

<sup>1</sup>Unless otherwise stipulated, the integrations in Eqs. (2, 1), (2.2) and in all subsequent equations is between infinite limits.

<sup>2</sup>When  $\varepsilon < 0$  the circuit around the poles of the integrand in (2.11) is given by the rule

$$w(\xi) = \lim_{\varepsilon \rightarrow 0} \int_{-\infty}^{\infty} \frac{e^{i q \xi} d\xi}{1 + \varepsilon(q + i\xi)^2}.$$

<sup>1</sup>A. S. Davydov, *Teor. Mat. Fiz.* **40**, 408 (1979).

<sup>2</sup>A. S. Davydov, *Zh. Eksp. Teor. Fiz.* **78**, 789 (1980) [*Soviet Phys. JETP* **51**, 397 (1980)].

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<sup>7</sup>A. G. Litvak and A. M. Sergeev, *Pis'ma Zh. Eksp. Teor. Fiz.* **27**, 549 (1978) [*JETP Lett.* **27**, 517 (1978)].

<sup>8</sup>M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions*, Dover, New York, 1965.

<sup>9</sup>S. I. Pekar, *Issledovaniya po elektronnoi teorii kristallov* (Studies of the electron theory of crystals) Gostekhizdat, Moscow, 1951 [English translation U. S. AEC Report AEC-tr-5575 (1963); German translation published by Akademie Verlag].

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## Nuclear spin-lattice relaxation in semiquantum liquids

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We calculate in this paper the temperature-dependence of the nuclear spin-lattice relaxation time  $T_1$  of semiquantum liquid hydrogen. We show that  $T_1$  is inversely proportional to the temperature  $T$ . Such a  $T$ -dependence of  $T_1$  is a universal consequence of the specific properties of semiquantum liquids. The results are in good agreement with experimental data on  $T_1$  measurements in liquid hydrogen.

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Andreev<sup>1-3</sup> has recently developed a theory of semiquantum liquids. These are liquids in the temperature range  $T_d \ll T \ll \Theta$  ( $\Theta$  is the Debye temperature and  $T_d$  determines the quantum indeterminacy of the energy which is connected with the delocalization of the particles in the liquid which is caused by tunnel transitions). In his papers he showed that semi-quantum liquids are characterized by a universal temperature-dependence of the thermodynamic quantities and the kinetic coefficients, which is caused by the specific properties of semi-quantum liquids. He showed, in particular, that the specific heat and the heat conductivity coefficient are proportional to the temperature  $T$  while the viscosity is inversely proportional to  $T$ , in good agreement with experimental data.

One must expect that the specific properties of semiquantum liquids will also appreciably alter the temperature-dependence of the spin-lattice relaxation (SLR) time of the nuclei. Indeed, for liquid hydrogen in the temperature range 14 to 20 K one observes experimentally an inverse temperature-dependence of the SLR time of the nuclei<sup>4</sup> and this cannot be explained on the basis of the theory of classical liquids. In this temperature range the hydrogen must have the properties of a semi-quantum liquid.

The aim of the present paper is the calculation of the nuclear SLR time of semi-quantum liquids using the theories developed in the above-mentioned papers.<sup>1-3</sup>

When evaluating the SLR time we use a model analogous to the one used by Andreev<sup>3</sup> when calculating the heat conductivity coefficient. For the sake of simplicity we shall assume that the particles have spin  $\frac{1}{2}$  (if the particle spin is larger than  $\frac{1}{2}$ , it will not lead to any alteration in principle of the arguments which follows below) and, by analogy with Ref. 3 that perturbation of the form

$$\mathcal{V} = -fq,$$

is applied to the system, where  $q$  is the operator of the flux of the Zeeman energy between excited states of the particles in the liquid with energies  $\varepsilon_1 \pm \hbar\omega_0$  and  $\varepsilon_2 \pm \hbar\omega_0$ , where  $\omega_0$  is the NMR frequency and  $f$  is a harmonic generalized force. To evaluate the current  $q$  we write down the Hamiltonian of the system:

$$H = H_1 + H_2 + H_{int},$$

$$H_1 = \sum_{\sigma} \varepsilon_{1\sigma} a_{1\sigma}^+ a_{1\sigma}, \quad H_2 = \sum_{\sigma} \varepsilon_{2\sigma} a_{2\sigma}^+ a_{2\sigma}, \quad (1)$$

$$H_{int} = J(a_{1+}^+ a_{2-} + a_{1-}^+ a_{2+} + a_{2+}^+ a_{1-} + a_{2-}^+ a_{1+}), \quad \varepsilon_{i\pm} = \varepsilon_i \pm \hbar\omega_0. \quad (2)$$

$J$  is the interaction constant, and  $a_{i\sigma}^+$  and  $a_{i\sigma}$  are the creation and annihilation operators for particles in states with energies  $\varepsilon_{i\sigma}$ . In contrast to Ref. 3 we retain only the interaction which causes the transition of particles between the levels  $\varepsilon_{1\pm} - \varepsilon_{2\mp}$ , i.e., the transition between the states is accompanied by a spin flip. As we are merely interested in the temperature dependence of the SLR time, we shall not define concretely the interaction mechanism.

To find the expression for the Zeeman energy flux, we write down the explicit form of the Zeeman energy operator

$$E_z = \frac{\hbar\omega_0}{2} (N_+ - N_-),$$

$$N_+ = a_{1+}^+ a_{1+} + a_{2+}^+ a_{2+}, \quad N_- = a_{1-}^+ a_{1-} + a_{2-}^+ a_{2-},$$

the change in which is given by the equation

$$\frac{dE_z}{dt} = \frac{\hbar\omega_0}{2} \left( \frac{dN_+}{dt} - \frac{dN_-}{dt} \right),$$

i.e., the Zeeman energy flux operator can be expressed by the formula

$$q = \frac{\omega_0}{2i} [N_+ - N_-, H_{int}] = -i\omega_0 J (a_{1+}^+ a_{2+} + a_{2+}^+ a_{1+} - a_{2-}^+ a_{1+} - a_{1+}^+ a_{2+}). \quad (3)$$

For the sake of argument, we assume that  $a_{i\sigma}^+$  and  $a_{i\sigma}$  satisfy boson commutation relations.

One easily obtains from (3) the matrix elements of the current operator between the states  $|1+\rangle$ ,  $|2-\rangle$ ;  $|1-\rangle$ ,  $|2+\rangle$ :

$$q_{1+, 2-} = q_{2+, 1-} = -q_{2-, 1+} = -q_{1-, 2+} = -i\omega_0 J. \quad (4)$$

The dissipation of the applied-field energy will have the form

$$\dot{E} = \{w_{1+, 2-} (\varepsilon_{1+} - \varepsilon_{2-}) (N_{2-} - N_{1+}) + w_{1-, 2+} (\varepsilon_{2+} - \varepsilon_{1-}) (N_{1-} - N_{2+})\}. \quad (5)$$

Under the condition  $\hbar\omega \ll T$  (where  $\omega$  is the frequency of the alternating field), taking into account that the transition probability caused by the perturbation  $\hat{V}$  has the form

$$w_{1+, 2-} = \frac{\pi |f|^2}{2\hbar} |q_{1+, 2-}|^2 \{ \delta(\hbar\omega + \varepsilon_{1+} - \varepsilon_{2-}) + \delta(\hbar\omega + \varepsilon_{2-} - \varepsilon_{1+}) \},$$

we get for (5)

$$\dot{E} = \frac{\pi |f|^2}{2\hbar T} |q_{1+, 2-}|^2 \hbar^2 \omega^2 \{ N_{1+} \delta(\hbar\omega + \varepsilon_{1+} - \varepsilon_{2-}) + N_{1-} \delta(\hbar\omega + \varepsilon_{1-} - \varepsilon_{2+}) \},$$

$$N_{i\sigma} = e^{-\varepsilon_{i\sigma}/T} [2 + e^{-\varepsilon_{i\sigma}/T} + e^{-\varepsilon_{i\sigma}/T} + e^{-\varepsilon_{i\sigma}/T} + e^{-\varepsilon_{i\sigma}/T}]^{-1},$$

$N_{i\sigma}$  is the occupation number for the state of energy  $\varepsilon_{i\sigma}$ . Following Ref. 3, we can easily establish a relation between the amplitude of the alternating field  $f$  and the temperature difference  $\delta T$  between the spin and the lattice system, which has the form

$$f = \delta T / i\omega T.$$

We shall thus finally have

$$\dot{E} = \frac{\pi \hbar \omega_0^2 J^2}{2T^2} (\delta T)^2 \{ N_{1+} \delta(\hbar\omega + \varepsilon_{1+} - \varepsilon_{2-}) + N_{1-} \delta(\hbar\omega + \varepsilon_{1-} - \varepsilon_{2+}) \}. \quad (6)$$

We denote by  $\rho(\varepsilon_1, \varepsilon_2) d\varepsilon_1 d\varepsilon_2$  the probability that a given particle of the liquid has not only a ground state, but also two close-lying excited states with energies  $\varepsilon_1$  and  $\varepsilon_2$ . For small  $\varepsilon_1$  and  $\varepsilon_2$  the probability  $\rho$  can, according to Refs. 3 and 5, be assumed to be a constant quantity.

Averaging (6) and taking into account that  $\hbar\omega \ll T$  we get finally an expression for the energy dissipation per unit volume of the liquid:

$$\dot{E} = \frac{\pi a \hbar \omega_0^2 n \rho J^2 (\delta T)^2}{2T^2}, \quad a = \int_0^\infty \frac{e^{-z}}{2+4e^{-z}} dz, \quad (7)$$

where  $n$  is the number of particles per unit volume of the liquid. On the other hand, from irreversible thermodynamics it is well known that the connection

between the energy dissipation and the rate of change of entropy is<sup>6</sup>

$$\dot{E} = -2\sigma T, \quad (8)$$

where  $2\sigma$  is the energy production function.

The non-equilibrium state of our system is characterized by a single macroscopic parameter—the  $z$ -component of the nuclear magnetization  $M$ , which satisfies the following relaxation equation:

$$\frac{dM}{dt} = -\frac{M - M_0}{T_1}, \quad (9)$$

where  $M_0$  is the equilibrium value of the nuclear magnetization and  $T_1$  the required nuclear SLR time. If we use the Curie law  $M = CH/T$  ( $C$  is the Curie constant,  $H$  is the external magnetic field, and  $T$  the spin temperature) we can rewrite (9) in the form

$$\frac{d\delta\beta}{dt} = -\frac{\delta\beta}{T_1}, \quad (10)$$

where  $\beta = 1/T$  is the inverse temperature of the spin system,  $\delta\beta = \delta T/T_0^2$ , and  $T_0$  is the lattice temperature. Using the well known thermodynamic relations between  $\sigma$  and the macroscopic parameter  $\delta\beta$  from Ref. 6, using (10) we can easily obtain an expression for the function  $\sigma$ :

$$\sigma = \frac{1}{2T_1} \frac{(\delta\beta)^2}{\langle (\delta\beta)^2 \rangle},$$

where  $\langle (\delta\beta)^2 \rangle$  is the mean square of the fluctuating parameter  $\delta\beta$  in the equilibrium state. Using next the well known thermodynamic formula<sup>6</sup>

$$\langle (\delta T)^2 \rangle = T^2/c_s,$$

and the expression for the specific heat of a non-degenerate paramagnet at high temperatures

$$c_s = n(\hbar\omega_0)^2/4T,$$

we finally get for  $\sigma$

$$\sigma = \frac{n(\hbar\omega_0)^2}{8T_1} \frac{(\delta T)^2}{T^2}.$$

Using this expression Eq. (8) will be of the form

$$\dot{E} = \frac{(\hbar\omega_0)^2}{4T_1 T^2} (\delta T)^2. \quad (11)$$

Comparing Eqs. (7) and (11) we get the following expression for the nuclear SLR time:

$$\frac{1}{T_1} = \frac{2\pi a \rho J^2}{\hbar} T. \quad (12)$$

We see from (12) that the nuclear SLR time is inversely proportional to the temperature  $T$ . Such a  $T$ -dependence of  $T_1$ , like the  $T$ -dependence of other kinetic coefficients, is a universal consequence of the specific properties of semi-quantum liquids. From the example of liquid hydrogen we see that (12) agrees well with experimental data.<sup>4</sup>

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<sup>3</sup>A. F. Andreev and Yu. A. Kosevich, Zh. Eksp. Teor. Fiz. 77,

## Increase of magnetization of a paramagnet by an alternating electric field

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A new effect connected with an increase of the magnetization of a paramagnet by an alternating electric field is observed. The experiment was performed on Al-O<sup>-</sup> centers in quartz by the EPR method. The dependences of the effect on the intensity and frequency of the electric field are studied at different temperatures and crystal orientations. An interpretation of the observed phenomenon is offered, with account taken of the fact that not only the electric dipole moment but also the spin moment is connected with the Al-O<sup>-</sup> centers. It is shown that the polarization of the electric dipoles by the alternating electric field is transferred to the spin system by relaxation processes. Regardless of the polarity of the electric field, the magnetization of the paramagnet increases in this case. The observed phenomenon is in essence a relaxational magnetoelectric effect.

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Hou and Bloembergen<sup>1</sup> have described an increase of the magnetization of a paramagnet by an electric field. They have shown that the gist of the phenomenon reduces to the magnetoelectric effect predicted earlier<sup>2,3</sup> and observed in antiferromagnets<sup>4</sup> and in ferromagnets.<sup>5</sup> The change of the magnetization in these experiments is attributed to the terms of order EH contained in the thermodynamic potential  $\Phi$  ( $E$  and  $H$  are the electric and magnetic field strengths). The effect takes place in both constant and alternating fields. Phenomena of this kind in paramagnets were investigated theoretically by Roitsin<sup>6</sup> by the spin-Hamiltonian method.

In the present study we have observed an appreciable increase (by approximately one order of magnitude) of the magnetization of a paramagnet by an alternating electric field. In its outward attributes, the observed phenomenon is similar to the magnetoelectric effect, but is substantially different with respect to the mechanism of its production and to its behavior. In particular, no increase of the magnetization was observed in a dc electric field in our experiments.

### THE EXPERIMENT

The measurements were performed by the EPR method in the 3-cm band at  $T = 4.2$  K and 1.5 K, on single-crystal  $\alpha$ -quartz with Al-O<sup>-</sup> centers. The center density was  $\sim 10^{18}$  cm<sup>-3</sup>. The paramagnetism of these samples is due to a hole (spin  $S = \frac{1}{2}$ ) localized on one of the two oxygen ions (which we shall label 1 and 2) of the AlO<sub>4</sub><sup>4-</sup> tetrahedron in which Si<sup>4+</sup> is re-

placed by Al<sup>3+</sup>.<sup>7</sup> The energy of hole localization on the two other oxygen ions is 30 meV higher.<sup>8</sup> Owing to the anisotropy of the  $g$ -factor, the holes in positions 1 and 2 produce two separately registered groups of EPR lines (six lines each, owing to the hyperfine interaction with the Al<sup>27</sup>). For the details of the EPR and structure of the center see Ref. 7.

In our experiments, graphite electrodes were deposited directly on the crystal, and the electric field was fed to them through clamp contacts. The electric field  $E$  was applied perpendicular to the threefold axis of the crystal,  $E \perp L_3$ . In addition, the condition  $E \parallel H_0$  was satisfied, where  $H_0$  is the external magnetic field. The dc electric field increased the intensity of one group of lines and decreased that of the other, so that the combined net magnetization of the system remained unchanged (for details see Ref. 9).

We have observed that when an alternating electric field of frequency  $\nu_E$  was applied to the crystal, the intensities of both groups of EPR lines increased to equal degrees. The effect is a maximum at  $E \perp L_2$  and is absent at  $E \parallel L_2$ , where  $L_2$  is the twofold axis of the defective tetrahedron in which Si<sup>4+</sup> is replaced by Al<sup>3+</sup>. In these experiments, the EPR signals were observed on an oscilloscope screen. The frequencies of the electric and magnetic fields were rational multiples, and the signals were obtained at the maximum amplitude of the electric field. The increase of the signal was independent of the polarity of the electric field. When an automatic plotter with a time constant larger than  $\nu_E^{-1}$  was used as a recorder, the averaged effect