# On the feasibility of production of microkelvin temperature by nuclear demagnetisation

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**Abstract.** Nuclear demagnetisation as a means of refrigeration has been proposed and achieved long ago. In this paper an attempt has been made to show that PrBe$_{13}$, a Van-Vleck paramagnet, can be used to produce a lattice temperature of 10 $\mu$K or lower. Such a calculation can be used for a computer simulation of the process.

## 1. Introduction

Adiabatic demagnetisation of nuclear spins as a means of producing very low temperature was proposed in the thirties. Some workers (Kurti *et al* 1956) succeeded in reducing the spin temperature of Cu-nuclei to about 1 $\mu$K. However, in order to use this process to refrigerate other systems, *e.g.*, conduction electrons in the same metal or liquid $^3$He, some difficulties must be overcome. Nuclear demagnetisation, in spite of having the same principle as that of demagnetisation of a paramagnetic salt like cerium magnesium nitrate CMN, has many practical differences. Even if the nuclear spin temperature is lowered to less than 1 $\mu$K, the lattice and the conduction electrons in thermal contact with the precooling stage (the mixing chamber of a dilution refrigerator or a Pomeranchuk cell) are not cooled at all and the nuclear spins are warmed back to the initial temperature. So, cooling of the lattice and conduction electrons along with the nuclei is imperative to obtain significant cooling of the system. As the nuclear magnetic moments are about 1/1836 of the electronic magnetic moments of magnetic ions in a paramagnetic salt, 1836 times the value of $H_i/T_i$ are required to obtain the same amount of entropy reduction in a system of nuclei as in the paramagnetic system. $H_i$ and $T_i$ denote the applied magnetic field and temperature respectively before demagnetisation.

The spontaneous ordering of nuclear spins due to their dipole-dipole interaction alone determines the ultimate temperature $T_d$ which can, theoretically, be reached by demagnetisation to $H_j = 0$. Their interaction energy is

$$\epsilon_d = \vec{\mu} \sum_{j=1}^{N} \left[ \frac{\vec{\mu_j}}{r_j^3} - \frac{3r_j(\vec{\mu_j} \cdot \vec{r_j})}{r_j^5} \right] = \mu h_d \tag{1}$$

where $h_d$ denotes the effective dipolar field. Now we can see that

$$T_d = \frac{\epsilon_d}{k_B} \propto \frac{\mu^2}{\gamma^3} \propto \frac{\mu^2}{\rho}$$

where $\rho$ is the density of spins. Equation (1) gives for nuclear spins $T_d \simeq 0.1\,\mu\text{K}$. For CMN, having the weakest dipole-dipole interaction among magnetic ions, $T_d = 1.9\,\text{mK}$ (Huiskamp and Lounasmaa 1973). Berglund *et al* (1972) using copper as the refrigerent have cooled liquid $^3\text{He}$ down to $0.7\,\text{mK}$ for the first time. But rare earth compounds have the following advantages over Cu (Bucher *et al* 1971): (1) Large cooling entropy ($\sim 10^2$ times); (2) Shorter spin-lattice relaxation time ($\sim 10^{-2}$ times in mK region); (3) Large resistivity ($10^3$—$10^5$ times larger) to reduce eddy current effects.

## 2. Formulation of the problem

Rare earth compounds containing non-Kramers ions are very interesting as regards nuclear demagnetisation. These ions have a singlet ground state and some of them with high Van Vleck susceptibilities can induce large hyperfine fields at the nucleus when a moderate external field is applied. This hyperfine enhancement can be as large as 400 or even more. Therefore, using these materials the experimental situation for nuclear demagnetisation can be improved. Very interesting results in hyperfine induced nuclear refrigeration have been obtained by Andres and Bucher (1970, 1972) using $PrPt_5$ and $PrTl_3$. Another material $PrCu_6$ seems to be promising.

Our interest is in $PrBe_{13}$. The Be-riched compounds $PrBe_{22}$ and $PrBe_{13}$ have extremely low electronic specific heat at the temperatures in which we are interested. Density of states per atom at the Fermi surface for $PrBe_{22}$ is three times that of Be-metal and in the case of $PrBe_{13}$ it is expected to be still higher but lower than the density of states of any metal or alloy. Moreover, magnetisation studies (Bucher *et al* 1975) have shown that $PrBe_{13}$ does not magnetically order down to a temperature $\sim 1\,\text{mK}$.

The relation between the initial and final nuclear spin temperature is (Huiskamp and Lounasmaa 1973)

$$T_{nf} = \left(\frac{H_{nf}^2 + h_d^2}{H_{ni}^2 + h_d^2}\right)^{\frac{1}{2}} T_{ni} \tag{2}$$

where $H_{nf}$ and $H_{ni}$ are the final and initial effective fields at the nuclei. Again $H_n$ is given by

$$H_n = (1 + K) H. \tag{3}$$

The Knight shift $K$ can be expressed as

$$K = \frac{A \, \chi(0)}{N g_J \, \mu_B \, g_N \, \mu_N}. \tag{4}$$

In eq. (4) $A$ denotes the hyperfine constant, $\chi(0)$ the Van Vleck susceptibility, $N$ the Avogadro number, $g_J$ the $g$-factor of $Pr^{3+}$, $\mu_B$ the Bohr magneton, $g_N$ the nuclear $g$-factor of Pr-nucleus and $\mu_N$ the nuclear magneton. Using eq. (4) in eq. (3) we get

$$T_{nf} = \left\{ \frac{H_f^2 (1 + K)^2 + h_d^2}{H_i^2 (1 + K)^2 + h_d^2} \right\}^{\frac{1}{2}} T_{ni}$$

$$= \frac{H_f}{H_i} T_{ni} \tag{5}$$

if we consider $H_i$ and $H_f$ much larger than $h_d$. Andres and Bucher (1970, 1972) have shown this linear relation to be valid for PrPt$_5$.

The time rate of change of nuclear spin temperature is given by

$$\frac{dT_n}{dt} = \frac{T_n}{k} (T_e - T_n) \tag{6}$$

where $T_e$ indicates the electronic temperature (same as lattice temperature). $k$ denotes the Korringa constant and is given by

$$k = \tau_1 T$$

$$= \left( \frac{g_S \mu_B}{g_N \mu_N} \right)^2 \frac{\hbar}{4\pi k_B} K^{-2} \{K(\alpha)\}^{-1} \tag{7}$$

where $\tau_1$ denotes the nuclear spin lattice relaxation time, $g_S$ denotes the electron spin $g$-factor, $k_B$ the Boltzmann constant and $K(\alpha)$ is the electron interaction factor.

Now the heat flow equations are

$$C_e \frac{dT_e}{dt} = \frac{dQ}{dt} - \frac{dQ_n}{dt} \tag{8 a}$$

and

$$C_n \frac{dT_n}{dt} = \frac{dQ_n}{dt} - T_n \left( \frac{\partial S}{\partial H} \right)_{T_n} \frac{dH}{dt} \tag{8 b}$$

where $dQ/dt$ is the rate of heating (per mole) to the electronic system and $dQ_n/dt$ is the rate of heat flow from electrons to nuclei. As for PrBe$_{13}$, the electronic specific heat $C_e = \gamma T$, has an extremely low value. The value of $\gamma$ for LaBe$_{13}$ and LuBe$_{13}$ have been experimentally found to be 0·58 and 0·60 respectively in mJ/°K$^2$ mole. For PrBe$_{13}$ we can take the former value for $\gamma$.

At $T \approx 1$ mK, $C_e = 0·58 \ \mu$J/° K mole

and

at $T \approx 1 \ \mu$K, $C_e = 0·58 \ n$J/°K mole.

For $dT_e/dt \sim 1 \ \mu K/$sec, the left hand side in eq. (8 a) is of the order of $10^{-12}$ Joule/sec in the first case and $10^{-15}$ Joules/sec in the second case. Neglecting the second term on the right hand side in eq. (8 b) we have

$$C_n \frac{dT_n}{dt} = \frac{dQ}{dt} \tag{9}$$

because the process is adiabatic and so is isentropic. The entropy is reduced during the process of magnetisation prior to adiabatic demagnetisation. Still a small amount of entropy reduction may be achieved. Substituting eq. (6) in eq. (9) we get

$$\frac{C_n T_n}{k}(T_e - T_n) = \frac{dQ}{dt} .$$  (10)

In this derivation we have assumed that the second term in the right hand side of eq. (8 b) is small compared to the first term which is sufficiently justified for our present isentropic demagnetisation purpose. However, an exact calculation is being done by the present authors taking into consideration the second term on the right hand side of eq. (8 b).

## 3. Numerical results

The nuclear specific heat $C_n$ per mole of Pr has been found to be (Lounasmaa 1967)

$$C_n = 850 \, T_n^{-2} \text{ mJ/mole } °K.$$

Using this expression for $C_n$ in eq. (10) we have

$$k \frac{dQ}{dt} = 850 \times 10^{-3} \left( \frac{T_e}{T_n} - 1 \right) \text{ Joules/sec.}$$  (11)

Berglund *et al* (1972) were able to design a nuclear refrigeration cryostat in which the best value of $dQ/dt$ attained was $10^{-9}$ Joules/sec. So, from eq. (11) we finally get

$$\frac{T_e}{T_n} = 1 + \frac{10^{-9} \, k}{850 \times 10^{-3}} .$$  (12)

Using the following numerical values (Genicon and Tournier 1976) $g_J = 0.8$, $g_N = 1.71$, $g_s = 2.0023$, $N = 6.023 \times 10^{23}$ $A/k_B = 52.5$ mK, $K(a) = 0.76$ (Riegel 1975), $\chi(0) = 0.047$ cm$^3$/mole (Bucher *et al.* 1975), we get

$$K = 8.94$$

and

$$k = 4.624 \times 10^{-8} \sec °K.$$

Using $H_i/H_f \sim 10^3$, $T_{nf} = 10^{-3} T_{ni}$.

For $T_{ni} = 10$ mK (dilution refrigerator or Pomeranchuk cell), $T_{nf} = 10 \, \mu K.$

Then

$$\frac{T_{ef}}{T_{nf}} = 1 + \frac{4.624 \times 10^{-17}}{850 \times 10^{-3}} .$$

Hence we can conclude that $T_{ef}$ is practically the same as $T_{nf}$. Thus using PrBe$_{13}$ as a refrigerent, it is possible to attain a lattice temperature as low as $10 \, \mu K$.

A rough estimate of order of magnitude of the nuclear ordering temperature in PrBe$_{13}$ have been done by Bucher *et al* (1975). Using the molecular field approximation they have found the value of this quantity to be $\sim 0.2$ mK. The applicability of this approximation in the case of rare earth beryllides is doubtful. Indirect exchange between nuclei through conduction electrons can in principle bring about an ordering, in addition to the dipole interactions considered in the present paper. However this exchange energy seems to be negligible (Genicon and Tournier 1976).

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