regarded as temperature independent, and justifies the way we have compared our results to antiferromagnetic resonance spectra at 77 °K.

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Magnetic Susceptibility of MnF_2 near T_N and Fisher's Relation

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Measurements of the parallel magnetic susceptibility χ of MnF_2 are reported for $60 < T < 80$ °K in temperature intervals of about 0.1 °K. A check on Fisher's relation, viz., $C_m \simeq A\,\partial(\chi T)/\partial T$, is made near the Néel temperature T_N using Teaney's measurements of the magnetic specific heat C_m. General features of Fisher's relation are found to be valid, except A is found to have considerable temperature dependence in the temperature range $|(T - T_N)/T_N| < 0.03$. It is shown that for $T > T_N$ the observed temperature dependence of A can in part be explained by including the effect of the uniaxial anisotropy on the temperature dependence of the transverse correlation functions.

I. INTRODUCTION

It has been shown by Fisher[1] that in a simple antiferromagnet with predominantly short-range interactions, the magnetic specific heat C_m is related to the parallel static susceptibility χ by the relation

$$C_m \simeq A \frac{\partial(\chi T)}{\partial T} . \quad (1)$$

Here T is the temperature and A is expected to be a slowly varying function of T near the Néel temperature T_N. Wolf and Wyatt[2] extended these calculations to include the long-range dipolar interaction in dysprosium aluminum garnet (DAG) and experimentally verified the Fisher relation, Eq. (1), in DAG. Similarly, the Fisher relation has been found to hold in $CoCl_2 \cdot 6H_2O$.[3] In both DAG and $CoCl_2 \cdot 6H_2O$—considered to be good examples of Ising antiferromagnets—the temperature dependence of A near T_N was found to be negligible.

In his paper Fisher[1] compared Eq. (1) with existing data on MnF_2 and MnO and found qualitative agreement. However, as he pointed out, more detailed measurements are needed near T_N for both these materials to allow for a quantitative check on Eq. (1). In the case of MnF_2, additional motivation for its study comes from the fact that this material is considered to be an ideal example of a uniaxial antiferromagnet and that the Fisher relation so far has been verified in Ising antiferromagnets only. Consequently, we have measured χ in MnF_2 at small temperature intervals around T_N and made a quantitative check on Eq. (1) using the specific-heat data of Teaney.[4] This comparison shows that although general features of Eq. (1) are valid for MnF_2, A is found to have considerable temperature dependence near T_N. It is proposed that at least part of the temperature dependence is due to the uniaxial anisotropy.

II. EXPERIMENTAL PROCEDURES

For magnetic-susceptibility measurements we designed and constructed a vibrating-sample magnetometer, which has several features in common with the one described by Foner and McNiff.[5] The details of this magnetometer will be described elsewhere.[6] A 0.529-g sample of MnF_2, obtained from Ventron Electronics Corp., was driven by means of a synchronous motor with a frequency of 1 Hz and an amplitude of about 1 in. The signal was picked up by means of two circular coils placed on either side of the moving sample such that the planes of the coils were parallel both to each other and to the direction of motion of the sample. The temperature of the sample was measured by means of copper-constantan thermocouple glued directly to the sample. The sample and coils were then enclosed by a stationary stainless-steel can, the interior of which could be evacuated and filled with gaseous helium for purposes of heat exchange. The output of the coils was measured using a Keithley model 155 null detector–microvoltmeter, the amplified output of which could be recorded on a chart recorder. At each temperature about ten readings were taken, the average value taken as the susceptibility at that temperature. The magnetometer was calibrated using a single-crystal disk of nickel. The thermocouple voltages were measured by means of a Leeds and Northrup K-5 potentiometer, using a liquid-nitrogen bath as reference. To control the temperature of the sample the magnetometer was immersed in a Dewar using liquid nitrogen as a coolant. The sample was first cooled to about 54 °K by pumping on the liquid-nitrogen chamber and freezing the nitrogen. Once this point was reached, the sample was then allowed to warm up slowly. The rate of warming was slow enough that readings at constant temperature could be made, each reading taking about 10–15 sec. The temperatures were converted from V to °K by means of thermocouple tables.[7] The magnetic field was generated by a Varian 12-in. electromagnet with a pole gap of 3.5 in. The field was set and controlled by means of a Varian Fieldial Mark I. For all susceptibility measurements a field of 1 kOe aligned parallel to the c axis (easy axis) of MnF_2 was used. The susceptibility of MnF_2 is sufficiently small so that the demagnetizing field, estimated to be about 1 Oe, was neglected. The relative and absolute accuracies of the susceptibility measurements are estimated to be 1 and 3%, respectively.

III. REVIEW OF FISHER'S THEORY

We first review, very briefly, the derivation of the Fisher relation, Eq. (1). This is done to bring out the approximation made in the derivation and for the purpose of discussion later in this paper. Fisher[1] assumed that the simple antiferromagnet is described by the Hamiltonian

$$\mathcal{H} = 2J \sum_{\langle ij \rangle} [S_i^z S_j^z + (1 - \alpha_0)(S_i^x S_j^x + S_i^y S_j^y)] , \quad (2)$$

where J is the exchange integral, S_i^x, S_i^y, and S_i^z are the x, y, and z components of the spin at lattice site i, and the sum is over all pairs of spins i, j. In Eq. (2), α_0 is the anisotropy parameter, so that $\alpha_0 = 0$ for the Heisenberg model and $\alpha_0 = 1$ for the Ising model. For MnF_2, $\alpha_0 \simeq 0.015$.[8] If all sites are assumed equivalent, and the sum is evaluated only over nearest-neighbor pairs, then the magnetic configuration energy U_m is given by the relation

$$U_m(T) = JNq[\langle S_0^z S_1^z \rangle + (1 - \alpha_0)(\langle S_0^x S_1^x \rangle + \langle S_0^y S_1^y \rangle)] . \quad (3)$$

In the case of MnF_2, it is actually the next-nearest-neighbor interaction which predominates, but this does not alter the derivation. In Eq. (3), N is the number of lattice sites and q is the number of neighbors S_1 to the spin S_0 at the origin. Assuming that the temperature dependence of the three correlation functions in Eq. (3) is the same

(an approximation strictly valid only if $\alpha_0 = 0$, and for $\alpha_0 = 1$ this approximation is unnecessary), we can write

$$U_m(T) = JNq(3 - 2\alpha_0)\langle S_0^z S_1^z \rangle . \quad (4)$$

The zero-field susceptibility for an antiferromagnet is given by

$$\chi = \frac{Ng^2\mu_B^2}{kT} \sum_{i=0}^{N} \langle S_0^z S_i^z \rangle , \quad (5)$$

where g is the electronic g value and μ_B is the Bohr magneton. Equation (5) may be written in the form

$$\chi T = (Ng^2\mu_B^2/k)[\tfrac{1}{3} S(S+1) + p\langle S_0^z S_1^z \rangle] . \quad (6)$$

Fisher argued that the higher-order correlations such as $\langle S_0^z S_2^z \rangle$, $\langle S_0^z S_3^z \rangle$, etc., are of the same order of magnitude and alternating in sign for antiferromagnets. Furthermore, they have essentially the same temperature dependence as $\langle S_0^z S_1^z \rangle$ near T_N,[1] so that Eq. (6) follows from Eq. (5). The factor p was introduced to account for the neglected terms in Eq. (6). It follows from the above arguments that p should be a slowly varying function of T on the order of unity. Taking the temperature derivative of Eqs. (4) and (16) and comparing the results gives

$$C_m \simeq \frac{Jkq(3 - 2\alpha_0)f}{g^2\mu_B^2} \frac{\partial}{\partial T}(\chi T) , \quad (7)$$

where any temperature dependence of p ($\equiv 1/f$) has been neglected. Equation (7) is equivalent to Eq. (1), with A given by

$$A = \frac{Jkq(3 - 2\alpha_0)f}{g^2\mu_B^2} . \quad (8)$$

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Our experimental results of the measurement of the parallel susceptibility of MnF_2 for $60 < T < 80$ °K are shown in Fig. 1. Our over-all results for $54 < T < 300$ °K compare well with those of Foner,[9] both in magnitude and temperature dependence. Note that the data in Fig. 1 were taken at temperature intervals of about 0.1 °K.

Comparison of Eqs. (4) and (6) suggests that a more meaningful quantity is the product χT rather than χ itself since χT is proportional to U_m. A plot of χT vs T is shown in Fig. 2. It should be noted that for a paramagnet obeying the Curie law, χT is independent of temperature. The slope of the χT -vs- T curve in Fig. 2 appears to be the largest for $67 < T < 68$ °K. According to Eq. (1) the maximum in the slope of χT should occur at $T = T_N$.

The slope of the χT curve in Fig. 2 was determined using a least-squares fit to the data. The data were fitted to a second-order polynomial in short intervals. At the midpoint of each interval the slope of the fitted curve was determined, and the interval was then slid by one data point. The method is repeated until all the data points are exhausted. To do these numerous calculations the procedure was written into a Fortran program which was run on the IBM 360/75 computer of the West Virginia University. The maximum slope is found to occur at $T = 67.29$ °K for a 20-point per interval fit[10] as shown in Fig. 3. The points in this figure correspond to the right-hand side of Eq. (7) with $f = 1.42$. (The reason for this choice of f is discussed later.) Values of other constants used for MnF_2 are $g = 2.0$, $q = 8$, $J = 1.76$,[11] and $\alpha_0 = 0.015$.[8] For comparison, the specific-heat data

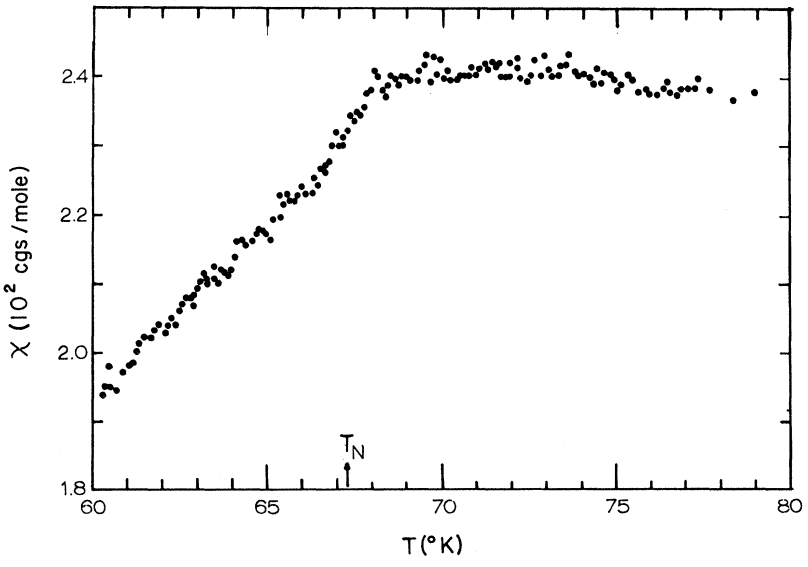

FIG. 1. Isothermal magnetic susceptibility of MnF_2 for $60 < T < 80$ °K. The arrow, marked T_N, locates the Néel temperature = 67.29 °K as measured in this experiment.

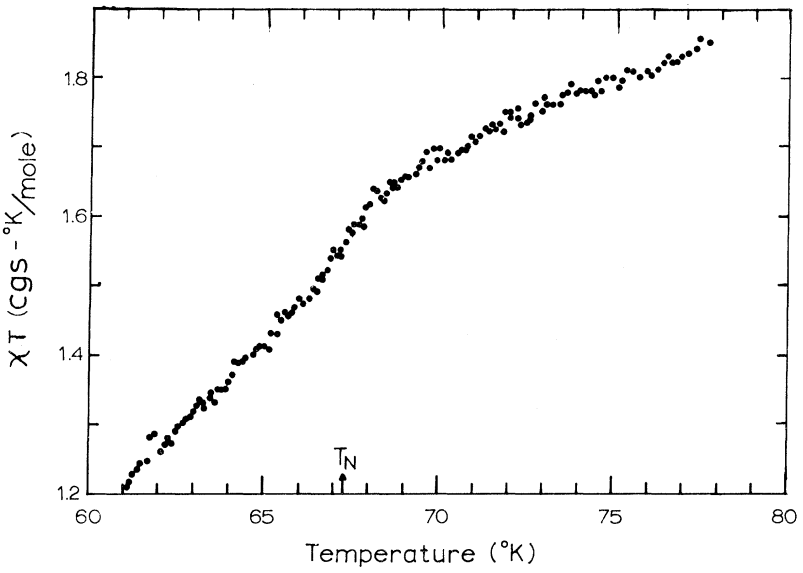

FIG. 2. Product χT (susceptibility times temperature) for $60 < T < 80$ °K as determined from Fig. 1.

of MnF_2 as reported by Teaney[4] are also shown in Fig. 3. Teaney notes that the Néel temperature for his two samples was different, perhaps indicating the effect of different amounts of impurities in the two samples. For the supposedly better sample he finds $T_N = (67.33 \pm 0.01)$ °K. From the maximum in $\partial(\chi T)/\partial T$ we find $T_N = (67.29 \pm 0.05)$ °K,[10] in reasonable agreement with his specific-heat measurements.

It is evident from Fig. 3 that there is an excellent agreement between the specific-heat and $\partial(\chi T)/\partial T$ curves for $|\epsilon| (= |1 - T/T_N|) > 0.03$. However, near T_N, and in particular above T_N, the disagreement between the two curves is considerably pronounced. In Fig. 3 and in deriving Eq. (7), we have assumed that f is temperature independent. However, Fig. 3 suggest that f is temperature dependent. One way of approximating the temperature dependence of f may be to take the ratio of the two curves in Fig. 3. The results of this procedure are shown in Fig. 4. Note that there is a noticeable peak in the value of f just below T_N and a probable dip for T just above T_N. We feel that an important cause of the peak in f is the lack of resolution in this region, which is evident from the rounding of the $\partial(\chi T)/\partial T$ curve near T_N.

The above procedure for finding the temperature dependence of f is not self-consistent, since Eq. (7) was derived by assuming that f is temperature independent. The same equation is then used to find the temperature dependence of f. To see whether the constant A [Eq. (8)], and therefore f, is indeed temperature dependent, the specific-heat

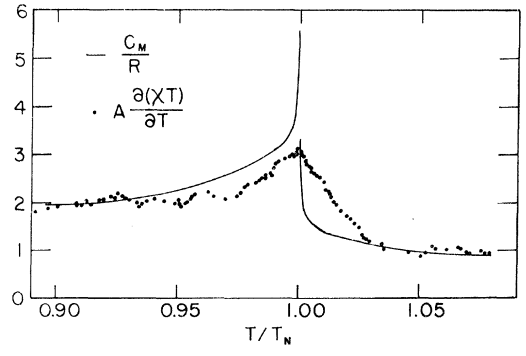

FIG. 3. Comparison of C_m/R and $(A/R)\,\partial(\chi T)/\partial T$ [see Eqs. (7) and (8)]. The smooth curve represents the magnetic specific-heat (C_m/R) data of Teaney (Ref. 4). The solid circles represents $(A/R)\partial(\chi T)/\partial T$ as obtained from Fig. 2 with $A/R = 3.3 \times 10^9$ $Oe^2/°K$ (see text).

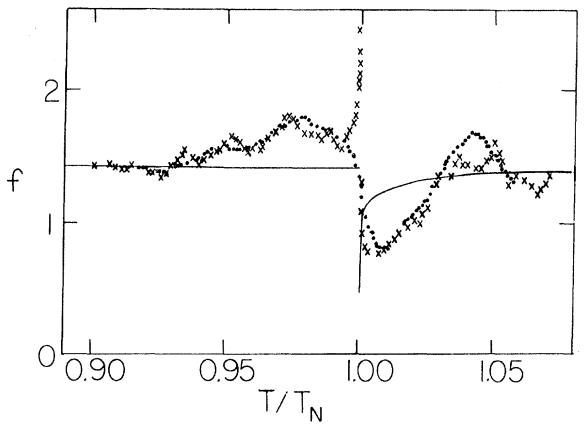

FIG. 4. The crosses represent the values of f as obtained from Fig. 3 and Eq. (7). The solid circles represent the value of f as determined from the slope of Fig. 5 [see Eq. (9)]. The solid line is drawn using Eq. (14), which includes the effect of uniaxial anisotropy for $T > T_N$.

data should be integrated to yield U_m, that is, $\Delta U_m(T) = \int_{T_1}^{T} C_m(T)\,dT$. From Eqs. (4) and (6) one can easily write

$$\int_{T_1}^{T} \frac{C_m(T)}{R}\,dT = \frac{JNq}{R}(3-2\alpha_0)\left[f(T)\left(\frac{k\chi T}{Ng^2\mu_B^2} - \tfrac{1}{3}S(S+1)\right)\right]_{T_1}^{T}, \tag{9}$$

where T_1 is an arbitrary starting temperature and the integral is evaluated for varying values of T from T_1 to the end of the data. If $\Delta U_m(T)$ is evaluated this way and is plotted against χT, it should yield a straight line of slope A if f is not a function of T. Such a plot of the experimental data is shown in Fig. 5. The observed deviation from a straight-line behavior near T_N indicates that A (and f) is temperature dependent. The solid line in Fig. 5 was drawn using a least-squares fit to the data points for $|\epsilon| > 0.03$. This procedure yields $A = (3.3 \pm 0.1)\times 10^9$ Oe^2/°K. (Note that the slope has been multiplied by the gas constant.) This value of A yields $f = 1.42$, the value used in the plot of Fig. 3. The above value of A is in excellent agreement with $A = (3.2 \pm 0.3)\times 10^9$ Oe^2/°K, the value obtained by Shapira *et al.*[12] from the study of the magnetic phase diagram of MnF_2.

The temperature dependence of $f(T)$ can be computed from the slope of Fig. 5. The results of this computation using a fit to a second-order polynomial as before are also shown in Fig. 4. It should be noted that the peak found by using the earlier method, Eq. (7), is no longer present. However, the large dip in the value of f above T_N is still present. A probable reason for this dip and its absence in the Ising antiferromagnets[2,3] is discussed below.

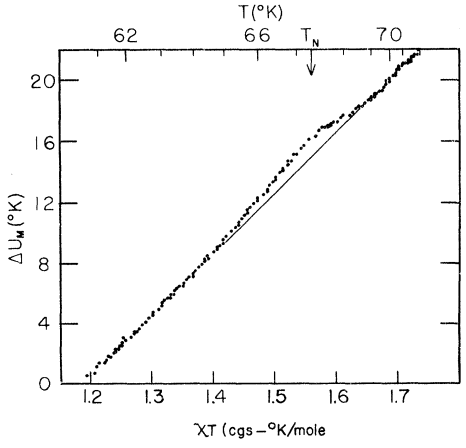

FIG. 5. Plot of $\Delta U_m(T) = \int_{T_1}^{T} [C_m(T)/R]dT$ vs χT [see Eq. (9)], where $T_1 = 60.76$ °K. The straight line represents the least-squares fit to the data for $T < 63.32$ °K and $T > 69.60$ °K.

V. EFFECT OF ANISOTROPY NEAR T_N

From the data presented above it is evident that f is temperature dependent in MnF_2, whereas it is temperature independent in the Ising antiferromagnets DAG[2] and $CoCl_2 \cdot 6H_2O$.[3] In DAG, where dipolar energy contributes about one-half the magnetic energy, Wolf and Wyatt included the long-range dipolar interaction. In MnF_2 the dipolar contribution to the magnetic energy is only about 1.5% of the exchange contribution, so that its omission could not be the reason for the observed discrepancy. Fisher's assumption of replacing the higher-order correlations by the factor p in Eq. (6) seems to be valid in $CoCl_2 \cdot 6H_2O$, and there is no reason to suspect why this should not hold true in MnF_2.

From the above discussion one reaches the conclusion that one approximation that might not be valid in MnF_2 is that of assuming that the temperature dependence of $\langle S_0^x S_1^x \rangle$ and $\langle S_0^y S_1^y \rangle$ is the same as that of $\langle S_0^z S_1^z \rangle$. Of course, this assumption is unnecessary for the Ising and Heisenberg models. Using the molecular-field approximation for $T > T_N$, Moriya[13] has shown that as a result of the uniaxial anisotropy, the perpendicular staggered susceptibility should diverge at a temperature $T_\perp = T_\parallel - \Delta$, where $T_\parallel = T_N$, and Δ is proportional to the anisotropy constant α_0, Eq. (2). For MnF_2 Moriya calculated $\Delta = 1.36$ °K. Neutron-scattering experiments of Dietrich[14] and Schulhof *et al.*[15] give $\Delta \simeq 1.3$ °K and $\Delta = 2.8$ °K, respectively. To see how this difference between $T_\parallel$ and $T_\perp$ affects the temperature depencence of A we follow a derivation similar to that given in Sec. III.

From Eq. (4), the specific heat $C_m(T)$ is given by $\partial U_m/\partial T$, i.e.,

$$C_m(T) = JNq\left(\frac{\partial}{\partial T}\langle S_0^z S_1^z\rangle + 2(1-\alpha_0)\frac{\partial}{\partial T}\langle S_0^x S_1^x\rangle\right), \tag{10}$$

where the x and y components of the correlation function have been assumed equivalent. Now it is reasonable to expect[16] that the correlation functions carry the specific-heat singularity as

$$\langle S_0^z S_1^z\rangle = \langle S_0^z S_1^z\rangle_c + \frac{E}{1-\alpha}|\epsilon|^{1-\alpha} + (\text{higher powers of } \epsilon) \tag{11a}$$

and

$$\langle S_0^x S_1^x\rangle = \langle S_0^x S_1^x\rangle_c + \frac{E}{1-\alpha}|\epsilon'|^{1-\alpha} + (\text{higher powers of } \epsilon'), \tag{11b}$$

where $\langle S_0^x S_1^x\rangle_c$ and $\langle S_0^z S_1^z\rangle_c$ are the values of the correlation functions at $T = T_\perp$ and $T = T_N$, respectively. The specific-heat index α is $\frac{1}{8}$ in three dimensions,[17] and $\epsilon' = (T - T_\perp)/T_\perp$ and $\epsilon = (T - T_N)/T_N$.

Note that in analogy with the neutron-diffraction data we have assumed that $\partial(\langle S_0^x S_1^x \rangle)/\partial T$ diverges at $T = T_\perp$ rather than at $T = T_N$. We have also made the simplifying assumption that $E_\parallel = E_\perp$ and $\alpha_\parallel = \alpha_\perp$. Substituting Eqs. (11) into Eq. (10), we get

$$C_m(T) = \frac{JNqE|\epsilon|^{-\alpha}}{T_N}\left(1 + 2(1-\alpha_0)\frac{T_N}{T_\perp}\left|\frac{\epsilon}{\epsilon'}\right|^{\alpha}\right). \tag{12}$$

Combining this with Eq. (6) yields (for $T > T_N$)

$$C_m(T) = \frac{Jkqf}{g^2\mu_B^2}\left(1 + 2(1-\alpha_0)\frac{T_N}{T_\perp}\left|\frac{\epsilon}{\epsilon'}\right|^{\alpha}\right)\frac{\partial}{\partial T}(\chi T). \tag{13}$$

Comparison of Eq. (13) with Eq. (7) suggests that the effective value of f has changed to f' given by

$$f' = f\,\frac{[1 + 2(1-\alpha_0)(T_N/T_\perp)|\epsilon/\epsilon'|^{\alpha}]}{3 - 2\alpha_0}. \tag{14}$$

Using Eq. (14), f' is plotted in Fig. 5 using $\alpha = \frac{1}{8}$ and $T_\perp = 64.5\,^\circ\mathrm{K}$.[15] A semiquantitative agreement is obtained with the observed decrease in the value of f.

For $T < T_N$ the situation is more complicated.[18] The neutron-diffraction data yield essentially a temperature-independent transverse staggered susceptibility. This behavior is not fully understood.[18] Therefore we have not attempted any calculations for $T < T_N$. However, it is evident that if $\langle S_0^z S_1^z \rangle$ and $\langle S_0^x S_1^x \rangle$ have the same temperature dependence below T_N, then $f' = f$. This behavior is shown in Fig. 5 by the solid line for $T < T_N$.

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[10]It should be pointed out that the position and the magnitude of the maximum in the slope of χT was slightly dependent on the number of points used for the fit. Due to some scatter in the data, 15- and 10-point fits yielded considerable scatter in the slope of χT. For this reason, the fewest number of points per interval was used such that a definite maximum in $\partial(\chi T)/\partial T$ could be resolved. Admittedly, this procedure produces error in the position of the maximum. From this work we estimate $T_N = (67.29 \pm 0.05)\,^\circ\mathrm{K}$.
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