Supplemental Material

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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I. SAMPLE INFORMATION

Three samples, containing GaAs/AlGaAs quantum wells (QWs), were used in this study. They labeled $S1$, $S2$, and $S3$ respectively.

Sample $S1$ was grown by metal-organic vapor phase epitaxy (MOVPE) and is among the highest quality samples ever reported by MOVPE growth [23]. The extraordinarily high quality was achieved through epitaxial growth on slightly misoriented substrates $\leq 0.6^o$ off-(100) GaAs substrates and a high V/III ratio for AlGaAs growth. The sample has three active quantum wells of widths 2, 5, 15 nm respectively and the $Al_{0.30}Ga_{0.70}As$ barriers. The three QWs can be assumed to be uncoupled. More details can be found in Ref. 23.

The sample $S2$ was again grown by MOVPE and was of relatively very poor quality. It had four independent GaAs QWs of thicknesses 2, 3, 5, and 10 nm respectively sandwiched between $Al_{0.3}Ga_{0.7}As$ layers. The separation between the QWs was 130 nm. These are the same samples as those used in Ref. 33.

A molecular beam epitaxy (MBE)-grown high quality sample, $S3$, was used for photoconductivity measurements. It had a 20−period 12 nm $Al_{0.2}Ga_{0.8}As$ / 10 nm-GaAs MQW-structure with 200 nm $Al_{0.2}Ga_{0.8}As$ cladding layers, followed by 200 nm low temperature-GaAs were grown by MBE and processed into long, 10 or 20 μm wide mesas and provided with lateral Au-contacts, using conventional photolithographic methods. Sandwiching the QWs between low temperature-GaAs is an important innovation that allows for a uniform *lateral* electric field to develop. Further details of the device are given in Ref. 24.

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II. THE URBACH FOCUS AND HOW IT ESTABLISHES THE ESSENTIAL ROLE OF PHONONS

The appearance of the Urbach focus in Fig. 1(c) [of the main paper] is spectacular and may seem surprising. But a moment's reflection tells us that the fact that there is a focus is an independent proof of the essential role of phonons. From Eqs. (1) and (2) (main text), the temperature-dependent Urbach edge fits the empirical form

$$\kappa(\hbar\omega, T) = \kappa_0 \exp\left[\frac{2\sigma_0}{\hbar\Omega_p}\left(\frac{\hbar\Omega_0 - \hbar\omega}{\coth(\frac{\hbar\Omega_p}{2k_BT})}\right)\right], \omega < \Omega_0 \quad (1)$$

Though we have presented an argument for the coth dependence of the Urbach slope in the main text, in Eq. (1), one expects that instead of the temperature-independent $\hbar\Omega_0$, there should be a temperature-dependent function that displaces each of the exponentials parallel to one another (on a linear-x and log-y scale) as the absorption edge shifts with temperature. Why then do the exponential fits of the absorption tails at different temperature all meet at a temperature-independent focal point despite the strong shift in the energy gap with temperature?

There have been a variety of (semi-)empirical relationships proposed to capture the temperature dependence of the energy gap $E_g(T)$. For our purpose we consider the one proposed by Cardona and coworkers [25],

$$E_g(T) = E_g(0) - C_2 \coth\left(\frac{\hbar\Omega_p}{2k_BT}\right) \quad (2)$$

$E_g(0)$ is the gap at zero temperature for the unperturbed crystal and $E_g(0) - C_2$ is the experimentally measured value of the gap at zero temperature. C_2 is the zero-temperature energy renomalization and $\hbar\Omega_p$ the characteristic phonon energy. But from above Eq. (1) and (2) [Supplement], we see that even after replacing $\hbar\Omega_0$ with $E_g(T)$, the functional form of Eq. (1) is unaltered. Furthermore, at absolute zero temperature the Urbach focus energy $\hbar\Omega_0$ corresponds to $E_g(0)$, without the energy renormalization C_2. Thus $\hbar\Omega_0$ is the energy gap of

the hypothetical 'bare' crystal with the electron-phonon interaction switched off. The magnitude of the zero-temperature bandgap renormalization due to zero-point phonons can be substantial [26]. The Urbach slope may roughly looked upon as the complementary effect, arising from the level broadening caused by the same electron-phonon interaction that causes the renormalization of the critical point energies. We should mention that the above argument for the temperature-independence of the Urbach focus energy has previously also been discussed by Johnson [27] and Cody [12].

III. NATURE OF THE INTERFACE DISORDER AND THE BEHAVIOR OF THE EXCITON LINEWIDTHS IN MAGNETIC FIELD

It is expected that the structural disorder resulting from interface roughness in QWs along with various compositional and topological imperfections will contribute to the Urbach tail at zero temperature, adding to the contribution from phonon scattering. Thus, Fig. 2 (main text) and Table I (main text), where the Urbach tail of different samples is discussed, is surprising. For all good quality QWs (QWs in $S1$ and $S3$), the low temperature Urbach slope is similar ($\sim$ 2 meV). The excitonic linewidth on the other hand scales inversely with the width of QWs due to the interface roughness disorder. But for QWs in $S2$, there is a strong dependence on the Urbach slope on the interface disorder.

To understand this a little better, we have plotted the magnetic field dependence of the exciton linewidth for QWs of comparable width from two different samples, one of good quality ($S3$) in [Fig. 1(a) Supplement] and one highly disordered ($S2$), in [Fig. 1(b) Supplement]. A contrary behavior of the magnetic field-dependent change of the exciton linewidth is observed in these two samples which may be understood as follows [33]. A magnetic field applied perpendicular to the plane of the QW adds a harmonic potential to the existing Coulombic potential of the excitons. This extra potential acts to reduce the exciton size. In the high magnetic field limit, the exciton size would approach the magnetic length, l_B, which would scale with the magnetic field B as $l_B \sim 1/\sqrt{B}$. This reduction of the effective exciton radius in the plane of the QW reduces the interface area which an exciton samples and consequently the interface disorder averaging. The excitonic linewidth would increase with the magnetic field in high quality QWs where the correlation lengths of the interface disorder (which inevitably exists due to AlAs forming only 30% of the interface in usual $Ga_{0.7}Al_{0.3}As$/GaAs heterostructures) are small and approach white-noise-like character [32]. Samples with an additional disorder arising from imperfect reactor conditions during growth, on the other hand, may have large correlation lengths for the interface disorder potential arising from mesoscopic inhomogeneity. In this case, shrinking of the exciton will lead to it effectively sampling a smaller part of this larger-scale inhomogeneity. As a consequence, the linewidth will decrease with increase in magnetic field [33].

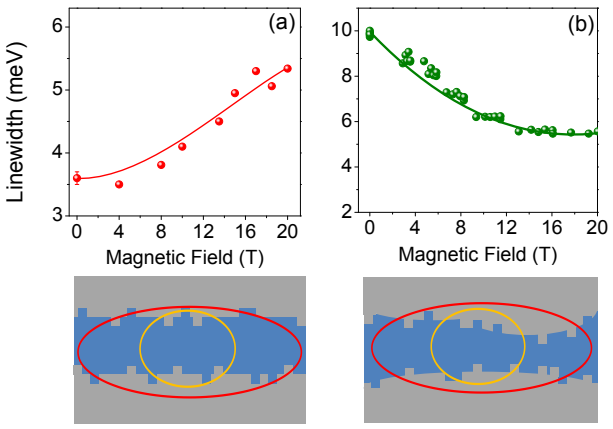

FIG. 1: [Supplement] Magnetic field dependence of linewidth of the heavy-hole exciton resonance for high quality (a) and poor quality (b) QWs show contrary behavior. Data in (b) is adapted from a figure in Ref. 33. The linewidth in (a) is inferred from photoconductivity data. The corresponding schematics below show the nature of interface disorder, only atomic-scale composition fluctuations in (a) and an additional mesoscopic inhomogeniety in (b). In presence of magnetic field, the exciton shrinks and consequently samples over a smaller region of the interface.

Thus Fig. 1 [Supplement] clearly distinguishes between the nature of disorder in high and low quality QWs. An important and perhaps unanticipated conclusion of Fig. 1 [Supplement] 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 do. This empirical observation is also qualitatively seen within the (rather simplistic) one-dimensional Anderson model [34]; Urbach energy is affected by the correlation lengths of the onsite disorder in a similar fashion [Fig. 1 of Ref. 27]. The same argument can also be used for semiconductor alloys where one would again expect that the Urbach edge in systems like $Al_xGa_{1-x}As$ will not be much affected whereas one will observe a large Urbach energy for mismatched systems like $In_xGa_{1-x}N$, $In_xGa_{1-x}As_{1-y}N_y$ or $InAs_ySb_{1-y}$, not just because of the larger onsite disorder but also due to their propensity for clustering. This is also observed [35].

IV. SHOULDN'T THE ZERO-POINT ELECTRIC FIELD IONIZE THE EXCITONS IN BULK GaAs?

The inferred electric field of 3 kVcm^{-1} is already close to the Coulombic field of the bound state $F_{coul} \approx E_B/(qa_B)$ for bulk GaAs. Why then does the optical absorption spectrum in high-purity bulk GaAs show clear excitonic resonances [28] up to around 2 00K (Fig. 2

[Supplement])? Unfortunately it is not very easy to compute the precise effect of the electric field on the linewidth because of the non-pertubative nature of the calculation.

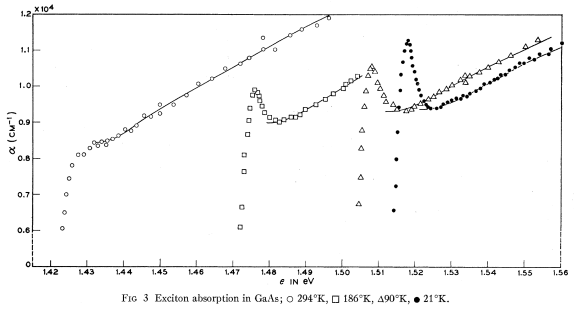

FIG. 2: [Supplement] Figure is reproduced from Ref 28.

But experimentally, we found the exciton resonance (in 12 nm QWs) to be surprisingly robust to an in-plane electric field. Fig. 3 [Supplement] shows electro-absorption (photoconductivity) data on 12 nm multi-QW GaAs sample. Even up to an externally applied field (over and above the internal field) of 9 kVcm^{-1}, the resonance is only marginally broadened.

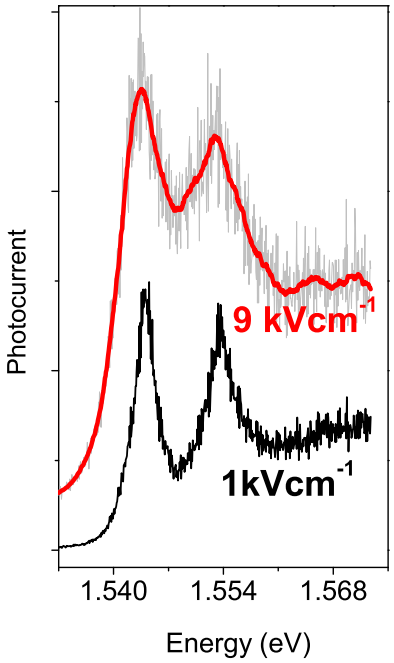

FIG. 3: [Supplement] Photoconductivity measurements at 4 K show that exciton resonance is only marginally broadened at high electric field. Photocurrent at high electric field was very noisy (perhaps due to the impact ionization) and hence had to be smoothened.

V. RELATIVE ROLES OF ACOUSTIC AND OPTICAL PHONONS IN THE URBACH TAIL

While the bulk of the paper is focussed on around the electric field from optical phonons, Fig. 1 (d) [main paper] pointed to the role of acoustic phonons. Perhaps this needs to be clarified.

At zero temperature, perturbation caused by the zero-point electric fields of all phonons affects the Urbach tails since the occupation factors do not come into the picture. In this sum of all contributions, the overwhelmingly dominant contribution is from the LO phonons' Frohlich coupling to band states. The zero-point acoustic phonons' contribution to the electric field (from piezo-electric coupling) is in comparison negligible. Hence the experimentally inferred electric field at 4 K indeed matches well with the field estimated from the optical phonons alone. This is the main conclusion of the work.

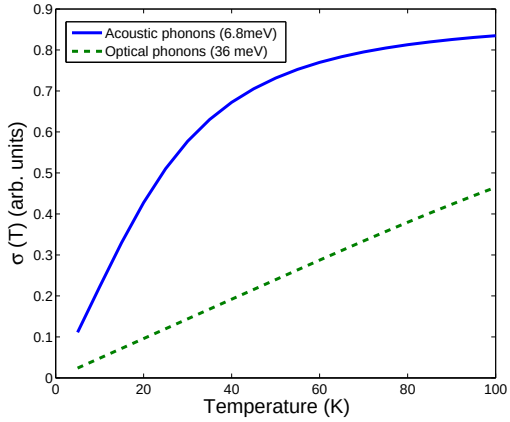

FIG. 4: [Supplement] Plot of $[2k_BT/\hbar\Omega_p]\tanh[\hbar\Omega_p/2k_BT]$ with $\hbar\Omega_p = 6.8$ meV (solid line) and 36 meV (dashed line) corresponding to acoustic and optical phonons respectively. The acoustic phonon contribution is selectively picked up in the temperature-dependent data. Note that due to the different strengths of the pre-factor σ_0, the optical phonon contribution will be further reduced, since $\sigma_0[2k_B/\hbar\Omega_p]$ scales inversely as the electric field.

But in Fig. 1(d) [main paper], the Urbach slope parameter $\sigma(T)$ in the temperature dependence of the absorption coefficient $\kappa(\hbar\omega, T)$

$$\kappa(\hbar\omega, T) = \kappa_0 \exp[-\sigma(T)(\hbar\Omega_0 - \hbar\omega)/k_BT],\ \omega < \Omega_0, \quad (3)$$

when fitted to the function

$$\sigma(T) = \sigma_0[2k_BT/\hbar\Omega_p]\tanh[\hbar\Omega_p/2k_BT]. \quad (4)$$

revealed a much smaller effective phonon energy. This, somewhat non-intuitive function, has the property that $\sigma \to 0$, as $T \to 0$. *Hence $\sigma(T)$ selectively picks out only the temperature-dependent contribution to the Urbach slope ignoring the zero-temperature background.* Moreover, $\sigma(T)$ is a monotonically increasing function with

$0 \leq \sigma(T) \leq \sigma_0$. $\sigma(T)$ scales as the phonon occupation but inversely as the strength of the effective phonons' electric field. In the range of temperatures studied (up to 100 K), number of optical phonons continues to be small, whereas acoustic phonons occupations are strongly temperature-dependent. The combination of these factors results in temperature dependence of $\sigma(T)$ to fit the acoustic phonon energies, even though the coupling is much weaker compared to optical phonons.

VI. REFERENCES

The references used for Supplemental Material are reproduced here from the main text. The numbers are the same as those in the main text.

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