Specific heat below 1 mK and the electric-field gradient in indium

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The specific heat of polycrystalline indium has been measured to well below 1 mK to determine the nuclear quadrupole interaction and to study the nuclear spin-spin interaction. The sign of the electric field gradient eq is found to be positive instead of the negative sign reported previously. This sign agrees with a recent calculation based on band theory. The local field coming from the nuclear spin-spin interaction is determined to be $b_{\rm int}=43\pm5$ mT in indium. [S0163-1829(96)04625-5]

I. INTRODUCTION

The nuclear spin system is extremely ''pure'' at very low temperature because the entropy due to the other degrees of freedom are no longer existing. Nuclear magnetic ordering in metals has been studied for Cu,[1] Ag,[2] Sc,[3] In in $AuIn_2$,[4] and hyperfine-enhanced magnetic systems.[5] These spin systems have no sizable nuclear quadrupole interaction except for Sc. Although the nuclear quadrupole interaction causes a limitation of the final temperature of nuclear demagnetization cooling, the quadrupole nuclear spin systems have unique anisotropy; Ising or planar anisotropy appears depending on the sign of the nuclear quadrupole coupling constant.[6] Among various pure metals, indium (In) has a large nuclear quadrupole interaction which is suitable for the study of anisotropic spin systems. Moreover, In has a large nuclear Curie constant, a small Korringa constant, and a high thermal conductivity. These properties are useful as a nuclear refrigerant as well with the large specific heat in the temperature range as discussed below.

For actual use of In as a coolant, it is also essential to know the detailed energy level diagram of the nuclear spin where the electric quadrupole coupling constant e^2qQ/h plays an important role. While the absolute value $|e^2qQ/h|$ of 45.2 MHz is obtained from the nuclear quadrupole resonance (NQR) at 4.2 K,[7–9] the sign has been still under controversy.

For the experimental study several works have been carried out to cool In in the millikelvin temperature range.[10–12] The first of them was reported by Symko.[10] The lowest temperature was about 2 mK because of poor thermal contact between the In sample and thermometer. Although it was not as low as expected, the experiment showed a possibility to cool other materials by demagnetization of In. Since then, Tang *et al.*[12] studied the specific heat of In in the temperature region between 1 and 10 mK and concluded the negative sign of eq. However, it is difficult to discriminate between the positive and negative signs, because the corresponding specific heat difference is very small in their temperature range. To determine the sign definitely, the specific heat has to be measured in a temperature region much lower than the characteristic temperature of the quadrupole coupling ($|e^2qQ/k_B|\cong2$ mK). Another method to determine the sign of eq is NQR below the characteristic temperature. The sign of eq in scandium metal has been recently determined by NQR at temperatures down to 0.1 mK.[13]

In this paper, we present results of specific heat measurements for polycrystalline In to well below 1 mK.[14] The sign of eq in In is obtained as positive instead of the negative sign reported previously. By establishing the sign of eq, we can discuss the nuclear spin-spin interaction of In from an analysis of the specific heat.

II. NUCLEAR QUADRUPOLE INTERACTION IN In

The indium nucleus with a nuclear spin $I=9/2$ has two isotopes of ^{115}In and ^{113}In with their natural abundance of 95.7% and 4.3%, respectively. Their magnetic moments are $^{115}\mu=+5.534\mu_N$ and $^{113}\mu=+5.523\mu_N$, and their quadrupole moments $^{115}Q=+0.83$ b and $^{113}Q=+0.82$ b.[15] Here μ_N is the nuclear magneton. The interaction between the electric quadrupole moment Q and the electric field gradient eq at the nucleus is expressed as

$$H_Q=\frac{e^2qQ}{4I(2I-1)}[3I_z^2-I(I+1)], \tag{1}$$

where the z axis is parallel to the c axis (the symmetry axis of the electronic field gradient). Since the crystal structure of In is body-centered tetragonal, eq does not vanish. The quadrupole interaction splits the spin state ($I=9/2$) into five degenerated levels ($m_z=\pm1/2,\pm3/2,\pm5/2,\pm7/2,\pm9/2$). For the case $eq>0$, the ground state is $m_z=\pm1/2$ and the energy splitting to the first excited state ($m_z=\pm3/2$) is $|e^2qQ/24k_B|=0.09$ mK, whereas for the case $eq<0$, the ground state is $m_z=\pm9/2$ and the energy splitting to the first excited state ($m_z=\pm7/2$) is $|e^2qQ/6k_{B|}=0.36$ mK. At temperatures low compared to the energy difference of two lowest doublets, the spin system shows anisotropy. For the positive quadrupole coupling constant, it has a planar anisotropy because the spins are constrained to lie within the xy plane. For the negative coupling constant, it would show an Ising anisotropy.[6]

The sign of eq in In has been studied experimentally by several methods so far. The first study is on the NQR frequency shift as a function of the solute concentration in In-rich alloys.[16,17] The second study is on γ-ray anisotropy from the isotope $^{114}In^m$ in In.[18] Another work to determine the sign of eq in In is from the specific heat measurements by Tang *et al.*[12] The negative sign is deduced from all these

experiments. However, the first method is somewhat indirect, since their analysis is based on the assumption that the field gradient eq does not depend on the particular impurities, but only on the lattice parameters. In the second experiment, the negative sign of e^2qQ is derived, but the sign of eq cannot be determined without knowledge of the sign of Q in $^{114}In^m$. The sign obtained by Tang *et al.*[12] is not convincing as will be discussed later.

From a theoretical point of view, there are two contributions to eq in In. The first contribution is from ionic cores in the tetragonal lattice. The second one is due to non-s character of the conduction electron. Theoretical calculation of the ionic contribution to eq gives a negative sign and only 20% of the absolute value.[8,9] Recently, Leiberich *et al.* calculated not only the ionic contribution to eq, but also the conduction electron contribution.[19] The conduction electron wave functions were determined by the use of augmented plane wave (APW) method. They obtained the results $eq(\text{theor})=+89.1\times10^{13}$ esu/cm^3, which sign is inconsistent with all the experimental results mentioned above. Its absolute value agrees with $|eq(\text{expt})|=82.08\times10^{13}$ esu/cm^3 derived from the NQR data on the assumption of $^{115}Q=+0.76$ b for In. This agreement suggests to us studying the nuclear quadrupole interaction in In carefully.

III. CALCULATION OF THE SPECIFIC HEAT OF In

The specific heat of In nuclear spin without spin-spin interactions is calculated numerically using a single-site Hamiltonian given by

$$H=H_Q-g_N\mu_N I_z B\cos\theta-g_N\mu_N I_x B\sin\theta. \tag{2}$$

Here H_Q is the quadrupole interaction given in Eq. (1). The second and third terms are the nuclear Zeeman energy where θ is the angle between the c axis and the direction of the applied magnetic field. The energy eigenvalues are calculated by a numerical diagonalization of the Hamiltonian. The molar specific heat $C(\theta)$ in the magnetic field is given by

$$C(\theta)=R\beta^2\left\{\sum\ (E_i^2e^{-E_i\beta}/Z)-\left(\sum\ E_ie^{-E_i\beta}/Z\right)^2\right\}. \tag{3}$$

where R is the gas constant, $\beta=I/k_BT$, E_i is the ith energy eigenvalue of Eq. (2), and Z is the partition function. We assume that we find all orientations of the crystal axis with respect to the applied field. For a polycrystal, we must therefore average $C(\theta)$ over all solid angles. The specific heat of each isotope is calculated independently. Although the difference of the specific heat is very small, the contributions of ^{115}In and ^{113}In are averaged with respect to the natural abundance. The calculated results using $|e^2qQ/h|=45.24$ MHz for In^{115} and $|e^2qQ/h|=44.60$ MHz (Ref. 7) for ^{113}In are shown in Fig. 1. The solid line is the specific heat for $eq>0$, and the dashed line represents that for $eq<0$. In a later section we use $\langle C(\theta)\rangle_{\text{ran}}$ as the averaged specific heat over random orientations for $eq>0$.

IV. EXPERIMENTAL DETAILS

A two-stage nuclear demagnetization scheme was used to cool the In sample. The first stage is effectively 24 mol of copper in the magnetic field. The copper stage was precooled down to 12 mK in 9 T by a home made dilution refrigerator. The second stage is In sample itself. The second stage magnet is a home made superconducting magnet with an outer Nb tube shield. The inner diameter of the magnet is 16 mm, and the field homogeneity is better than 1% over the sample (1.5 cm long). Its remanent field is less than 1 mT after demagnetization from 1.2 T. This magnet was installed in vacuum space and thermally anchored to the mixing chamber. The polycrystalline indium rod of 99.99999% purity,[20] 12 mm diameter, and 15 mm long was cut along the cylinder axis. Two of the half cylinders were soldered to the Ag thermal link with In itself as solder. To reduce eddy current heating, slots were cut on the sample. The thickness of the fins is about 3 mm. The amount of In sample is 0.113 mol. A pure Pt wire wound noninductively around a small Ag post is used as a heater. The thermal link and the heater post were bolted to the Ag base plate with Ag-Si screws. The Ag base plate is thermally connected with the Cu stage through a tin heat switch and is supported mechanically by four Vespel SP-22 rods. The temperatures of both nuclear stages were measured with Pt NMR thermometers after calibration against a ^{3}He melting curve thermometer (^{3}He MCT).[21,22]

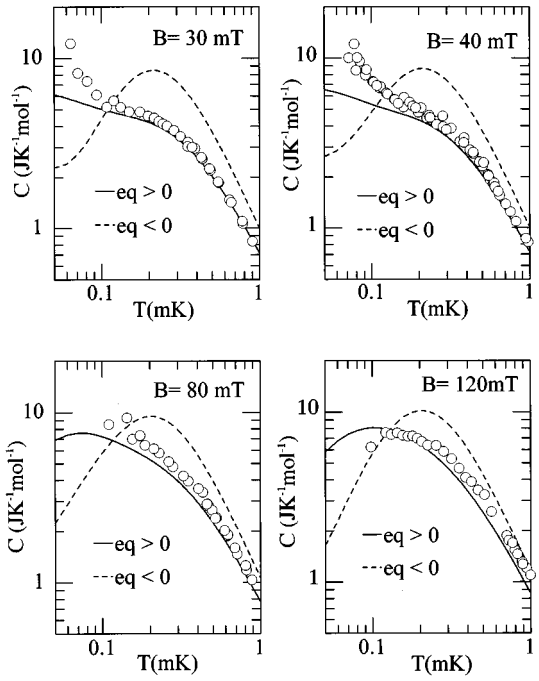

FIG. 1. Specific heat of In in fields of 30, 40, 80, and 120 mT. The solid lines are the calculated specific heat for the positive eq, and the dashed lines are for the negative eq in the field.

The In sample itself was demagnetized typically from $T_i=0.22$ mK and $B_i=1.2$ T to final measuring fields, for example, at $B_f=30$ mT and $T_f=61$ μK. A typical sweep time from 1.2 T to 30 mT was about 8 h. Since the bulk of In and Ag was demagnetized, eddy current heating due to demagnetization was not negligible. Hence the rate was chosen to minimize the total heat leak due to the external heat leaks and the eddy current heating. After the demagnetization, the specific heat was measured using the adiabatic heat pulse method. In order to keep In in the normal state, a magnetic field larger than B_c (29.3 mT) was applied. The addendum

consists mainly of Ag parts of about 240 g. The addendum specific heat has been measured to be too small compared with the sample specific heat below 20 mK.

V. RESULTS AND DISCUSSION

A. Sign of *eq*

The measured specific heat per mole in fields of 30, 40, 80, and 120 mT are shown by open circles in Fig. 1. The calculated specific heat for $eq<0$ (dashed line) has a maximum around 0.2 mK, while the calculated specific heat for $eq>0$ (solid line) flattens below 0.3 mK. This difference is obvious in the lower magnetic fields. The experimental data clearly show positive eq for In (the sign of Q in In is positive). At the same time a rapid increase of the specific heat is seen below 0.1 mK. This will be discussed later. The positive sign is consistent with the theory of Leberich *et al.*,[19] but not with the former specific heat experiment.[16] The discrepancy is explained as follows. Since the difference in the calculated specific heat for both signs of eq is small in the temperature region above 1 mK, the data of Tang *et al.* can be explained by both signs of eq with a slightly different absolute value. In addition, the ^{3}He MCT scale they used is an old one. Namely, the fixed points adopted by them are the superfluid A transition ($T_A=2.709$ mK) and the magnetic ordering transition ($T_S=1.082$ mK). These temperatures are (10–15)% higher than those now accepted ($T_A=2.49$ mK and $T_S=0.93$ mK).[21] Therefore it is not adequate to determine the sign from their specific heat data. The positive eq obtained in our experiment means that the ground state of the nuclear spin is $m_z=\pm 1/2$ and has the planer anisotropy.

B. Specific heat below 0.1 mK

The specific heat results in low magnetic fields seem to increase rapidly at low temperatures below 0.1 mK as shown in Fig. 1. One interpretation of this rapid increase might be attributed to a nuclear spin ordering in In. However, the increase can be well explained by the temperature difference between In nuclei and the Pt NMR thermometer as discussed below. In the present heat pulse method, the specific heat is obtained as the ratio of the applied heat input $\Delta Q_{\rm ap}$ and the temperature increase $\Delta T_{\rm Pt}$. The measured specific heat $C_{\rm meas}$ is given by

$$C_{\rm meas}=\Delta Q_{\rm ap}/\Delta T_{\rm Pt}. \tag{4}$$

$C_{\rm meas}$ is equal to the real specific heat of In ($C_{\rm In}$) as long as $T_{\rm Pt}=T_{\rm In}$. However, the sample stage is not completely adiabatic or a small heat leak $\dot{Q}$ flows into In nuclei. This heat leak causes the temperature difference between the temperature of the In nuclear spin ($T_{\rm In}$) and that of the Pt thermometer ($T_{\rm Pt}$).

There are four possible origins which may cause the temperature difference in our system. The first one is the coupling between the In nuclear spin and conduction electron. It is negligible because of the estimated relaxation time from the small Korringa constant $T_1T=0.08$ sec K.[23] The second origin is the boundary thermal resistance between In and the Ag thermal link. If we assume that the Wiedemann-Franz law holds even in the present low-temperature region, thermal conductance of a metal can be estimated from its electric resistance. The electric resistance of the Ag-In boundary is obtained to be about 1 nΩ from our measurement at 4.2 K. The third one is the thermal conductance through the thermal link. The residual resistivity ratio (RRR) of our Ag thermal link is about 1300. Then, for the cross section $A=0.08$ cm^2 and length $L=7$ cm, the electric resistance is 100 nΩ. The last possible origin is the press contact of the Ag thermal link against the support plate with six 3-mm-diam screws of Ag-Si.[24] Its electronic resistance is estimated to be on the order of 10 nΩ.[24] Although these four thermal resistances are connected in series, the temperature difference is mainly determined by the Ag thermal link itself. The electrical resistance of 100 nΩ corresponds to the thermal conductance $0.25T$ (W/K) based on the Wiedemann-Franz law.

Now we assume that $\dot{Q}$ enters into one end of the thermal link with the thermal conductance $\kappa_0 TA/L$, the temperature difference between In and the thermometer can be derived from the equation

$$\dot{Q}=\frac{\kappa_0 A}{2L}\,(T_{\rm Pt}^2-T_{\rm In}^2). \tag{5}$$

Therefore the specific heat and temperature of In are expressed as Eq. (6) on the condition of $2\dot{Q}/K_0 \ll {T_{\rm meas}}^2$ and $\Delta T_{\rm Pt} \ll T_{\rm meas}$, where $K_0=\kappa_0 A/L$ and $T_{\rm meas}$ is the averaged temperature of the thermometer before and after a heat pulse,

$$\begin{aligned} C_{\rm In}&=C_{\rm meas}\{1+(\dot{Q}/K_0){T_{\rm meas}}^{-2}\}^{-1},\\ T_{\rm In}&=T_{\rm meas}-(\dot{Q}/K_0)/T_{\rm meas}. \end{aligned} \tag{6}$$

Thus the temperature difference between the sample and thermometer causes the T^{-2}-dependent deviation of the specific heat from the real value. A similar phenomenon has already been reported by Krusius and Pickett.[25] Their T^{-6} deviation to the measured specific heat $C_{\rm meas}$ of arsenic was detected below 26 mK because the thermal resistance has a T^{-3} dependence which is due to the boundary resistance between metal and insulator. The observed heat leak to our second stage at the field 30 mT is 0.12 nW. The corrected specific heat for the finite thermal conductivity at 30 mT is shown in Fig. 2 for several thermal conductance values. A sharp increase of the specific heat disappears for the thermal conductance $K_0T=0.09$ T (W/K). Almost the same result is obtained for 40 mT data. This thermal conductance value is smaller by a factor of about 3 than that estimated above by the use of the Wiedermann-Franz law. Since a similar deterioration of the thermal conductivity in pure metals has been observed by several groups,[26,27] it seems reasonable to expect that the rapid increase of the specific heat below 0.1 mK is due to the finite thermal conductivity and does not arise from a nuclear ordering of In nuclei.

C. Nuclear spin-spin interaction from ''high-temperature'' specific heat measurements

Since the value of e^2qQ is definitely determined for In as in the above, one can exactly calculate the specific heat of the quadrupole interaction for polycrystalline In. Hence the nuclear spin-spin interaction can be derived from the difference (ΔC) between the measured specific heat ($C_{\rm meas}$) and

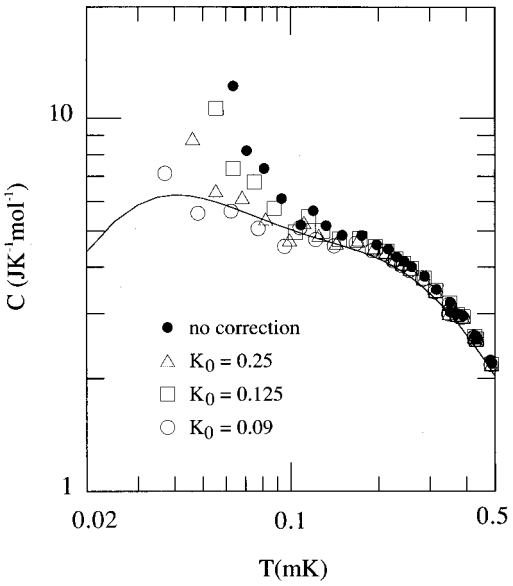

FIG. 2. Specific heat of In in a field of 30 mT using the correction of the heat leak described in the text for various thermal conductivities. The solid line is the calculated specific heat for the positive eq at 30 mT.

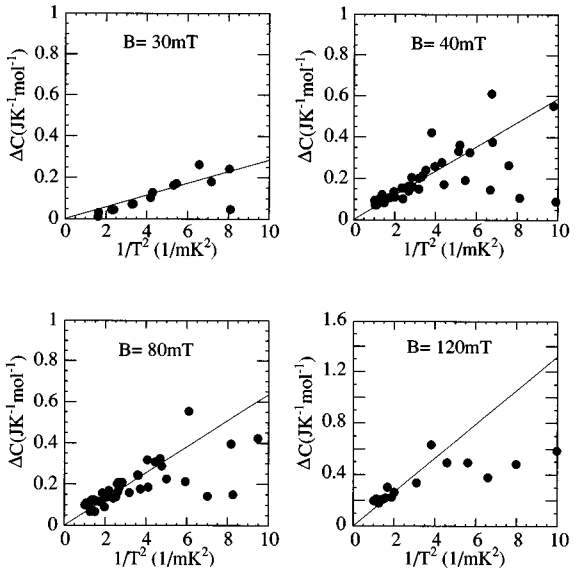

FIG. 3. Difference (ΔC) between the measured specific heat and the calculated one $\langle C(\theta)\rangle_{\rm ran}$ vs I/T^2 in various fields. Solid lines are the fitted lines using Eq. (7) for $T>0.5$ mK.

the calculated one ($\langle C(\theta)\rangle_{\rm ran}$). From the high-temperature approximation, ΔC is expressed as

$$\Delta C = C_{\rm meas} - \langle C(\theta)\rangle_{\rm ran} = (\lambda/\mu_0) b_{\rm loc}^2/T^2. \tag{7}$$

where λ is a nuclear Curie constant per mole, μ_0 is the permeability of free space, and $b_{\rm loc}$ is a local field. When the $b_{\rm loc}$ comes from the nuclear spin-spin interaction alone, $b_{\rm loc}$ is equal to $b_{\rm int}$:

$$b_{\rm int} = \sqrt{\frac{I(I+1)}{2g_N^2\mu_N^2}\sum_j (A_{ij})^2}, \tag{8}$$

where A_{ij} is a nuclear spin-spin interaction between i and j site defined by

$$H_{\rm int} = \sum_{i>j} A_{ij} I_i I_j . \tag{9}$$

The local field $b_{\rm loc}$ can be derived from ΔC vs T^{-2} plot as shown in Fig. 3. The field dependence of derived $b_{\rm loc}$ is shown in Fig. 4. If the contributions of the quadrupole specific heat and the Zeeman specific heat are extracted in Eq. (7) properly, the derived $b_{\rm loc}$ should not show any field dependence. The obtained field dependence of $b_{\rm loc}$ can be understood as follows. In our calculation of the specific heat, we assumed that our sample consists of many randomly oriented grains. On the other hand, it is well known that a well-annealed high-purity polycrystalline metal (RRR $\sim$10 000) has large grains of cm sizes. Hence it is natural to assume that our In sample also consists of a few large grains. Therefore, to estimate the real local field ($b_{\rm int}$), we should use the averaged specific heat $\langle C(\theta)\rangle_{\rm sample}$ according to the distribution of orientation of grains in the sample. The relation between $\langle C(\theta)\rangle_{\rm sample}$ and $b_{\rm int}$ is given by

$$C_{\rm meas} - \langle C(\theta)\rangle_{\rm sample} = (\lambda/\mu_0) b_{\rm int}^2/T^2. \tag{10}$$

Although we do not know the real distribution of orientation in our sample, an extrapolation to zero field in Fig. 4 gives us the real local field of $b_{\rm int}=43\pm5$ mT because there is no preferred axis in zero field.

Information about the distribution of crystal orientations in our sample can be obtained from Fig. 4. $b_{\rm loc}$ is found to increase as the field is applied. By comparison of Eqs. (7) and (10), it becomes clear that $\langle C(\theta)\rangle_{\rm sample}$ should be larger than $\langle C(\theta)\rangle_{\rm ran}$. Therefore the distribution of crystal orientations against the applied field in our polycrystal is not random. The distribution is biased towards $\theta=90°$ (perpendicular to the c axis) because $C(\theta)$ increases as θ increases and has a maximum at $\theta=90°$ at temperatures to which the high-

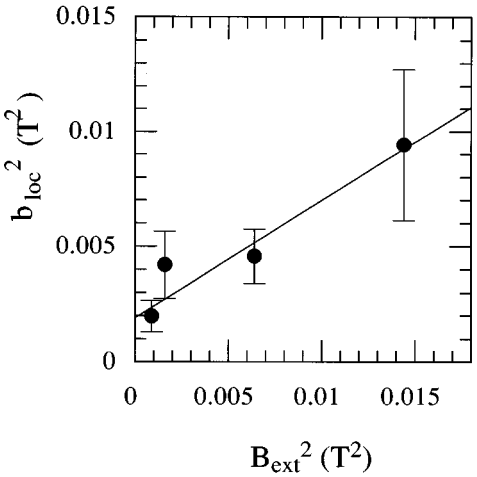

FIG. 4. External field dependence of the local field. $(b_{\rm loc})^2$ is plotted against $(B_{\rm ext})^2$, because the specific heat on the high-temperature approximation is expressed as a function of $(B_{\rm eff}/T)^2$, where $B_{\rm eff}^2 = b_{\rm int}^2 + b_{eqQ}^2 + B_{\rm ext}^2$ and b_{eqQ} is the local field corresponding to the nuclear quadrupole interaction.

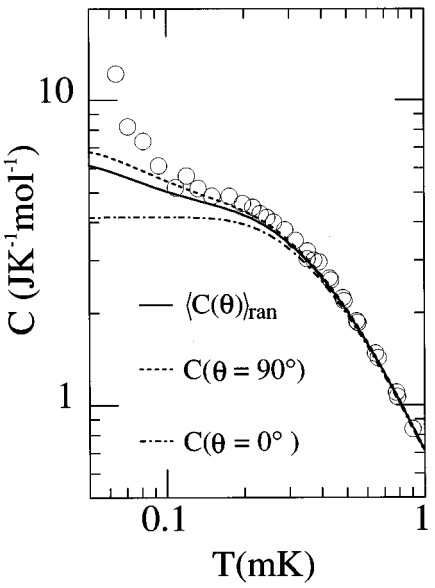

FIG. 5. Calculated specific heats $C(\theta=0°)$, $C(\theta=90°)$, and $\langle C(\theta)\rangle_{\rm ran}$ with the experimental result at 30 mT. $\langle C(\theta)\rangle_{\rm sample}$ lies between $C(\theta=90°)$ and $\langle C(\theta)\rangle_{\rm ran}$.

temperature approximation can be applied. For the different preferred orientation ($\theta=0°$), the $b_{\rm loc}$ should decrease as the field is increased. To clarify the relation between $C(\theta=0°)$, $C(\theta=90°)$, and $\langle C(\theta)\rangle_{\rm ran}$ in a field of $B=30$ mT, these are shown in Fig. 5 with experimental data. $\langle C(\theta)\rangle_{\rm sample}$ lies between $C(\theta=90°)$ and $\langle C(\theta)\rangle_{\rm ran}$.

Using $b_{\rm int}=43\pm5$ mT, the nuclear spin-spin interaction A_{ij} can be estimated to be $|A_{ij}|/k_B=1.6\pm0.2$ μK from Eq. (8). Here we assumed the same interaction value A_{ij} for the four nearest neighbors and the eight next-nearest neighbors, because the distance to the nearest neighbors is close to that of the next-nearest neighbors. The dipole interaction coefficient of In, $(\mu_0 h^2/4\pi)\gamma^2/r^3/k_B$, is of the order 0.01 μK, where γ is a gyromagnetic ratio of the nucleus and r is a separation of the nearest neighbors in In. Therefore the Rudermann-Kittel exchange interaction is dominant in A_{ij}.

Our estimate of the Rudermann-Kittel interaction using the Fermi wave vector in the free electron model for In suggests a ferromagnetic spin ordering. The ordering temperature is given as $T_C^{\rm MF}=z|A_{ij}|I(I+1)/3k_B$ at zero field in the molecular field approximation, where $z=12$ is used as the coordination number. Because of the large nuclear spin $I=9/2$ and the strong interaction A_{ij}, a high ordering temperature $T_C^{\rm MF}=160$ μK is predicted. However, the simple molecular field approximation usually gives $T_C^{\rm MF}$ higher than the real T_C because of neglect of the short-range magnetic ordering. The temperature range studied in the field $B=30$ mT is down to 64 μK, which goes much below the simple estimated $T_C^{\rm MF}=160$ μK. There can be two interpretations of our measured specific heat below 100 μK. The first one tells us that we observed the high-temperature tail of the nuclear magnetic transition specific heat peak. The second interpretation assumes certain thermal resistance between the platinum NMR thermometer and the In sample with measured heat leaks. The obtained data can be almost explained by the second interpretation, and yet we may have seen the precursor of the nuclear ordering at the lowest temperatures. Then the real situation of the magnetic ordering has to be studied under three conditions: The specific heat has to be measured in temperatures lower than 64 μK. The heat leaks to In samples should be small enough. The magnetic field which destroys the superconductivity of In should be applied in parallel to the c axis since the ground state of the In spin ($m_z=\pm1/2$) has planar anisotropy (the c axis is a hard axis). Hence a single crystal should be used for further study.

VI. CONCLUSION

We have measured the specific heat of polycrystalline In well below 1 mK. The positive sign of eq is definitely determined. From the high-temperature specific heat analysis ($T>0.5$ mK), the nuclear spin-spin interaction $|A_{ij}|/k_B=1.6\pm0.2$ μK and the corresponding the local field $b_{\rm int}=43\pm5$ mT are derived. Information on the crystal orientation of our polycrystal sample is obtained also. The real nuclear magnetic ordering with the planer anisotropy should appear below 64 μK.

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