Evidence for a temperature-dependent tunneling parameter

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Measurements are reported of the integrated intensity of a microwave absorption line in RbCl:Ag^+ (which corresponds to an on-center to off-center transition) as a function of temperature and hydrostatic pressure. The integrated intensity is found to decrease very rapidly with temperature, much more rapidly than population effects would predict. We interpret these results in terms of a polaronlike dressing of the tunneling matrix element which has a temperature dependence of the form $\Delta^2=\Delta_0^2 e^{-(T/T_0)^2}$. For this absorption line, T_0 is about 10 K near 1 kbar and decreases with increasing hydrostatic pressure.

I. INTRODUCTION

RbCl:Ag^+ is a model off-center system and has been extensively studied since it was first discovered.[1-3] Although the Ag^+ ion substitutionally replaces the Rb^+ ion, it occupies off-center positions which are located along the twelve $\langle 110 \rangle$ directions in the crystal, as a result of its smaller ionic radius. Reorientation from one off-center position to another takes place via a tunneling process at low temperatures. As a result of the off-center displacement, the defect carries an electric dipole moment p ($p=0.78$ e Å) (Refs. 2 and 3) and the twelve off-center states are split when an electric field E is applied. Microwave transitions between these states are called paraelectric resonance (PER). At $E=0$, tunneling lifts the twelvefold degeneracy. Measurements clearly show that the dominant tunneling parameter is the second-neighbor tunneling parameter[2] μ which corresponds to a rotation of the off-center dipole by 90°. When hydrostatic pressure P is applied, μ increases and reaches 31 GHz at $P=1.85$ kbar. Over this range p decreases by roughly 60%.

A more striking effect when pressure is applied[4] is the sudden decrease[5] in the paraelectric resonance intensity (for electric-dipole transitions between the states split by E) above $P\approx 1.3$ kbar (for $T=4.2$ K) and the subsequent discovery of a new resonance that is very strongly pressure dependent[6] (only observe over the range $1.0<P<1.3$ kbar). We have interpreted these results in terms of a two-configuration model for Ag^+ in RbCl: over a limited range of lattice parameter, corresponding to a small range in hydrostatic pressure, both on-center and off-center configurations for the Ag^+ ion can exist. Some of the Ag^+ ions are off-center as observed at low pressures and some are on-center. As the pressure is increased, the lowest state of the on-center configuration decreases rapidly in energy and lies below the off-center tunneling states for $P>940$ bar. Above 1.3 kbar, the system is essentially[4,5] in the on-center state at 4.2 K although a few centers are still thermally excited to the off-center states even at 2.0 kbar. The new resonance line has been identified as a transition between an on-center ground state and the lowest energy of the (excited) tunneling states. A similar two-configuration situation was found in Kl:Ag^+ by Sievers and co-workers at zero hydrostatic pressure.[7,8]

We began a series of experiments to look for possible excited-state transitions between the tunneling states and higher on-center states. The initial experiments were carried out at slightly higher temperatures (6–8 K) to assure that the low-lying excited states were thermally populated. The unexpected result was the discovery that the on-center to off-center transition intensity is extremely temperature dependent. Over the range 4–15 K the signal intensity of this resonance line decreases by nearly 2 orders of magnitude, an effect that is inconsistent with population changes. This paper is a report on these new results.

In Sec. II we describe briefly the apparatus and the samples. We present examples of the data in Sec. III and provide the relevant background and some possible explanations for a strong temperature dependence in Sec. IV. The analysis of the data is given in Sec. V and a discussion of the results and the conclusions is presented in Sec. VI.

II. APPARATUS AND SAMPLES

A. Spectrometer and cryostat

The microwave spectrometer for high-pressure paraelectric resonance studies has been described previously.[9] With this system, the low-temperature probe can be moved vertically a distance of 30 cm for changing the cell pressure as follows: (1) the probe is raised so that the high-pressure cell is no longer covered by liquid helium; (2) a heater on the pressure cell is turned on to raise the temperature of the solid helium pressure medium above the melting point; (3) the helium gas pressure in the cell is changed to the desired value; and (4) the cell is recooled at constant pressure just enough to solidify the helium. The temperature is then cycled above and below the melting point several times [repeating step (4)] to verify that the cell is completely full. Pressure corrections are made

for lower temperatures assuming constant volume cooling.

For measurements at 4.2 K or lower, the cell is lowered into the liquid helium, and the helium vapor pressure is used to determine the temperature. For higher temperatures, the cell sits above the liquid helium and the waveguide structure below the cell serves as a cold link. The heater on the cell is then controlled to obtain the desired temperature which is measured using a germanium resistance thermometer.

B. Samples and procedures

The crystals of RbCl:Ag^+ (Czochralski grown) were produced at the crystal-growth facility at the University of Utah. The concentration of Ag^+ ions in the samples varied from 60 to 120 ppm. All measurements were taken on ⟨100⟩-oriented thin plates with thickness ranging from 0.25 to 0.5 mm. A swept high voltage, up to 5 kV maximum, was applied across the sample to provide electric fields from 0 to 200 kV/cm.

Measurements were made of the signal intensity as a function of temperature at several pressures in the range 1.0–1.3 kbar and at microwave frequencies between 50 and 150 GHz. After setting the pressure and temperature, the spectrometer was set up at a particular frequency and the electric field swept slowly from $E=0$ to some maximum value. Standard lock-in detection was used; consequently the observed signal is the derivative of the microwave absorption. Typical peak-to-peak E-field modulation amplitudes were 1–3 kV/cm. In general, relative intensity measurements are difficult to make accurately; the most reliable results were obtained by operating at a fixed microwave frequency and varying the temperature.

III. DATA PRESENTATION

The data are collected as swept electric-field derivative spectra for a series of fixed frequencies ν, pressures P, and temperatures. A typical spectrum is illustrated in Fig. 1

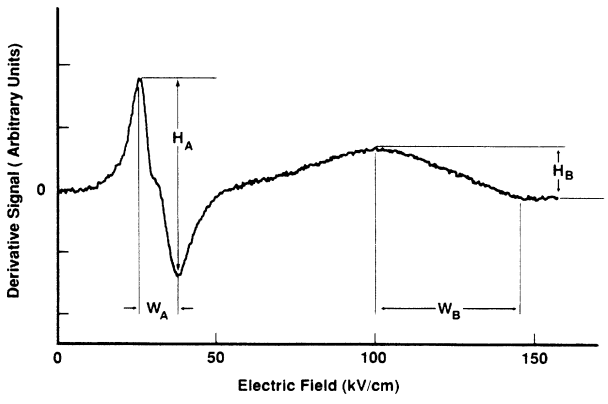

FIG. 1. A plot of the derivative of the microwave absorption as a function of electric field. The low-field line is labeled A and the high-field line is labeled B. The peak-to-peak heights and widths are shown for a trace at $\nu=67.82$ GHz, $P=1.12$ kbar, and $T=4.2$ K.

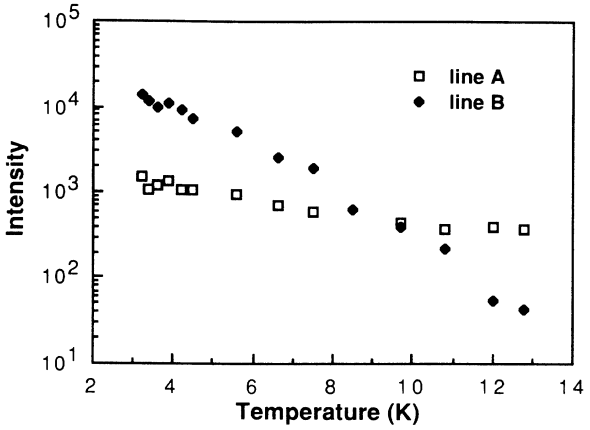

FIG. 2. A comparison of the temperature dependence of the intensities I_A and I_B at $\nu=68.05$ GHz and $P=1.105$ kbar. The intensities are in arbitrary units.

for $\nu=67.82$ GHz $P=1.12$ kbar, and $T=4.2$ K. Similar traces were taken for many different temperatures and microwave frequencies. The lower field line (A) is the normal paraelectric resonance transition between the tunneling states while the broad, higher-field resonance (B) is the on-center to off-center transition. We denote the peak-to-peak height as H and the peak-to-peak width as W as indicated. The integrated intensity I is then proportional to HW^2. In the following graphs we plot HW^2 as a function of T for fixed ν and P.

Figure 2 compares the temperature dependence of the intensities I_A and I_B at $\nu=68.05$ GHz and $P=1.105$ kbar. Above 4 K, I_A decreases with temperature some-

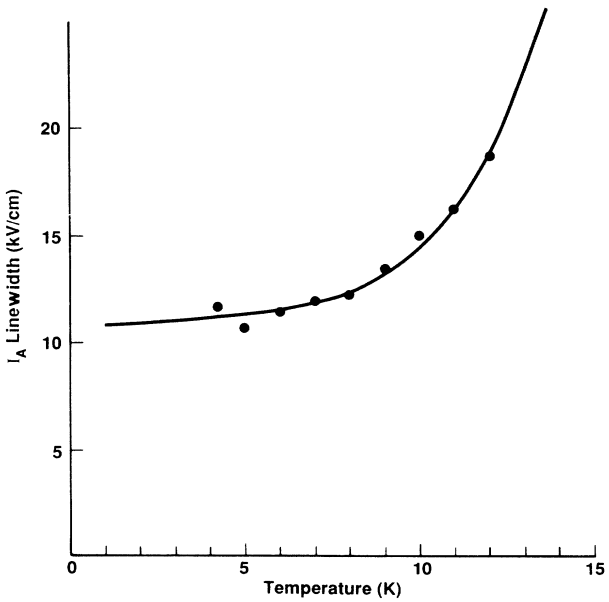

FIG. 3. A plot of the linewidth as a function of T for line A at $\nu=57.06$ GHz and $P=1.12$ kbar. The smooth average curve was used to determine W in calculations of intensity.

what more rapidly than expected from simple population effects. I_B, on the other hand, decreases catastrophically, in contrast to simple population considerations which only predict a decrease of order 4.

Two problems arise in intensity measurements as a function of temperature: (1) small changes in the resonator Q or the coupling with T can change the effective gain and (2) the widths of the lines increase rapidly at high temperatures and errors in the width increase. Since the widths enter the intensity expression quadratically, uncertainties in this parameter are a major source of scatter in the intensity measurements. To minimize this scatter, we first plot the linewidth as a function of temperature and draw a smooth average curve through the data (see Fig. 3). One way to do so is to fit the data to the expression

$$W = W_0 + \alpha T + \beta T^n \ .$$

W_0 is the static broadening at low T, αT is expected for one-phonon relaxation processes, and a βT^n dependence is expected for multiphonon processes. The value of W from the average curve is then used in calculating I. This greatly decreases the scatter in plots of I versus T. This approach was followed in all the I versus T data presented.

The amplitude problem (effective gain problem) can be reduced by comparing to a standard signal. Since the behavior of the tunneling levels is quite well understood (we address this assumption later) we use the intensity of line A as our reference for studying line B. We then plot the ratio of intensities I_B/I_A as a function of T as shown in Fig. 4.

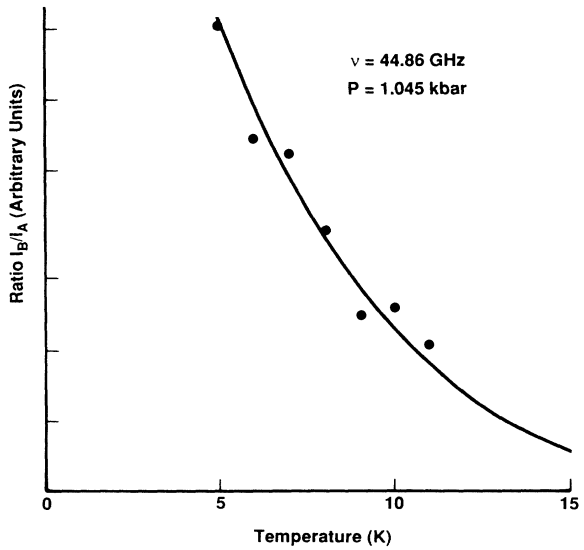

FIG. 4. The ratio r of the intensities I_A/I_B as a function of temperature for $\nu = 44.86$ GHz and $P = 1.045$ kbar. The line is a calculated fit as outlined in Sec. IV.

IV. BACKGROUND AND MODELS

As pointed out in the introduction, the RbCl:Ag^+ is a well-studied $\langle 110 \rangle$ off-center system. In the absence of any applied field the degeneracy of the twelve equivalent states is lifted by tunneling. Although in principle four different types of tunneling processes can occur[10] [nearest neighbor η (60°), next nearest neighbor μ (90°), third nearest neighbor ν (120°), and fourth nearest neighbor σ (180°)], μ dominates for all values of the hydrostatic pressure and increases with P approximately linearly above 1 kbar according to[11]

$$|\mu| = 0.0295(P - 800) \qquad (1)$$

for P in bars and μ in GHz.

In a $\langle 100 \rangle$ applied field E_{100}, this tunneling multiplet splits into three multiplets, each with four states; the middle multiplet is independent of E_{100}. In principle these multiplets are also split by tunneling, however, because the tunneling parameters are small relative to kT, we ignore the tunneling splitting in the electric-field multiplets when calculating populations.

In addition, an on-center state exists that decreases rapidly in energy as P increases and is below the tunneling states for $P > 900$ bar. We treat this state as the singlet ground state of a 3D harmonic oscillator, similar to the on-center system observed by Sievers and coworkers[7,8] in KI:Ag^+ at $P = 0$ and $T \approx 4$ K. ϵ_1 denotes the splitting between the centroid of the tunneling states and the on-center state and varies with pressure;[6]

$$\epsilon_1 = 0.86(P - 940) \ , \qquad (2)$$

with P in bars and ϵ_1 in GHz. The resultant simplified energy-level diagram for our model, for a pressure of roughly 1.24 kbar, is given in Fig. 5 where most tunneling splittings are ignored, and the field-independent middle multiplet is shown as a dotted line.

For simplicity, we describe the states of the on-center configuration as a set of harmonic oscillator (HO) levels for a 3D well. If n_1 is the population in the ground state, then the population in all the HO excited states is $n_1(1-X)^{-3}$, $X = e^{-\hbar\omega_0/kT}$, where $\hbar\omega_0$ is the harmonic-oscillator splitting. For reasonable values of $\hbar\omega_0$, only the first few HO states are populated for $T \leq 15$ K.

Defining the energy of the microwave photon as $\hbar\omega$ we can write the total population in the ground state at the resonant electric field for line A as

$$n_1 = \frac{N_0}{[(1-X)^{-3} + 4e^{-\epsilon_1/kT}(e^{\hbar\omega/2kT} + 1 + e^{-\hbar\omega/2kT})]} \ . \qquad (3)$$

The integrated intensity of a resonance transition between energy levels E_a and E_b is

$$I_{ab} \propto (n_a - n_b)|M_{ab}|^2 \ , \qquad (4)$$

where n_a and n_b are the populations and M_{ab} is the matrix element. For example, for the six-level system[12] used to describe KCl:OH^-,

$$I_{ij} \propto [f_j(E,T) - f_i(E,T)] \left[\Delta \frac{p(\cos\theta_j - \cos\theta_i)}{h\nu} \right]^2 ,$$

where $f_j(E,T)$ is the probability of the jth state being occupied for a given E and T, Δ is the tunneling parameter, p is the dipole moment, and θ_j is the angle between the applied electric field and the off-center dipole orientation j.

A similar expression is obtained in perturbation theory for the twelvefold tunneling system, when one tunneling parameter dominates. We assume in the following analysis that for E_{100},

$$I_A = I_{24} \propto p_A^2 \mu^2 f_1(E)(n_2 - n_4) \quad \text{(line } A\text{)} , \tag{5}$$

$$I_B = I_{12} \propto p_B^2 \Delta^2 f_2(E)(n_1 - n_2) \quad \text{(line } B\text{)} , \tag{6}$$

where Δ is the on-center to off-center tunneling parameter, $f_1(E)$ and $f_2(E)$ are functions of electric field that are nearly independent of Δ or μ, and p_A and p_B are the effective dipole coupling parameters, i.e., $p_a = (p_2 - p_4)\cos\theta$ where θ is the angle between the directed dipole and the E_{100} electric field. (Note that $p_B = p_A/2$). With this approximation, the intensities at the resonant electric field are

$$I_A = \frac{N e^{-\epsilon_1/kT}(e^{\hbar\omega/2kT} - e^{-\hbar\omega/2kT})\mu^2 p_A^2}{(1-X)^{-3} + 4e^{-\epsilon_1/kT}(e^{\hbar\omega/2kT} + 1 + e^{-\hbar\omega/2kT})} , \tag{7}$$

$$I_B = \frac{N'(1 - e^{-\hbar\omega/kT})\Delta^2 p_B^2}{(1-X)^{-3} + 4e^{-\epsilon_1/kT}(e^{(\epsilon_1 - \hbar\omega)/kT} + 1 + e^{-(\epsilon_1 - \hbar\omega)/kT})} , \tag{8}$$

and

$$R = \frac{I_B}{I_A} = C e^{(2\epsilon_1 - \hbar\omega)/2kT} e^{[-T^2(1/T_{0B}^2 - 1/T_{0A}^2)]} Z(\omega, T) , \tag{9a}$$

$$Z(\omega,T) = \left[\frac{(1-X)^{-3} + 4e^{-\epsilon_1/kT}(e^{\hbar\omega/2kT} + 1 + e^{-\hbar\omega/2kT})}{(1-X)^{-3} + 4e^{-\epsilon_1/kT}(e^{(\epsilon_1 - \hbar\omega)/kT} + 1 + e^{-(\epsilon_1 - \hbar\omega)/kT})} \right] \tag{9b}$$

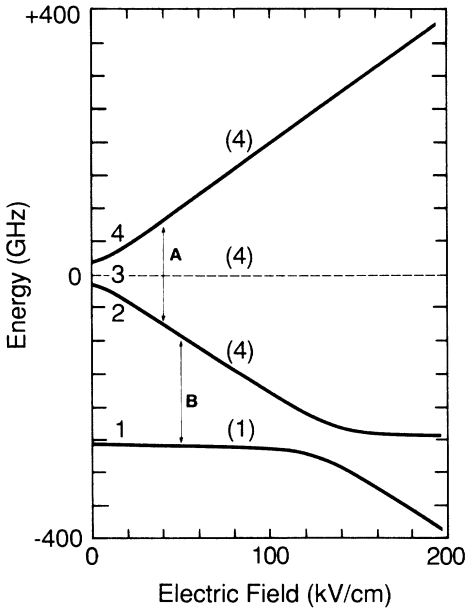

FIG. 5. The simplified energy-level diagram for a ⟨100⟩ electric field and a hydrostatic pressure of 1.24 kbar. Note that levels 2, 3, and 4 are each fourfold degenerate. Level 3 is not involved in the allowed transitions (for $\mu \neq 0$; η, ν, $\sigma = 0$) and is shown as a dotted line.

where N and N' are constants (for a particular value of E) which include $f_1(E)$ and $f_2(E)$, $X = \exp(-\hbar\omega_0/kT)$ as before, C includes N and N', Δ_1^2 and μ_1^2 are defined below [Eq. (10)], and we note that the splitting of the tunneling levels 2 and 3 at the electric field corresponding to resonance B is $\Delta\epsilon = \epsilon_1 - \hbar\omega$ (see Fig. 5).

The question now is how can the intensity of line B have such a strong temperature dependence? One possibility is to have a large degeneracy of excited states close to the tunneling states in energy. Then when the temperature is raised, the ground state is depleted and the intensity of line B decreases. In general, the presence of such states will also decrease the intensity of line A, but as a result of the high degeneracy of the tunneling multiplet (12) it can be a smaller effect for some choices of the excited states. One possible explanation for these states is that the excited on-center states lie rather close to the tunneling states (i.e., $\hbar\omega_0$ is comparable to ϵ_1). This possibility, and an arbitrary, highly degenerate excited state, were considered in the analysis.

A more interesting possibility is that the effective tunneling parameter is temperature dependent. Several theoretical treatments of the polaron dressing of these matrix elements[13–15] yield a Debye-Waller-like factor

$$\Delta^2 = \Delta_0^2 e^{-W_0} e^{-(T/T_0)^2} = \Delta_1^2 e^{-(T/T_0)^2} ,$$

where Δ_0 is the bare tunneling parameters, W_0 is a temperature-independent constant, Δ_1 is the value of Δ at low temperatures, and T_0 is a parameter than depends on

the potential-well parameters and the coupling of the defect to the lattice vibrations. That Δ should be temperature dependent can be seen from two perspectives. First, as the temperature increases, the probability of thermal activation over the barriers increases and the ion spends less time in a potential well. Consequently, the probability of tunneling is reduced. Alternatively,[16] as the temperature is increased, the potential wells begin to fluctuate and for a transition from E_1 to E_2 via a photon of energy $\hbar\omega$, the energies $(E_1+\hbar\omega)$ and E_2 are not always equal. When they are not equal, the probability of a transition is greatly decreased.

For the RbCl:Ag^+ system we have two different tunneling parameters that are important and hence in principle two different parameters, T_{0A} and T_{0B}. We therefore use the following forms for the two most important tunneling parameters of this system:

$$\Delta^2=\Delta_1^2 e^{-(T/T_{0B})^2}, \qquad \mu^2=\mu_1^2 e^{-(T/T_{0A})^2}. \tag{10}$$

To have a very strong T dependence for line B, but not line A, requires $T_{0B}<T_{0A}$. As will be seen in the next section, a range of values for T_{0A} can explain the data because the ratio R [Eq. (9)] becomes insensitive to T_{0A} for $T_{0A}>40$ K and $T\leq 14$ K.

V. DATA ANALYSIS

A. Intensity of line A

In Fig. 6 we show the intensity of line A as a function of T for $P=1.32$ kbar and $\nu=68.82$ GHz. For this pressure, I_A first increases rapidly with T, reaches a maximum, and then decreases slowly at high T. The low-T regime is dominated by the fact that the on-center state for this pressure is lower in energy than the tunneling states by an energy ϵ_1. The temperature at which the maximum occurs is determined by the value of ϵ_1 [Eqs. (2) and (5)]. Above the maxima the intensity is expected to decrease from the usual population considerations. The data, however, always decrease faster with T, and I_A is about 25–30 % lower at 15 K than anticipated.

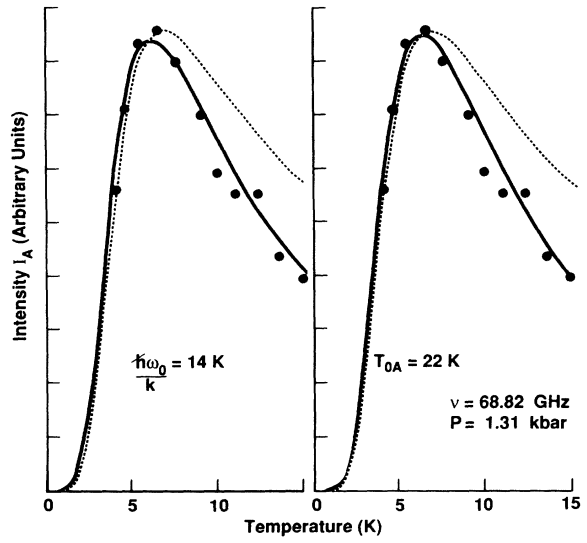

FIG. 6. A plot of I_A as a function of temperature for $P=1.316$ kbar and $\nu=68.82$ GHz. In (a) we have assumed no temperature dependence in μ and fit the data to Eq. (7) with $\hbar\omega_0/k=14$ K. In (b) the HO excited states were removed ($\hbar\omega_0/k=1000$ K) and the data fit using $T_{0A}=22$ K.

The additional temperature dependence can be accounted for in several ways. First, if we assume that μ is independent of T, we can fit the data by including the on-center HO excited states using $\hbar\omega_0/k\approx 14$ K as shown in Fig. 6(a). Clearly for this model several HO excited states are populated at $T\approx 15$ K (because of the large degeneracies of the excited states even the level at $\hbar\omega_0/k\approx 70$ K is appreciably populated). However, this value of $\hbar\omega_0/k$ seems rather low compared to the value of 24 K obtained for the on-center Ag^+ ion in KI.

Over the limited temperature range of the data we can also obtain a reasonable fit using a single excited state about 13 K above the ground state, with a degeneracy of roughly 10. However, there is no obvious motivation as to why one excited state would exist and we do not pursue such a model further.

A second alternative is that the matrix element μ is temperature dependent. If we ascribe all the additional temperature dependence to the parameter T_{0A} [Eq. (10)] we obtain $T_{0A}\approx 22$ K as shown in Fig. 6(b). However, it is possible that both $\hbar\omega_0$ and T_{0A} play a role; we cannot separate these effects for line A with the present data.

B. Ratio of intensities I_B/I_A

The intensity I_B decreases very rapidly with T (Fig. 2) such that even the ratio R decreases by a large factor from 4 to 15 K. In this case, inclusion of the HO levels has a rather small effect on R as seen in Fig. 7 for temperature-independent tunneling parameters. This is expected from an examination of Eq. (9); at $T\rightarrow\infty$ or $T\rightarrow 0$, the factor $Z(\omega,T)$ [Eq. (9b)] is 1. Only at intermediate temperatures (5 K) is there a small deviation. The more striking result in Fig. 7 is that the shape of the curve is wrong. We can, however, obtain a good fit to the ratio data if we let $T_{0B}\approx 8$ K and assume T_{0A} is large. We believe this is clear evidence that the effective tunneling parameters are indeed temperature dependent as predicted. The effect of T_0 on the relaxation rate for NaBr:F^- has been observed by Kanert *et al.*[17]; to our knowledge the present measurements are the first time that such a temperature dependence has been observed so directly. In the rest of the analysis, we will ignore the $1/T_{0A}$ term in Eq. (9). From part A above, $T_{0A}\geq 22$ K; consequently for these values, T_{0B} might be $\approx 10\%$ high under this approximation, an error less than or equal to our uncertainty in T_{0B}.

In Fig. 8 we show fits of the data to Eq. (9) for a range of frequencies and pressures. For the higher-pressure data the ratio drops rapidly with T and then becomes constant at higher temperatures; however, the position of the line designated as line B also shifts with T for these pressures. We believe that a third weak line, corresponding to the transitions for n_2 to n_3 and n_3 to n_4 in Fig. 5

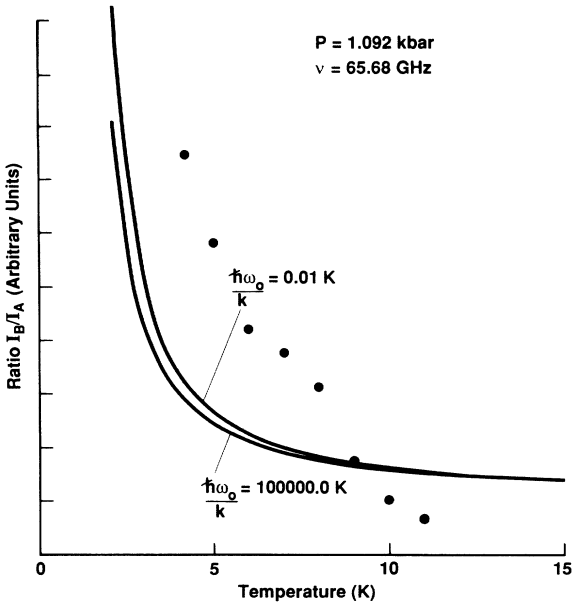

FIG. 7. A comparison of the experimental values of R ($\nu=65.68$ GHz, and $P=1.092$ kbar) as a function of T to the prediction of Eq. (9) when the tunneling parameters are held independent of temperature. The curves for $\hbar\omega_0/k=0.01$ and 10^5 K indicate that R is insensitive to this parameter.

has become strong enough to be observed at these pressures; it becomes easily observed at somewhat higher pressures.[5] As a result of the very broad linewidths at the higher temperatures, and the low signal-to-noise ratio, it is very difficult to separate these two lines for intensity determinations. We note that for the sum I_c of the intensities for the transitions n_2-n_3 and n_3-n_4, the ratio I_c/I_A becomes constant, independent of T for $\hbar\omega/kT<1$. For

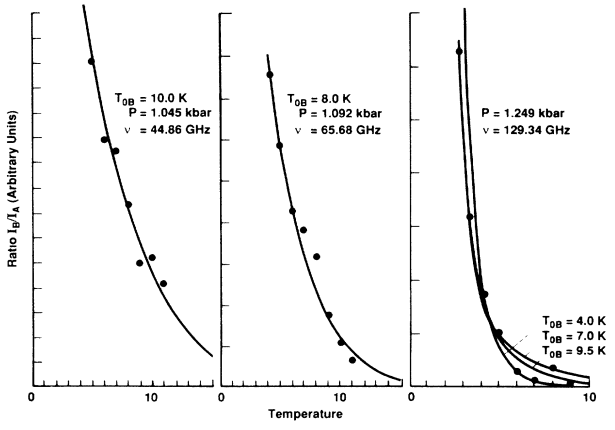

FIG. 8. Fit of R to Eq. (9) for several data sets. The best fit of T_{0B} is indicated. At the higher pressures, another weak line is present which masks the on-center to off-center transition above 7 K. For $P=1.25$ kbar, a T-independent term has been subtracted from R to account for this additional line.

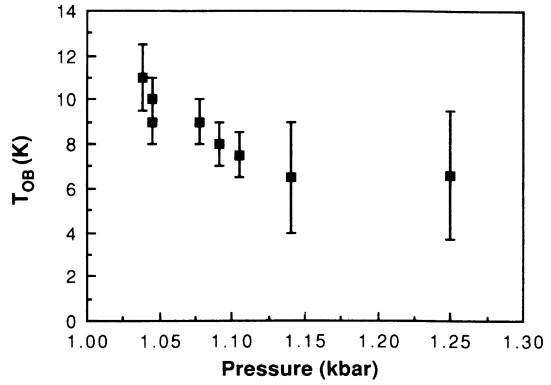

FIG. 9. A plot of T_{0B} as a function of hydrostatic pressure.

the data at the higher pressures, we therefore estimate T_{0B} from the low-T data. In this case the error in T_{0B} is large because the T dependence of R is dominated by the factor $\exp[(\epsilon_1-\hbar\omega/2)/kT]$ in Eq. (9) when ϵ_1 becomes large and T is low.

In Fig. 9 we plot the values of T_{0B} as a function of P from 1.0 to 1.25 kbar. There is a clear tendency for T_{0B} to decrease with increasing pressure. This is not unexpected if the increasing pressure reduces the barrier and softens the potential between the on-center and off-center sites. Then the probability for thermal activation should increase, and the temperature, at which thermal vibrations begin to modulate the potential significantly, will decrease.

VI. DISCUSSION OF RESULTS AND CONCLUSIONS

We have observed a very rapid decrease in the intensity of the on-center to off-center transition which we have interpreted in terms of a Debye-Waller-like temperature dependence of the tunneling parameter. Fits to the data yield values of T_{0B} near 9 K but there is a clear indication that T_{0B} is decreasing as P increases. For the normal PER transition between the tunneling states the temperature dependence of the intensity is not as strong and a temperature dependence for μ, the second-nearest-neighbor tunneling parameter, would require that $T_{0A}\approx 22$ K. However, the latter effect could also be explained in terms of excited on-center states with $\hbar\omega_0/k\approx 14$ K. To clarify this aspect, ir measurements at $P\approx 1.3$ kbar are needed to measure $\hbar\omega_0$ directly.

The effects of a T-dependent tunneling matrix element have previously been observed indirectly in the T dependence of the relaxation.[17] The present measurements indicate that it can be observed directly and will control the matrix elements of transitions induced by phonons and oscillating electric fields. For glasses[18] this may have a very important consequence if the value of T_0 for such systems is low for the rapidly tunneling atoms. Then tunneling splittings and matrix elements can no longer be treated as constants, independent of temperature. It appears unlikely from our investigations of tunneling systems in the alkali halides that these effects will be impor-

tant at temperatures well below 1 K. However, a rapid decrease in the magnitude of a tunneling parameter at higher temperatures would lead to peaks or plateaus in quantities such as the specific heat or the thermal conductivity.

ACKNOWLEDGMENT

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