The solution is

$$\gamma = -2\{R(x,0)[f(x)+g(x)]\}^{-1} . \quad (7.2)$$

The right-hand side has a positive minimum of 3.1 at $x \simeq 1.2$, giving a critical value

$$r_s \simeq 9.4 . \quad (7.3)$$

At this point the electron gas presumably becomes ferromagnetic. If we had used instead of $f(x)$ and $g(x)$ the Hubbard interpolation function $h(x)$ in (7.1), we would have obtained a critical value $r_s \simeq 3.0$.

VIII. SUMMARY

The results obtained in this work are summarized as follows.

(i) A dielectric function for all momentum transfers is derived.

(ii) The separate contributions to the correlation energy from the singlet and triplet states are calculated.

(iii) The stability of the paramagnetic ground state is put in evidence.

(iv) The limit of stability is estimated.

Although our approximation procedures greatly simplify the problem and lead to reasonable results, their accuracy and range of validity are hard to assess. This lack of knowledge precludes us from studying the effects of the cubic and quartic Hamiltonians. It is perhaps more appropriate to investigate at this stage the other properties of the electron gas in the present approximation.

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Nuclear-Magnetic-Resonance Study of the Hydrogen Nucleus in OH^- in the Potassium Chloride Lattice*

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Experimental data are presented on the nuclear-magnetic-resonance signal from the hydrogen magnetic-dipole moment in the OH^- ion substituted for Cl^- in a KCl lattice. The recovery rate of the signal after saturation is measured for OH^- concentrations down to 1.2×10^{19} OH^-/cm^3. This rate is used to deduce a displacement distance $= 0.4\times10^{-8}$ cm for the hydrogen nucleus from the halogen vacancy site when the OH^- ion has $\langle 100\rangle$-directed localized states in a rigid lattice and a correlation time $= 4\times10^{-8}$ sec. The effects of librating states of the OH^- and lattice relaxation are estimated. A discussion of the dependence of the displacement distance on the correlation time is included.

I. INTRODUCTION

In recent years a great deal of experimental and theoretical work has been done on alkali halides with OH^- ions substituted as an impurity.[1] The OH^- is known to go into the KCl lattice by substituting for Cl^- ion, and the potential minima of the OH^- ion are believed to be such that alignment of the OH^-

along a ⟨100⟩ crystallographic direction is favored. The ⟨100⟩-directed states of the OH^- ions in zero electric field are still not well understood. Recent literature[2,3] has reported theoretical values for the equilibrium displacement of the OH^- ion's center of mass from the site center varying from $0.078-0.3\times10^{-8}$ cm. In an effort to gain more information about the OH^- ion in zero applied electric field, we have studied the postsaturation recovery of the nuclear-magnetic-resonance (NMR) signal of the hydrogen nuclear magnetic moment in the OH^- ion.

The dominant recovery mechanism for the net magnetization of the hydrogen nuclear magnetic moments is the fluctuation of the magnetic-dipole–magnetic-dipole interaction between the hydrogen nuclear moment and the nuclear magnetic moments of neighboring ions. Fluctuations result from changes in the relative positions of the hydrogen nucleus and its neighbors due to transitions of the OH^- ion from one directed state to another. We shall show in Sec. III that the recovery rate is proportional to $(\omega_H^2\tau^2+1)^{-1}\tau$, where ω_H is the Larmor frequency of the hydrogen nucleus in our dc magnetic field and τ is the electric-dipole-lattice relaxation time. For dc magnetic fields appropiate with our radio-frequency magnetic field which oscillates at 30 MHz, the electric-dipole relaxation times implied from the electrocaloric effect[4] imply a maximum of $(\omega_H^2\tau^2+1)^{-1}\tau$ vs τ near liquid-helium temperatures in a KCl lattice. The recovery of the net magnetization of the hydrogen nuclear magnetic moments due to this mechanism should therefore be most effective at temperatures of a few deg K.

II. EXPERIMENT

The signal observed in a rapid passage through the resonant field of a collection of nuclear magnetic moments is proportional to the net magnetization M of the resonant moments along the dc field at the beginning of the pass.[5,6] Since the resonant nuclei are a minority-spin system, the experimental conditions of the pass can be arranged so that the net magnetization of the resonant nuclei along the dc field is zero at the end of a pass.[7] Varying the time between the first and a subsequent pass then allows one to sample the recovery of M.

The spectrometer used in this experiment is of a fairly standard crossed-coil design. In order to minimize signals from hydrogen nuclei in the coil forms, a press-fit set of coils using only Teflon and copper was designed. Since the recovery time between passes is as long as 26 min, the radio-frequency field is turned off between passes through the resonant dc field. The voltage step at the output of the detector caused by turning on the radio-frequency field is damped at a point immediately after the detector with a metal-oxide-semiconductor field-effect-transistor switching arrangement which shorts the input to the audio amplifier for the first 0.1 sec of the time that the radio-frequency field is on. The signal is fed to a voltage-to-frequency converter and stored in quarters of a multichannel scaler.[8] Since signals after each of four recovery times are stored in sequence, long-term drifts in the receiver amplifiers do not effect the relative size of the four signals integrated in the course of several days of running time. Possible problems from long-term drifts in the current-controlled dc magnetic field are avoided by triggering the multichannel–scaler-channel advance with a separate spectrometer system using the F^{19} resonance in C_6F_6.

Figure 1 is a typical set of signals. In spite of extensive measures taken to remove unwanted hydrogen nuclei from the forms which support our transmitting and receiving coils, a small background signal was detected with no sample in the coil. Measurements made on this signal show that it has a relaxation time much shorter than the shortest recovery time used in our experiment. The rigorous corrections to the relative sizes of the measured signals, which are therefore possible, were made in all the data to be reported in this paper.

III. THEORY

A. Reorientation of OH^-: Correction Time

For lattice temperatures between 1.57 and 2.14 °K, we find experimentally that the recovery of M becomes independent of OH^- concentration at concentrations below 2×10^{19} OH^-/cm^3. We interpret this to mean that OH^--OH^- interaction effects on the energy eigenstates of most of the OH^- ions present at average concentrations below 2×10^{19} OH^-/cm^3 are significantly less than kT for these temperatures. This observation is in agreement with other experiments which bear on the OH^--OH^- interaction ef-

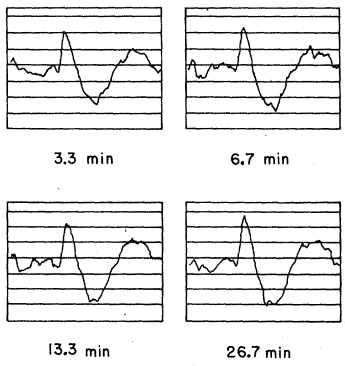

FIG. 1. Signals obtained at a temperature of 1.92 °K from a crystal with 1.2×10^{19} OH^-/cm^3 after 3.3-, 6.7-, 13.3-, and 26.7-min recovery times. The trace shown was taken from the display of a multichannel scaler which had summed 49 passes.

fects.[9] Rough estimates of the OH^--OH^- electric-dipole interaction[10] indicate that the electric-dipole interaction at concentrations $\approx 10^{19}$ OH^-/cm^3 must be much larger than the tunneling splitting due to the overlap of the ⟨100⟩-directed states in KCl.[11] When the OH^--OH^- electric-dipole or the stress-elastic-dipole interactions cause perturbations greater than the tunneling splitting but less than kT, a rigorous theoretical treatment becomes prohibitively difficult. Early experimental data[12] on the dielectric relaxation of the OH^- in KCl has indicated that interaction effects might be approximated by an ensemble of OH^- systems with single electric-dipole-lattice relaxation times. For concentrations $\approx 10^{19}$ OH^-/cm^3, these relaxation times appear to be in the range from 8×10^{-9} to 8×10^{-8} sec. Early electrocaloric measurements[13] yield similar results if one is willing to extrapolate the results obtained to very short-switching times. More recent work[14] done on the induced optical dichroism of the uv absorption of OH^- dipoles in several alkali halides with longer relaxation times than OH^- in KCl has indicated, at least for the systems studied, that internal perturbations in the alkali halide lattice may be expected to produce localized ⟨100⟩ dipole states even in the limiting case of no applied electric field. If these directed states, rather than the tunneling states which are often used to describe OH^- ions in KCl at very low concentrations, are appropriate, one would expect a relaxation behavior in zero applied electric field to be similar to that found with an applied electric field sufficiently small, that the splitting of the directed states of the OH^- by electric fields is less than kT.

This case has been discussed in theoretical work work[15] based on the assumption that only 90° one-phonon reorientations are important in the reorientation of the OH^- in KCl for temperatures of 4.2 °K and lower. The work finds that for the case in which the splitting of the directed states of the OH^- by electric fields is less than kT, each of the four possible 90° transitions from a given ⟨100⟩ state has a transition probability per unit time W which is approximately independent of both the direction and magnitude of the electric field at an OH^- site; the electric-dipole-lattice relaxation with an applied electric field can be described by a single exponent with time constant $\tau = 1/4W$. τ is also equal to the correlation time for the single-exponential reduced-correlation function[16] implied by the above approximations. Since our data shows no departure from the single-exponential recovery of the net magnetization M that one expects with a single-exponential reduced-correlation function, the use of a single correlation time τ is at least sufficiently accurate for our experiment. We shall show in Sec. IV B that our results are not critically dependent on the exact value of τ.

B. Spin-Lattice Relaxation of Hydrogen Spin System

In a first calculation we make two questionable but greatly simplifying assumptions: The position of the hydrogen nucleus when the OH^- is in a ⟨100⟩ state is assumed to be localized at a distance d from the lattice site of the OH^- ion. Furthermore, in what we shall call the rigid-lattice model we assume fixed positions for the potassium and chlorine nuclei which are nearest neighbors and next-nearest neighbors to the OH^- ion's lattice site. We note that we are thus neglecting direct modulation of the magnetic-dipole–magnetic-dipole interaction by the lattice vibrations, but this mode is known to be exceedingly inefficient in causing magnetic relaxation at temperatures ≈ 4 °K, so its omission is not a serious error. Direct modulation of the magnetic-dipole–magnetic-dipole interaction by the stretching vibration of the OH^- is neglected for the same reason.

The resonant field of potassium or chlorine nuclei is never attained during our experiment. Furthermore, either spin-flip terms couple nuclei near the OH^- ion to more distant nuclei to form subsystems at thermal equilibrium with the lattice, or electric field gradients from the OH^- ion's electric-dipole moment will couple the electric-quadrupole terms of the neighboring spins's Hamiltonians directly to the lattice. We therefore assume that these spins remain in thermal equilibrium with the lattice throughout the experiment.

When the time dependence of M due to the fluctuating magnetic-dipole–magnetic-dipole interaction is calculated under the usual assumptions,[17] with the additional effects of restrained motion incorporated (the average of the magnetic-dipole perturbation terms is no longer zero[18]), and the high-temperature ($kT_{\text{lattice}} \gg \hbar\omega_H$) approximation[19] is made, we find

$$\frac{dM}{dt} \simeq -N\hbar\gamma_H(\langle I_z\rangle - \langle I_z\rangle_{eq}) \sum_\kappa \frac{\tau}{1+\tau^2(\omega_H-\omega_\kappa)^2} g_{1\kappa} + \frac{\tau}{1+\tau^2\omega_H^2} g_{2\kappa} + \frac{\tau}{1+\tau^2(\omega_H+\omega_\kappa)^2} g_{3\kappa} \quad , \qquad (1)$$

where $\hbar I \equiv$ spin angular-momentum operator of the hydrogen nuclei; $\langle I_z\rangle \equiv \mathrm{T}\,[(\text{spin-density matrix})\times I_z]$; $\langle I_z\rangle_{eq} \equiv \mathrm{T}\,[(\text{thermal-equilibrium spin-density matrix})\times I_z]$; N is the number of OH^- ions; γ_H the hydrogen gyromagnetic ratio; ω_H the Larmor frequency of the hydrogen nucleus; ω_κ the Larmor frequency of the κth neighbor nucleus; and $g_{1\kappa}(d)$, $g_{2\kappa}(d)$, $g_{3\kappa}(d)$ are complex geometric factors proportional to the average fluctuation of the squared magnitude of the magnetic-dipole–magnetic-dipole interactions between the hydrogen nuclear magnetic moment and the κth neighboring nuclear magnetic moment.

With $M(t=0)=0$, the above equation integrates to

$$M(t) = (1 - e^{-t/T_1})(N\hbar\gamma_H \langle I_z \rangle_{eq}) \equiv (1 - e^{-t/T_1}) M_{eq} \quad . \quad (2)$$

The spin-lattice relaxation time T_1 is given by

$$(T_1)^{-1} = \sum_\kappa \{\tau[1 + \tau^2(\omega_H - \omega_\kappa)^2]^{-1} g_{1\kappa} + \tau(1 + \tau^2\omega_H^2)^{-1} g_{2\kappa} + \tau[1 + \tau^2(\omega_H + \omega_\kappa)^2]^{-1} g_{3\kappa}\} \quad . \quad (3)$$

For a KCl lattice all the ω_κ are small compared to ω_H, so we write

$$(T_1)^{-1} \approx \tau(1 + \tau^2\omega_H^2)^{-1} \sum_\kappa (g_{1\kappa} + g_{2\kappa} + g_{3\kappa}) \quad . \quad (4)$$

The geometric factor $g \equiv \sum_\kappa (g_{1\kappa} + g_{2\kappa} + g_{3\kappa})$, with κ summed over first- and second-nearest neighbors has been evaluated with a computer program for several values of the parameter g and directions of the dc field along the ⟨100⟩, ⟨110⟩, and ⟨111⟩ crystallographic axes. The 12 second-nearest-neighbor terms were included in the sum because the relatively large Cl gyromagnetic ratio, which is contained in the geometric factors $g_{i\kappa}$ to the second power, partially offsets the effect of the relatively large distance of the second-nearest neighbors which tends to make these terms small. For values of d from 0 to 0.8×10^{-8} cm, the second-nearest-neighbor terms contributed about one-third of the net value of g. The calculated variation in g with changes in the direction of the dc magnetic field was about one part in 10^4 for the values of d implied by our measured T_1, and less than 2% for all the values of d that were tried. What calculated variation there is between values of T_1 for different directions of the dc field is far below changes that would be detectable with our spectrometer. Figure 2 is a plot of g as a function of d fitted to the points we calculated for the rigid-lattice model with sharply localized states.

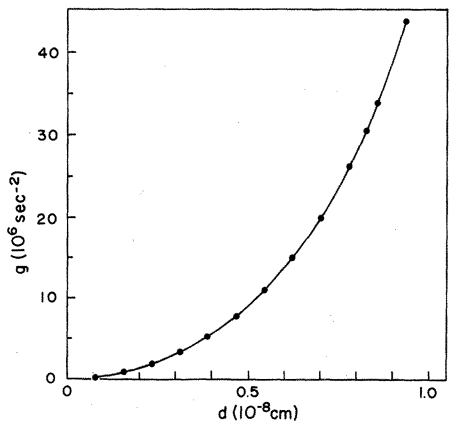

FIG. 2. Geometric factor g as a function of the distance d of the hydrogen nucleus from the OH^- lattice site for the rigid-lattice model with sharply localized states.

TABLE I. Experimental results for the hydrogen spin-lattice relaxation time T_1 as a function of temperature and direction of the dc magnetic field. The correlation time τ is assumed to be equal to 4×10^{-8} sec. $\omega_H = 184.25 \times 10^6$ rad/sec, the value corresponding to the dc magnetic field during recovery.

OH^-/cm^3 (10^{-19})	Temp (°K)	Magnetic dc field direction	T_1 (min)	$(T_1\tau)^{-1}\{\tau^2\omega_H^2+1\}$ (10^6 sec^{-2})
3.3	4.0	⟨100⟩	1.6	14.4
3.3	2.14	⟨100⟩	5.9	3.9
3.3	1.92	⟨100⟩	8.4	2.7
3.3	1.57	⟨100⟩	10.6	2.2
1.9	1.92	⟨100⟩	3.6	6.4
1.9	1.92	⟨110⟩	3.8	6.1
1.9	1.57	⟨100⟩	3.8	6.1
1.2	1.92	⟨100⟩	3.3	7.0
1.2	1.92	⟨110⟩	3.7	6.2
1.2	1.92	⟨111⟩	3.2	7.2

IV. RESULTS AND CONCLUSIONS

Usable data were obtained from three crystals[20] with average OH^- impurity concentrations of 3.3, 1.9, and $1.2\times10^{19} OH^-/cm^3$. The concentrations were determined by measuring the absorption constant at room temperature of the main stretching band of the OH^- ion.[21] Table I lists our results for T_1 together with the resultant experimental value for $g = (T_1\tau)^{-1}(\tau^2\omega_H^2+1)$. $\omega_H = 184.25\times10^6$ rad/sec, the value corresponding to the dc magnetic field during recovery. We took $\tau = 4\times10^{-8}$ sec as a reasonable value for the correlation time.[22,23]

A. Sharply Localized States in a Rigid Lattice

As long as the effects of OH^--OH^- interactions are sufficiently small to allow the approximation in Sec. III A, which leads to a single-exponential reduced-correlation function, the product $(T_1\tau)^{-1} \times (\tau^2\omega_H^2+1)$, which is equal to $g(d)$ for a single-exponential reduced-correlation function, should remain constant for different samples and different temperatures of the same sample. As is shown in Table I, the interaction effects are clearly visible in the heavily doped crystal. Since g is a strongly varying function of d in the range where $g \approx 5\times10^6$ sec^{-2}, each of the numbers $(T_1\tau)^{-1}(\tau^2\omega_H^2+1)$ obtained for the less heavily doped crystals give essentially the same value of d: From Table I and Fig. 2 we read $d = 0.4\times10^{-8}$ cm for the displacement of the hydrogen nucleus in the sharply localized ⟨100⟩-directed states of the OH^- ion in a rigid KCl lattice. With a distance of 0.9×10^{-8} cm between the center of mass and the hydrogen nucleus in the OH^- ion,[24] this number places the center of mass of the OH^- ion 0.5×10^{-8} cm from the halogen vacancy site and

in a direction opposite to the direction of the hydrogen nucleus.

B. Uncertaintly of the Value of τ

The principal uncertainty in the value of d obtained from our measurement stems from doubt about the appropriate value of τ. Since $\tau^{-1}(\tau^2\omega_H^2+1)$ is at most a linear function of τ, while $g(d)$ is a much more rapidly varying function of d, the value of d obtained from the relation

$$(T_1\tau)^{-1}(\tau^2\omega_H^2+1)=g(d)$$

will not be critically dependent on the exact value of τ. We illustrate this dependence in Fig. 3, which is a plot of the values of d obtained from Fig. 2 as a function of various assumed values of τ. We note that the entire range of "reasonable" values for τ, that is $\tau=8\times10^{-9}$ to 8×10^{-8} sec, give surprisingly small values of d.

C. Nonlocalized States and Nonrigid Lattice

In this section we investigate contributions to g either due to motion of ions surrounding the OH^- site or due to a nonlocalized $\langle100\rangle$ state of the OH^- ion. Since our experiment measures only one parameter, T_1, we are unable to isolate these contributions. The NMR data do, however, give a useful independent check of various models put forward for the OH^- ion's motion, and we can place upper bounds on what effect lattice relaxation or nonlocalized $\langle100\rangle$ states of the OH^- could have on the calculation of g.

The effect of allowing the $\langle100\rangle$ states of the OH^- ion to be nonlocalized was investigated by using the model of Klein[25] for representative terms in g, as is outlined below. Since the pure librational

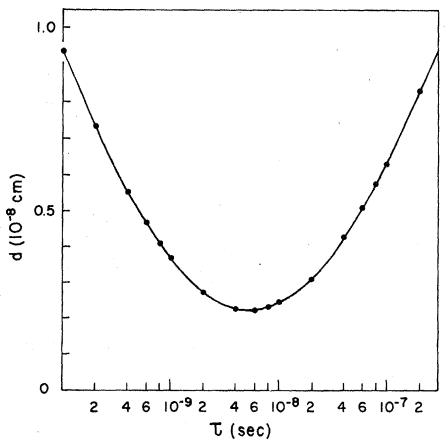

FIG. 3. Distance d of the hydrogen nucleus from the OH^- lattice site obtained from the measured spin-lattice time of the hydrogen spin system as a function of the correlation time assumed in the calculation.

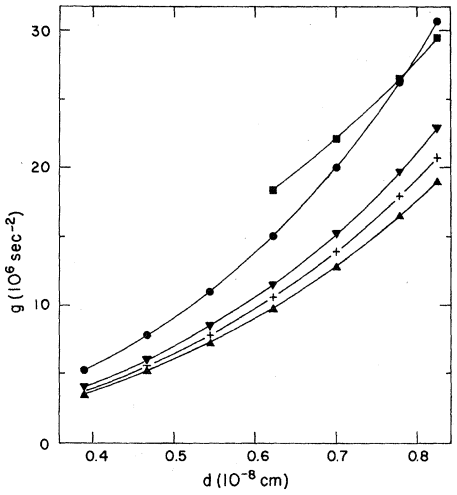

FIG. 4. Geometric factor g as a function of the distance d of the hydrogen nucleus from the OH^- lattice site for a relaxed lattice. Closed circle: no displacement of the neighboring ions; closed triangle, plus, and closed inverted triangle: displacement of the potassium ion nearest the hydrogen end of the OH^- ion away from the ion by 0.28, 0.22, and 0.16×10^{-8} cm, respectively; closed square: displacement of the potassium ion nearest the hydrogen end of the OH^- ion away from the ion by 0.22×10^{-8} cm, displacement of the four potassium ions perpendicular to the OH^- ion's symmetry axis toward the ion by 0.125×10^{-8} cm, and displacement of the potassium ion nearest the oxygen end of the OH^- ion toward the ion by 0.50×10^{-8} cm. A curve corresponding to the closed square, but without the displacement of the potassium ion nearest the oxygen end of the OH^- ion, was also calculated and was found to lie just under the curve of pluses.

mode oscillates at $\omega\gg\omega_H$, the librational motion does not constitute a separate relaxation mode. What this motion does do is require that we replace the sharply localized position coordinates in g by values which are averaged over the librational motion of the OH^- ion in each of the $\langle100\rangle$-directed states. Using the simple harmonic-oscillator states of Klein and his value of $\langle\theta^2\rangle^{1/2}\approx0.24$ rad, we estimate from the changes in representative terms of g, that the net effect of the librational motion is a 30% reduction of the theoretical value of g at a given d. Use of our experimental T_1 with this corrected g implies a value of d which is about 10% larger than the value obtained from the uncorrected g.

We have calculated the effect of radial relaxation of the nearest-neighbor potassium ions on g in a manner similar to the approach used for the librating OH^- ion. Since the time necessary for the lattice to relax is ~ (phonon frequencies)$^{-1}$, which is much less than the characteristic time τ for a transition from one relaxed $\langle100\rangle$ state, the effects of lattice relaxation can be accounted for by simply replacing the rigid-lattice coordinates of the neighboring potassium ions by their relaxed values. The

displacements estimated by Hartel[26] from experimental data imply a change in g of less than 1%. The theoretically computed neighboring-ion displacements suggested by Quigley and Das[27] for the OH^- ion center-of-mass displacement that they compute imply a change in g of about 4%. The resultant g at the center-of-mass displacement that they suggest would correspond to a $T_1 = 0.85$ min, a value much smaller than the 3.6 min value we find experimentally. Figure 4 is a plot of $g(d)$ for several different assumed sets of neighboring-ion displacements. With the exception of the one point corresponding to the neighboring-ion displacements suggested by Quigley and Das at the OH^--ion center-of-mass displacement that they compute (closed square at $d = 0.83 \times 10^{-8}$ cm), these sets of neighboring-ion displacements were selected to illustrate those sets of displacements which produce a large change in g. As one would expect from the definition of g, they are large highly unsymmetric displacements. Inspection of Fig. 4 shows that all the neighboring-ion displacements proposed to date cause change in g which are much smaller than the 30% reduction in g for a given d that we estimate to be caused by librational motion of the OH^- ion.

We conclude that the dependence of g on d is so strong compared to the effect of any librational motion or lattice relaxation proposed so far that one will find the value of d as determined by the correct τ and our measured T_1 using sharply localized ⟨100⟩-directed states of the OH^- ion in a rigid KCl lattice to be at least 90% of the actual distance between the OH^- lattice site and the hydrogen nucleus in the OH^- ion.

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