

Interacting Drift-Diffusion Theory for Photoexcited Electron-Hole Gratings in Semiconductor Quantum Wells

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Phase-resolved transient grating spectroscopy in semiconductor quantum wells has been shown to be a powerful technique for measuring the electron-hole drag resistivity ρ_{eh} , which depends on the Coulomb interaction between the carriers. In this Letter we develop the interacting drift-diffusion theory, from which ρ_{eh} can be determined, given the measured mobility of an electron-hole grating. From this theory we predict a crossover from a high-excitation-density regime, in which the mobility has the “normal” positive value, to a low-density regime, in which Coulomb drag dominates and the mobility becomes negative. At the crossover point, the mobility of the grating vanishes.

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The phenomenon of Coulomb drag, whereby an electronic current driven in a quasi-two-dimensional electron gas drags carriers in an adjacent quantum well, creating a potential difference or a current in the latter has been a topic of intense interest in condensed matter physics for the past two decades [1–10]. The effect was first predicted, theoretically, by Pogrebinskii [1] and Price [2], but the field exploded only in the early 1990s, following the first successful realization of independently contacted bilayer structures [11]. Since then the field has flourished, due to the insights it affords on the nature of Coulomb correlations, nonequilibrium fluctuations, and quantum coherence between spatially separated systems. While the original experiments were done on electronic bilayers [3], it was soon realized that electron-hole bilayers offer even more interesting scenarios [7,8,10,12]. For example, electrons and holes can condense in an excitonic superfluid, with or without a magnetic field [10,13–15], and the occurrence of such a condensation should lead to striking manifestations in Coulomb drag experiments [15]. More recently, drag effects have also been investigated for different spin populations in a single quantum well (spin Coulomb drag) [16–21], in bilayer graphene [22,23], in topological insulators [24], and in trapped cold atoms [25].

The study of drag effects in electron-hole bilayers is notoriously complicated by the fact that, in order to host carriers of opposite polarities, the two layers must be held at different chemical potentials, while their spatial separation is of the order of 20–30 nm. In contrast to this, a nonequilibrium nonhomogeneous distribution of electrons and holes can be rather easily achieved in a single layer with a laser of frequency larger than the band gap, which creates an equal number of electrons and holes, in addition to the carriers (say, electrons) that are already present at equilibrium.

In a recent series of experiments [20,26,27] the interference between two laser beams coming from different directions and polarized in the same direction has been

exploited to create a transient electron-hole ($e-h$) grating, i.e., a spatially periodic modulation of the electron and hole densities on the surface of an n -type GaAs quantum well. Electron-hole drag manifests itself in quite a striking and direct way in the mobility of the $e-h$ grating under the action of an electric field [27,28]. The crucial observation is that the drift velocity of the $e-h$ grating—a collective formation—differs, in general, from the drift velocity of the background majority carriers (electrons). If the background carriers were absent (e.g., in an intrinsic material) then the drift velocity would vanish, since the net electric force on a neutral object is zero. In the opposite limit, in which the $e-h$ grating is a small perturbation on the background electron density, the mobility depends crucially on the rate of momentum transfer between electrons and holes [28]. Let us assume at first that this can be neglected. Then the constraint of charge neutrality causes the dense electrons to follow the few holes rather than the other way around. Under these conditions the grating moves with the drift velocity of the holes, and the mobility is positive [29]. Coulomb collisions between counterflowing electrons and holes produce the equivalent of an electric field that pushes the holes in the direction opposite to the external field. Which of the two fields ultimately dominates depends on the mobility of the electrons: if that mobility is sufficiently high the holes will inevitably reverse their direction of flow. The mobility of the grating will then be negative and approach the mobility of the electrons when the latter is large.

The possibility of observing anomalous (i.e., negative) mobility for an electron-hole packet in n -type semiconductors was first pointed out by McLean and Paige [30] and recently confirmed by Yang *et al.* [27] in n -GaAs. However, these authors relied for their analysis on a simplified phenomenological model in which the $e-h$ grating and the background electrons are treated as separate entities. The $e-h$ grating, being a neutral entity, does not directly respond to the electric field. Hence, it is

impossible, within this model, to describe the competition between Coulomb drag and the normal ambipolar mobility of the grating: the resulting formulas are correct only in the limit of strong Coulomb drag and cannot be used to predict the crossover between the normal and the “anomalous” (i.e., Coulomb drag-dominated) regimes.

In this Letter, we develop a full-fledged interacting drift-diffusion theory from which we derive general formulas for the mobility and the diffusion constant of the e - h grating. These formulas not only enable us to extract precise values of the Coulomb drag resistivity from Doppler velocimetry data, but they also lead to a striking prediction: the e - h grating mobility can be driven through a sign reversal by changes in excitation intensity, temperature, or background density. With an appropriate choice of parameters, the e - h grating can be made stationary in the presence of an electric field.

Our starting point is the drift-diffusion equation for electron and hole densities (n and p , respectively), in which, however, we take into account the presence of the off-diagonal homogeneous conductivity σ_{eh} (this is what makes our theory interacting, whereas, in the “standard” theory, σ_{eh} is set to zero). Thus, we have

$$e\partial_t \begin{pmatrix} -n \\ p \end{pmatrix} = -\nabla \cdot \left[\begin{pmatrix} \sigma_{ee} & \sigma_{eh} \\ \sigma_{he} & \sigma_{hh} \end{pmatrix} \begin{pmatrix} \mathbf{E}_e^e \\ \mathbf{E}_h^e \end{pmatrix} \right], \quad (1)$$

where the effective electric fields are expressed as $\mathbf{E}_{e/h}^e = \mathbf{E} \pm \frac{1}{e} \nabla \mu_{c/v}$. Here, \mathbf{E} includes both external and built-in electric fields. μ_c and μ_v are chemical potentials of electrons and holes, respectively, not to be confused with the homogeneous mobilities μ_e and μ_h , which will appear later. By substituting the electric fields, the continuity equations become

$$e\partial_t n = \nabla \cdot [eD_{ee}\nabla n + eD_{eh}\nabla p] + \mathbf{E} \cdot \nabla \sigma_e + \sigma_e \nabla \cdot \mathbf{E}, \quad (2)$$

$$e\partial_t p = \nabla \cdot [eD_{hh}\nabla p + eD_{he}\nabla n] - \mathbf{E} \cdot \nabla \sigma_h - \sigma_h \nabla \cdot \mathbf{E}, \quad (3)$$

where $\sigma_e = \sigma_{ee} + \sigma_{eh}$ and $\sigma_h = \sigma_{hh} + \sigma_{eh}$. The diffusion matrix elements are $D_{\alpha\beta} = (-1)^{\delta_{\alpha\gamma}} \sigma_{\alpha\gamma} (\chi^{-1})_{\gamma\beta} / e^2$ (sum over γ implied), where $\chi_{\gamma\beta} = \partial n_\gamma / \partial \mu_\beta$ is the susceptibility matrix [31].

Following the standard procedure [29] we combine Eqs. (2) and (3) to cancel the space charge term $\nabla \cdot \mathbf{E}$, and only then impose the charge neutrality condition $\delta p = \delta n$, where δn and δp are the deviations of the corresponding densities from equilibrium and we assume ($|\nabla \delta n|/n \ll 1$). The result is

$$\partial_t (\delta n) = -\mu_a \mathbf{E}_{\text{ext}} \cdot \nabla \delta n + D_a \nabla^2 \delta n, \quad (4)$$

where the e - h grating mobility and diffusion constant are defined by

$$\mu_a = \frac{[\sigma_e(\partial_n + \partial_p)\sigma_h - \sigma_h(\partial_n + \partial_p)\sigma_e]}{e(\sigma_e + \sigma_h)}, \quad (5)$$

$$D_a = \frac{(\sigma_e D_h + \sigma_h D_e)}{(\sigma_e + \sigma_h)}. \quad (6)$$

Here $D_h = D_{hh} + D_{he}$ and $D_e = D_{eh} + D_{ee}$. By taking into account the electron-hole Coulomb drag, the conductance matrix can be expressed as [18]

$$\hat{\sigma} = \left(\rho_e \rho_h + \rho_{eh} \rho_h \frac{\mu_h}{\mu_e} + \rho_{eh} \rho_e \frac{\mu_e}{\mu_h} \right)^{-1} \times \begin{pmatrix} \rho_h + \rho_{eh} \frac{\rho_h}{\rho_e} \frac{\mu_h}{\mu_e} & -\rho_{eh} \\ -\rho_{eh} & \rho_e + \rho_{eh} \frac{\rho_e}{\rho_h} \frac{\mu_e}{\mu_h} \end{pmatrix}, \quad (7)$$

where the resistivities of electrons and holes are defined as $\rho_e = 1/(n\mu_e e)$ and $\rho_h = 1/(p\mu_h e)$, respectively (these resistivities can be straightforwardly measured in homogeneous dc transport experiments in which the carriers are either electrons or holes). ρ_{eh} stands for the cross resistivity due to electron-hole Coulomb drag. By substituting the conductivity matrix elements into Eqs. (5) and (6), we obtain

$$\mu_a = \frac{-\mu_e(n-p)/n}{1 + (\mu_e/\mu_h)(\rho_e/\rho_{eh}) + (p/n)(\mu_e/\mu_h)} \left\{ 1 - \frac{\rho_e}{\rho_{eh}} \left[\frac{1 + e\rho_{eh}(p\mu_e + n\mu_h) - enp(\mu_e + \mu_h)(\partial_n + \partial_p)\rho_{eh}}{1 + \rho_e/\rho_h + e\rho_{eh}\mu_h(n-p)^2/n} \right] \right\}, \quad (8)$$

$$D_a = \frac{D_e p \mu_h + D_h n \mu_e + e\rho_{eh} \mu_e \mu_h (n-p)(D_h n - D_e p)}{p \mu_h + n \mu_e + e\rho_{eh} \mu_e \mu_h (n-p)^2}. \quad (9)$$

These two equations are the main results of this Letter. For orientation we now discuss some limiting cases, taking, for definiteness, the n -type case, i.e., $n > p$: (i) Weak Coulomb drag limit ($\sigma_{eh}, \rho_{eh} \sim 0$):

$$\mu_a \approx \frac{n-p}{n/\mu_h + p/\mu_e}, \quad (10)$$

$$D_a \approx \frac{D_e p \mu_h + D_h n \mu_e}{p \mu_h + n \mu_e}. \quad (11)$$

These results agree with the well-known expressions for the e - h grating mobility and diffusion constant in the absence of Coulomb drag [29,32]. The e - h grating mobility depends on the difference of electron and hole densities. In

the n -type case, μ_a is positive; therefore, the grating moves in the direction of the electric field. (ii) Weak pumping regime ($\delta n \ll n$, $\rho_h \gg \rho_e$):

$$\mu_a \approx \left(\frac{\rho_e}{\rho_{eh}} - 1 \right) \frac{\mu_e}{1 + (\mu_e/\mu_h)(\rho_e/\rho_{eh})}, \quad (12)$$

$$D_a \approx D_h \approx \frac{k_B T}{e} \frac{\mu_e \rho_e / \rho_{eh}}{1 + (\mu_e/\mu_h)(\rho_e/\rho_{eh})}, \quad (13)$$

where the minority carriers are assumed to be nondegenerate. Similar to Eq. (3) of Ref. [28], the expression of the mobility here predicts a sign reversal of μ_a at $\rho_e = \rho_{eh}$. Specifically, μ_a becomes negative when the Coulomb drag resistivity is larger than the ordinary resistivity of the electrons. One notices that the ratio D_a/μ_a in this case is only dependent on ρ_e/ρ_{eh} and temperature, which suggests a neat way to extract the drag resistivity in experiment [27]. (iii) Strong pumping case, i.e., $\delta n \sim n$ and, consequently, $p \sim n$. In this case, we find that the expression in square brackets in Eq. (8) is larger than (1) (the derivative of Coulomb drag with respect to the density is negative), which suggests that μ_a can be positive even for $\rho_e < \rho_{eh}$. If the carrier density due to the injection is much larger than the one from doping, then one has $n \approx p$, resulting in $\mu_a \approx 0$.

In order to test the predictive power of Eqs. (8) and (9) we need a reasonable model for the Coulomb drag resistivity ρ_{eh} and the homogeneous drag-free mobilities $\mu_e = e/[m_e^* \sum_i (\tau_i^e)^{-1}]$ and $\mu_h = e/[m_h^* \sum_i (\tau_i^h)^{-1}]$, where m_e^* and m_h^* are the effective masses of electrons and holes. In

two-dimensional polar materials the dominant scattering mechanisms are [33] (i) polar-optical phonon scattering [33]

$$\tau_{\text{OP}}^{-1} = \frac{1}{4\pi\epsilon_0} \frac{e^2 \pi \sqrt{2m^* \omega_0 / \hbar} N_{\text{LO}}}{2\hbar} \left(\frac{1}{\kappa_\infty} - \frac{1}{\kappa_0} \right), \quad (14)$$

where the temperature dependence comes from the longitudinal optical phonon number N_{LO} , (ii) acoustic phonon deformation potential scattering [34]

$$\tau_D^{-1} = \frac{\Xi^2 k_B T m^*}{D v_{sl}^2 \hbar^3} \frac{3}{2a}, \quad (15)$$

and (iii) ionized impurity scattering, which, in the random phase approximation, has the form

$$\tau_{\text{imp}}^{-1}(\epsilon_k) = \frac{n_i e^4}{16\pi \hbar E_F \kappa_0^2 \epsilon_0^2} \int_0^{2\pi} d\theta (1 - \cos\theta) \times \left(\frac{\kappa_D}{k_F} + \sqrt{2\epsilon_k (1 - \cos\theta) / E_F} \right)^{-2}, \quad (16)$$

where the screening wave vector is $\kappa_D = \frac{e^2}{2\epsilon_0 \kappa_0} \frac{1}{\hbar^2 \pi} [m_e^* (1 - e^{-\hbar^2 n \beta \pi / m_e^*}) + m_h^* (1 - e^{-\hbar^2 p \beta \pi / m_h^*})]$ and $E_F(k_F)$ is the Fermi energy (wave vector). κ_0 and κ_∞ in these equations stand for the dielectric function in the static and high frequency limits, respectively. ϵ_0 is the permittivity in vacuum and $\hbar\omega_0$ is the optical-phonon energy. In Eq. (15), D and v_{sl} represent the crystal density and longitudinal sound velocity, respectively. a is the effective well width. Ξ is the deformation potential constant. Finally, the resistivity due to electron-hole Coulomb drag is obtained from [18]

$$\rho_{eh} = \frac{\hbar^2}{e^2 n p k_B T} \frac{1}{(2\pi)^2} \int_0^\infty dq \frac{q^3}{2} \int_0^\infty d\omega \text{Im} Q_{0e} \text{Im} Q_{0h} [(1 + \kappa_D/q)^2 \sinh^2(\hbar\omega/2k_B T)]^{-1}, \quad (17)$$

where $Q_{0e/h}$ is the temperature-dependent Lindhard function for electrons or holes times the Fourier transform of the Coulomb interaction (see Ref. [35]). By taking the electron doping density $n_0 = 1.9 \times 10^{11} \text{ cm}^{-2}$ and fitting the low temperature electron mobility measured in the experiment by Yang *et al.* [27] in 9 nm GaAs quantum wells, $\mu_e(5 \text{ K}) = 5.5 \times 10^5 \text{ cm}^2/\text{V}$, we determine the effective impurity density to be $n_i = 0.005 n_0$. Unfortunately, the same model, when applied to holes, leads to a hole mobility about 5 times larger than the one implied by the experimentally measured values of μ_a and D_a . This discrepancy may be attributed to the band structure or additional scattering mechanisms for holes. We have found that rescaling μ_h by a factor 0.2 at all temperatures leads to results in excellent agreement with experiment. The following calculations are based on this rescaling.

In Fig. 1(a), we plot the electron resistivity ρ_e as a function of temperature, as well as the electron-hole drag resistivity ρ_{eh} in the weak pumping regime. It is seen that the electron-hole drag resistivity is smaller than the

electron resistivity both at high and low temperature. However, at intermediate temperature, one has $\rho_{eh} > \rho_e$. The two curves intersect at 1.5 and 385 K. For comparison, we also plot the electron resistivity (green dotted curve) for a more strongly disordered system, in which $n_i = 0.2 n_0$. In this case, the regime in which $\rho_{eh} > \rho_e$ is significantly narrower, due to the increase of electron resistivity, but still clearly visible (the electron-hole Coulomb drag is essentially independent of impurity concentration).

Figure 1(b) shows the spin-grating mobility μ_a calculated from Eq. (8) in the infinitesimal pumping regime. Two changes in sign of μ_a vs temperature are visible: they correspond to the two crossings between ρ_{eh} and ρ_e . In this figure, we also plot the experimentally determined μ_a (blue squares). We find that the theoretical results are in excellent agreement with the experiment in the temperature range 20–150 K, in which range the e - h drag dominates and the e - h grating mobility is negative. However, the experimental data do not show any sign reversal. On the high temperature end, it simply appears that the experiment

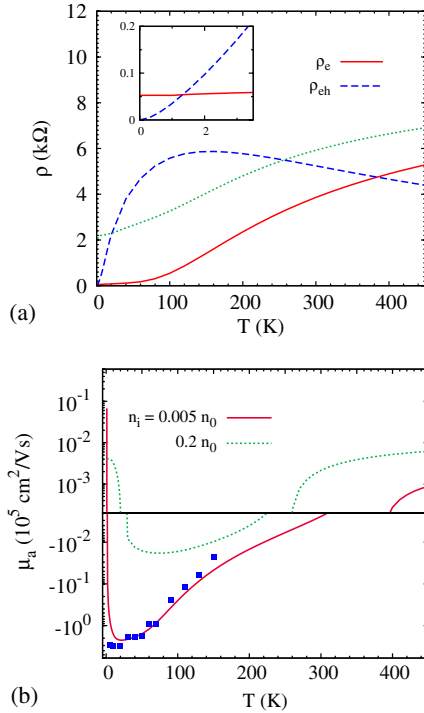


FIG. 1 (color online). (a) ρ_e (red solid curve) and ρ_{eh} (blue dashed curve) as functions of temperature with $n_i = 0.005n_0$. The green dotted curve represents ρ_e with $n_i = 0.2n_0$. Inset: zoom in on the low temperature region $T < 3.5$ K. (b) Grating mobility as a function of temperature. The blue squares are experimental data from Ref. [27]. The apparent breaks in the curves as they cross zero are artifacts of the logarithmic scale.

did not reach sufficiently high temperature. A high-temperature crossover was, in fact, observed in Ref. [28]. At low temperature, the reason for the discrepancy may lie in an undesired laser-induced heating of the electrons.

In Fig. 2, we plot the e - h grating mobility as a function of pumping intensity at 20 and 200 K. Neglecting the equilibrium hole density, we have $p = \delta n$ and $n = n_0 + p$. Then, the pumping intensity can be quantified by the ratio p/n_0 . We find that the mobility at first increases with increasing pumping intensity and becomes positive at

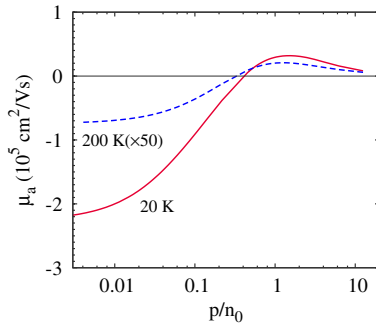


FIG. 2 (color online). Grating mobility as a function of pumping intensity (the latter quantified by the ratio p/n_0) at 20 and 200 K.

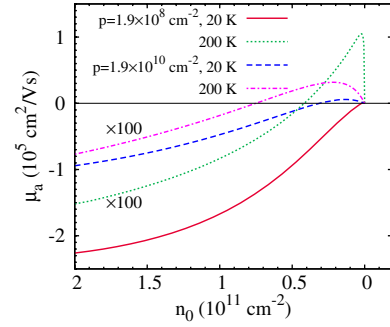


FIG. 3 (color online). Grating mobility as a function of equilibrium electron density.

large excitation levels. After reaching the maximum value, the mobility begins to decrease upon further increase of the pumping intensity, and eventually approaches zero. This behavior is consistent with our discussion above.

Another way to change the sign of the e - h grating mobility is by tuning the equilibrium electron density by means of electrostatic gating. In Fig. 3, we show μ_a vs n_0 for different temperatures and pumping intensities. In all cases, the grating mobility at first increases with decreasing background density and reaches a maximal value. This can be understood by the quick increase of the electron resistivity, since ρ_e/ρ_{eh} increases. As the background electron density further decreases to a negligible value, we fall back into the “strong pumping limit” ($n \approx p$) and the mobility decreases towards zero.

As a last point, we observe that the e - h grating diffusion constant remains essentially identical to the hole diffusion constant D_h (see Fig. 4), both being substantially reduced by Coulomb drag caused by interaction with the background electrons. We notice that, as expected, the effect

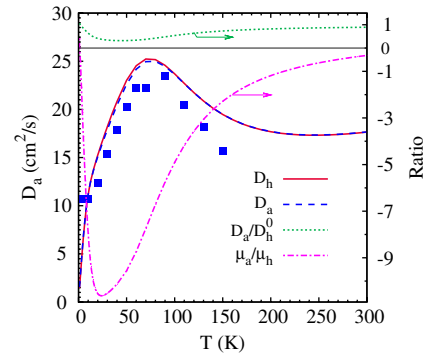


FIG. 4 (color online). Ambipolar diffusion constant (blue dashed curve) and hole diffusion constant (red solid curve) as a function of temperature for $n_b = n_0$ in the weak pumping case. Notice that the two curves are essentially identical. The filled squares represent the experimental data for the ambipolar diffusion constant reported by Yang *et al.* [27]. The green dotted curve shows the ratio between the ambipolar diffusion constants with and without Coulomb drag, while the pink chain curve shows the corresponding ratio for the e - h grating mobility.

of Coulomb drag on the grating diffusion constant is much less dramatic than its effect on the mobility. The theoretical results show good agreement with the experiment.

In summary, we have developed a drift-diffusion theory for the calculation of the mobility and diffusion grating of an electron-hole grating in a single electronic layer, taking fully into account the effect of Coulomb drag between electrons and holes. Our formulas show that the mobility of an electron-hole grating is a sensitive probe of electron-hole drag. Further, due to its dependence of electron-hole drag, the mobility of the grating can be driven through changes of sign by changing temperature, excitation power, or background electron density.

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