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Nuclear Magnetic Relaxation in Solid Para-Deuterium

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In this note we point out the difference of the nuclear relaxation mechanism between solid ortho-H_2 and para-D_2 below T_λ. It has been investigated theoretically by a number of authors[1] that the low-lying rotational excited states of solid ortho-H_2 or para-D_2 can be described in terms of a quasi-particle of librational excitation; this quasi-particle may be called a libron. The present author estimated the nuclear magnetic spin-lattice relaxation time T_1 of solid ortho-H_2 below T_λ on the basis of the inelastic scattering of librons by nuclear spins.[2] The relaxation mechanism assumed is the intramolecular magnetic dipole coupling and $\boldsymbol{I}$-$\boldsymbol{J}$ coupling modulated by the intermolecular electric quadrupole interaction which gives rise to librons. However, for para-D_2 another relaxation mechanism has to be taken into account in addition to the mechanism assumed for ortho-H_2. This mechanism arises from the coupling between the electric quadrupole moment of deuteron and the field gradient at its position.

The perturbation Hamiltonian H' responsible for the nuclear relaxation for a homonuclear $^1\Sigma$ diatomic molecule in the subspace $J=1$ can be written as[3]

$$h^{-1}H' = -d\boldsymbol{I}^2\boldsymbol{J}^2 - (c-1.5d)\boldsymbol{I}\cdot\boldsymbol{J} + 3d(\boldsymbol{I}\cdot\boldsymbol{J})^2, \quad (1)$$

$$c = \mu_i H'(I_i h)^{-1}, \quad (2)$$

$$hd = \left(\frac{\mu_i^2}{I_i^3 r^3}\right)\frac{I(I+1)+4I_i(I_i+1)}{5(2I-1)(2I+3)} - \frac{eQ(\partial^2 V/\partial Z^2)}{10I_i(2I_i-1)}\left[1-\frac{I(I+1)+4I_i(I_i+1)}{(2I-1)(2I+3)}\right], \quad (3)$$

where $\boldsymbol{I}$ is the resultant nuclear spin angular momentum in units of $\hbar$ and $\boldsymbol{J}$ the molecular rotational angular momentum in units of $\hbar$. μ_i is the nuclear magnetic moment and I_i the corresponding nuclear spin. H' is the magnetic field due to the molecular rotation. The quantity r is the distance between the two nuclei, Q is the nuclear quadrupole moment. $\partial^2 V/\partial Z^2$ is the field gradient along the molecular axis. For hydrogen molecule, Q is zero. We see from Eq. (1) that the temperature dependence of T_1 below T_λ is the same for both ortho-H_2 and para-D_2 except their magnitude.

From the molecular beam experiment,[3] the coefficients c and d are known as

$$c = 113.8\ \text{Kc/sec}, \quad d = 57.68\ \text{Kc/sec} \quad \text{for } H_2, \quad (4\cdot a)$$

$$c = 8.78\ \text{Kc/sec}, \quad d = 25.24\ \text{Kc/sec} \quad \text{for } D_2. \quad (4\cdot b)$$

Hence, the perturbation Hamiltonian is

$$h^{-1}H' = -57.68\boldsymbol{I}^2\boldsymbol{J}^2 - 27.28\boldsymbol{I}\cdot\boldsymbol{J} + 173.04(\boldsymbol{I}\cdot\boldsymbol{J})^2 \quad \text{for } H_2, \quad (5\cdot a)$$

$$h^{-1}H' = -25.24\boldsymbol{I}^2\boldsymbol{J}^2 + 29.07\boldsymbol{I}\cdot\boldsymbol{J} + 75.72(\boldsymbol{I}\cdot\boldsymbol{J})^2 \quad \text{for } D_2. \quad (5\cdot b)$$

If we use Eq. (5·b) instead of Eq. (5·a) and repeat the same calculation as the one made in the case of ortho-H_2, we can get the T_1 for para-D_2. The result shows that the T_1 below T_λ for para-D_2 is 5.1 times larger than that of ortho-H_2.

In addition, for para-D_2 the most dominant contribution in Eq. (5·b) comes from the coupling between the electric quadrupole moment of deuteron and the field gradient, because in Eq. (3) the quadrupole effect is 22.51 Kc/sec. We, therefore, conclude that the dominant relaxation mechanism of solid para-D_2 is the electrostatic quadrupole interaction rather than the mechanism assumed for ortho-H_2. The result obtained from the numerical calculation for ortho-H_2 by the present author shows that the T_1 just below T_λ is about 10 sec.[2] On the other hand, a recent experimental result by White et al. on 93% para-D_2 at 2.0°K shows very long relaxation time T_1, i.e. 195 ± 10 sec.[4] In this case, T_λ is 3.4°K.

The theoretical T_1 for this experiment is given by

$$\left\{T_1 \text{ of ortho-}H_2 \text{ at temperature } T = \frac{2.0}{3.4}\times 4.2^\circ\text{K}\right\}\times 5.1\,, \qquad (6)$$

where 4.2°K is the theoretical T_λ given by Okada, Matsuda and the present author.[1] The result is 560 sec.

Comparing the theoretical and experimental results, it seems to be consistent to use Eq. (1) as a perturbation Hamiltonian for nuclear magnetic relaxation for both ortho-H_2 and para-D_2.

We still do not have precise experimental data available for the temperature dependence of T_1 well below T_λ for both cases. Further experiment is to be hoped.

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