Fermi-Edge Singularities In Al\textsubscript{x}Ga\textsubscript{1-x}As Quantum Wells: Extrinsic Versus Many-Body Scattering Processes

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A Fano resonance mechanism is evidenced to control the formation of optical Fermi-edge singularities in multi-subband systems such as remotely doped Al\textsubscript{x}Ga\textsubscript{1-x}As heterostructures. Using Fano parameters, we probe the physical nature of the interaction between Fermi sea electrons and empty conduction subbands. We show that processes of extrinsic origin like alloy-disorder prevail easily at 2D over multiple diffusions from charged valence holes expected by many-body scenarios.

Drastic manifestations are expected from the interaction of a magnetic or charged impurity with a Fermi sea of electrons \cite{1}. Since the successful explanation of the divergent X-ray absorption edges of simple metals \cite{2} by Mahan \cite{3} and Nozières et al. \cite{4} thirty years ago, a Fermi sea of electrons under optical excitation has been recognized as a model system to study these issues experimentally. Indeed a many-body electronic state can develop in presence of the positively charged electronic vacancy (or core hole) involved in optical processes, which multiply scatters conduction electrons throughout and along the Fermi surface. This induces divergences at the Fermi-edge of optical spectra, the so-called Fermi-edge singularities (FES). FES are by essence highly sensitive to phase space restrictions such as hole localization and reduced dimensionalities: they cannot form without either strong hole localization or the dimensionality being unity \cite{5}.

Electron systems embedded in semiconductor heterostructures have therefore attracted much attention to test these predictions \cite{6}. FES were first observed by Skolnick et al. \cite{7} in the low-temperature Photoluminescence (PL) spectrum of a remotely doped In\textsubscript{0.47}Ga\textsubscript{0.53}As/InP quantum well (QW). Emphasis was put on the anomalous temperature dependence of the singularity and on the localization of valence holes by alloy fluctuations. More recently, experiments by Chen et al. \cite{8} in In\textsubscript{0.15}Ga\textsubscript{0.85}As/AlGaAs QWs with weaker hole localization also put forward the tunability of FES, by bringing the first empty QW subband into resonance with the Fermi level. It was then proposed that empty conduction subbands could act as additional scattering channels for Coulomb processes, in qualitative agreement with many-body multi-subband numerical calculations in the infinite hole-mass approximation \cite{9}.

Although this interpretation is widely referred to, no experiments have been undertaken to test many-body schemes beyond qualitative agreement. While attempting to do so with Al\textsubscript{2}Ga\textsubscript{1-x}As QWs, it appeared to us that non-coulombian intersubband scatterings can induce FES as well. The scope of this Letter is to describe this issue quantitatively. Our experiments demonstrate that extrinsic processes like alloy-disorder can prevail easily over multiple Coulomb scatterings of Fermi sea electrons predicted in the framework of many-body scenarios.

The paper is organized as follows. We show that the formation of FES in Al\textsubscript{x}Ga\textsubscript{1-x}As QWs is governed by a Fano resonance mechanism \cite{10} between Fermi-sea electrons and discrete excitonic transitions associated with empty conduction subbands. This model is first validated by the existence of scaling properties of PL spectra when FES are enhanced by reduced intersubband spacings. It is then used to gain insight into the microscopic nature of conduction Intersubband Couplings (ICs) at work. Indeed, the stronger the ICs, the more divergent the FES, so that one can probe directly in experiments the efficiency of extrinsic scattering processes such as alloy-disorder, remote-doping disorder or artificial ICs in lateral superlattices \cite{11}. Experimental results fall in close agreement with microscopic Fano calculations. We demonstrate that alloy-disorder is the dominant contribution to observed FES in Al\textsubscript{x}Ga\textsubscript{1-x}As QWs.

Samples investigated in this work are remotely doped Al\textsubscript{1-x}Ga\textsubscript{x}As/Al\textsubscript{0.33}Ga\textsubscript{0.67}As QWs grown on GaAs substrates by molecular beam epitaxy. They are all of same thickness L\textsubscript{z}=25nm, spacers (in the range 5-8 nm) and sheet density n\textsubscript{s} \approx 8.10\textsuperscript{11}cm\textsuperscript{-2}, but vary in their aluminium content x (2.3% \leq x \leq 7.1%). Confined 2D-levels are represented in fig. 1a. The asymmetry of the QW potential originates in the dipole formed by remote ionized dopants (z > 0) and the Degenerate Electron Gas (DEG) partially filling the first conduction subband E\textsubscript{1}. This confers a PL activity to the two first subbands E\textsubscript{1} and E\textsubscript{2} with photocreated valence holes localized on potential fluctuations at the top of the heavy-hole HH\textsubscript{1} subband (fig. 1b). We perform PL spectroscopy at 1.8K. Samples are optically excited by a Ti:Sa laser at 1.7 eV, with a \approx 1 W.cm\textsuperscript{-2} density so as to avoid inhomogeneous heating of the DEG within the 40\mu m\textsuperscript{2} laser spot. The DEG PL (E\textsubscript{2}HH\textsubscript{1}) extends from lowest wavevector transitions at E\textsubscript{g} up to Fermi wave-vector transitions at E\textsubscript{F}+E\textsubscript{F} (E\textsubscript{F} \approx 25meV). Without any influence of E\textsubscript{2}HH\textsubscript{1}, its oscillator strength decreases monotonously with energy, because of hole localization and indirect optical processes \cite{12} (see spectrum * in fig.1c). The PL of...
the empty QW subband E₂ exhibits a dominant excitonic feature E₂ₓ of high oscillator strength and discrete character, visible either by thermal activation above E₀+Eₓ in PL, or in PL Excitation spectra. We define ∆=E₂ₓ - E₀-Eₓ. Variations of ∆ are achieved along a given sample by use of the flux gradients of effusion cells in the epitaxy chamber. We stop the wafer rotation during the spacer layer growth between dopants and the QW. The thinner the spacer, the stronger the electric field at the QW interface. This tunes the E₂-E₁ separation, while Eₓ hardly changes under illumination [13]. As seen from PL spectra of sample A (x=7.1%) in fig.1c, a FES forms and develops [3] when ∆ is decreased to zero [4].

![Diagram](image)

**FIG. 1.** a) Schematic 2D-confined conduction and valence envelope-functions for E₁, E₂ and HH₁. Remote δ-doping is achieved on the z > 0 side. b) In-plane k-space band-structure showing the PL recombination of Fermi-sea electrons with photocreated holes at T=0K. Localized HH₁ states are sketched by a k-space extended wave-function Ψ_{loc}. c) 1.8 K PL spectra of sample A recorded for various ∆ values.

To interpret these data, we consider the Fano resonance model depicted in fig.2a. E₂ₓ is taken as a discrete level coupled with a matrix-element W to the continuum of E₁HH₁ electron-hole pairs. By assuming an infinite hole mass, all physical parameters simply refer to the conduction band : W equals to the IC between E₁ and E₂, and the E₁HH₁ continuum is populated by the E₁ Fermi-Dirac electrons. We take a parabolic dispersion and a constant PL oscillator strength for E₁. Fano [10] gave an analytical description of the spreading of a discrete level coupled to a continuum. Optical FES occur (see fig.2a) due to the partial filling of E₁HH₁ near E₂ₓ, and get more divergent as ∆ is reduced. A qualitative agreement with experiments [3] is thus obtained, without any particular assumption on the physical nature of W.

The validation of the Fano resonance model comes from the scaling property evidenced in fig.2a that all PL spectra for various ∆ should display a common envelope lineshape when plotted with E₂ₓ as the origin of energies. The comparison with experimental data is not straightforward because E₁-E₂ varies experimentally, while the E₁HH₁ PL oscillator strength intrinsically depends on energy [12]. We compensate for these nominal E₁HH₁ PL variations by dividing all data of fig.1c by the E₁HH₁ spectrum with ∆ → ∞ (⋆ in fig.1c). Data are then plotted in fig. 2b with E₂ₓ as the origin of energies. Remarkably, all spectra superpose on each other in the range of populated electron states [13]. No clear transition exists from convergent to divergent Fermi edges in fig 2b. This means that FES only appear in raw PL data when the intrinsic PL decay of E₁HH₁ at Eₓ gets balanced by the positive slope of the Fano envelope for small ∆ values.

![Diagram](image)

**FIG. 2.** a) Fano resonance model, where E₂ₓ is seen as a discrete level in the E₁HH₁ continuum (top). In the coupled system (bottom), a FES appears, due to the partial filling of E₁ (dark grey). 2Γ is the resonance width. b) Points : data of fig.1c, normalized by the PL spectrum with ∆ → ∞ (⋆ in 1c) and plotted with E₂ₓ as the origin of energies. Lines : fits using a Fano profile (q=12.2 and Γ=0.60 meV) and Fermi-Dirac electrons (T=9.0±1K). c) FES “enhancement factor” (points, from ref.[8]) in an In₀.15Ga₀.85As QW, fitted (line) by a Fano profile with Γ=0.66 meV and q=6.6.

In order to get a quantitative analysis of the envelope profile, we assume a statistical disorder property for the interaction W : < W >= 0 and < W² ≠ 0. This accounts for alloy disorder, which is later evidenced to dominate FES in sample A. The normalized PL intensity can then be derived analytically from ref. [10] :

\[ I(E) = f(E) \cdot \frac{q^2 + (E - E_{2x})^2/\Gamma^2}{1 + (E - E_{2x})^2/\Gamma^2} \]

where E is the PL energy : f(E) the occupation number in the E₁ subband ; Γ is the observed half width
at half maximum of the resonance, only related to the statistical squared interaction average $<\mathcal{W}^2>$ and the density of states $D$ of the $E_1$ conduction subband by $\Gamma = \pi <\mathcal{W}^2> D$; $q^2$ is the experimental oscillator strength of the excitonic resonance relative to the continuum, inversely proportional to $<\mathcal{W}^2>$ and $D^2$. It depends also strongly on wavefunction overlaps through the ratio of the oscillator strengths of $E_2HH_1$ and $E_1HH_1$. $q^2$ is therefore quite sensitive to small geometrical fluctuations from sample to sample.

Our data are nicely fitted by this model (fig. 2b), with Fano parameters $\Gamma = 0.60$ meV and $q = 12.2$. All fits use Fermi-Dirac distributions of electrons with an effective temperature $T = 9.0 \pm 1$K. This shows that electrons are indeed thermalized, though not with the lattice at 1.8 K due to the incomplete and slow relaxation of photogenerated electrons above the $E_2$ subband edge. The consistency of small $\Delta$ data with our fit indicates that the $E_{2x}$ resonance remains weakly populated enough to stay in the “atom-like” regime \[10\]. Also, taking $f(E) = 1$ in the Fano lineshape formula, we can check that it fits the FES “enhancement factor” (FES PL intensity divided by its $\exp - \infty$ value) measured by Chen et al. in In$_{0.15}$Ga$_{0.85}$As QW structures \[8\]. Their data are indeed nicely reproduced with $\Gamma = 0.66$ meV and $q = 6.6$ (fig.2c).

Before quitting the phenomenological level, we focus on the temperature dependence of FES in systems with explicit intersubband interaction. Observed thermal quenchings are expected from many-body theories \[7\], where an actual occupation number discontinuity is required to enhance multiple Coulomb scatterings at $E_F$. Here or in ref. \[8\], the FES disappears, only because the PL relative minimum between $E_g + E_F$ and $E_{2x}$ vanishes with raised temperatures \[12\].

Up to this point, we successfully assessed the model with respect to $\Delta$ and temperature variations. We now analyse the microscopic origin of the Fano parameter $\Gamma$. On the theoretical side, $\Gamma$ only depends on the strength $W$ of intersubband couplings and can be computed for a given microscopic process. Experimentally, the Fano model predicts that the stronger $\Gamma$, the more divergent the Fermi-edges at fixed $\Delta$. By designing appropriate samples, one can thus test: i) whether extrinsic scattering processes like alloy-disorder play an effective role on the formation of FES; ii) if experimental variations of $\Gamma$ correlate with microscopic calculations.

We display in fig.3a the PL spectra of remotely doped quantum wells C, B and A of with QW aluminium content $x$ equal to 0.023, 0.044 and 0.071 respectively, taken at fixed $\Delta = 4.3 \pm 0.1$ meV. As seen from the global PL lineshapes, the localization of photogenerated valence holes remains constant, dominated by the roughness at the QW interface (see fig. 1) rather than by random alloy potential fluctuations. Enhancement of many-body processes due to hole localization \[11\] can therefore be excluded. Nevertheless, FES get more pronounced with increased alloy concentration $x$, while the excitonic resonance broadens. Fitted $\Gamma$ Fano parameters \[20\] linearly increase with $x$ within experimental uncertainty (fig.3b).

This explains the quadratic enhancement of FES visible from fig.3a when $\Gamma$ is linearly increased in the regime where $\Delta / \Gamma \gg 1$. It also demonstrates that alloy disorder is the dominant contribution to the IC parameter $W$.

![Graph showing FES enhancement](image)

**FIG. 3.** a) 1.8K PL of samples C, B and A taken at $\Delta = 4.3 \pm 0.1$ meV. The Fermi-edge regions are zoomed in. b) Fitted Fano parameters $\Gamma$ as a function of alloy content. Microscopic calculation are given without (dotted line) and with (full line) a residual disorder $\Gamma_r = 0.2$ meV (see text).

To quantify this, we calculate $\Gamma$ microscopically in the infinite-hole-mass approximation \[19\] :

$$\Gamma_{alloy} = x(1 - x)m\bar{\Omega}_0\delta V^2 / L_z h^2$$

This applies for a square quantum well of width $L_z$, a conduction electron mass $m$, $\delta V$ being the conduction band offset between pure AlAs and pure GaAs, and $\bar{\Omega}_0$ the crystal cell volume. With $L_z = 18$ nm (representative of the confinement length of $E_1$ and $E_2$ wave-functions), $m = 0.07 m_0$ and $x = 71\%$, we obtain $\Gamma = 0.61$ meV. This quantitative agreement is striking for such a simple model. The fit from fig.3b is achieved by introducing a residual scattering $\Gamma_r = 0.2$ meV in the pure GaAs limit and thus taking a Fano parameter $\Gamma^2 = \Gamma^2_r + \Gamma^2_{alloy}$. We can also estimate $\Gamma_{alloy} \approx 0.5$ meV for the In$_{0.15}$Ga$_{0.85}$As QWs of ref. \[8\], in close agreement with either our fit (fig.2c) or the empirical two-level coupling (0.6 meV) measured by Chen et al.

We now focus on ICs induced by random positioning of ionized dopants. Strictly speaking, $W$ now depends on the occupation of $E_1$ states. We nonetheless assume $W$ equal to its value for Fermi wave-vector ($k_F$) electrons. $\Gamma$ can then be computed \[10\] and gets proportional to $-2k_F.\bar{\Omega}_0$ in the limit of a thick spacer layer $\bar{\Omega}_0$, with a prefactor of $\approx 60$ meV for a square QW structure of width $L_z = 20$ nm and a sheet density $n_s \approx 10^{12}$ cm$^{-2}$. Due to high doping ($k_F \geq 10^8$ cm$^{-1}$), this predicts a poor efficiency even for very shallow spacers. This is already visible from fig. 2b where indirect optical processes \[13\]...
- of same physical origin - only affect small wave-vector
conduction states. Also, we measured no increase of $\Gamma$ in
a sample similar to C but with a 2.5 nm spacer.

We finally mention the case of FES in tilted lateral
superlattices, where artificial intersubband couplings are
created by a non separable 1D periodic confinement be-
tween the growth ($z$) and an in-plane ($x$) direction. This
has been shown to promote optical FES [11]. In fact, our
data also fit to a Fano scheme [11] with parameter $\Gamma=3.2$
meV. The larger strength of ICs compared to 2D QWs
explains the formation of pronounced FES even for large
$\Delta$ parameters [11]. The experimental $\Gamma$ value matches a
microscopic calculation, using a typical value of 30 meV
for the peak-to-peak lateral confinement amplitude [22].

Microscopic calculations of Fano parameters fall there-
fore in quantitative agreement with our experiments,
where ICs have been varied in physical nature and over
one decade in amplitude. This demonstrates that non-
coulombian scattering processes can efficiently control
the formation of FES in multi-subband semiconductor
structures, in the simple picture of a band-structure par-
tative analogy was underlined in early many-body inter-
actions issues [13] since disorder gets generally enhanced,
and thus

$\Gamma_{\text{alloy}} = 1.0$ meV, this criterium discards random alloy-
permation $\Delta$ spectra. This is more relevant, since for instance the
measured directly from the E
continuum (visible in fig.2b where fits sys-
emis a low-energy peak of the DEG PL, all the more pronounced with thinner
spacer layers. This signs the increased efficiency of indi-
rect optical processes for less remote dopants, while they
are taken constant in the normalization procedure.

A strong population of E
atom-like excitonic character, both in PL and PL excitation spectra :
T.A. Fisher, P.E. Simmonds, M.S. Skolnick, A.D. Martin
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(1989) and references therein.

The same happens when $\Delta$ is reduced at fixed tempera-
ture (see e.g. $\Delta = 2.9$ and 2.25 meV in fig.1c and 2b).

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ductor structures”, Les Editions de Physique (1988).

In fig.3b, $\Gamma$ is fitted from $\Delta \to 0$ PL data, rather than
measured directly from the E$_{2X}$ resonance width in large
$\Delta$ spectra. This is more relevant, since for instance the high-energy side of E$_{2X}$ is affected by the luminescence of the E$_2$HH$_1$ continuum (visible in fig.2b where fits sys-
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This enables experiments similar to ref. [8], where Chen
et al. used an in-plane magnetic field to tune $\Delta$.

Reported $\Delta$ values stem from the fit in fig.2b (see text).
They fall in close agreement with magneto-optical PL
data, which provide an actual measurement of $E_{\text{g}} + E_F$.

Small discrepancies occur around the low-energy peak
of the IP signal, rather than

A strong population of E$_{2X}$ would alter its atom-like exi-
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