

Spin-lattice relaxation of nuclei in semiquantum liquid hydrogen

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A concrete nuclear relaxation mechanism is proposed. This mechanism explains in a natural way the anomalous temperature and concentration dependence of the spin-lattice relaxation time of nuclei in liquid hydrogen at low temperatures.

The dependence of the nuclear spin-lattice relaxation time T_1 on the temperature and on the concentration (K) of orthohydrogen in the liquid was studied experimentally at low temperatures in Refs. 1 and 2. Various attempts^{2,3} have been made to explain the experimental results in the theory of classical liquids, but these theories do not give a satisfactory explanation of the results, as was pointed out in Ref. 2.

Recent years have furnished solid support for the model developed by Andreev.^{4,5} According to this model, the elementary excitations in semiquantum liquids are, along with phonons, sets of two-level systems with a broad spectrum of splittings and with a nearly flat distribution in splitting energy, $\nu(\epsilon) = \nu_0 = \text{const}$. This model was used in Ref. 6 to calculate the nuclear spin-lattice relaxation time of semiquantum liquids. It was shown that T_1 is inversely proportional to the temperature of the liquid. This is a universal property of semiquantum liquids and agrees well with the experimental results of Ref. 1.

In Ref. 6 we did not specify the particular mechanism for the interaction of the nuclear spins with the elementary tunneling excitations (the two-level systems). The question of the dependence of T_1 on the concentration K was thus left open. In the present paper we wish to discuss an effective interaction which leads to a coupling of the nuclear spins with the pseudospins of two-level systems.

We describe the elementary excitations by the Hamiltonian

$$\mathcal{H} = \sum_n \epsilon_n S_n^z, \quad (1)$$

where S is the pseudospin of the two-level system, and ϵ_n is the splitting energy of the two-level system of index n . The Hamiltonian of the Zeeman energy of the nuclei is

$$\mathcal{H} = \omega_0 \sum_i I_i^z, \quad (2)$$

where ω_0 is the NMR frequency of the hydrogen nuclei, and I is the nuclear spin.

In second-order perturbation theory, the interaction of the nuclear spins with phonons and of the pseudospins with phonons leads to an indirect interaction of the nuclear spins with the pseudospins of the two-level systems. We know,² however, that the direct interaction of nuclear spins with phonons is ineffective. A mechanism far more effective for coupling nuclear spins with phonons is the indirect mechanism due

to the coupling of the nuclei of ortho-hydrogen molecules with the rotational angular momentum of the molecules and the coupling of the rotational angular momentum with phonons; i.e., the indirect interaction can occur in the scheme $I \sim L \sim \text{phonons}$.

In the absence of an interaction between the ortho-hydrogen molecules and the surrounding molecules, the states of the former would of course be triply degenerate in the direction of the angular momentum (for ortho-hydrogen in the ground state, we have $L = 1$). The interaction of an ortho-hydrogen molecule with the surrounding molecules lifts this degeneracy, however, as was shown in Ref. 7. The typical intervals found for the splittings ΔE are ~ 1.5 K. The magnitude of the splitting can be written in the form

$$\Delta E = \alpha + \beta K. \quad (3)$$

Expression (3) states that the splitting ΔE stems from two different factors. The first term, α , describes the splitting of the states of the ortho-hydrogen molecule due to its interaction with surrounding para-hydrogen molecules (obviously, α is independent of the concentration of ortho-hydrogen molecules). The second term, βK , describes the splitting of the states $L_z = 0, \pm 1$ due to the coupling of the ortho-hydrogen molecule with the surrounding ortho-hydrogen molecules. This splitting is obviously proportional to K . Since the mechanisms for the para-ortho and ortho-ortho interactions are different, the splitting ΔE will depend on K .

Let us write that part of the complete Hamiltonian of the system which we will need in order to derive the indirect nuclear-phonon interaction:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1, \quad (4)$$

$$\mathcal{H}_0 = \omega_0 \sum_i I_i^z + \Delta E \sum_i L_i^z + \sum_p \omega_p a_p^+ a_p,$$

$$\mathcal{H}_1 = \mathcal{H}_{IL} + \mathcal{H}_{L\phi}, \quad (5)$$

$$\mathcal{H}_{IL} = A \sum_i (I_i \cdot L_i) \quad \mathcal{H}_{L\phi} = \sum_{ip} C_i^m(p) L_i^m (a_p - a_{-p}^+).$$

For simplicity, we are assuming that the splitting spectrum of the states with $L_z = 0, \pm 1$ is equidistant. Here $\sum_p \omega_p a_p^+ a_p$ is the Hamiltonian of the phonon system in the second-quantization representation, ${}^p \mathcal{H}_{IL}$ is the Hamiltonian of the interaction of the nuclear spin with the rotational angular momentum, and $\mathcal{H}_{L\phi}$ is the Hamiltonian of the interaction of the rotational angular momentum of the ortho-hydrogen molecules with phonons.²

Eliminating the orbital states in second-order perturbation theory in the operator \mathcal{H}_1 , we find the Hamiltonian of the indirect nuclear-phonon interaction:

$$\mathcal{H}_{I\phi} = \frac{A}{2\Delta E} \sum_{ip} \{ I_i^+ L_i^z C_i^+(p) (a_p - a_{-p}^+) + I_i^- L_i^z C_i^-(p) (a_p - a_{-p}^+) \}. \quad (6)$$

This interaction will evidently lead to a coupling of the nuclear spins with the pseudo-spins of the two-level systems through the phonon field. This coupling may be either a "single-mode" or a "two-mode" coupling. The single-mode coupling is \mathcal{H}_{IS} ⁽¹⁾

$= \sum_{ij} J_1 S_i^+ I_j^-$, and the two-mode coupling is

$$\mathcal{H}_{IS}^{(2)} = \sum_{ijk} J_2 (I_i^+ S_j^- S_k^+ + I_i^- S_j^+ S_k^-).$$

It is easy to see, however, that the spin-lattice relaxation time calculated for the nuclei from $\mathcal{H}_{IS}^{(1)}$ will be proportional to the number of two-level systems at the NMR frequency, while the relaxation due to the interaction $\mathcal{H}_{IS}^{(2)}$ will be more effective, since two-level systems with all possible splittings will contribute to the relaxation in this case.

We see from (6) that the constant of the effective nucleus-phonon interaction is proportional to the reciprocal of ΔE , so that the interaction constant J_2 will also depend on the reciprocal of $\Delta E = \alpha + \beta K$. Because of this reciprocal dependence, the spin-lattice relaxation time of the nuclei will decrease with increasing concentration of the ortho-hydrogen in the liquid. This conclusion agrees with the experimental results of Ref. 1.

In summary, the anomalous temperature and concentration dependence of the spin-lattice relaxation time of hydrogen nuclei at low temperatures can be explained from a common standpoint by the relaxation mechanism proposed here, which is based on the arguments of Refs. 4–6.

¹M. Bloom, *Physica* **23**, 378 (1957).

²A. Abragam, *The Principles of Nuclear Magnetism*, Clarendon Press, Oxford (1961) (Russ. transl. IL, Moscow, 1963).

³T. Moriya, *Prog. Theor. Phys.* **18**, 567 (1957).

⁴A. F. Andreev, *Pis'ma Zh. Eksp. Teor. Fiz.* **28**, 603 (1978) [*JETP Lett.* **28**, 556 (1978)].

⁵A. F. Andreev and Yu. A. Kosevich, *Zh. Eksp. Teor. Fiz.* **77**, 2518 (1979) [*Sov. Phys. JETP* **50**, 1218 (1979)].

⁶L. L. Buishvili and A. I. Tugushi, *Zh. Eksp. Teor. Fiz.* **79**, 1898 (1980) [*Sov. Phys. JETP* **52**, 958 (1980)].

⁷T. Nakamura, *Prog. Theor. Phys.* **14**, 135 (1955).

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