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SPECIFIC HEAT ANOMALY AND NUCLEAR RESONANCE IN CRYSTALLINE HYDROGEN IN CONNECTION WITH NEW DATA ON ITS STRUCTURE

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FOLLOWING Keesom,[7] the interpretation of experimental data on the specific heat anomaly[1,2] and nuclear resonance has been based[3-6] so far on a close-packed hexagonal lattice structure for crystalline hydrogen. Recently Lazarev[8] and co-workers have found an error in this previous work and showed that the x-ray lines obtained could be explained either by a hexagonal lattice, deviating a little from close-packing ($a = 3.7$, $c = 6.42$), or by a tetragonal lattice ($a = 4.5$, $c = 3.68$). It was therefore necessary to determine the degree of agreement of the experimental data on the specific heat anomaly with the new crystal lattices ascribed to hydrogen. Since the structure is not yet resolved unambiguously, it is no less important to calculate the anisotropy of nuclear resonance in a hydrogen single crystal* and to find out whether the structure can be elucidated by the nuclear resonance method.

1. From Hill and Ricketson's experimental data,[1] Nakamura[2] found that within a certain temperature range the dependence of the anomalous specific heat, C_V, on the temperature T and concentration ρ is given by

$$C_V = \frac{R}{T^2}(\alpha\rho + \beta\rho^2), \tag{1}$$

where $\alpha = 1.1$ and $\beta = 15.7$. He also obtained a similar formula theoretically, and found the coefficients α and β to be very sensitive to the crystal structure. For a close-packed hexagonal lattice Nakamura obtained α strictly equal to zero, with $\beta = 20$ to the first approximation and 18 to the second. For the newly determined tetragonal lattice we calculated for α and β values in better agreement with experiment: $\alpha = 0.3$ and $\beta = 18$ to the first approximation and 16 to the second.

2. In Van Vleck's formula[9] for the second moment of a resonance line due to intermolecular† dipole-dipole interaction, the crystal structure is taken into account through the sum

$$\sum_k r_{ik}^{-6}(3\cos^2\theta_{ik} - 1)^2, \tag{2}$$

where r_{ik} is the distance between the i-th and k-th molecules, and θ_{ik} is the angle between the magnetic field and the vector θ_{ik}. We have calculated the sum for an arbitrary magnetic field direction and obtain the following expressions for tetragonal and hexagonal lattices:

$$\sum_k r_{ik}^{-6}(3\cos^2\theta_{ik} - 1)^2 = c^{-6}(9.2 + 0.88\cos^2\theta + 0.24\cos^4\theta), \tag{3}$$

$$\sum_k r_{ik}^{-6}(3\cos^2\theta_{ik} - 1)^2 = a^{-6}(11.2 - 16.4\cos^2\theta + 19.7\cos^4\theta), \tag{4}$$

where θ is the angle between the field and the fourfold or corresponding sixfold axis. The considerable difference between the anisotropies for a tetragonal (3) and hexagonal (4) lattice can conveniently be used to decide the structure of solid hydrogen.

3. Moriya and Motizuki[6] proposed a theory of spin-lattice relaxation in solid hydrogen. Because of the difficulty of the calculation, the relaxation time, T_1, was derived for the simplest case corresponding to the magnetic field parallel to the sixfold axis. Since the determination of the orientation of a single crystal under experimental conditions presents considerable difficulty, we considered it necessary to carry through the cumbersome calculation and obtained T_1 for the general case of an arbitrary field orientation both for the hexagonal and for the tetragonal lattice. We do not give here the final formula, which is complicated. It has been established that the anisotropy of T_1 is not more than 5%, so that in measurements of T_1 on single crystals or polycrystalline material one would expect practically identical results.

4. The crystal lattice of deuterium was first determined by Kogan, Lazarev, and Bulatova[8] (tetragonal, a = 3.35, c = 5.79) and it seemed useful to explore the possibility of confirming these results by data on the anisotropy of nuclear resonance. We have limited ourselves to examining orthodeuterium, as the intensity of the resonance is almost an order of magnitude greater than for paradeuterium. The rotational state of orthodeuterium has spherical symmetry, so that intramolecular broadening should be absent. According to Hatton and Rollin[3] one can also neglect the line broadening caused by the quadrupole moment of the deuterium nucleus. The summation (2) leads to considerable anisotropy:

$$\sum_k r_{ik}^{-6}(3\cos^2\theta_{ik} - 1)^2 = a^{-6}(8.32 - 3.2\cos^2\theta). \qquad (5)$$

In conclusion I would like to express my sincere thanks to B. G. Lazarev for his interest in the work.

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[*] The present work was started in connection with an experimental search for anisotropy in nuclear resonance in hydrogen, carried out by A. A. Galkin and I. V. Matyash, to whom we are grateful for suggesting the problem.

[†] The measurements of the specific heat anomaly[1] and Nakamura's theory[2] indicate that in this temperature range an appreciable anistropy of molecular orientation is already starting. The intramolecular resonance line broadening connected with this can, in principle, be calculated from Moriya and Motizuki's theory,[6] but such a calculation is extremely unwieldy. We should point out that the experimental data given by Sugawara et al.,[10] with values of the second moment determined on polycrystalline material, indicates the relatively small intramolecular broadening, at least for small ortho-hydrogen concentrations ($\rho \sim$ 10–20%).

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DENSITY OF CHARGED PARTICLES IN THE CHANNEL OF A SPARK DISCHARGE

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IN order to determine the density of charged particles in the channel of a spark discharge we have investigated the shape of the He II line λ = 4686 A produced in a discharge in helium. Under the experimental conditions in the present work (p = 1.5 — 12 atmos, C = 0.05 μf, U = 2 — 12 kv, L = 0.18 — 3.6 μh) up to 0.3 μsec after the initiation of the discharge only the spark lines of helium (4686 and 3203 A) are radiated; however, the shapes of these lines could not be examined quantitatively because of smearing. The line shapes were recorded by means of a photoelectric system in which traces are made at two different instants of time after the initiation of the discharge.[1,2] It has been established that at the beginning of the discharge the 4686 A line is highly broadened and shifted toward the red, although there is no noticeable asymmetry. The displacement was measured with respect to the position of the same line at later instants of time, when the line exhibits essentially no displacement (t ≈ 1 μsec). It is reasonable to assume that the red shift of the line is due to the quadratic Stark effect.[3] The absolute values of the displacement (up to 8 A) and the half width (up to 50 A) indicate that in the initial stages of the discharge the density of charged particles is quite appreciable.

This density can be estimated by three methods:

1. Using the shape of the 4686 A line computed by Unsöld on the basis of the Holtsmark theory for the linear Stark effect it is possible to find the density of charged particles N by matching (at the wings of the line) the experimental and theoretical shapes (cf. for example, references 5 and 6).

It should be noted that for line shapes which