# Experiments on the nuclear spin exchange interaction in solid $^3$He at high molar volumes

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A reanalysis of previously published data is presented to show that the Weiss constant of solid $^3$He is negative at a molar volume of 24.50 cm$^3$/mole. The phenomenological fit of Johnson and Cohen to a wide range of experimental data suggested that it would be positive at $v = 24.25$ cm$^3$/mole. Also new data on the low-field isochoric pressure $P_v(T)$, are presented for molar volumes of 24 cm$^3$/mole and 24.5 cm$^3$/mole.

The purpose of this comment is to present some experimental data that have a bearing on some ideas recently put forward by Johnson and Cohen.[1,2] These authors have shown that essentially all experimental data on solid $^3$He at a molar volume of 24.25 cm$^3$ (on the melting curve at $T=0$) can be fit using a two-parameter isotropic Heisenberg Hamiltonian,

$$\mathcal{H} = -2J_1 \sum_{\substack{i<j \\ \mathrm{nn}}} \vec{I}_i \cdot \vec{I}_j - 2J_2 \sum_{\substack{i<j \\ \mathrm{nnn}}} \vec{I}_i \cdot \vec{I}_j ,$$

where $J_1$ is the nearest-neighbor exchange energy and $J_2$ is the next-nearest-neighbor exchange energy. The first sum on the right is over nearest neighbors (nn) only and the second is over next-nearest neighbors (nnn), and $\vec{I}_i$ is the nuclear-spin operator for the $i$th lattice site.

The values of $J_1$ and $J_2$ derived by Johnson and Cohen are quite different from values that could be described as commonly accepted. They require $J_1 > 0$ and $J_2 < 0$ at all molar volumes. The values that have been used previously to fit data at lower molar volumes are $J_1 < 0$, $J_2 > 0$,[3,4] and $J_1 < 0$, $J_2 < 0$.[5] These latter sets of values have also received support from microscopic calculations. Johnson and Cohen point out in order for their two-parameter fit to work for all molar volumes the Weiss constant $\Theta$ must change sign at some molar volume between 24 and 24.25 cm$^3$/mole. They indicate that such a sign change is unlikely and that therefore either the two-parameter fit is inappropriate or the high-temperature data are incorrect or incorrectly analyzed.

This intriguing suggestion moved us to begin experiments on the exchange interaction in solid $^3$He at molar volumes greater than 24 cm$^3$/mole as well as to carefully review susceptibility data previously published by one of us in that region.[6] We will first discuss this reanalysis.

The susceptibility measurements published in[6] 1969 give a Weiss constant $\Theta$ of $-4.9 \pm 2.0$ mK at a quoted molar volume of 24.2 cm$^3$/mole. The molar volume measurement in that experiment was made at the high-temperature freezing point of the solid because at that time it was felt that the low-temperature melting curve of solid $^3$He was somewhat uncertain. Since the melting curve is well established to quite low temperatures now, we decided to see if we could reanalyze the 1969 data in terms of the melting point of the sample on the low-temperature side of the melting-curve minimum. The melting of the sample at the low-temperature point was much easier to determine than at the high-temperature point because it was possible to use the difference in the nuclear susceptibilities of the solid and the liquid rather than the difference in the spin-lattice relaxation times.

Our review of the data shows that the sample quoted as 24.2 cm$^3$/mole in those 1969 data melted between 0.055 and 0.051 K. Using the melting-curve data published by Trickey, Kirk, and Adams[7] we find that this places the molar volume of that sample between 24.49 and 24.51 cm$^3$/mole. Thus the previous data, when corrected, gives a value $\Theta = -4.9 \pm 2.0$ mK at $v = 24.50$ cm$^3$/mole. The uncertainty in the molar volume is less than 0.03 cm$^3$/mole even when the uncertainty in the temperature measurements is included.

Although this does not give a very precise value for the Weiss constant the sign seems unambiguous. This data when coupled with the susceptibility data of Sites, Osheroff, Richardson, and Lee[8] at $v = 24.1$ cm$^3$/mole and Johnson and Wheatley[9] at $v > 24$ cm$^3$/mole would appear to make it very unlikely that the Weiss constant is positive at $v = 24.25$ cm$^3$/mole.

We have also made preliminary measurements of the isochoric pressure $P_v(T)$ at low magnetic field between 0.4 and 0.04 K. The pressure and nuclear susceptibility were measured simultaneously, but the lack of reliable temperature measurements below 0.35 K has forced us to use the susceptibility as a thermometric parameter. This was done by measuring the Curie constant at 0.4 K and assuming a Weiss constant consistent with previous measurements.[6,8-11] The measurements have been performed at molar volumes of 23.86, 23.94, and 24.50 $cm^3$/mole, the uncertainty in each being less than 0.02 $cm^3$/mole. The pressure was measured with a capacitance strain gauge[12] and the nuclear susceptibility was measured by cw NMR techniques.

If we follow Cohen and Johnson[1] and write

$$P_v(T) = \left(\frac{3A}{v}\right)\frac{a}{T} + \cdots , \quad (1)$$

where $A = 8.206 \times 10^7$ $cm^3$ atm/mole K then our measurements determine the parameter $a$. The data at 23.86 and 23.94 $cm^3$/mole serve to check the consistency of our data with previous measurements of $a$.[11] The results are shown in Table I for the case where the Weiss constants are chosen to be consistent with previous measurements and with the results of the first part of this comment. The values of $a$ are derived from least-squares fits to the data and the errors quoted are the standard deviations. The error in parentheses for 24.50 $cm^3$/mole is that due to the uncertainty in the Weiss constant. The values of $a$ obtained from a fit to the data of Panczyk and Adams[11] at 23.86 and 23.94 $cm^3$/mole are 6.30 and 6.96 $mK^2$, respectively. Our value of $a$ at 24.50 $cm^3$/mole is larger than a simple extrapolation of that data would predict (i.e., $a \simeq 18$ $mK^2$). Given the uncertainty in the Weiss constant and the limited temperature range over which data can be taken, further experiments are needed to test the significance of this.

TABLE I. Summary of results of isochoric pressure measurements.

| Molar volume ($cm^3$/mole) | Assumed Weiss constant (mK) | $a$ ($mK^2$) |
|---|---|---|
| $23.86 \pm 0.01$ | $-2.38$ | $6.50 \pm 0.49$ |
| $23.94 \pm 0.01$ | $-2.52$ | $6.82 \pm 0.70$ |
| $24.50 \pm 0.02$ | $-5.0$ | $26.10 \pm 4.00$ ($\pm 2.15$) |

We are improving the thermometry and refrigeration systems in our experiment in order to be able to test the high-temperature expansions for the thermodynamic properties of solid $^3He$ over the widest possible range of temperatures, magnetic fields, and molar volumes.

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