Electron-electron scattering in far-infrared quantum cascade lasers

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A large depolarization shift indicates a strong nonequilibrium intersubband electron-electron scattering (Γ) in tunneling structure. For a far-infrared subband separation Δ~10 meV, the rate Γ scales with the upper-subband occupation and is never significantly reduced by screening. Despite this nonradiative decay a finite population inversion can be maintained. Finally, the applied bias changes the wave-function symmetry so as to cause a dramatic variation of the electron-electron scattering rate.

In the quantum cascade laser 1 a nonequilibrium current injection exclusively into the upper subband of a multiple-level tunneling structure provides a finite intersubband population inversion and actual lasing (at midinfrared frequency ω~300 meV). This seminal achievement culminates a search begun in 1971 with the proposal by Kazarinov and Suris 2 and soon after the groundbreaking work by Esaki and Tsu. 3 There is also a considerable interest in the far-infrared (or Terahertz) regime and we mention in particular the observation 4 of spontaneous intersubband emission in superlattices excited by a current flow. Adapting the quantum-cascade-laser design 1 we presently investigate the population inversion in a tunneling structure with far-infrared subband separation, Δ~11 meV.

The prospect of such far-infrared stimulated emission is raised by the small intersubband decay, 1/τ~0.03 meV, observed 5 at temperatures T=50 K and at weak optical pumping. 6 This small decay is possible because the optical-phonon frequency (ΩLO~36 meV in GaAs) exceeds the subband separation, Δ~11 meV, and optical-phonon emission processes are inhibited 5 at temperatures below the activation energy ΩLO, Δ=25 meV.

We find, however, that the current injection results in a strong nonequilibrium electron-electron intersubband scattering (Γ) which we evaluate for a complete upper-subband occupation below the emitter chemical potential. This scattering Γ is never significantly reduced by screening and, unlike the near-equilibrium electron-electron scattering, 7 is not inhibited by the Pauli exclusion (as we can assume population inversion.) In this letter we (1) identify a simple scaling of the nonequilibrium electron-electron scattering rate (Γ) with the upper-subband occupation, (2) predict a very strong intersubband decay ~2Γ~1.0 meV for an upper-subband sheet density N_L~10^{11} cm^{-2} comparable to that in the midinfrared quantum cascade laser 1 (3) demonstrate that a smaller population inversion density (~0.17×10^{11} cm^{-2}) can be maintained at a moderate tunneling current density, and finally (4) predict for the electron-electron scattering a dramatic bias dependence arising from the so-called quantum-confined Stark effect. 8

A far-infrared quantum-cascade-laser design. The bottom panel of Fig. 1 illustrates the intersubband scattering (Γ) between two upper-subband electrons [E_2+E_i(k)] to two lower-subband electrons [E_1+E_i(k)]. For the current-injected upper-subband occupation density n_2 the lower-subband occupation n_1 results from the net nonradiative decay n_2 Γ_{nr}.

The subband occupation densities are determined from the two-level rate equation involving additional tunneling rates 9 —Γ_0, Γ_1— illustrated in Fig. 1:

\[
\frac{dn_2}{dt} = (N_L-n_2)\Gamma_0 n_1 n_2 \Gamma_1,
\]

There is no current injection into the lower subband because the lower band edge of the emitter is raised above E_1. The steady-state solution of Eq. (1) thus yields n_2 = N_L (1 - Γ_{nr}/Γ_1), and population inversion requires Γ_2~Γ_1. The lower-level escape rate Γ_1~0.5 meV is significantly larger than the decay rate 1/τ~0.03 meV measured under a weak optical pumping. 6 Here we investigate the far-infrared quantum-cascade-laser scattering to test if population inversion can be maintained. 10

Effective Coulomb interaction. The characteristic in-plane momentum transfer q~√2m_e Δ together with the zero-frequency background dielectric constant ԑ_0 provides a natural scaling of the effective Coulomb interaction (e^2/ԑ_0 q^2 Δ)U(q), where U(q) is a dimensionless matrix element introduced below. For the screened U(q) and unscreened U^0(q) matrix element we find (a) a moderate q variation, (b) a correspondingly moderate dependence on an effective Thomas-Fermi wave vector q_{TF}, and (c) a numerical value of U^0(q=0) that can be estimated from experiments. 5

The screened effective dimensionless matrix element is defined

U(q) = U_L U(q) = 2π \int dx_2 \int dx_1 \Psi_2(x_2) \Psi_1(x_1) q \phi(x_1)(x_2)

\times \exp(-\sqrt{q^2+q_{TF}^2}/\sqrt{\phi(x_1)(x_2)}).

(2)
in terms of the resonant-level wave functions\textsuperscript{12} and Thomas-Fermi wave vector $q_{\text{TF}}$. The unscreened interaction matrix element $U^0(q)$, the $q_{\text{TF}}\rightarrow 0$ limit of (2), is finite at $q=0$. The moderate variation of $U^0(q)$, namely $U^0(q_{\Delta})\approx U^0(0)/4$, can be deduced analytically for a square quantum well with infinite barriers. That the screening is ineffective in modulating the nonequilibrium electron-electron scattering follows directly from the observation $U(q) = U^0(\sqrt{q^2 + q_{\text{TF}}^2})$ because the estimated Thomas-Fermi screening wave vector remains smaller than the characteristic momentum transfer,\textsuperscript{11} $q_{\text{TF}} < q_{\Delta}$.

Finally, the strength of the effective nonequilibrium electron-electron interaction is evident from the observed\textsuperscript{3} large equilibrium depolarization shift\textsuperscript{13,14} $\Delta^*-\Delta \approx 2$ meV of the absorption peak, $\Delta^*$, from the far-infrared subband separation $\Delta \approx 11$ meV at sheet density $N_s \approx 10^{11}$ cm$^{-2}$. In particular, neglecting the coupling to other quantum levels, we have\textsuperscript{11,14}

$$\left(\Delta^*\right)^2 - \Delta^2 = 2N_s(e^2/\epsilon_0\Delta) U^0(q = 0).$$

For a finite occupation density $n_2 \leq N_{s}\approx N_{s}\approx 10^{11}$ cm$^{-2}$ we thus expect the strong interaction $N_s(e^2/\epsilon_0\Delta) U^0(0) \approx 2$ meV.

**Scaling of electron-electron scattering.** We use the Fermi golden rule\textsuperscript{15} to evaluate the total rate $\Gamma$ for two (opposite-spin\textsuperscript{16}) upper-subband electrons to decay to subband $E_1$. For complete upper-subband occupation (i.e., $n_2 = N_{s}$) at zero temperature we obtain $\Gamma$ as a sum over the in-plane momentum transfer $q$ of the squared matrix element $|U(q)|^2$ weighted by the phase-space contribution $|P(q)|$ introduced below. Because, however, $|U(q)|^2$ exhibits only a moderate $q$ variation and because the scattering phase space is dominated by the contribution at $q_{\Delta}$, we can approximate\textsuperscript{17}

$$\Gamma \approx \frac{\text{Ry}^*}{\pi} \left| \frac{\mu_2}{\Delta} \right| |U(q_{\Delta})|^2 I_p(\mu_2/\Delta),$$

where

$$I_p(\mu_2/\Delta) = \int dq \frac{q}{k_{\mu_2}} P(q) = I_p(0) = 0.785$$

represent a dimensionless integrated phase-space measure essentially independent of $\mu_2/\Delta$.

The top panel of Fig. 2 verifies linear-in-$\mu_2$ scaling of $\Gamma$, as expressed in Eqs. (4) and (5). For the unscreened interaction the linear scaling is nearly exact and closely approximated by $\text{Ry}^*/\pi^2(\mu_2/\Delta)|U^0(q_{\Delta})|^2 I_p(0)$. For the screened rate ($q_{\text{TF}} > 0$), there is some deviation arising from the increasing screening of squared matrix element $|U(q)|^2$. At most, that screening causes a factor of 2 reduction even at $\mu_2 > \Delta$.

To explain the central result Eq. (4) we consider the phase-space contribution at momentum transfer $q$ (see also Ref. 15),

$$N_s^2 \mu_2^{-1} P(q) = \frac{1}{2} \sum_{k'k} \Theta(\mu_2 - E_i(k))$$

$$\times \Theta(\mu_2 - E_i(k')) 2\pi \delta(E_i - E_i).$$

The displayed one-half factor arises because we only consider direct scattering between opposite-spin electrons.\textsuperscript{16} The energy difference, $E_i - E_i$, between the final and initial state depends on $q$ and on the initial in-plane momenta, $k$ and $k'$. In Ref. 11 we show that the weighted dimensionless phase-space contribution, $(q/k_{\mu_2})^2 P(q)$, (a) has the domain $-1 \leq q/k_{\mu_2} - \sqrt{1 + (q_{\Delta}/k_{\mu_2})^2} \leq 1$, (b) is always strongly peaked at the characteristic momentum transfer $q_{\Delta}$ with constant maximum value $(q_{\Delta}/k_{\mu_2})^2 P(q_{\Delta}) = 8/(3\pi^2)$, and consequently, (c) results in an almost $\mu_2/\Delta$-independent integrated phase-space measure, $I_p(\mu_2/\Delta) = I_p(0)$.

The key observation is (b), which follows from the assumed quadratic subband dispersion, $E_i(k)$. Specifically, at
FIG. 2. Top panel shows the approximate scaling, solid curve, with electron occupation \( \mu_2/\Delta \) of unscreened (screened) nonequilibrium scattering rate \( \Gamma \), dashed–single-(double-) dotted curve. Screening causes at most a factor of 2 reduction of \( \Gamma \) even at \( \mu_2/\Delta \). The interaction matrix elements are evaluated at \( V_{\text{sym}} \) where the rate \( \Gamma_{22-21} \) dotted curve, essentially vanishes. Bottom panel demonstrates \( 19 \) that a finite population inversion (left axis) \( n_2-n_1 \approx 0.17 \times 10^{10} \text{ cm}^{-2} \) can be maintained at a moderate current density (right axis) \( J = e \Gamma_e(N_L-n_2) \) despite the strong intersubband scattering. Note, however, that the population inversion quickly saturates and eventually decreases, whereas the current density \( J \approx e N_L(1-n_2/N_L) \) shows a faster-than-linear increase with \( \mu_2/\Delta \).

The characteristic momentum transfer, \( q = q_{\Delta} \), the \( \delta \)-function argument, \( (E_f-E_i) \), in Eq. (6) reduces to \( (q_\Delta(k_y-k_y^i)/m^*_{e}) \), where we have chosen the \( y \) direction to be parallel to the in-plane momentum transfer. The phase-space contribution, Eq. (6), then scales as \( m^*_{e}k^3_{y}/q_{\Delta} \) and (upon extracting \( N^2_L \mu_2^{-1} \varepsilon m^*_{e}k^3_{2} \)) we arrive at the constant value \( (q_{\Delta}/k_{\mu_2})P(q_\Delta) = 8/(3\pi) \).

A finite population inversion. The bottom panel of Fig. 2 estimates the population inversion \( n_2-n_1 \) (left axis) and the current density \( J = e \Gamma_e(N_L-n_2) \) (right axis). These estimates are based on the steady-state solution of Eq. (1) using the simple assumption

\[
\Gamma_{\text{sc}}(n_2) = \Gamma_{\text{sc}} + \frac{2}{N_L} \left( \frac{n_2}{N_L} \right) \frac{Ry^*}{\pi} \frac{\mu_2^2}{\Delta} |U^0(q_{\Delta})|^2 |I_p(0)|, \tag{7}
\]

for the total intersubband decay rate at voltage drop \( V_{\text{sym}} \). We assume \( 1 \) in Eq. (7) the total single-electron decay rate \( \Gamma_{\text{sc}} \) bounded by the value, \( \Gamma_{\text{sc}} = 0.03 \text{ meV} \), measured \( 2 \) at weak optical pumping and \( T = 50 \text{ K} \). The estimate, \( \Gamma_{\text{sc}}(n_2) - \Gamma_{\text{sc}} \) for the electron-electron decay results as follows. The scattering \( 2 \gamma_{22-21} \) can be neglected at \( V_{\text{sym}} \). The scattering \( \Gamma \) removes electrons at a time but is reduced by the partial upper-subband distribution \( f_2(k) = (n_2/N_L) \Theta(\mu_2 - E_i(k)) \). Finally, we approximate the resulting electron-electron decay \( 2(n_2/N_L)\Gamma \) by the scaling result (solid curve in top panel) for \( \Gamma \).

The current injection in the midinfrared quantum cascade laser \( 1 \) maintains a population inversion \( n_2 - n_1 \approx 10^{11} \text{ cm}^{-2} \) which requires \( \mu_2 \approx 5 \text{ meV} \) and \( \Gamma_e > \Gamma_{c2} \). In the present far-infrared structure the resulting strong decay \( 2 \Gamma_e \approx 1.0 \text{ meV} \) would eliminate such a population inversion. Nevertheless, Fig. 2 demonstrates \( 19 \) that a smaller population inversion, \( n_2 - n_1 \approx 0.17 \times 10^{10} \text{ cm}^{-2} \) can be maintained at current densities comparable to the midinfrared quantum cascade laser. \( 1 \)

However, also note the population inversion, \( n_2 - n_1 \), quickly saturates and eventually decreases whereas the current density, \( J = e \Gamma_e(N_L-n_2) \), shows a faster-than-linear increase with \( \mu_2/\Delta \). A choice of \( \Gamma_e > \Gamma_{c2} \approx 1.0 \text{ meV} \) (not shown) does not increase the maximum population inversion and causes a strongly nonlinear rise of the current with \( \mu_2/\Delta \). The electron-electron scattering thus forces a non-trivial optimization of \( \Gamma_e/\Gamma_{c2} \) and \( \mu_2/\Delta \).

FIG. 3. Dramatic voltage-drop variation (top panel) of electron-electron scattering rates, \( \Gamma \) and \( \gamma_{22-21} \), explained (bottom panel) by the quantum-confined Stark effect \( 3 \) on the wave-function overlap and symmetry. The bias dependence of \( \Gamma_e \) and \( \gamma_{22-21} \) reflects the wave-function–symmetry dependence of the characteristic matrix element \( |U(q_{\Delta})|^2 \) and \( |U_{22-21}(q_{\Delta}/\sqrt{2})|^2 \), respectively. In particular, the matrix element \( U(q_{\Delta}) \) and thus \( \Gamma_e \) enhance at \( V_{\text{sym}} \) because of the increased wave-function overlap. In contrast, the matrix element \( U_{22-21}(q_{\Delta}/\sqrt{2}) \) and thus \( \gamma_{22-21} \) are strongly reduced close to \( V_{\text{sym}} \) but increase dramatically when, for \( V \neq V_{\text{sym}} \), the wave-function symmetry is lost. Finally, the upper panel shows the combined electron-electron scattering rate, \( 2\Gamma + \gamma_{22-21} \), which also exhibits a significant wave-function–symmetry variation.
Wave-function-symmetry dependence. The bottom panel of Fig. 3 shows the so-called quantum-confined Stark effect\(^1\) of the bias voltage on the subband separation and on the wave-function overlap and symmetry.\(^2\) The minimal subband separation occurs at voltage drop \(V_{sym}\) (vertical dotted line). The dipole matrix element, \(|\langle \Psi_2 | x | \Psi_1 \rangle|\) (solid curve), enhances at \(V_{sym}\) with the increased wave-function overlap. In contrast, the center-of-charge separation, \(|\langle \Psi_2 | x | \Psi_2 \rangle\rangle - (\langle \Psi_1 | x | \Psi_2 \rangle)\rangle\) (dashed curve) vanishes at \(V_{sym}\) but rapidly changes with \(V-V_{sym}\), a variation reflecting the loss of wave-function inversion symmetry.

The top panel of Fig. 3 shows the dramatic voltage-drop dependence of both scattering rate \(\Gamma\) (solid curve) and of \(\Gamma_{2-21}\) (dashed curve).\(^\dagger\) This variation reflects the wave-function-symmetry dependence of the characteristic matrix elements for a constant ratio \(\mu_2/\Delta = 1/2\); see Eq. (4) (cf. Ref. 17). In particular, the matrix element \(U(q_{x})\), containing an even number of upper-level wave functions, can never be zero, and in fact enhances at \(V-V_{sym}\). In contrast, the characteristic matrix element \(U_{22,21}(q_{x}/V^2)\), containing three upper-level wave functions, must vanish close to \(V_{sym}\) (due to the near-exact wave-function inversion symmetry), but increases rapidly with the finite charge inversion symmetry. Finally, the top panel of Fig. 3 shows the total electron-electron decay, \(21' + \Gamma_{2-21}\), which also depends significantly on the wave-function-inversion symmetry. Ensuring an upper-subband current injection at \(V\neq V_{sym}\), may thus enhance the population inversion beyond the value, \(n_2 - n_1 = 0.17 \times 10^{11} \text{ cm}^{-2}\), estimated in Fig. 2.

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\(9\) We estimate \(E\) \(\approx\) \(2.0\) meV for the structure shown in Fig. 1.

\(10\) The strong nonequilibrium electron-electron scattering may also affect the study [see, for example, A. N. Korotkov, D. V. Averin, and K. K. Likharev, Phys. Rev. B 49, 7548 (1994)] of possible continuous Bloch oscillations in a two-level tunneling structure.


\(12\) We thank Dr. B. Galdrikian for kind permission to use his Schrödinger-solver code.


\(15\) The rate \(\Gamma\) is defined by \(\{\{\{\{\{\{N_f\}f\}f\}f\}f\}f\}f\}f\}f\) \(= \Sigma_g (\epsilon_g / e g_d )^3 U(q)^2 N_f^2 \mu_2^2 P(q)\) with phase-space contribution \(N_f^2 \mu_2^2 P(q)\) listed in Eq. (6).

\(16\) We assume for the same-spin scattering that the moderate variation of \(U(q)\) causes the direct and exchange contribution to approximately cancel.

\(17\) We also evaluate (for \(n_2 = N_f\)) the scattering \(\Gamma_{2-21}\) between two-subband \(E_2\) electrons of which only one decays to subband \(E_1\). This rate can be approximated as Eq. (4) but with a different characteristic matrix element \(U_{22,21}(q_{x}/V^2)\) (defined by three upper-level wave functions) and phase-space measure (Ref. 11) \(I_{2-21} - (\mu_2/\Delta)\). There is no scaling of \(\Gamma_{2-21}\) because the Pauli exclusion (within subband \(E_2\)) restricts the phase space \(I_{2-21}\) for \(\mu_2/\Delta < 1/2\); see Ref. 11.

\(18\) The intersubband decay due to impurity, interface defect, and acoustic-phonon scattering remains at temperature \(T < 50\) K strictly bounded by the experimental value \(1/\tau = 0.03\) meV. We estimate the decay due to thermally activated optical-phonon emission bounded at \(10^{-3}\) meV for \(T > 25\) K.

\(19\) We take \(\Gamma_{1,2} = 0.51(1.01)\) meV (estimated at \(V_{sym}\)) and use \(\Gamma = \Gamma_{c,2}/4\).

\(20\) J. Faist, F. Capasso, A. L. Hutchinson, L. Pfeiffer, and K. W. West, Phys. Rev. Lett. 71, 3573 (1993); this paper reports and explains the corresponding voltage-drop dependence of the optical selection rules.