Letters to the Editor

Experiments on Spin Temperature

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WE have performed experiments, to be described below, which may be explained only by utilizing the concept of spin temperature. The first group, which may be described as experiments in nuclear calorimetry, are studies in the changes of polarization produced by the thermal mixing of the spin systems of two nuclear species contained in the same sample. The process of mixing is performed by taking the sample from the strong magnetic field H_0 into the earth's field and returning it into the field H_0 in a time short compared to the spin-lattice relaxation time. The two spin systems, isolated by their different Larmor frequencies in the strong field, are no longer isolated in the weak field and find a common temperature. Hence this process is irreversible in the Ehrenfest sense and thermodynamically reversible only in very special cases.

These experiments are summarized in Table I, where M_0 and M_0' stand for the equilibrium polarizations of the species in question. The sample used was a single crystal of LiF, for which, in H_0, $T_1(\text{Li}^7)\cong 4.5$ min, $T_1(F^{19})\cong 1.4$ min, and in the earth's field, $T_1(\text{common})$ $\cong 6$ sec. The observations were made with a Varian V-4200 spectrometer, adjusted for U-mode observation with $H_1=1$ gauss at 8 Mc/sec, with which the level populations could be examined, reversed by fast passage, or saturated.

TABLE I. Summary of nuclear calorimetric experiments. The observed values, which represent the average of several measurements, are reproducible to about 10%.

Expt.	Polarization before mixing		Polarization after mixing	
	Fluorine	Lithium	Fluorine	Lithium
(a)	M_0	M_0'	$0.95\,M_0$	$0.95\,M_0'$
(b)	M_0	Saturated	$0.42\,M_0$	$0.51\,M_0'$
(c)	Saturated	M_0'	$0.42\,M_0$	$0.43\,M_0'$
(d)	M_0	$-M_0'$	$0.27\,M_0$	$0.20\,M_0'$
(e)	$-M_0$	M_0'	$0.05\,M_0$	$0.00\,M_0'$
(f)	Saturated	$-M_0'$	$-0.16\,M_0$	$-0.17\,M_0'$
(g)	$-M_0$	Saturated	$-0.29\,M_0$	$-0.34\,M_0'$
(h)	$-M_0$	$-M_0'$	$-0.71\,M_0$	$-0.73\,M_0'$

The results of the experiments described by Table I can be understood in terms of ordinary calorimetry by assigning to each system a specific heat proportional to $\gamma^2 I(I+1)$ and by making allowance for the relaxation which occurs during the course of the experiment and for the incomplete reversal of the magnetization by the fast-passage operation. This simple analysis is valid as long as the mixing field is high compared to the internal local fields because the energy of the spin-spin interaction may then be neglected Experimentally we have found that mixing begins to take place in a field of 75 gauss which is about ten times the rms local field. Experiments (a) and (h) can be recognized as the reversible processes observed by Pound[1] and Pound and Purcell[2] and appear here as a special case of the mixing of two systems at the same temperature. Experiments (d) through (h) are believed to be the first examples of calorimetry carried out with systems at negative temperatures.[2]

The results of Table I suggest that it is possible to "pump" a nuclear system with a long spin-lattice relaxation time into a polarized state by cooling it at regular intervals by thermal contact with a system which has a shorter T_1. This was demonstrated by using a powdered sample of CsCl for which $T_1(\text{Cs})\cong 9$ min, $T_1(\text{Cl})\cong 3\,5$ sec in strong fields, and $T_1(\text{common})$ $\cong 20$ sec in the earth's field. Commencing with Cs unpolarized, the sample was quickly removed from and restored to the strong field at six-second intervals, chosen to allow time for the Cl to polarize, for a period of two minutes, after which the Cs showed a polarization of $0.7\,M_0$, which otherwise would have taken about ten minutes to obtain. It is interesting to speculate that in favorable cases it may be possible to measure the magnetic moment and thermal relaxation time of an isotope which is not otherwise observed by measuring its heating effects upon an observed isotope.

An experiment of quite a different kind demonstrates that, for the case of equally spaced levels, the mechanism of mutual spin-flips seeks in a time $\sim T_2$ to establish a spin-temperature for the spin system. This mechanism, unfortunately overlooked by Proctor and Robinson,[3] has been shown to account for the complete saturation of the nuclear magnetic energy levels of Na^{23} in NaCl caused by intense ultrasonic waves at twice the Larmor frequency. This may be understood in the following step-wise fashion: the pure quadrupole transitions caused by the ultrasonic waves tend to saturate the levels related by $\Delta m=2$, resulting in a polarization $0.2\,M_0$. However, in a time $\sim T_2$ a Boltzmann population is re-established, allowing further ultrasonic saturation to take place. Equations (4) and (6) of reference 3, describing the saturation for the cases of dipolar and quadrupolar relaxation, become identical and correspond to $M/M_0=(1+12WT_1/7)^{-1}$, where W is the quadrupole transition probability. This expression has been experimentally verified in this laboratory for all values of M.

Further experimental results and a theoretical discussion will be published at a later time.

[1] R. V. Pound, Phys. Rev. **81**, 156 (1951).
[2] E. M. Purcell and R. V. Pound, Phys. Rev. **81**, 279 (1951).
[3] W. G. Proctor and W. A. Robinson, Phys. Rev. **104**, 1344 (1956).

Unified Theory of Interacting Bosons

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R ECENT work has contributed to the understanding of properties of helium II. Yet there is room for a unified theoretical approach to the problem of interacting bosons for both solid and liquid states. In particular the liquid is like the solid as regards cohesive energy and packing. One is interested in computing these from first principles, as well as in the connection between the vibration spectrum of the solid and the excitation spectrum of the liquid, the elucidation of the liquid solid transition under pressure, scattering of excitations, etc. The treatment of these diverse phenomena requires construction of wave functions for the system of interacting bosons, which are reasonably accurate and simple, from a unified point of view. In the following we report on one such unified approach, based on an elementary physical picture. The procedure is an outgrowth of the small-oscillation theory of the interaction of a particle and a scalar field.[1]

The Hamiltonian for a system of bosons of mass M with the two-body interaction potential V (thus neglecting the detailed electronic structure of the atoms) will be written in the formalism of second quantization:

$$H = \frac{\hbar^2}{2M}\int \nabla\psi^\dagger\cdot\nabla\psi d^3x + \tfrac{1}{2}\int\int \psi^\dagger(\mathbf{x})\psi^\dagger(\mathbf{x}')$$
$$\times V(\mathbf{x}-\mathbf{x}')\psi(\mathbf{x})\psi(\mathbf{x}')d^3x d^3x',$$

where $\psi(\mathbf{x})$, $\psi^\dagger(\mathbf{x})$ are operators satisfying the Bose-Einstein commutation rules such as $[\psi(\mathbf{x}), \psi^\dagger(\mathbf{x}')] = \delta(\mathbf{x}-\mathbf{x}')$. Associated with H are the Heisenberg equations of motion for the time-dependent operators $\psi(\mathbf{x},t)$ and $\psi^\dagger(\mathbf{x},t)$:

$$\frac{\partial\psi}{\partial t} = \frac{i}{\hbar}[H,\psi]; \quad \frac{\partial\psi^\dagger}{\partial t} = \frac{i}{\hbar}[H,\psi^\dagger].$$

We may consider H as governing the motion of a nonlinear, three-dimensional classical wave field. The commutation rule for the canonically conjugate variables,

$$q(\mathbf{x}) = (\psi+\psi^\dagger)(\hbar/2)^{\frac{1}{2}} \quad\text{and}\quad p(\mathbf{x}) = (\psi-\psi^\dagger)(\hbar/2)^{\frac{1}{2}}i,$$

is replaced by the Poisson-bracket relation. $\hbar$ is retained insofar as it enters in the kinetic energy of the field, describing the DeBroglie wave character of the associated particles. The classical equation of motion has special, exact, separable solutions of the form $\psi = f(\mathbf{x}) \times \exp(-iEt/\hbar)$, where $f(\mathbf{x})$ and E are obtained from the nonlinear eigenvalue problem

$$Ef = -\frac{\hbar^2}{2M}\nabla^2 f + \int V(\mathbf{x}-\mathbf{x}')\,|f(\mathbf{x}')|^2 d^3x'\cdot f(\mathbf{x}),$$

$$\int |f|^2 d^3x = N.$$

We note that there is always a solution of uniform density, namely,

$$f(x) = \left(\frac{N}{L^3}\right)^{\frac{1}{2}}, \quad E = \frac{N}{L^3}\int V(\mathbf{x})d^3x.$$

But if $V(\mathbf{x})$ is negative in some region of space, there may be other solutions, such as a periodic solution with E lower than for the uniform solution. In the classical theory there are solutions in the vicinity of each exact solution in which the field carries out small oscillations. Writing $Q = (\varphi+\varphi^\dagger)(\hbar/2)^{\frac{1}{2}}$, $\psi = e^{-iEt/\hbar}[f(\mathbf{x})+\varphi(\mathbf{x},t)]$ and linearizing the equation of motion, we obtain

$$+\hbar^2\ddot{Q} = +\frac{\hbar^2}{2M}\left(\nabla^2 - \frac{\nabla^2 f}{f}\right)\left\{-\frac{\hbar^2}{2M}\left(\nabla^2 - \frac{\nabla^2 f}{f}\right)Q(\mathbf{x},t)\right.$$
$$\left.+2f\int V(\mathbf{x}-\mathbf{x}')f(\mathbf{x}')Q(\mathbf{x}')\cdot d^3x'\right\}.$$

The character of the oscillation spectrum depends on $V(\mathbf{x})$ and on the underlying solution $f(\mathbf{x})$. For the uniform solution one obtains Bogolyubov's[2] spectrum yielding phonons for long wavelengths, free-particle behavior for short wavelengths.

The classical analysis may be used to find eigenfunctions by noting that three elementary canonical transformations are involved; a time-dependent one introducing a phase, a linear shift $\psi\rightarrow f+\psi$, and a normal-mode transformation. These are then simple unitary transformations in the quantum theory, the first adding a term to H. We are thus led to the set of approximate eigenfunctions,

$$\Phi = \exp(S_2)\cdot\exp(S_3)\Phi_0(\cdots N_k\cdots).$$

Φ_0 are eigenfunctions of the number operators $N_k = \psi_k^\dagger\psi_k$. [$\psi_k$ is a Fourier component of $\psi(\mathbf{x})$.]

$$S_2 = \int \{f(\mathbf{x})\psi^\dagger(\mathbf{x}) - f^*(\mathbf{x})\psi\}d^3x,$$

and S_3 is a quadratic form in ψ, $\psi^\dagger$. From the set we construct eigenfunctions of the total number and