured the temperature dependent electrical resistivity of single and polycrystal samples of UBe_{13} in high magnetic fields. Two maxima in the resistivity are observed at T_{M1} and T_{M2}. T_{M1}, the temperature of the colder maximum, increases quadratically with magnetic field H, a field dependence previously observed under hydrostatic pressure. The high temperature maximum at T_{M2} emerges in fields above about 4 T and increases linearly with H, a behavior which may be due to a sharpening of the crystal field levels associated with a depression of the Kondo effect by high magnetic fields. [S0163-1829(96)05634-2]

The unusual behavior of the heavy-fermion compound UBe_{13} in both its superconducting and normal states continues to attract both experimental and theoretical attention.[1] A fundamental understanding of the possibly unconventional superconducting state of UBe_{13} must be built upon an understanding of the normal state, a normal state which appears similar to that of the better understood cerium compounds, particularly $CeCu_2Si_2$. Two quantities which highlight the similarity between UBe_{13} and $CeCu_2Si_2$ are the specific heat[2] and the electrical resistivity.[3]

The temperature dependence of the electrical resistivity, $\rho(T)$, of UBe_{13} has been described as one of the most striking of the heavy-fermion systems.[4] At high temperatures the slope is negative, characteristic of the single-ion Kondo effect. As the temperature is lowered a ''shoulder'' near 10 K is followed by a broad maximum near $T_{M1} \simeq 2.5$ K. At this maximum the value of $\rho \simeq 200\ \mu\Omega$ cm is close to the so-called unitary limit where each atom scatters resonantly.[5] Below T_{M1} the decrease in $\rho(T)$ is interrupted by the transition to superconductivity near $T_c \simeq 0.9$ K. The resistivity at T_c is still enormous (on the order of 100 $\mu\Omega$ cm), a fact which calls some of our most fundamental assumptions about superconductivity in UBe_{13} into question.[6]

The similarity in the shape of $\rho(T)$ for UBe_{13} to that of $CeCu_2Si_2$ has led to a similar interpretation of the two maxima (the second, at T_{M2}, associated with the shoulder near 10 K): The maximum at T_{M1} being due to the onset of coherent scattering from a Kondo lattice[7,8] which develops at low temperatures, although it seems clear that other mechanisms besides the Kondo effect could be operating.[9] Other proposals for the ground state of UBe_{13} are also consistent with the existence of the maximum at T_{M1}: Knetsch and co-workers[10] have proposed a model in which spin fluctuations coexist with a single-ion Kondo background, the maximum at T_{M1} being due to a ''freezing'' of the spin fluctuations. Another proposal associates this maximum with about 5% of the U ions being split by about 7 K off a nonmagnetic ground state.[11] This latter proposal assumes that this maximum in $\rho(T)$ is associated with the same mechanism which produces a Schottky-like maximum in the specific heat observed at the same temperature, an association which may not be simple.[11] A nonmagnetic ground state has been proposed by Cox[12] with a quadrupolar Kondo model which is consistent with several thermodynamic measurements on UBe_{13}.[13] However, recent nonlinear susceptibility measurements show that the low-energy excitations in this material are dipolar,[14] a result consistent with the specific-heat results discussed above,[8] inelastic neutron-scattering results,[15] and the scenario proposed by Knetsch and co-workers.[10]

The maximum at T_{M2}, again in analogy to $CeCu_2Si_2$, has been identified as resulting from the ''freezing out'' of scattering into a crystal-field level.[16] The existence of crystal-field states is consistent with the well-defined Schottky anomaly centered near 70 K.[8] The clear signature of such states is absent in photoemission experiments,[17] however, crystal-field levels are strongly broadened by the Kondo effect[18] which clearly exists in UBe_{13}. This broadening also acts to reduce T_{M2} from the 180 K one might expect based on the specific-heat measurements discussed above.

Doping on the uranium site rapidly suppresses the maximum at T_{M1} and moves the maximum at T_{M2} to higher temperatures,[19] effects similar to those observed under hydrostatic pressure.[20] Thorium is a particularly interesting dopant because of its affect on the superconducting phase diagram.[1] An extensive study of $\rho(H,T)$ of pure and thoriated UBe_{13} focusing on the low-temperature behavior has been made by Knetsch and co-workers.[10]

Despite the similarity in behavior between $CeCu_2Si_2$ and UBe_{13} described above, the applicability of $CeCu_2Si_2$ theories to UBe_{13} have been called into question by Andraka and co-workers: Measurements of the longitudinal magnetoresistance of UBe_{13} to 15 K are well described by a scaling relation of the form $(T+T^*)^{\beta}/H$ with $T^* = -0.75$ K and $\beta = 0.6$ (Ref. 23), the scaling dimension β being somewhat similar to that expected for a magnetic two-channel Kondo effect[22] while the negative value of T^* suggests the presence

of ferromagnetic correlations.[23] Similar measurements on $CeCu_2Si_2$ to 25 K show a scaling consistent with the single-ion Kondo effect ($\beta=1$ and T^* positive), a scaling which breaks down below 5 K.[24] This discrepancy suggests a fundamental difference between the low-temperature states of these compounds.

The emergence of the warmer maximum at T_{M2} in high fields is apparent in the isothermal magnetoresistance data of Brison *et al.*[25] However, it is not clear from this isothermal data at what temperature and/or magnetic field the maximum emerges or to what extent (if any) it is affected by further increases in field. Since this maximum is thought to be associated with potential underlying mechanisms of the heavy-fermion state itself (crystal fields and the Kondo effect) we decided to quantitatively characterize it. Since we are unaware of any quantitative characterization of the colder maximum at T_{M1} at ambient pressure we incorporate a study of it also.

In this paper we report data taken on three samples of pure UBe_{13}, two polycrystals and one single crystal. The polycrystalline samples were prepared by arc-melting appropriate quantities of the constituent elements in an argon arc furnace as described elsewhere, these samples exhibit superconducting onset temperatures of 0.97 K and 0.94 K for polycrystal sample Nos. 1 (Ref. 21) and 2 (Ref. 19), respectively. The single-crystal sample was grown from pre-arc-melted UBe_{13} embedded in Al flux as described elsewhere,[3] this sample has a superconducting onset temperature of 0.88 K and was oriented with its [100] axis parallel to the external field. Both isothermal and isofield resistivity measurements were made on the single crystal and on polycrystal sample no. 1 from 2 to 50 K and from 0 to 11 T. A less extensive set of measurements, focusing on the temperature dependence of ρ in the vicinity of the maxima were made on polycrystal sample no. 2 and on a polycrystal sample of $U_{1-x}Th_xBe_{13}$ with $x=0.03$ (Ref. 19) for comparison purposes.

The temperature was measured using a carbon glass resistor with the manufacturers calibration. No correction for magnetoresistance was made, $\Delta T/T<3\%$ for this particular resistor in fields below 12 T for temperatures above 4.2 K. Temperatures below 4.2 K were checked against the vapor pressure of ^{4}He. Measurements of the electrical resistivity were made using a standard four-lead phase-sensitive technique with the wires attached to the sample with silver paint. The excitation current density was approximately 0.5 A/cm^2 at 229 Hz and was perpendicular to the magnetic field, the noise was always less than about 15 nV. A geometric factor for converting the measured electrical resistance to electrical resistivity was calculated assuming the value of the resistivity to be 107 $\mu\Omega$ cm at 300 K.[4] Polycrystal sample no. 2 was measured to temperatures of about 2 K and in fields up to 18 T at the National High Magnetic Field Laboratory Pulse Facility at Los Alamos National Laboratory where resistance measurements were made with a Linear Research LR-700 bridge and a Cernox thermometer was used without correction for magnetoresistance.

The transverse magnetoresistance of our single crystal and polycrystal sample no. 1 exhibits the scaling in the longitudinal magnetoresistance mentioned above for a very high purity, very long-time annealed single crystal of UBe_{13} (a result which suggests that the distinction between longitudinal and transverse measurements of the resistivity is negligible). This agreement confirms the results of Andraka and Stewart[23] as well as the quality of these two samples. This agreement also shows that magnetoresistance measurements on older single crystals (of which ours is representative) agree with measurements on newer, higher-quality samples. This scaling does not directly impact our discussion of maxima in the resistivity since the magnetoresistance, defined as $[\rho(H,T)-\rho(0,T)]/\rho(0,T)$, does not exhibit maxima.

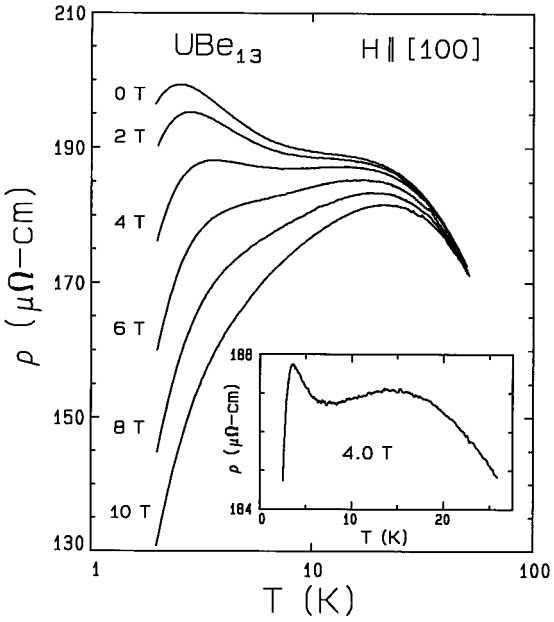

FIG. 1. The resistivity of a single crystal of UBe_{13} in several magnetic fields shown. The crystal is oriented with the field parallel to [100]. The temperature is plotted on a logarithmic axes. The inset shows the data at 4 T plotted on a linear temperature scale.

Our measurements of $\rho(T)$ for the single-crystal sample at several fixed fields are shown in Fig. 1 where T is plotted on a logarithmic scale. The low-temperature and high-temperature maxima at T_{M1} and T_{M2}, respectively, both increase with H and coexist over a narrow range of magnetic fields near 4 T as shown in the inset of Fig. 1. In order to quantitatively characterize the position and width of each resistivity maximum we choose a Gaussian function for convenience:

$$\rho(T)=A\ \exp-2\left(\frac{T-T_M}{\Delta T}\right)^2,$$

where T_M is the temperature at which $\rho(T)$ is a maximum, ΔT characterizes the width of the maximum, and A is a constant. We Taylor expand the Gaussian about T_M discarding terms of order T^3 and higher. The resultant power series is fit to the data in the vicinity of each maximum (a range of approximately 0.6 K centered on T_{M1} and a range of about 7.0 K centered on T_{M2}) using a standard linear regression routine which yields values (and errors) for T_M and ΔT. For the narrow range of magnetic fields where both maxima exist in $\rho(T)$ the data used in determining T_{M1} and T_{M2} do not overlap (see the inset of Fig. 1).

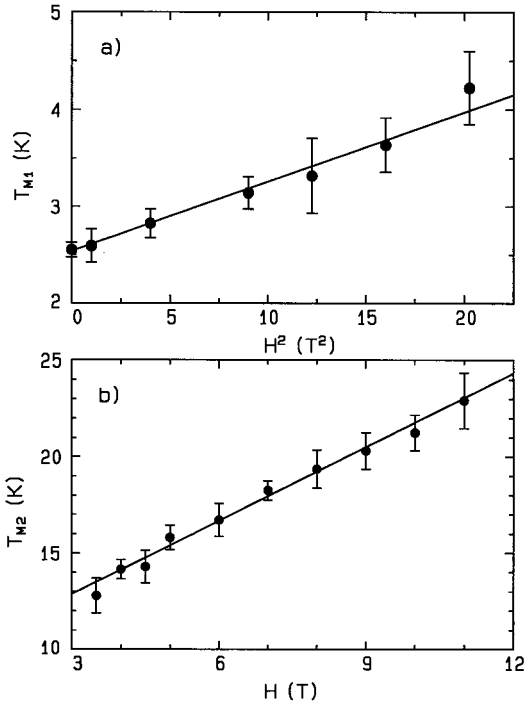

FIG. 2. The field dependence of the two maxima in the resistivity of UBe_{13}. (a) The position of the low-temperature maximum, T_{M1}, plotted against the square of the magnetic field. The solid line represents a linear regression fit. (b) The position of the high-temperature maximum, T_{M2}, plotted against the magnetic field. The solid line represents a linear regression fit.

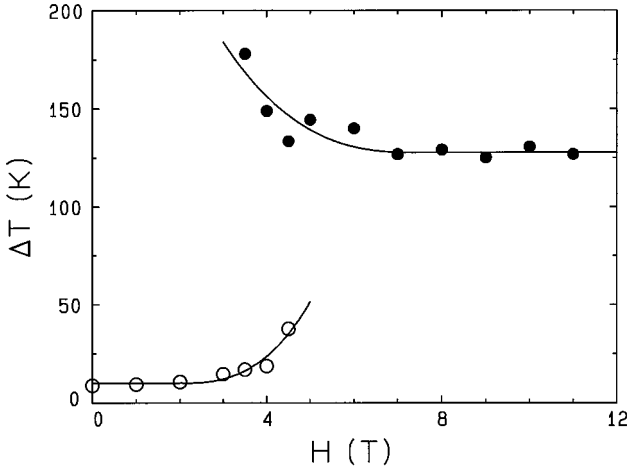

FIG. 3. The widths of the two maxima in the resistivity of UBe_{13} (see text). The open circles represent the width of the low-temperature maximum. The solid circles represent the width of the high-temperature maximum. The solid lines are guides to the eye.

As shown in Fig. 2(a) T_{M1}, the temperature of the colder maximum, increases quadratically with H. (This quadratic dependence on H also appears to exist, at least qualitatively, after subtracting the resistivity of a 6% thoriated sample as performed and described by Knetsch and co-workers.[10] As shown in Fig. 2(b) T_{M2}, the temperature of the warmer maximum, increases linearly with H. (Data on polycrystal sample no. 2 show that this linearity continues to at least 18 T.) The field dependence of the widths of both maxima are shown in Fig. 3 where the low- and high-temperature maxima are denoted by open circles and solid circles, respectively, the solid lines are guides to the eye. The peak at T_{M1} broadens appreciably with field before vanishing near 5 T. The peak at T_{M2} narrows somewhat with increasing field and tends towards field independence above about 7 T.

The data shown in Figs. 2 and 3 are from the single-crystal sample (data from both polycrystal samples behave in a similar fashion), the solid lines in Fig. 2 represent linear regression fits to the functional form

$$T_{Mn}(H) = T_{\mathrm{on}} + \alpha_n H^{2/n},$$

where $n=1$ or 2 for the low- or high-temperature maximum, respectively. The fit results for T_{on} and α_n are summarized in Table I for our three pure UBe_{13} samples along with the results of similar fits for the single maximum exhibited by the thoriated sample ($n=2$ only).

We first consider the low-temperature maximum at T_{M1}. This maximum is clearly destroyed by high magnetic fields as can be seen by the data in Fig. 1 and by the diverging width shown in Fig. 3. The H^2 dependence we observe for this maximum has also been found by Thompson *et al.* under a hydrostatic pressure of 14.8 kbar.[4] The value of α_1 we determined for our polycrystalline samples is about 40% larger than that determined by Thompson *et al.*[4] indicative of a pressure dependence to this parameter. We also note that Thompson *et al.* observe this maximum in fields above 6 T whereas our maximum has disappeared by 5 T, perhaps there is a connection between a lower value of α_1 and the survival of this maximum to higher fields. None of the proposals for the origin of this maximum discussed above includes a quantitative prediction for the field dependence of T_{M1} (to the best of our knowledge).

We now turn to the high-temperature maximum at T_{M2}. In this case a magnetic field of order 4 T or higher is necessary for the ''shoulder'' in $\rho(T)$ to emerge as a maximum. As the field increases this maximum sharpens somewhat, the width tending towards field independence above about 7 T as shown in Fig. 3. Such behavior suggests that the magnetic field depresses a magnetic scattering mechanism (such as the spin fluctuations suggested by Knetsch and co-workers[10]), a scattering mechanism which obscures this maximum in low fields. Although this maximum is rather broad the high-field independence of its width indicates that the depression of the scattering mechanism discussed above is not responsible for the field dependence of T_{M2} itself.

If we examine the mechanisms thought to be responsible for the existence of this maximum it is clear that Zeeman splitting between the crystal-field levels should act to decrease T_{M2} with increasing field[26] while suppression of the Kondo scattering by high fields should act to increase it. A theory of the electrical resistivity and magnetoresistance of cerium compounds incorporating the influence of both crystal-field and Kondo effects does indeed yield maxima in the resistivity the temperature of which increases with magnetic field.[27] In this theory, depending upon the choice of parameters, two maxima can exist in the resistivity. It is interesting to note that the colder of the two maxima usually exhibits a marked field dependence in fields comparable to those in our study while the warmer maximum shows no

TABLE I. Fit parameters for the resistivity maxima in pure and thoriated UBe_{13} (see text).

	T_{01} (K)	α_1 (K/T^2)	T_{02} (K)	α_2 (K/T)
Single crystal	2.52 ± 0.09	0.07 ± 0.01	9.5 ± 0.6	1.25 ± 0.07
Polycrystal no. 1	2.19 ± 0.04	0.042 ± 0.004	12.4 ± 0.6	1.07 ± 0.06
Polycrystal no. 2	2.59 ± 0.03	0.047 ± 0.005	11.2 ± 1.2	1.14 ± 0.11
$U_{1-x}Th_xBe_{13}$ ($x=0.03$)	–	–	33.0 ± 0.5	0.4 ± 0.1

significant field dependence at all. To the extent that this model might be pertinent to uranium compounds in general and UBe_{13} in particular (see above) one might therefore look to *differences* between UBe_{13} and $CeCu_2Si_2$ as a guide to possible origins of the field dependence of T_{M2} we observe in UBe_{13}.

One such difference exists between the crystal-field splitting in zero field and the temperature at which the ''shoulder'' appears in the resistivity. For $CeCu_2Si_2$ the shoulder appears in the vicinity of 130–150 K (Ref. 28), comparable to the observed crystal-field splitting of 140 K (Ref. 16). However, in UBe_{13} we find the shoulder near 10 K (see Table I) while the crystal-field splitting is more than an order of magnitude larger [180 K (Ref. 8)]. As mentioned above crystal field levels are strongly broadened by the Kondo effect, a broadening which acts to reduce T_{M2} from that expected based on the specific-heat measurements.[18] We therefore propose that the field dependence of T_{M2} which we observe is due to a sharpening of the crystal-field levels resulting from a depression of the Kondo effect by high magnetic fields. Such a sharpening may be detectable by photoemission and/or inelastic neutron-scattering measurements in high fields. Extrapolating from our measurements of T_{02} and α_2 (see Table I), T_{M2} doubles from its zero-field value in a field of about 9 T. If a doubling of T_{M2} results from a halving of the width of the crystal-field levels the proposed experiments should be feasible. Another prediction resulting from this hypothesis is that T_{M2} for $CeCu_2Si_2$ should not significantly increase with field since the agreement between crystal-field splittings and the temperature of the resistivity shoulder suggests that its crystal-field levels are not appreciably broadened to begin with. We are unaware of any data in the literature with which to test this last prediction.

Lastly, we comment on the influence of thorium doping on T_{M2}. It is tempting to attribute the increase in T_{M2} with thorium concentration to a depression of the Kondo effect by the addition of nonmagnetic impurities. Indeed, a concentration of about 13% thorium is sufficient to increase T_{M2} to about 50 K (Ref. 19). However, the resistivity of $U_{0.1}Th_{0.9}Be_{13}$ shows an abrupt drop, presumably due to crystal fields, at a temperature near 60 K (Ref. 29). It appears that the influence of thorium doping on this feature (in zero field) is confined to concentrations below about 10%. (We find it interesting that a thorium concentration of about 10% also marks the upper boundary for the unusual superconductivity exhibited by the $U_{1-x}Th_xBe_{13}$ system.) It is also not clear why α_2 for the thoriated sample is less than half that observed for the pure samples (see Table I). Additional measurements on thoriated samples and an examination of their field dependence may be useful in determining the influence of nonmagnetic impurities on the Kondo effect and crystal-field levels of this system.

In conclusion, we have measured the temperature-dependent electrical resistivity of UBe_{13} in high magnetic fields. Two maxima are observed at T_{M1} and T_{M2}, respectively. T_{M1} is observed to increase quadratically with H, a field dependence observed earlier under hydrostatic pressure. T_{M2} increases linearly with H, a behavior which may be due to a sharpening of the crystal-field levels associated with a depression of the Kondo effect by high magnetic fields.

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[1] For a review see N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by A. Gschneidner, Jr. and L. Eyring (Elsevier, Amsterdam, 1991), Vol. 14, p. 343.

[2] H. Rietschel, B. Renker, R. Felten, F. Steglich, and G. Weber, J. Magn. Magn. Mater. **76&77**, 105 (1988).

[3] H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **50**, 1595 (1983).

[4] J. D. Thompson, M. W. McElfresh, J. O. Willis, Z. Fisk, J. L. Smith, and M. B. Maple, Phys. Rev. **B 35**, 48 (1987).

[5] M. B. Maple, J. W. Chen, S. E. Lambert, Z. Fisk, J. L. Smith, H. R. Ott, J. S. Brooks, and M. J. Naughton, Phys. Rev. Lett. **54**, 477 (1985).

[6] P. Coleman, Physics B **206&207**, 872 (1995).

[7] F. G. Aliev, N. B. Brandt, R. V. Lutsiv, V. V. Moschalkov, and S. M. Chudinov, JETP Lett. **35**, 539 (1982).

[8] R. Felten, F. Steglich, G. Weber, H. Reitschel, F. Gompf, B. Renker, and J. Beuers, Europhys. Lett. **2**, 323 (1986).

[9] James S. Schilling, Phys. Rev. B **33**, 1667 (1986).

[10] E. A. Knetsch, G. J. Nieuwenhuys, J. A. Mydosh, R. H. Heffner, and J. L. Smith, Physica B **186-188**, 251 (1992); E. A. Knetsch, Ph.D. thesis, Leiden, 1993.

[11] J. S. Kim and G. R. Stewart, Phys. Rev. B **51**, 16 190 (1995).

[12] D. L. Cox, Phys. Rev. Lett. **59**, 1240 (1987); D. L. Cox, Physica C **153**, 1642 (1988); D. L. Cox, J. Magn. Magn. Mater **76**, 53 (1988).

[13] M. McElfresh, M. B. Maple, J. O. Willis, D. Schiferl, J. L. Smith, Z. Fisk, and D. L. Cox, Phys. Rev. B **48**, 1039 (1993).
[14] A. P. Ramirez, P. Chandra, P. Coleman, Z. Fisk, J. L. Smith, and H. R. Ott, Phys. Rev. Lett **73**, 3018 (1994).
[15] G. H. Lander, S. M. Shapiro, C. Vettier, and A. J. Dianoux, Phys. Rev. B **46**, 5387 (1992).
[16] S. Horn, E. Holland-Moritz, M. Loewenhaupt, F. Steglich, H. Scheuer, A. Benoit, and J. Flouquet, Phys. Rev. B **23**, 3171 (1981).
[17] E. Wuilloud, Y. Baer, H. R. Ott, Z. Fisk, and J. L. Smith, Phys. Rev. B **29**, 5228 (1984); J. W. Allen, J. Magn. Magn. Mater. **76**, 324 (1988).
[18] S. Kashiba, S. Maekawa, S. Takahashi, and M. Tachiki, J. Phys. Soc. Jpn. **55**, 1341 (1986).
[19] J. L. Smith, Z. Fisk, J. O. Willis, B. Batlogg, and H. R. Ott, J. Appl. Phys. **55**, 1996 (1984).
[20] M. C. Aronson, J. D. Thompson, J. L. Smith, Z. Fisk, and M. W. McElfresh, Physica B **163**, 515 (1990).
[21] J. S. Kim, B. Andraka, and G. R. Stewart, Phys. Rev. B **44**, 6921 (1991).
[22] A. Tsvelik, J. Phys. C **18**, 159 (1985).
[23] B. Andraka and G. R. Stewart, Phys. Rev. B **49**, 12 359 (1994).
[24] Bohdan Andraka, Phys. Rev. B **52**, 16 031 (1995).
[25] J. P. Brison, O. Laborde, D. Jaccard, J. Flouquet, P. Morin, Z. Fisk, and J. L. Smith, J. Phys. (Paris) **50**, 2795 (1989).
[26] K. R. Lea, M. J. M. Leask, and W. P. Wolf, J. Phys. Chem. Solids **23**, 1381 (1962).
[27] Y. Lassailly, A. K. Battacharjee, and B. Coqblin, Phys. Rev. B **31**, 7472 (1985).
[28] See, for example, U. Rauchschwalbe, Physica B **147**, 1 (1987), and references therein.
[29] B. Andraka, W. W. Kim, J. S. Kim, and G. R. Stewart, Phys. Rev. B **44**, 5040 (1991).