Since $\nabla T_L \to 0$ at $\dot{Q} = \dot{Q}_c$, the "crossing phenomenon" can be expressed as $\nabla T_\Omega(\Omega = 10) = \nabla T_L(\Omega = 0)$. If this is a general result then when combined with Eqs. (1) and (6) it gives a condition for $V_c(\Omega)$, namely, $\kappa L_0(T, V_c) = 3\Omega$. Recalling[9] that $b = (\kappa/2\Omega)^{1/2}$, this condition can be written as

$$(L_0^{1/2} b)_c = (\tfrac{3}{2})^{1/2} = 1.22. \tag{7}$$

This is essentially the same critical length condition [Eq. (4)] that was used above in the analysis of the critical heat currents (Fig. 2). Notice also that the sharpness of the transition in rotating channels is simply a consequence of the larger value of L_0 or of ∇T_L at $\dot{Q}_c$ (solid circles in Fig. 2).

The concepts developed for describing superfluid turbulence in small channels, as expressed in Eqs. (1), (2), and (3), have been successfully applied to data obtained in channels an order of magnitude larger. In nonrotating channels the temperature gradient is in good quantitative agreement with Eqs. (1) and (2), and the critical heat flux for different temperatures channel sizes and shapes agree with the critical length condition in Eq. (3). When this condition is naturally extended to channels in rotation [Eq. (4)] it also agrees quantitatively with the magnitude and $\Omega^{1/2}$ dependence of W_c, the temperature dependence of W_c, and the "crossing phenomenon". Considering the range of data described and the good quantitative agreement obtained, it must be concluded that the physical ideas expressed by Eqs. (1), (2), and (3) are essentially correct.

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[6] The velocity V is related to the heat flux W by $V = W/\rho_s ST$.

[7] Actually, α appears to be a weak function of d [D. R. Ladner, Ph.D. thesis, Ohio State University, 1979 (unpublished)]. A value of 2.5 is used for α in the present paper, but the conclusions are not strongly dependent on this choice.

[8] It should be noted that Eq. (2) presumably describes homogeneous turbulence and is not valid in the critical region ($V_c < V \leq 1.2\, V_c$). The hysteresis effects (Ref. 5) associated with the critical region do indicate that L_0 is discontinuous at V_c, but the exact form of the discontinuity is not known. The Vinen formula for L_0 reduces to Eq. (2) for $V > 1.2\, V_c$ and gives $(L_0^{1/2} d)_c = 2.0$.

[9] There may be a constant of order unity in this expression for b depending on the geometrical arrangement of the lines and the precise meaning of "average separation."

[10] It is interesting to note that the single length obtained by the "parallel addition" of b and d, namely $bd/(b+d)$, gives both Eq. (6) and the $\Omega = 0$ critical condition, Eq. (3).

Experimental Test of Theoretical Models for Urbach's Rule at Excitonic Absorption Edges

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It is shown that electroabsorption measurements provide an elegant test to decide between the two most favored theories of Urbach's rule at excitonic absorption edges, namely the model of the field-ionized exciton by Dow and Redfield and the model of the momentarily trapped exciton by Sumi and Toyozawa. Experiments performed at the intrinsic Urbach tails of the ionic insulators CuCl and TlCl give support to the field-ionization model.

In a large variety of insulating and semiconducting materials the spectral dependence of the absorption coefficient α at the low-energy tail of the fundamental optical absorption edge can be expressed by the empirical relation

$$\alpha = \alpha_0 \exp[\sigma(\hbar\omega - E_0)/k_B T], \tag{1}$$

where $\hbar\omega$ is the photon energy, T is the temperature, k_B is Boltzmann's constant, and α_0, σ, and E_0 are fitting parameters. This dependence, which is known as Urbach's rule,[1] has been confirmed experimentally as a kind of universal law[2] and has stimulated many efforts for its theoretical explanation.[3] In the past, two competing theories of exponential absorption edges have found particular attention, the model of the electric-field-ionized exciton by Dow and Redfield (DR)[4] and the model of the momentarily trapped exciton by Sumi and Toyozawa (ST).[5]

Both theories stress different aspects of exciton phonon coupling: In the DR theory, the tailing of the optical absorption edge is ascribed to field ionization of exciton states by electric microfields from lattice vibrations or charged impurities. The tunneling of the electron away from the hole through the Coulomb barrier leads to an exponentially decaying density of states below the absorption threshold, resulting in Urbach's rule. In this model, the influence of the phonon field on the relative motion of the electron-hole pair and therefore exciton *interband* scattering is the dominant mechanism.

The ST theory, on the other hand, discusses the center-of-mass motion of the exciton in the phonon field, where exciton *intraband* scattering is important. Urbach's rule follows from an interplay between the mobile nature of the exciton and the tendency of the vibrating lattice to localize the exciton momentarily. The exponential line shape of the exciton absorption observed on the low-energy side can be regarded as intermediate between the Lorentzian line shape of the mobile exciton and the Gaussian shape of the trapped exciton.

Despite this clear-cut classification of possible mechanisms for Urbach's rule, only a few experiments as to the specific nature of the interaction mechanisms have been reported. Investigations[6] of the absorption edges of GaAs and ZnS showed that the electric field of charged impurities has an influence on the breadth of the edge. In several semiconductors a broadening of the edge was observed in domains of intense acoustic flux.[7] The domain-induced absorption is due to ionization of excitons in the large piezoelectric microfields of these domains.[8]

These experiments demonstrated that absorption edges in semiconductors are very sensitive to electric microfields of various origin and showed that "extrinsic" exponential tails can be caused by ionization of the exciton. However, they could not prove rigorously that "intrinsic," phonon-induced Urbach tails are caused by the same mechanism. In addition, since these experiments were performed in semiconductors with comparatively shallow excitons, they do not allow one to conclude that field ionization is equally important in more ionic insulators. It is known from emission spectra that self-trapping of excitons plays an essential role in strongly ionic substances like the alkali halides. On the other hand, the large electric fields from LO phonons in strongly ionic crystals may compensate for the deeper binding energy of the excitons, so that field ionization is also possible. Therefore the validity of the DR or the ST model of Urbach's rule for intrinsic absorption edges remains an open question, in particular for ionic insulators.

This Letter describes an experimental test which uses the changes of the absorption edge in an external electric field to decide whether interband or intraband exciton scattering is dominant in the Urbach tail region. The method is based on the fact that the external electric field primarily influences the relative motion of the electron-hole pair and thus interferes with the internal field mechanism, whereas it does not act on the exciton's center-of-mass motion. Therefore, different absorption changes should occur depending on the mechanism which is responsible for the Urbach tail.

We first discuss the absorption changes expected on the basis of the ST theory, where exciton intraband scattering is regarded as the dominant mechanism. In this model, the external field F_e will only polarize the relative motion of the electron-hold pair leading to a Stark effect of the exciton spectrum. For a hydrogenic exciton spectrum the absorption edge of the lowest exciton band will be shifted by $\Delta E_0 = \Delta E_{1s} = -9E_B F_e^2/8F_I^2$. Here, E_B is the exciton binding energy and F_I the ionization field defined by $ea_B F_I = E_B$, where a_B is the exciton Bohr radius and e the elementary charge. Simultaneously, the polarization will lead to a decrease of the oscillator strength f_{1s} of the 1s exciton and cause a corresponding change of the prefactor α_0 in Urbach's rule by $\Delta\alpha_0/\alpha_0 = \Delta f_{1s}/f_{1s} = -43F_e^2/32F_I^2$. As an additional indirect effect of the field, the exciton-phonon coupling constant for intraband scattering in the 1s exciton dispersion band will be decreased by the polarization leading to a change of the steepness parameter σ in Urbach's rule. The change of σ can be approximately related to the change in oscillator strength through $\Delta\sigma/\sigma = -2\Delta f_{1s}/f_{1s}$. Taking three effects together, we expect a relative change

$$\frac{\Delta\alpha}{\alpha} = \left[\frac{9}{8}\frac{\sigma}{k_BT}E_B - \frac{43}{32}\left(1 + 2\ln\frac{\alpha_0}{\alpha}\right)\right]\frac{F_e^2}{F_I^2} \qquad (2)$$

of the absorption constant in the Urbach tail region. For fixed α the quantity $\Delta\alpha/\alpha$ should vary linearly with the logarithmic slope σ/k_BT of the edge and therefore linearly with reciprocal temperature.

In the DR model, the absorption constant α is an average of the field-dependent exciton absorption $\alpha(F)$ performed over a Gaussian-Maxwellian distribution $P_0(F)$ of microfields of strength F. On the low-energy side of the exciton the spectral dependence of $\alpha(F)$ can be expressed as $\alpha(F) \propto \exp[C(\hbar\omega - E_0)/F]$, where C is a constant. In the performing of the average, the exponential character of the edge is well preserved[4] and α can be described by $\alpha \propto \exp[C'(\hbar\omega - E_0)/F_{\mathrm{rms}}]$, where C' is a renormalized constant and F_{rms} is the average microfield. If we compare with Urbach's rule, the relation $C'/F_{\mathrm{rms}} = \sigma/k_BT$ should hold. As Dow and Redfield showed,[4] the temperature dependence $F_{\mathrm{rms}} \propto k_BT$ can be explained by taking into account the nonuniformity of the microfields, which leads to a specific cutoff procedure in the summation over the different phonon contributions.

In the presence of an additional external electric field F_e we have to replace the distribution $P_0(F)$ by a distribution $P(F)$ which results from the superposition of the external and the internal fields. The new distribution is given by

$$P(F) = P_0(F)\left[1 - \frac{3}{2}\frac{F_e^2}{F_{\mathrm{rms}}^2} + \frac{3}{2}\frac{F_e^2F^2}{F_{\mathrm{rms}}^4}\right]$$

which is exact to second order in the external field. The relative change of the absorption constant calculated with this distribution can be approximated analytically by

$$\frac{\Delta\alpha}{\alpha} = \frac{3^{1/3}}{2}\left(\frac{C}{C'}\right)^{2/3}\left(\ln\frac{\alpha_0}{\alpha}\right)^{2/3}\frac{F_e^2}{F_{\mathrm{rms}}^2}. \qquad (3)$$

This equation allows us to determine the average internal field F_{rms} from the field-induced absorption changes, if the DR model is valid. One would then expect that $\Delta\alpha/\alpha$ varies proportionally to the square $(\sigma/k_BT)^2$ of the logarithmic slope, in view of the above mentioned relation $C'/F_{\mathrm{rms}} = \sigma/k_BT$.

As an application of these results we have studied the influence of an external electric field on the Urbach tails of the ionic insulators CuCl and TlCl which are ideally suited for this purpose. Both compounds possess well-known exciton states with extended Urbach tails. Concerning the binding energies E_B of the 1*s* excitons, CuCl with $E_B = 190$ meV (Ref. 9) resembles strongly ionic compounds with deep excitons, whereas TlCl with $E_B = 11.3$ meV (Ref. 10) is closer to the more covalent compounds with shallow excitons. The study of extremely ionic compounds like the alkali halides becomes difficult because of the large values of F_I and F_{rms}, and the investigation of semiconductors is complicated by photoconductivity and sample heating.

Samples of CuCl and TlCl with thicknesses ranging from 50 to 200 μm were prepared by solidification of the material between quartz plates in vacuum. With these samples the Urbach tails could be observed in the region 10^1 cm^{-1} $< \alpha < 10^3$ cm^{-1} and in a temperature range between 80 and 400 K. We find the following parameters of the exponential edges: $\alpha_0 = 5\times10^4$ cm^{-1}, $\sigma = 1.6$, $E_0 = 3.230$ eV for CuCl, and $\alpha_0 = 2\times10^4$ cm^{-1}, $\sigma = 1.2$, $E_0 = 3.425$ eV for TlCl. The electric-field-induced changes of the absorption constant were measured in a transverse field configuration using techniques of modulation spectroscopy and synchronous photon counting. Absorption changes quadratic in the external field F_e were observed in all cases up to the largest fields applied (9 kV/cm for CuCl and 5 kV/cm for TlCl).

Figure 1 shows that the field-induced absorption changes $\Delta\alpha/\alpha$ for fixed α vary quadratically with the logarithmic steepness σ/k_BT of the edge and

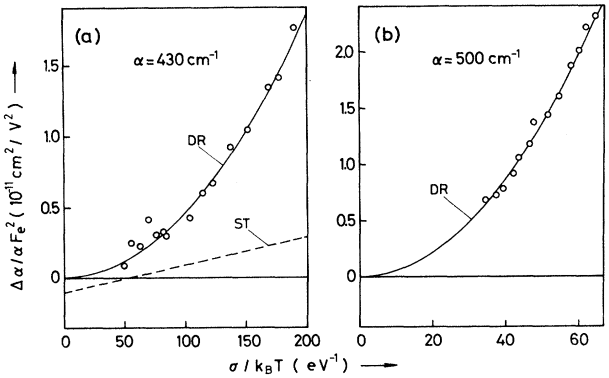

FIG. 1. Relative change $\Delta\alpha/\alpha$ of the absorption constant by an external electric field F_e as a function of σ/k_BT for fixed α at the excitonic absorption edges of (a) CuCl and (b) TlCl. Solid lines are fits with a quadratic dependence predicted for the DR model. Dashed line for CuCl denotes predictions on the basis of the ST model (for TlCl this line is far off scale).

thus follow the predictions of the DR model. The dashed line in Fig. 1(a) shows the effect linear in σ/k_BT expected for CuCl on the basis of the ST model and calculated from Eq. (2) with known parameters of the Urbach edge, the exciton binding energy, and the ionization field. The ionization field for the CuCl 1s exciton was determined experimentally from the quadratic Stark shift measured in the central part of the exciton band.[11] With $\Delta E_{1s}/F_e^2 = -2\times10^{-14}\ e\cdot\text{cm}^2/\text{V}$ and $E_B = 190$ meV,[9] we find $F_I = 3.3\times10^6$ V/cm. For TlCl the ionization field can be estimated from the exciton parameters applying the relation $F_I = 2\,E_B^2\,\epsilon/e^3$, where ϵ is the dielectric screening constant of the exciton. With static screening [$\epsilon = 37.6$ (Ref. 10)] and $E_B = 11.3$ meV (Ref. 10), we find $F_I = 6.7\times10^4$ V/cm and $\Delta\alpha/\alpha F_e^2$ would be of the order of -2×10^{-9} cm^2/V^2, which is far out of the scale of Fig. 1(b). Hence, despite some approximations made above, the large discrepancies between the measured $\Delta\alpha/\alpha$ and the values predicted for the ST model also favor the DR model.

Since the ratio C/C' is known from the field-averaging procedure ($C/C' = 1.4$ in the experimentally relevant region of the spectrum), the average internal field $F_{\rm rms}$ can be determined with the help of Eq. (3) from the quadratic absorption changes. We find, at 300 K,

$F_{\rm rms} = 1.3\times10^6$ V/cm for CuCl,

$F_{\rm rms} = 0.4\times10^6$ V/cm for TlCl.

These values should be compared with the average fields calculated from lattice dynamics. Similar to Dow and Redfield[4] the temperature-dependent cutoff wave vector q_c in the summation of the phonon contributions over momentum space may be estimated in the following way. To permit the tunneling, the kinetic energy of the electron-hole relative motion gained by localization in a potential trough of length $\Delta x = \pi/q_c$ via the uncertainty relation $\Delta x\,\Delta p \geqslant \hbar$ must not exceed the potential energy $eF_{\rm rms}/q_c$. This condition leads to q_c depending on $F_{\rm rms}$ with the result, after summation over the LO phonon contributions,[12]

$$F_{\rm rms} = \frac{4}{3}\pi\,\frac{e\mu}{\hbar^2}\,\frac{\epsilon_0-\epsilon_\infty}{\epsilon_0\epsilon_\infty}\,k_BT, \qquad (4)$$

where μ is the reduced effective mass of the exciton and ϵ_0 and ϵ_∞ are the static and high-frequency dielectric constants, respectively. With $\epsilon_0 = 7.9$ (Ref. 13), $\epsilon_\infty = 3.6$ (Ref. 14), $\mu = 0.39$ (Ref. 15) for CuCl and $\epsilon_0 = 37.6$ (Ref. 10), $\epsilon_\infty = 5.1$ (Ref. 10), $\mu = 0.24$ (Ref. 10) for TlCl we obtain at $T = 300$ K the theoretical values $F_{\rm rms} = 1.2\times10^6$ V/cm for CuCl and $F_{\rm rms} = 0.8\times10^6$ V/cm for TlCl, in reasonable agreement with the experimental values found above.

In summary, we have shown experimentally that field ionization of excitons is the dominant mechanism for Urbach's rule at the phonon-induced absorption edges of the ionic insulators CuCl and TlCl. Together with the investigations cited above on extrinsic absorption edges of more covalent semiconductors this gives support to the conjecture that field ionization is a really universal mechanism for exponential absorption edges.

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