

FIG. 1. Variation of the spin resonance second moment, σ^2 , with the ratio, ρ , of M centers to F centers.

strengths for the M and F centers in their usually recognized bands.² Second moment measurements are notoriously lacking in precision and the present report must be considered preliminary.

If we let ρ be the ratio of the total number of M centers to F centers in a given sample and σ^2 , σ_F^2 , σ_M^2 be the second moments of the observed and individual spin resonances, we can use the relation

$$\sigma^2 = \sigma_F^2 + \left[\frac{\rho}{1+\rho} \right] (\sigma_M^2 - \sigma_F^2). \quad (1)$$

It is safe in the samples measured to assume a homogeneous distribution of both types of centers. Then ρ can be expressed in terms of the measured optical absorption constants $\mu_M(\nu)$, $\mu_F(\nu)$, and the unknown oscillator strengths f_F , f_M . The absorption of the M center is anisotropic³ and, since unpolarized light was used, f_M must be considered to be the average over the polarization angle.

$$\rho = (f_F/f_M) \left[\int \mu_M d\nu / \int \mu_F d\nu \right].$$

It is possible by very prolonged x-ray exposures (up to 2 weeks at room temperature) to vary the second factor from 0.038 to 0.078. The resolved structure measurements provide a point at $\rho=0$. This is sufficient to permit a best choice of (f_F/f_M) which makes the plot of σ^2 vs $\rho/(1+\rho)$ a straight line whose slope is $(\sigma_M^2 - \sigma_F^2)$. This plot is shown in Fig. 1 and is drawn for $f_F/f_M=10$. Taking f_F as approximately 0.8,⁴ we have 0.08 for f_M . The value of σ_M^2 is around 10 500 gauss². At this stage of the investigation, the probable error in f_F/f_M can only be guessed at as around 25%. On the other hand, σ_M^2 would not change by more than 10% for either limit.

From the only previous theory for the M center,⁵ the magnitude of σ_M^2 is reasonable. However, if one carries through the formalism of Inui *et al.* for the coefficients of the LCAO (linear combination of atomic orbitals) in LiF, the results indicate a strongly resolved quartet of equally intense lines. This is in direct con-

tradition to the experimental results obtained which should certainly have revealed this at very high values of ρ . One expects a lower oscillator strength for the M center than for the F center because of the smaller overlap of the excited and ground states. However, it was not expected that the difference would be so large. The investigation is now being continued, using independent methods, to measure the hyperfine interactions and thereby check the determination of f_M .

I would like to thank Dr. C. K. Jen and Mr. B. S. Gourary for many informative discussions in the course of this work.

† This work supported by the Bureau of Ordnance, Department of the Navy.

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³ M. Ueta, J. Phys. Soc. Japan **7**, 107 (1952); D. Compton and W. Lamb (private communication).

⁴ R. H. Silsbee, Phys. Rev. **103**, 1675 (1956); C. J. Rausch and C. V. Heer, Phys. Rev. **105**, 914 (1957).

⁵ Inui, Uemura, and Toyazawa, Progr. Theoret. Phys. Japan **8**, 355 (1952).

Double Transitions in the Infrared Spectrum of Solid Hydrogen*

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(Received April 17, 1957)

IN two previous communications^{1,2} the infrared fundamental band of liquid and solid hydrogen at low resolution was described; it has now been recorded with a grating spectrometer and further interpretation of its structure is possible.

Figure 1(a) shows the band in solid normal hydrogen as previously recorded using a prism spectrometer with a spectral slit width of 15 cm^{-1} . The three main groups of maxima were correlated with the Q branch and the $S(0)$ and $S(1)$ lines of the collision-induced spectrum of the gas.¹ The broad maxima Q_R , $S_R(0)$, and $S_R(1)$ are undoubtedly summation tones of translational lattice frequencies with the molecular vibration and rotation, and correspond to a normal second-order crystal spectrum modified by the free molecular rotation peculiar to solid hydrogen.

The sharp components, on the other hand, are most probably due to induction by the molecular quadrupole moments. Figure 1(b) shows the new detail of the structure obtained using a grating spectrometer with a spectral slit width of 2 cm^{-1} . The Q branch has two components with frequencies near the $Q(0)$ and $Q(1)$ Raman frequencies of the solid,³ and the weak lines designated $S_1(0)$ and $S_1(1)$ occur at the calculated positions of the corresponding Raman lines. Each of these lines, therefore, originates in a transition in a

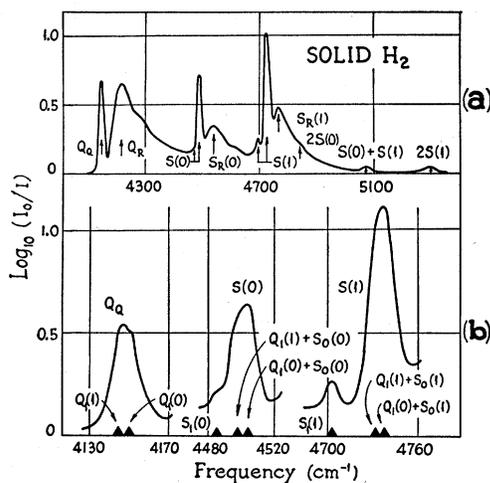


FIG. 1. (a) The fundamental infrared absorption of solid normal hydrogen observed by using a spectral slit width of 15 cm^{-1} . (b) The sharp components of the fundamental band observed with a spectral slit width of 2 cm^{-1} . The frequencies of the expected transitions are shown on the abscissa axis. Absorption at $Q_1(1)$, $Q_1(0)$, $S_1(0)$, and $S_1(1)$ results from a rotation-vibration transition in a single molecule, whereas absorption at $Q_1(1)+S_0(0)$, etc., results from simultaneous vibrational and rotational transitions in a pair of molecules.

single molecule. The strong S lines, however, have frequencies which correspond to a vibrational transition in one molecule accompanied by a rotational transition in the ground vibrational state of a second molecule. For each rotational transition there are two possibilities for the accompanying vibrational transition. For example, in the case of $J=0 \rightarrow J=2$ these are $Q_1(0)+S_0(0)$ and $Q_1(1)+S_0(0)$, where the subscripts 0 and 1 indicate $\Delta v=0$ and $\Delta v=1$, respectively; these transitions are separated in frequency from one another and also from the single transition, $S_1(0)$, because of the vibration-rotation interaction. The frequencies of the expected transitions are indicated on the abscissa axis of Fig. 1(b). The agreement of the measured and calculated frequencies shown in Table I confirms the validity of the assignments.

At any given molecule in the solid the electric field due to the regularly spaced but presumably randomly oriented neighboring molecules is rapidly fluctuating about zero value. Also the dipoles induced in neighboring molecules by the quadrupole of the given molecule cancel one another on the average. The induced absorption for a single molecule should therefore be very weak. The situation is similar to a highly compressed gas where, in ternary and higher order collisions, the different induced moments tend to annul one another.⁴ However, it has been pointed out by van Kranendonk⁵ that for simultaneous transitions involving a pair of molecules the cancellation effect is not operative; in compressed gases, liquids, and solids, where a given molecule is in a more or less symmetrical environment, double transitions should be more probable than single transitions. The correctness of this view, which would be

TABLE I. Measured and calculated frequencies.

Frequency of maximum (cm^{-1})	Assignment	Calculated frequency ^a (cm^{-1})
4148	$Q_1(1)$	4144
4152	$Q_1(0)$	4150
4486	$S_1(0)$	4486
4503	$\{Q_1(1)+S_0(0)$ $Q_1(0)+S_0(0)$	4498
4702	$S_1(1)$	4702
4736	$\{Q_1(1)+S_0(1)$ $Q_1(0)+S_0(1)$	4731
		4736

^a The frequencies of the $Q_1(1)$ and $Q_1(0)$ lines were observed in the Raman effect of the solid³; the other frequencies have been calculated by using the rotational constants for the gas [C. Cumming, Ph.D. thesis, University of Toronto (unpublished)].

difficult to show in the gas at room temperature where the induced band is rather diffuse, is well confirmed by the above analysis of the spectrum of the solid.

In liquid hydrogen the lines due to quadrupole induction are somewhat more diffuse than in the solid; however, the greater intensity of the double transitions as compared with the single transitions is as marked as for the solid.

Preliminary experiments on the absorption of liquid and solid hydrogen in the overtone region have shown that the intensity of the double vibrational transition is much greater than that of the pure overtone; in the compressed gas at a density of 426 Amagat units⁶ at 80°K , the difference between the two bands is much less marked.⁷

* This research was supported by a grant from the National Research Council of Canada.

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² Hare, Allin, and Welsh, *Phys. Rev.* **99**, 1887 (1955).

³ Allin, Feldman, and Welsh, *J. Chem. Phys.* **24**, 1116 (1956).

⁴ D. A. Chisholm and H. L. Welsh, *Can. J. Phys.* **32**, 291 (1954).

⁵ J. van Kranendonk, *Gordon Conference: "Infrared Spectra and Intermolecular Forces,"* 1956 (unpublished).

⁶ One Amagat unit of density is the density of the gas at a pressure of one international atmosphere and at 0°C .

⁷ Welsh, Crawford, MacDonald, and Chisholm, *Phys. Rev.* **83**, 1264 (1951).

Measurement of the Spin and Gyromagnetic Ratio of $\text{As}^{76}\dagger$

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(Received April 15, 1957)

THE spin and gyromagnetic ratio of 27-hour As^{76} have been measured by using the γ -ray anisotropy induced by several types of radio-frequency magnetic fields applied to a silicon crystal containing 5×10^{10} As^{76} atoms.

A silicon crystal doped with 3×10^{16} As^{75} atoms per cc was irradiated in the Brookhaven reactor and subsequently annealed in vacuum for 5 hours at 1100°C . The sample was placed in a rectangular TE_{10n} mode cavity resonant at $24\,000 \text{ Mc/sec}$. The H -plane sides