vicinity of $16\ \mathrm{cm}^2/\mathrm{V}$ sec were obtained during studies where the ions were considerably older. The reason that Biondi and Chanin[4] did not detect the ion having a mobility of $16\ \mathrm{cm}^2/\mathrm{V}$ sec was most probably that the discharge pulses used in their studies were shorter than those at which this ion appeared in the present experiment. The value of $20.9\ \mathrm{cm}^2/\mathrm{V}$ sec reported by Tyndall could probably refer to an impurity ion such as the neon ions, since it was found, during the present studies, that this ion was the dominant ion when commercial helium was not further purified by means of the cataphoretic-segregation process.[15]

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CHARACTERISTIC TEMPERATURE DEPENDENCE FOR LOW-LYING LATTICE RESONANT MODES*

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Recent investigations of impurity-induced lattice absorption in the far-infrared spectral region have demonstrated that sharp absorption lines can be associated with lattice resonant modes.[1-3] The experiments have clearly shown that a heavy impurity ion or light impurity ion coupled with weak force constants can activate a low-frequency infrared-active mode in alkali-halide crystals.

We wish to call attention to a marked temperature dependence of the resonant-mode absorption strength which has been observed. Interpreting our experimental results, we find that (1) the observed temperature dependence probably is characteristic of lattices which contain low-frequency resonant modes, and (2) a temperature-dependent contribution to the static dielectric constant should occur. Neither of these results has been previously predicted.

The experimental apparatus for the far-infrared measurements has been described elsewhere.[1] The experimental results are shown in Fig. 1 for a resonant mode absorption produced by a heavy impurity (KI:Ag$^+$) and also by a light impurity (KBr:Li$^+$). In Fig. 1(a) the absorption coefficient in the neighborhood of the resonant-mode absorption for KI:Ag$^+$ is shown for three different temperatures. As the sample temperature is increased the eigenfrequency decreases slightly and the half-width increases; however, the largest change occurs in the absorption strength ($\int A d\omega$ [cm^{-2}]). The exponential form of the temperature dependence of the absorption strength is shown in Fig. 1(b) where a semilog plot of the strength versus square of temperature can be fitted to a straight line. A similar temperature dependence is found for KBr:Li$^+$ and is shown in Fig. 1(c). Finally, the general nature of this temperature dependence is indicated by the similarity of the temperature dependence of the hydride and deuteride local-mode absorption strengths[4] in KCl:H$^-$ and KCl:D$^-$, as shown in Fig. 1(d), to the resonant-mode cases mentioned above.

To interpret the temperature dependence of the absorption coefficient we shall restrict

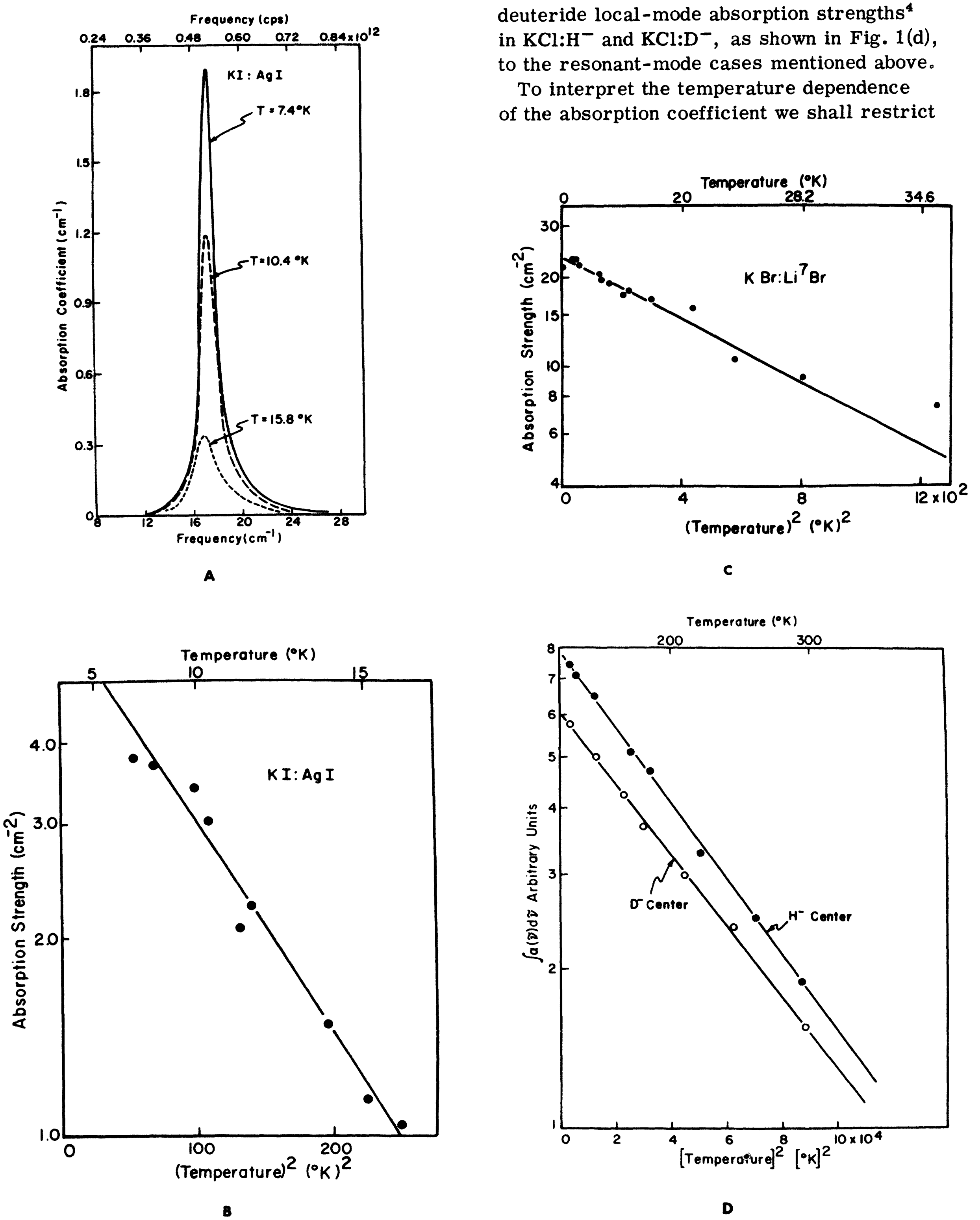

FIG. 1. (a) The temperature dependence of the far-infrared absorption coefficient for KI:Ag$^+$ (1.5×10^{18} Ag$^+$ per cm^3). The ordinate is the impurity-induced absorption coefficient. (b) The temperature dependence of the integrated absorption coefficient for KI:Ag$^+$, (c) for KBr:Li$^+$ (1.3×10^{18} Li$^+$ per cm^3), and (d) for KCl:H$^-$ and KCl:D$^-$.[4]

ourselves to the cases of a sharp resonant mode at very low frequencies or a localized mode far above the Debye cutoff frequency. For such an impurity mode, the Einstein oscillator model holds in first approximation.[3,5] Thus, we can divide the crystal into two parts, the one being associated with the impurity (the i system) and the other with the rest of the lattice (the L system). The Hamiltonian of the total system describing the motion of the ions can be written as the sum of three terms: the Hamiltonian of the i system, the Hamiltonian of the L system, and the interaction term which we shall call $V(r,R)$, where r and R are a set of coordinates in the i system and in the L system, respectively. Then if $V(r,R)=0$, the L system can be described by a set of harmonic oscillators. We look for eigenstates of the Hamiltonian of the form $|\alpha n\rangle = |\alpha\rangle |n\rangle$, where α and n specify eigenstates of the i system and the L system, respectively, and take the interaction to be of the form

$$\langle\alpha|V(r,R)|\alpha\rangle = \sum_\lambda C_{\alpha\lambda}\omega_\lambda Q_\lambda, \tag{1}$$

where ω_λ is the eigenfrequency of the mode λ in the L system and Q_λ is its normal coordinate. $C_{\alpha\lambda}$ is an interaction constant. The first assumption stems from the experimental facts[3] that the Einstein model holds for the i system, and the second is due to the assumption that $\langle\alpha|V(r,R)|\alpha\rangle$ is linear in strain operators in the L system. We neglect higher order coupling here. The eigenstates of the L system can then be described in terms of a set of displaced harmonic oscillators.

Using the above preliminaries we study the absorption coefficient. We shall suppose that the resonant mode or the localized mode is optically active, which, if we make use of a simple harmonic model for the crystal, yields the following typical resonance form for the absorption coefficient:

$$A_0(\omega) = \frac{B(\omega)}{\pi}\,\frac{\Gamma_{0/2}}{(\omega-\omega_0)^2+\Gamma^2_{0/4}}, \tag{2}$$

where ω_0 is the eigenfrequency of the resonant mode or the localized mode and Γ_0 is the width, which is to be taken as zero for the localized mode. $B(\omega)$ is a function of frequency which may be different for different properties of impurity modes. When we introduce the interaction between the i system and the L system, Eq. (2) may be modified as follows[6]:

$$A(\omega) = \frac{B(\omega)}{\pi}\sum_{n,n'}\rho(n)\frac{|\langle n'|n\rangle|^2(\Gamma_{0/2})}{(\omega-\omega_0+\omega_{n'}-\omega_n)^2+\Gamma_0{}^2/4}, \tag{3}$$

where $\hbar\omega_{n'}$ and $\hbar\omega_n$ are the energies of the L system in the final state and in the initial state, respectively, and $\rho(n)$ is the probability that the L system is initially in the state $|n\rangle$. In the evaluation of the square of the matrix element, $|\langle n'|n\rangle|^2$, we retain only terms which are quadratic in the displaced harmonic oscillator coordinates and neglect higher orders. An approximate expression of $A(\omega)$ near $\omega=\omega_0$, which is the absorption coefficient of the zero-L-phonon line, is given by[7]

$$A(\omega) = \exp\left\{-\frac{1}{\hbar M}\sum_{\hbar}\frac{(\langle n_\lambda\rangle+\frac{1}{2})}{\omega_\lambda} \times C_{\alpha\lambda}-C_{\alpha'\lambda})^2\right\}A_0(\omega), \tag{4}$$

where M is the mass of the atom in the L system and $\langle n_\lambda\rangle = \{\exp(\hbar\omega_\lambda/k_{\rm B}T)-1\}^{-1}$ is the average occupation number of the mode λ at temperature T. $k_{\rm B}$ is the Boltzmann constant. For the resonant mode in which Γ_0 is finite, Eq. (4) only holds approximately. If we assume that $(C_{\alpha\lambda}-C_{\alpha'\lambda})^2$ is constant and consider the low-temperature region where $T\ll\Theta_0$, where Θ_0 is the Debye temperature, then Eq. (4) becomes[8]

$$A(\omega) = \exp\{-\beta(1+\gamma T^2)\}A_0(\omega), \tag{5}$$

and the second term in the exponent yields the typical T^2 dependence.

A detailed discussion of the coefficients β and γ is omitted here.

The temperature dependence of the resonant-mode absorption cofficient given in Eq. (5) should produce an impurity-induced temperature-dependent contribution to the static dielectric constant, $\Delta\epsilon_1{}^i$, since from the Kramers-Kronig relations[9]

$$\Delta\epsilon_1{}^i = \frac{2}{\pi}\int_0^{\omega>\bar{\omega}}\frac{\epsilon_2{}^i(\omega')}{\omega'}d\omega', \tag{6}$$

where $\epsilon=\epsilon_1-i\epsilon_2$ and $\epsilon_2{}^i(\omega')$ describes only the low-frequency impurity-induced absorption contribution. The integral in Eq. (6) encompasses both the sharp resonant line at ω_0 and the important low-frequency region of the one-

L-phonon spectrum at $\bar{\omega}$. The intrinsic lattice absorption from the transverse optic branch of the L system is far removed from the low-frequency region considered here. As the temperature of the system increases from absolute zero, the absorption strength is transferred from the frequency region around ω_0 to the frequency region around $\bar{\omega}$, and the center of gravity of the total absorption strength shifts with temperature. As long as the center of gravity of the low-frequency impurity-induced absorption strength is temperature dependent, Eq. (6) guarantees that the static dielectric constant is also temperature dependent. An interesting consequence of this temperature dependence is that a characteristic electrocaloric effect[10] should occur for this system. This effect should be quite modest for KI:Ag$^+$ and KBr:Li$^+$ but should become measurable for systems with lower frequency impurity modes.[11]

In summary, then we conclude that the Debye-Waller factor, which appears in the zero-phonon lines of neutron diffraction, x-ray diffraction, the Mössbauer effect,[8] and optical absorption of some defects in solids,[12] also exists for the zero-L-phonon line of impurity-activated infrared absorption in alkali-halide crystals. The appearance of the Debye-Waller factor in this latter problem arises from random thermal modulation of impurity modes by lattice phonon modes. The low frequency of the resonant mode insures that the influence of this modulation can also be seen as a temperature-dependent contribution to the static dielectric constant.

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MAGNON-INDUCED ELECTRIC DIPOLE TRANSITION MOMENT*

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Recently Halley and Silvera[1] suggested a mechanism of inducing the electric dipole transition moment at an ion site in a magnetic material by magnon excitation. Assuming this mechanism, they interpreted the far-infrared absorption peak of FeF$_2$ at 154 cm^{-1}. The same mechansim was assumed by Greene et al.[2] in the interpretation of the magnon side band in the visible absorption spectrum of MnF$_2$.

In this Letter we will point out that these spectra as well as the far-infrared absorption[3] of CoF$_2$ should be, as shown at the end of this Letter, interpreted by another mechanism, which was originally considered to explain[4] the magnon side band of the $^3A_2 \rightarrow {}^1E$ line in KNiF$_3$ and was successfully applied to the Mn-