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Comment on "Thermal Boundary Resistance between Solid He^3 and Cerium Magnesium Nitrate"

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I have reanalyzed the thermal-relaxation-time experiments made by Reinstein and Zimmerman on a mixture of cerium magnesium nitrate and solid He^3, which supposedly showed a spin-spin thermal contact across the boundary. It has been found that some of the assumptions made in the original interpretation are suspect. A new interpretation, based upon a magnetic-field–dependent phonon bottleneck in cerium magnesium nitrate, is presented.

A recent paper by Reinstein and Zimmerman[1] described some measurements of the thermal relaxation time between powdered cerium magnesium nitrate (CMN) and *solid* He^3. The results were interpreted in such a way that the contribution to the thermal conductance across the boundary between the paramagnetic CMN and paramagnetic solid He^3 due to magnetic coupling was deduced. This is the same contribution that has been of great interest for thermal contact between paramagnetic solids and *liquid* He^3 at very low temperatures.[2,3] It will be argued here that some of the assumptions used in the interpretation of the data are not valid. Briefly, these are assumptions concerning the ratio of solid He^3 and CMN heat capacities, the spin-lattice relaxation time in solid He^3, and the magnetic field dependence of the phonon bottleneck in CMN. It will be shown that when more realistic assumptions are made, no information about the thermal boundary resistance can be learned from the data. Instead the data will be interpreted in terms of a magnetic-field–dependent phonon bottleneck in CMN.

Reinstein and Zimmerman cooled an experimental chamber which had an upper space of volume 0.40 cm^3, filled 68% by volume with 55-μm CMN powder and 32% with solid He^3 (23.9 cm^3/mole), and a lower space containing 2.15 cm^3 of solid He^3. The decay time for a magnetically induced temperature difference between CMN and the solid He^3 was measured as a function of temperature from 0.045 to ~0.1 K, and in magnetic fields of 0, 55, 137, and 178 G. In addition to the solid-He^3 experiment, measurements were also made when the sample chamber contained liquid He^3.[4] Table I shows decay times, taken from smoothed

TABLE I. A comparison of the relaxation times (in seconds)—see text.

Temp. (mK)	Field (G)	Measured liquid He^3	Measured solid He^3	Expected solid He^3	Total solid He^3	Limited solid He^3	Incomplete solid He^3
50	0	3	3	7	3	1.5	3
	50	6	5	14	5.5	2	5.5
	137	9.5	7	34	7	1	7
	178	12	8	45	7.5	1	8
100	0	0.75	0.75	3	0.75	0.55	0.75
	55	0.75(?)	0.9	5	0.7(?)	0.4(?)	0.75(?)
	137	1.3	1.2	16	1.1	0.25	1.25
	178	1.6	•••	23	1.2	0.2	1.5

curves through the data, for both liquid He^3 and solid He^3, at 0.05 and 0.1 K, and in fields of 0, 55, 137, and 178 G. Straight lines through the points at 0.05 and 0.1 K on a log-log plot of decay time versus temperature represent the data within the scatter.

If the heat capacity (joules/kelvin) of the CMN is C_S and of the He^3 is C_H, and if the thermal resistance (kelvins/watt) between the two is R, then the thermal decay time is

$$\tau = R[C_S C_H/(C_S + C_H)]. \quad (1)$$

The resistance was taken to be the sum of the CMN phonon-bottleneck resistance (R_{PB}) and the thermal boundary resistance (R_{TBR}). Thus,

$$\tau = (R_{PB} + R_{TBR})[C_S C_H/(C_S + C_H)]. \quad (2)$$

It was assumed that $C_H \gg C_S$. Hence the relaxation time reduces to

$$\tau = (R_{PB} + R_{TBR})C_S = \tau_{PB} + R_{TBR}C_S, \quad (3)$$

where τ_{PB} is the phonon-bottleneck time in CMN. Furthermore, it was assumed that τ_{PB} was independent of magnetic field, as expected theoretically if the bottlenecked-phonon lifetime is field independent,[5] and as shown experimentally by Mess[6] for one sample in which the bottlenecked-phonon mean free path was a small fraction of the sample size. Thus, since C_S is field dependent, the variation of the relaxation time τ with field enabled the thermal boundary resistance to be measured. The measured thermal boundary resistance was assumed to be due to two *parallel* resistances: the usual phonon resistance (acoustic mismatch)[7] and the spin-spin resistance.[3] The two contributions were separated by an analysis technique that used the temperature and field dependence of the measured thermal boundary resistance. The values of the phonon thermal boundary resistivity were found to be

$$\rho_p = 5500/T^3 \text{ and } 5700/T^3 \text{ cm}^2 \text{ K W}^{-1}$$

for the liquid- and solid-helium experiments, respectively. These values are to be compared with the theoretical acoustic-mismatch value of $150/T^3$ cm^2 K W^{-1} for a CMN–liquid-He^3 interface ($\sim 40/T^3$ cm^2 K W^{-1} for a CMN–solid-He^3 interface). They are to be compared with a measurement, in the same temperature range, for a single-crystal–liquid-He^3 interface of $55/T^3$ cm^2 K W^{-1} and with a value deduced for powder of the same size in liquid He^3 of $3/T^3$ cm^2 K W^{-1}.[8] Hence the value deduced by Reinstein and Zimmerman for the CMN–liquid-He^3 interface is 35 times the theoretical value (140 times for the solid), 100 times that measured for a single crystal of CMN, and ~ 2000 times that expected for a 55-μm powdered sample. It should be pointed out that measured thermal boundary resistances are never greater than acoustic-mismatch theory.[7,9] The area calculations could have been in error, but hardly by more than a factor of 3 or so. Thus, in this experiment the analysis and assumptions used produced an absurd result; in fact the usual phonon thermal boundary resistance would have been negligible compared with the phonon-bottleneck resistance. The validity of some of the assumptions made in deriving Eq. 3 will now be questioned.

While for this experiment and previous similar experiments involving CMN–liquid-He^3 mixtures it was true that the helium heat capacity was significantly greater than the CMN heat capacity, this assumption is not automatically valid when the cell contains solid He^3. In the temperature range 0.05 to 0.1 K the heat capacity of the solid is $\lesssim 1\%$ of that of the liquid.[10,11] Table II summarizes the relevant heat capacities for the Reinstein and Zimmerman cell. The first two columns for C_H show the heat capacity of the liquid

and solid He^3 in intimate contact with the CMN; the third column shows that of the total solid He^3 in the cell. The remaining columns show the heat capacity of the CMN in magnetic fields of 0, 55, 137, and 178 G. It is seen that for the solid He^3, $C_H \gg C_S$ is only even approximately valid provided that *all* the He^3 comes to equilibrium with the CMN.

For the solid-He^3 experiment Eqs. (2) and (3) should have included the He^3 spin-lattice thermal resistance. While it appears that the spin-lattice problem is not yet properly understood for solid He^3, it is known that the relaxation time in the temperature range below 0.2 K is of order minutes.[12] While in the upper space the presence of the paramagnetic CMN may substantially reduce this time, there is no reason to expect a reduction in the lower space which contains the bulk of the solid He^3. With definition of this spin-lattice time as τ_3 and the associated resistance as R_3, Eqs. (2) and (3) become[13]

$$\tau = (R_{PB} + R_{TBR} + R_3)[C_S C_H/(C_S + C_H)], \quad (2a)$$

$$\tau = \tau_{PB}\frac{C_H}{C_S + C_H} + R_{TBR}\frac{C_S C_H}{C_S + C_H} + \tau_3\frac{C_S}{C_S + C_H}. \quad (3a)$$

Since C_S is not negligible compared to C_H, all three terms in Eq. (3a) are field dependent. R_{TBR} can no longer be uniquely determined from the field dependence of τ in the straightforward way suggested by Eq. (3).[14]

Finally, the assumption that the phonon bottleneck in the CMN is field independent is questioned. This reduces to the question of whether the mean free path of the phonon is independent of frequency, since the bottlenecked-phonon frequency increases with magnetic field. Now it happens that for this powder in zero field, the mean free path is equal to half the powder size.[1,4,8] Further, the bottlenecked-phonon wavelength in zero field is also equal to half the powder size. In a situation where the phonon wavelength is equal to the mean free path and within a factor 2 also equal to the powder size, it seems presumptuous to assume that the mean free path will not change with frequency.

In this section, first the liquid-He^3 experiments will be analyzed and then an attempt will be made to understand the solid-He^3 experiment. As shown in Table II, the liquid in intimate contact with the CMN has a significantly greater heat capacity than the CMN. Also, the thermal diffusivity and spin diffusion constants lead to short relaxation times within the upper liquid He^3 (~ milliseconds for distances ~ 50 μm).[15] Thus the error in describing the relaxation by Eq. (2) will be small. If the thermal boundary resistance is taken equal to the expected value, then it can be neglected ($R_{TBR} < 0.1\% R_{PB}$). Hence the measured relaxation time reduces to the phonon-bottleneck time in CMN which then becomes field dependent. There are several reasons why the mean free path of the bottlenecked phonon may be frequency and/or field dependent. For instance, it may be that specular reflection is more likely as the wavelength becomes a smaller fraction of the particle size.

Given that the liquid-He^3 experiment determined τ_{PB}, then, for selected values of τ_3, Eq. (3a) can be used to estimate the relaxation time for the solid experiment. Again R_{TBR} will be neglected. Several possibilities and their results are considered: (a) Assume that the spin-lattice time τ_3 is 100 sec. The results of substituting into Eq. (3a) are shown as expected-solid-He^3 times in Table I. The agreement with the measured-solid-He^3 times is very poor. (b) Neglect τ_3 (i.e. $\tau_3 \ll 10$ sec). The results are shown as total-solid-He^3 times. The agreement is good. One possible mechanism for negligible He^3 relaxation time is that the energy does transfer from CMN to He^3 via spin-spin coupling, and then reaches the bulk of the He^3 via spin diffusion. However, the spin diffusion coefficient of solid He^3 is much too small to allow this.[16] (c) Assume that the spin-lattice time for the bulk of the solid He^3 is so

TABLE II. A comparison of heat capacities. C_H and C_S are the helium and CMN heat capacities, respectively.

	C_H (μJ/K)			C_S (μJ/K)			
Temp. (mK)	Upper liquid	Upper solid	Total solid	Zero field	55 G	137 G	178 G
50	3200	16	350	14	33	127	205
100	6400	8	180	3.5	8	32	51

long that only the upper solid He^3 in intimate contact with the CMN comes to equilibrium. The results are shown as limited-solid-He^3 times, and the agreement is poor. (d) Assume that only 90% of the He^3 in intimate contact with the CMN was converted to solid. The results shown as incomplete-solid-He^3 times show good agreement at both temperatures and all fields.

In summary, it is believed that the experiment of Reinstein and Zimmerman did not show magnetic thermal contact between solid He^3 and CMN. It would have been shorted by the usual Kapitza resistance. It has been shown that the results for the liquid- and solid-He^3 experiments can be explained by a field-dependent CMN phonon bottleneck, and for the solid experiment either incomplete conversion of liquid to solid or highly anomalous spin-lattice thermal relaxation in the solid He^3. It should be clear from this analysis that any future experiments should be made with a homogeneous mixture of CMN and solid He^3, at a sufficiently low temperature that spin-lattice resistances and phonon thermal boundary resistance are large, and with CMN powder dimension sufficiently small that spin diffusion times are short. The conditions are not too rigorous and the effect is well worth looking for.

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