Measurements of the Electric Field of Zero-Point Optical Phonons in GaAs Quantum Wells Support the Urbach Rule for Zero-Temperature Lifetime Broadening

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We study a specific type of lifetime broadening resulting in the well-known exponential "Urbach tail" density of states within the energy gap of an insulator. After establishing the frequency and temperature dependence of the Urbach edge in GaAs quantum wells, we show that the broadening due to the zero-point optical phonons is the fundamental limit to the Urbach slope in high-quality samples. In rough analogy with Welton's heuristic interpretation of the Lamb shift, the zero-temperature contribution to the Urbach slope can be thought of as arising from the electric field of the zero-point longitudinal-optical phonons. The value of this electric field is experimentally measured to be 3 kV cm^{-1}, in excellent agreement with the theoretical estimate.

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In close analogy with free-space theories, quasiparticles and elementary excitations in condensed matter systems are also described by quantum fields. The zero-temperature ground state of the solid (and liquid helium) is then the vacuum for the relevant interacting fields. The mode structure of the vacuum and the zero-temperature dynamics of interacting fields is essential to understanding a host of phenomena in condensed matter systems—from zero-temperature phase transitions, to cavity quantum electrodynamics with quantum dots, to electron dephasing in mesoscopic systems. Relevant to the context of this work, it has been pointed out that for good agreement with experiment, the zero-point motion of atoms must also be accounted for in the renormalization of the band energies in semiconductors [1]. But the zero-point fluctuations almost always appear only as ingredients for the consistent theory, not amenable to direct measurement [2].

In this Letter, we study a specific type of lifetime broadening resulting in the "Urbach tail" [4–15] in GaAs quantum wells (QWs) at low temperatures. We propose that there is a fundamental zero-temperature contribution to the Urbach slope resulting from the electric field of the zero-point optical phonons. The value of this electric field is estimated experimentally.

Urbach tail.—While the band theory of solids predicts that the density of states of insulators must have a sharp cutoff at the band edge, experiments have long shown a tail in the absorption spectrum that extends into the theoretically forbidden gap [4]. This departure from ideality would not have been so interesting if it were not for the near universality of the form of the joint density of states (JDOS) $\kappa(\hbar\omega)$ within the energy gap, both spectrally,

$$\kappa(\hbar\omega) = \kappa_0 \exp[-\sigma(T)(\hbar\Omega_0 - \hbar\omega)/k_B T], \qquad \omega < \Omega_0, \tag{1}$$

and in its temperature dependence,

$$\sigma(T) = \sigma_0[2k_B T/\hbar\Omega_p]\tanh[\hbar\Omega_p/2k_B T]. \tag{2}$$

The characteristic energies $\hbar\Omega_0$ and $\hbar\Omega_p$ and the two prefactors, σ_0 and κ_0, are the only material-specific quantities in Eqs. (1) and (2). Six decades have accumulated overwhelming experimental evidence that this relationship, the Urbach-Martienssen rule [4–15], holds in materials ranging from glasses with extreme topological disorder [12,13] to compositionally pure single crystals [15], in a range of energy gaps between 150 meV [16] and 10 eV [9].

An interesting consequence of this empirical rule (to be put on more rigorous ground below) is that the Urbach slope is finite even at zero temperature. But before delving into that, we will first need to establish the Urbach rule in GaAs QWs because this has previously not been done. After establishing the full Urbach rule [6] [Eqs. (1) and (2)] and the existence of the Urbach focus, we will study the effect of structural (interface roughness) disorder on the band tail parameters in different quality QWs and finally show that in high-quality samples, the fundamental limitation indeed comes from the zero-point phonons, which is then systematically studied.

Measurements.—The usual procedure to infer the JDOS from experiments is through optical absorption measurement [12]. Unfortunately, the small thickness of QWs rules out a direct absorption measurement with a large dynamic range. Hence, other spectroscopic techniques have been

used in the past to indirectly infer the absorption spectrum (see, e.g., Refs. [17–21]). Given the very large dynamic range made possible in modern photoluminescence (PL) measurements, PL is a good choice [17,18,20] to infer the absorption spectrum of high-quality samples where at low temperatures the Stokes shift is negligible. A prescription for determining the absorption coefficient $\kappa(\hbar\omega)$ from the spontaneous emission spectrum $\mathfrak{F}(\hbar\omega)$ measured by PL can be inferred from the relationship between Einstein A and B coefficients [21],

$$\mathfrak{F}(\hbar\omega) = \frac{n^2[\hbar\omega]^2}{\pi^2\hbar^3c^2}\kappa(\hbar\omega)f_c(1-f_v), \tag{3}$$

where f_c and f_v are the occupation probabilities for the conduction and valence band states and they control the high-energy region of the spectrum. Under low optical excitation, the low-energy tail of the absorption coefficient is related to the emission spectrum through $\kappa(\hbar\omega) = C'\mathfrak{F}(\hbar\omega)[\hbar\omega]^{-2}$, where the constant C' is estimated by normalizing the inferred absorption coefficient with the literature value at one specific energy (say, the band edge) [21]. We shall follow this prescription and use the PL spectra to infer the JDOS. It is emphasized that use of the van Roosbroeck-Shockley relation in this context will be incorrect [21]. For electroabsorption measurements, JDOS was inferred from photoconductivity measurements. Details of the samples are given in the Supplemental Material [22].

Urbach rule in GaAs/AlGaAs QWs.—Figure 1(a) shows the measured PL spectrum at 4 K at an excitation power of 10 μW for a QW of width 15 nm. Figure 1(b) shows that the extracted absorption spectrum from such PL spectra are insensitive to the excitation power within the range of the powers used in our measurements. The low-energy tails of the inferred absorption spectra [Fig. 1(c)] measured at different temperatures can be fitted with exponentials. When extrapolated, these fitted lines meet at a point showing the existence of the Urbach focus, as predicted by Eq. (1). The Urbach focus is usually interpreted as the zero-temperature band gap of the disorder-free virtual crystal and is hence larger than the measured gap ([12,16]; also see the Supplemental Material [22]). In Fig. 1(d), the Urbach slope parameter $\sigma(T)$ is plotted and fitted to Eq. (2). Figures 1(c) and 1(d) establish the Urbach rule in GaAs QWs.

The inferred $\hbar\Omega_p \approx 7$ meV is much smaller than the optical phonon energy (36 meV) in GaAs. This was unexpected but is easily understood in hindsight. The characteristic phonon energy determined from the temperature dependence of $\sigma(T)$ will be biased toward lower-energy phonons having the greatest change in the average number in the range of temperatures where the measurement is made (4–100 K) [6,29]. In this temperature range, the population of optical phonons (and its change) remains negligible. The characteristic energy of about 7 meV is attributed to one of the confined or interface acoustic phonon modes [30]. The phonon energy inferred from other QWs also had values between 5 and 7 meV.

Disorder and Urbach energy.—Figure 2 shows the Urbach edge measured in eight different GaAs/AlGaAs QWs in three different samples of varying quality grown in different reactors [31]. The zero of the energy has been

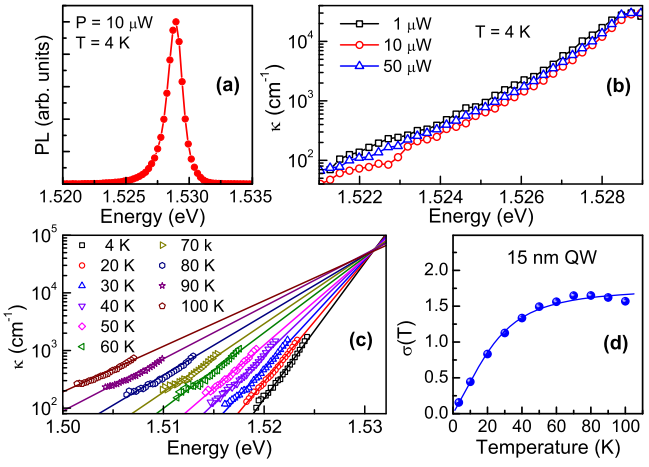

FIG. 1 (color online). (a) Photoluminescence spectrum of a 15 nm wide high-quality GaAs/AlGaAs QW. (b) The absorption spectra inferred from such PL spectra obtained at three different excitation powers. (c) Low-energy tails of the absorption spectra have a range where they are exponential. Furthermore, the curves measured at different temperatures extrapolate to a focus. (d) Temperature dependence of σ. Solid line is the fit to Eq. (2) with $\hbar\Omega_p = 6.8$ meV and the prefactor $\sigma_0[2k_B/\hbar\Omega_p] = 0.0443$.

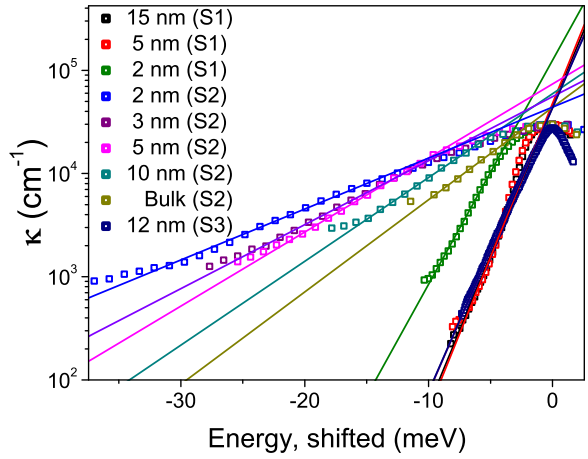

FIG. 2 (color online). Absorption spectra of different QW samples (inferred from PL in samples S1 and S2 and photoconductivity in sample S3) measured at 4 K. Sample S1 has MOVPE-grown high-quality three uncoupled QWs of widths 15, 5, and 2 nm. Sample S2 has metal-organic vapor phase epitaxy (MOVPE)-grown poor-quality four uncoupled QWs of widths 10, 5, 3, and 2 nm. Sample S3 is a high-quality molecular beam epitaxy (MBE)-grown sample with 50 uncoupled QWs each of thickness 12 nm.

TABLE I. Urbach energies in different QWs at 4 K.

Sample No.	S1	S1	S1	S3	S2	S2	S2	S2
QW width (nm)	15	5	2	12	10	5	3	2
Urbach energy (meV)	1.8	2.0	2.0	1.6	5.5	6.9	7.7	8.2

arbitrarily shifted in each case to facilitate a rough comparison of slopes. The actual values of the Urbach energy E_u [defined via a fit to the band tail of the absorption $\kappa \sim \exp(\hbar\omega/E_u)$] are listed in Table I. It is striking that while the low-temperature Urbach energy is roughly similar for the QW in high-quality samples (S1 and S3), the four QWs of the poor-quality sample (S2) show a strong dependence of the Urbach energy on well width, suggesting an influence of disorder, which is negligible in the high-quality samples. The disorder is clearly linked to interface roughness since the Urbach energy scales inversely as the well width. All of the four QWs are contained in the same sample, so the other disorder should be the same in all four QWs in sample S2. These observations suggest that there is a threshold for disorder, beyond which band tailing becomes significant.

An elegant way of characterizing interface roughness disorder is through the magnetic field dependence of the excitonic linewidth [32,33]. Following the more detailed discussion in the Supplemental Material [22], an important and perhaps unanticipated conclusion of Fig. 2 is that short-range-correlated (atomic scale) fluctuations of the interface do not much affect the Urbach energy, whereas mesoscopic scale correlations in the interface disorder greatly affect the Urbach energy [33–35].

Origin of band tail and zero-point phonons.—Figure 2 and Table I show that there is a nearly universal value of the Urbach energy ($E_U \sim 2$ meV) at low temperature for good quality GaAs/AlGaAs QWs, independent of their width. Short-range-correlated fluctuations present in good quality samples do not affect the Urbach energy. Thus, apart from these extrinsic disorder-related effects, there is also perhaps a fundamental intrinsic contribution to the Urbach energy. In typical GaAs QWs at low temperature, the electron-hole interaction is strong enough to form hydrogenic bound states (excitons), the polaronic coupling is weak, and topological disorder is low. The band tails are then believed to be caused by the modification of the excitonic resonance by intraband phonon scattering. In contrast with other arguments for band tails (e.g., Lifshitz theory [11]) there is no localization of excitons here.

This mechanism for band tailing was first proposed by Dow and Redfield using the following argument [6,36]. (i) In the presence of an electric field, the normally expected Franz-Keldysh broadening of the absorption edge transforms into an exponential tail when the excitonic effects are strong. Specifically, a part of the absorption spectrum below the excitonic resonance is described by

$$\kappa(\hbar\omega, F) \sim \exp[C(\hbar\omega - \hbar\Omega_0)/F], \tag{4}$$

where F is the average electric field experienced by excitons [14,29,36]. Equation (4) was inferred by making fits to the numerical solution of the single particle Schrödinger equation for hydrogenic excitons [36]. (ii) Polar-optical phonons are the source of an average nonzero electric field which the excitons experience, and proper averaging over the distribution (assumed Gaussian) of these internally generated electric fields should give the Urbach rule.

While this line of reasoning is now thought to be essentially correct, Dow and Redfield could not very cleanly arrive at Eqs. (1) and (2) from Eq. (4). Furthermore, Eq. (2) could only be inferred in its high-temperature limit, which is not relevant for our purpose here.

Within more microscopic descriptions [8,14,29], the band tails are understood as the usual lifetime broadening depicted by the imaginary part of the exciton self-energy, but where the non-Markovian character of the scattering processes gives an exponential rather than a Lorentzian broadening. While such microscopic theory does not seem to be directly amenable to experimental testing, one may examine a more rigorous version of the Dow-Redfield model itself [29]. Schäfer and Wegener make the interesting observation that the exciton-phonon interaction, under certain approximations, just manifests as an effective electric field for excitons [29]. The leading order term of the exciton-phonon self-energy in real space $\Sigma(\vec{r}, \vec{r}')$ (due to the long wavelength phonon modes under the approximation of locality and after tracing out the fermionic variable) turns out to have an explicit *linear* dependence on the position coordinate,

$$\Sigma(\vec{r}, \vec{r}') = \frac{e^2\Omega M}{8\pi\epsilon\epsilon^*\hbar}(1 + 2N_{\mathrm{LO}})|\vec{r}|\delta(\vec{r} - \vec{r}'). \tag{5}$$

Here, $\Sigma(\vec{r}, \vec{r}')$ is real. On dimensional grounds, one can simply read off an average internal electric field F_{in},

$$F_{\mathrm{in}} = \frac{e\Omega M}{8\pi\epsilon\epsilon^*\hbar}\coth\left(\frac{\hbar\Omega}{2k_BT}\right). \tag{6}$$

Here, M is the atomic mass, ϵ_0 the vacuum permittivity, and $1/\epsilon^* = 1/\epsilon(\infty) - 1/\epsilon(0)$, where $\epsilon(0)$ and $\epsilon(\infty)$ are the static and high-frequency dielectric constants, respectively. The $\coth(\hbar\Omega/2k_BT)$ term is just $(1 + 2N_{\mathrm{LO}})$, where N_{LO} is the phonon occupation factor. If one treats $\Sigma(\vec{r}, \vec{r}')$ as a perturbation on the exciton Hamiltonian, the arguments around Eq. (4) may still be used to explain the band tail, but now one has explicitly derived the full Urbach rule, Eqs. (1) and (2). Comparing these equations with Eqs. (4) and (6), the relationships between the slope parameter σ, the electric field F_{in}, and the constant C can be easily made: $\sigma/k_BT = C/F = 1/E_U$. E_U is the Urbach energy mentioned in Table I. Note that while the above expression is very approximate (even the first-order exciton-phonon virtual scattering is neglected), it is also attractive for this very reason; it does not depend on these details. Band tailing can still be looked upon as a result of broadening

due to the finite lifetime of hydrogenic bound states that are tunnel coupled with the continuum, just that the line shape is empirically determined from numerical calculations and not derived.

Also note that there is a finite Urbach energy even at zero temperature [in Eq. (6), $\coth(\hbar\Omega/2k_BT) \to 1$ as $T \to 0$]. This electric field $F_{\text{in}}(T=0)$ due to the zero-point vacuum fluctuations of the phonon fields gives a lower bound on the Urbach energy in an ideal disorder-free sample. All polar phonon modes, in particular the 36 meV polar-longitudinal-optical phonons, which were not visible in the low-temperature measurements [Fig. 1(c)], will also contribute to the zero-point electric field. These would, in fact, have the leading contribution.

Figure 3(a) shows the photoconductivity spectra in a 12 nm multi-QW sample measured at different values of in-plane electric field with the sample at 4 K. Photoconductivity is preferred over PL because the signal is actually enhanced on application of the electric field; the PL signal, on the other hand, rapidly quenches in even small electric fields. We see that there is a progressive broadening of the absorption tail in qualitative agreement with assertion (i). It is also worth comparing our Fig. 3(a) with the numerically generated Fig. 2(a) of Ref. [36].

This external electric field F_{ex} supplements the electric field of the phonons F_{in}, which, as discussed above, is always present and is in fact non-negligible even at zero temperature [7,37]. One may look at the electron-phonon self-energy [Eq. (5)] as an additional term in the effective exciton Hamiltonian, on the same footing as an externally applied electric field. Hence, the two may be combined into a single term by simple addition, and Eq. (4) may be modified to read

$$\kappa(\hbar\omega, F) \sim \exp[C(\hbar\omega - E_0)/(|F_{\text{in}}| + |F_{\text{ex}}|)]. \quad (7)$$

In general, F_{in} would also have contributions of electric fields due to charged impurities and dislocations, if any. Figure 3(b) is Fig. 3(a) replotted with, for clarity, only a few data points depicting the natural logarithm of the absorption coefficient $\ln[\kappa(F_{\text{ex}}, \hbar\omega)]$ for different values of the applied electric field. Good linear fits demonstrate the exponential behavior in accordance with Eq. (7) and thus experimentally validate the numerical calculations of Dow and Redfield [29,36]. Values of the slopes are mentioned in the caption of Fig. 3.

Furthermore, if this model is correct, the slope of the natural logarithm of the absorption coefficient (at a fixed electric field) as a function of photon energy $\hbar\omega$ in the band tail region [Eqs. (4) and (7)] must obey the relationship

$$\left[\frac{\partial}{\partial(\hbar\omega)}\ln\{\kappa(\hbar\omega, F)\}\right]_{F\text{fixed}}(|F_{\text{in}}| + |F_{\text{ex}}|) = C. \quad (8)$$

A check of the model is attempted in Fig. 3(c), where we have plotted the left-hand side of Eq. (8) for different values of the externally applied electric fields. F_{in} was kept as a parameter that was adjusted so that the deviation in the value of the left-hand side of Eq. (8) for different F_{ex} was minimized [Fig. 3(c)]. This also gives an estimate for the values of the constant C appearing in Eq. (6), $C = 2100$–$2150\ \text{kV cm}^{-1}\ \text{eV}^{-1}$, and the internal electric field due to the zero-point phonon modes, $F_{\text{in}} \approx 3\ \text{kV cm}^{-1}$ at zero temperature. This number is readily compared with expectation for Fröhlich coupling of 36 meV (417 K) LO phonons in GaAs [38] since the parameters for GaAs are well known. One finds that the theoretical estimate based on Eq. (6) gives $F_{\text{in}}(T=0)$ is $2.97\ \text{kV cm}^{-1}$ [38], in fortuitously good agreement with the experimental value.

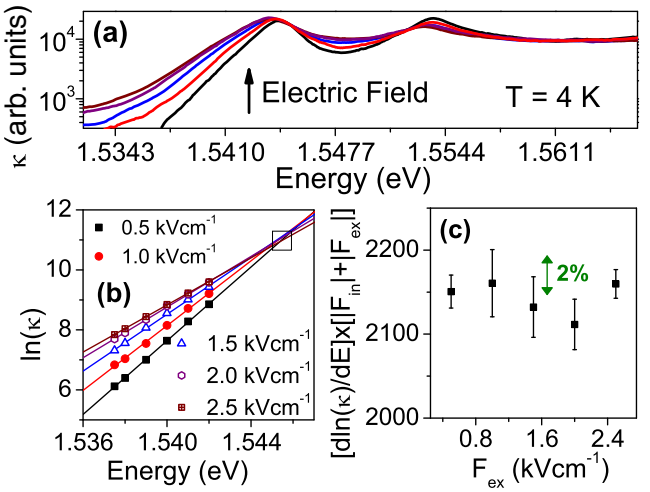

FIG. 3 (color online). (a) Electroabsorption spectra at different F_{ex} electric fields (inferred from photoconductivity) at 4 K. The values of F_{ex} are the same as in (b). Each of the curves has been arbitrarily scaled such that the absorption coefficients have the same value at 1.560 eV. (b) Natural logarithm of the low-energy part of the heavy-hole excitonic resonance plotted versus the photon energy for different values of F_{ex}. The linear fits to the curves yield the slope to be $614 \pm 6, 540 \pm 10, 473 \pm 8, 422 \pm 6$, and $393 \pm 3\ \text{eV}^{-1}$ for $F_{\text{ex}} = 0.5, 1, 1.5, 2, 2.5\ \text{kV cm}^{-1}$, respectively. The location of the Urbach focus E_0 [see Eq. (4)] is depicted by a square. (c) The slope of the logarithm of the absorption tail times the total electric field ($|F_{\text{in}}| + |F_{\text{ex}}|$) for different values of the external field F_{ex} is approximately constant if $F_{\text{in}} = 3\ \text{kV cm}^{-1}$. The error bars are the errors in the fits in (b).

Conclusions.—Let us recall that the argument presented here is very similar to the heuristic calculation by Welton for the Lamb shift [39], where also the energy shift between the $2^2P_{1/2}$ and $2^2S_{1/2}$ levels could be interpreted as being due to the zero-point electric fields. While the Lamb shift is the splitting of the energy levels, the Urbach tail is a complementary effect affecting the linewidth due to the tunnel coupling of the bound states with the continuum in the presence of an electric field. Since we are dealing with hydrogenic excitons, this atomic physics analogy is rather close and the finite temperature Urbach rule may thus (roughly) be looked upon as the broadening caused by the blackbody shift first discussed by Auluck and Kothari [40].

We should also mention that apart from the intrinsic interest in this problem, understanding the origin and the

fundamental limits on the band tails is also relevant in a wide range of contexts—from the feasibility of laser cooling of semiconductors by anti-Stokes PL [41], to providing allowed states for nonradiative Auger processes [42], and at a somewhat more fundamental level, the limitations in treating the semiconductor ground state at par with the quantum electrodynamic vacuum in the context of phenomena like virtual photoconductivity and the Unruh effect [43].

On a parting note, let us quote Jaffe [3], "Certainly there is no experimental evidence for the reality of zero-point energies in quantum field theory (without gravity)." While we also subscribe to this view in the context of vacuum quantum field theories, unlike, e.g., the electromagnetic fields, phonons emerge out of *real* harmonic oscillators and we have shown that they have measurable consequences even at zero temperature.

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[22] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.114.047402, which includes Refs. [23–28], for details of the samples, physical interpretation of the Urbach focus, and the effect of the quantum well interface roughness on the Urbach energy. The effect of the zero-point electric field on the exciton resonance and the relative roles of optical and acoustic phonons are also discussed.

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