Nonlocal Photorefractive Screening from Hot Electron Velocity Saturation in Semiconductors

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Intervalley scattering of hot electrons during high-field transport in transverse-field photorefractive quantum wells induces a nonlocal optical response in which photoinduced changes in the refractive index are spatially shifted relative to the optical stimulus, providing an avenue for optical gain. We demonstrate that the onset of the photorefractive phase shift coincides with the onset of velocity saturation. This nonlocal response is the high-resistivity consequence in semi-insulating semiconductors of the Gunn effect mechanism.   [S0031-9007(96)01508-6]

Optically induced refractive index gratings that respond nonlocally to spatially varying optical illumination are the source of nonreciprocal energy transfer between two coherent laser fields during two-wave mixing. This is one method to generate optical amplification without stimulated emission, among others [1]. Physical mechanisms that produce a nonlocal photorefractive response are therefore of both fundamental as well as practical interest [2]. Nonreciprocal energy transfer and record photorefractive gain were discovered in photorefractive quantum well structures [3] operating in the transverse-field geometry [4] under applied electric fields larger than 3 kV/cm. Standard mechanisms for the nonlocal response, such as diffusion fields [5–7], are negligibly small in this geometry, making the origin of the nonreciprocal energy transfer a mystery. Recently, a heuristic mechanism for this nonlocal photorefractive response was proposed that relies on the quenching of dielectric relaxation by intervalley electron scattering and non-Ohmic transport [8], but conclusive experimental verification of this mechanism has been lacking. In this Letter, we present direct experimental evidence for the high-resistivity analog of the Gunn effect in semi-insulating semiconductors that produces *static* space-charge that responds nonlocally to a steady, spatially sinusoidal illumination pattern.

In drift-dominated photorefractive effects, spatially inhomogeneous illumination produces photoinduced space charge that screens an applied field [9]. For the low-intensity photorefractive effects described here, the space-charge is trapped at deep-level defects, rather than being present as free carriers. The defects pin the Fermi level near midgap, making the material high resistivity with slow dielectric relaxation, which allows the space-charge to accumulate under low illumination intensities [10]. The total field inside the photorefractive crystal can be expressed in terms of the applied field E_0 and an effective dielectric function $\varepsilon(K)$ with the incident intensity $I(r) = I_0 + \sum_K I(K)e^{iKr}$ as

$$E(r) = -E_0 \sum_K \frac{e^{iKr}}{\varepsilon(K)} \frac{I(K)}{I_0 + I_s}, \tag{1}$$

where $I(K)$ is the Fourier component of the intensity pattern, I_0 is the total beam intensity, and I_s is the saturation intensity [11]. Under standard conditions (low fields and high defect densities) the effective dielectric function is independent of K and is purely real. The total internal field in this case responds locally to the light intensity. However, when $\varepsilon(K)$ becomes a function of K and becomes complex, the dielectric function is nonlocal, causing a spatial shift between the intensity pattern and the resultant internal field. We show below that velocity saturation caused by intervalley scattering of photoinduced carriers is one such mechanism that causes nonlocal screening.

One of the best-known examples of screening in semiconductors is the Gunn effect [12,13], caused by electron velocity saturation at high fields due to electron heating and scattering from the high-mobility direct-gap minimum to the low-mobility indirect conduction band minima. The electron temperature during high-field transport is determined by a balance between Joule heating of the electrons due to the field and energy relaxation. The electron temperature $T_e = T_0 + 2e\mu E^2 \tau_r / 3k_B$, where T_0 is the lattice temperature, μ is the field-dependent mobility, and τ_r is the energy relaxation time. Gunn effect domains do not occur in semi-insulating GaAs, because there is insufficient free-electron density to screen the applied field [14]. However, the electrons in semi-insulating GaAs can experience the same intervalley scattering and velocity saturation as in doped materials, contingent on the assumption that the energy relaxation time τ_r is much smaller than the electron trapping time. The electron energy relaxation time is temperature dependent, but is approximately $\tau_r = 200$ fs at

room temperature [15]. As shown below, the electron trapping time in our samples is approximately 100 ps. Therefore, the hot-electron picture can remain valid even in the case of semi-insulating semiconductors. It is also important to point out that the electron temperature is independent of carrier density, because it depends only on the intrinsic properties of mobility and energy relaxation, but is independent of extrinsic properties such as carrier concentration. The most important consequence of electron velocity saturation in semi-insulating materials is its effect on dielectric relaxation. Because the photorefractive effect is a dynamic balance between photogeneration and dielectric relaxation, the velocity saturation is expected to play an important role in photorefractive screening.

Using small signal analysis, we have previously shown that the dielectric relaxation rate, when velocity saturation is present, is given by [8]

$$\Gamma_{\text{die}} = \frac{en_0}{\varepsilon\varepsilon_0}\left[\frac{dv(E)}{dE} + i\,\frac{d\mu(E)}{dE}E_D\right], \qquad (2)$$

where n_0 is the carrier density, $\mu(E)$ is the field-dependent mobility, and E_D is the diffusion field. Small signal analysis is valid when the depth of modulation of the illumination is weak, i.e., when $I(K)/I_0$ from Eq. (1) is small. When the electron velocity saturates under high field, the first term in Eq. (2) vanishes, and the dielectric relaxation rate is significantly reduced relative to the zero-field value. This reduction of the dielectric relaxation causes an imbalance between the formation of trapped space charge (that screens the applied field), and the relaxation of that charge. Therefore electrons are depleted from the high-intensity regions and accumulate in the low-intensity regions, causing a spatial shift of the total internal electric field relative to the intensity pattern.

To test the nonlinear transport hypothesis as the source of the photorefractive phase shift, we measured two-wave and four-wave mixing in three different proton-implanted photorefractive quantum well and epilayer structures [16]. These structures were designed to each have different transport nonlinearities from different Γ-L valley energy separations $\Delta E_{\Gamma L}$, which produce three distinct signatures for the phase shift based on different degrees of velocity saturation. One of the structures was a multiple quantum well sample with quantum-confined excitons, similar to the original structures that exhibited the phase shift [3]. In addition, GaAs and $Al_{0.2}Ga_{0.8}As$ epilayers without quantum wells were also studied. The GaAs epilayer has the largest Γ-L valley energy separation $\Delta E_{\Gamma L} = 284$ meV. The multiple quantum well structure with 60 periods of 7.5 nm GaAs wells and 10.0 nm $Al_{0.1}Ga_{0.9}As$ barriers has an intermediate Γ-L valley energy separation $\Delta E_{\Gamma L} = 224$ meV in the barrier. The Γ-L separation in the barrier is important in the quantum well structure because of real-space transfer of hot electrons from the well to the barrier [13]. The smaller Γ-L separation in the barrier then dominates the intervalley scattering. The $Al_{0.2}Ga_{0.8}As$ epilayer

has the smallest Γ-L valley energy separation (of our three structures) of $\Delta E_{\Gamma L} = 163$ meV [17]. These quantum well and epilayer structures were chosen to provide a trend of decreasing Γ-L valley separation. It is important to point out that smaller Γ-L separations lead to *weaker* nonlinear current-voltage behavior at room temperature. This is because in materials with small Γ-L energy difference there can already be an appreciable thermal L-valley population at room temperature, leading to a weaker contrast in the low-field vs high-field mobility for these structures. The weaker mobility contrast in the AlGaAs also leads to weaker gain saturation than in GaAs.

We are able to discount trap limitation as a possible mechanism for the photorefractive phase shift by monitoring the relative defect concentrations in the three samples. The trend in trap concentrations was verified by carrier lifetime measurements using pump-probe measurements [18]. The purpose of these measurements was solely to monitor the trend in defect concentration, and not to predict actual carrier lifetimes during photorefractive mixing. The AlGaAs epilayer had a 160 ps lifetime, the multiple quantum well structure had a 143 ps lifetime, and the GaAs epilayer had a 48 ps lifetime. This monotonic decrease reflects increasing defect concentrations, which would produce decreasing photorefractive phase shifts under the trap limitation mechanism, opposite to the trend observed for the nonlinear transport mechanism. Therefore the sample lifetimes were specifically controlled to give a clear and unambiguous experimental differentiation between the trap-limitation mechanism and the nonlinear-transport mechanism.

We performed two-wave and four-wave mixing experiments under identical conditions for all samples using a CW Ti-sapphire laser pumped by an argon ion laser, with the pump and probe beams incident symmetrically on the device without focusing. The beam propagation direction was along the [100] growth axis of the layers, the beams were s-polarized, and the electric field was applied in the plane of the epilayer in the perpendicular [110] direction. Two-wave mixing measures the cross-modulation ratio, denoted by γ, between the pump and signal beams. Four-wave mixing measures the diffraction efficiency, denoted by η, from the induced gratings. By combining the two-wave and four-wave mixing measurements at a wavelength where the electroabsorption vanishes, the phase shift is obtained as [4]

$$\sin(\phi) = \frac{\Delta n}{|\Delta n|}\frac{\gamma}{2\sqrt{\beta\eta}}, \qquad (3)$$

where $\beta = I_2/I_1$ is the ratio of beam intensities. We selected the wavelength for each sample that coincided with the lowest-energy electroabsorption zero crossing. The refractive index changes are nearly a maximum at this wavelength.

Two-wave and four-wave mixing efficiencies for the GaAs epilayer, the AlGaAs/GaAs multiple quantum well,
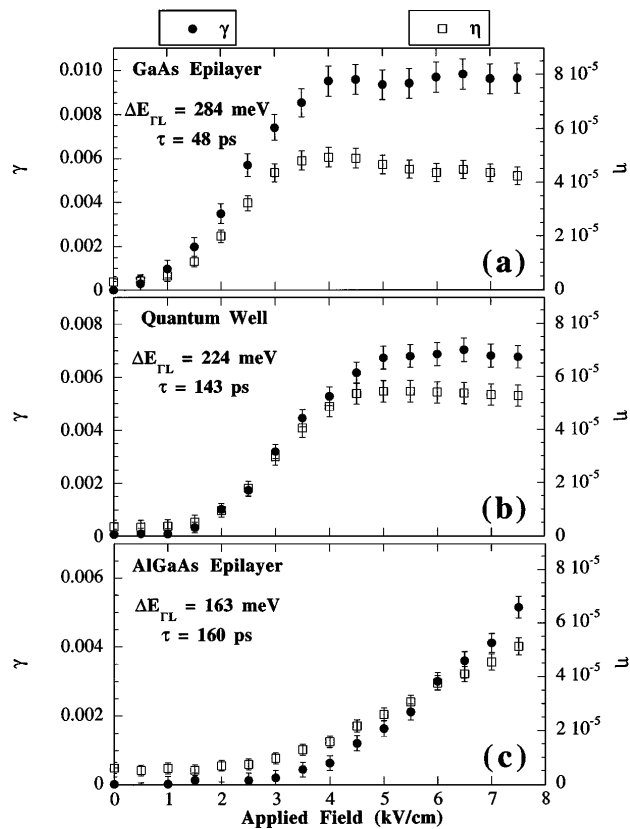
FIG. 1. Two-wave and four-wave mixing efficiencies as functions of applied electric field in three structures with decreasing intervalley scattering energies: (a) an epilayer of GaAs; (b) a 60 period 7.5 nm GaAs/10 nm $Al_{0.1}Ga_{0.9}As$ multiple quantum well structure; and (c) an $Al_{0.2}Ga_{0.8}As$ epilayer. The mixing experiments were performed with a fringe spacing of 10 μm.
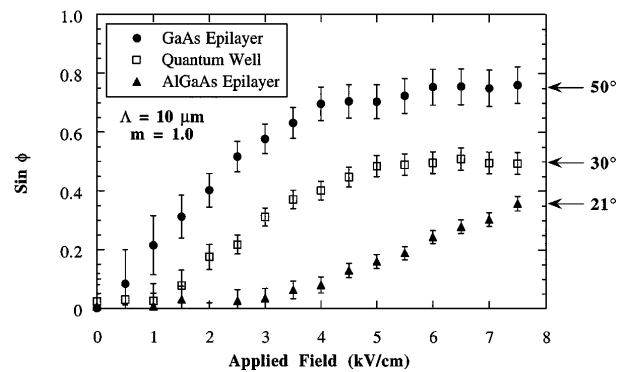

FIG. 2. Sine of the photorefractive phase shift vs applied field for the three different structures in Fig. 1. The GaAs epilayer has the smallest saturation field and the largest photorefractive phase shift, corresponding to the strongest transport nonlinearity.

and the AlGaAs epilayer are shown in Fig. 1 as functions of applied field up to 8 kV/cm. For all measurements, the fringe spacing is 10 μm, the beam ratio β is approximately equal to unity, and a total intensity of 20 mW/cm^2 was incident on the devices. This beam ratio gives a modulation depth of $m \approx 1$. The AlGaAs epilayer (smallest nonlinear transport) shows the smallest saturation of the mixing efficiencies with field, while the GaAs epilayer (strongest nonlinear transport) exhibits strong saturation of the mixing amplitudes for fields above 4 kV/cm. The multiple quantum well sample exhibited behavior intermediate between the two epilayers.

The phase shift is extracted from the two-wave and four-wave mixing efficiency for each structure using Eq. (3). The resulting $\sin(\phi)$ is shown in Fig. 2 as a function of applied field. The GaAs epilayer has a significant photorefractive phase shift that increases for fields up to 4 kV/cm, and saturates at a value of $\phi = 50°$. The AlGaAs epilayer, which is expected to have the smallest nonlinear transport based on intervalley scattering, has a phase shift of only $\phi = 21°$ at 8 kV/cm. The multiple quantum well structure saturates at an intermediate phase shift of $\phi = 30°$ for fields above 5 kV/cm. The phase shifts and the satu-

ration fields in each of the structures follow a monotonic decreasing trend from the GaAs epilayer, through the multiple quantum well to the AlGaAs epilayer, reflecting the monotonic trend in the Γ-L energy separation.

Although the experiments were performed at high-modulation depth to obtain a good signal-to-noise ratio, the theoretical predictions were derived using a small-modulation approximation [8]. We therefore repeated the mixing experiments on the GaAs sample using a modulation index as small as $m = 0.57$, which was approximately the signal-to-noise limit of the four-wave mixing experiment. At this reduced modulation index, the phase shift was again measured to be approximately $\sin(\phi) = 0.9$, which is consistent with the high-modulation value. It has also been pointed out that small modulation results are semiquantitatively valid for modulation close to unity [19]. The mobility-lifetime products were also found for a low applied field using photoconductivity measurements, with 2.1×10^{-7} cm^2/V for GaAs epilayer, 3.1×10^{-7} cm^2/V for the multiple quantum well, and 1.0×10^{-7} cm^2/V for the AlGaAs epilayer. This small range of the mobility lifetime products will not mask the role of the transport nonlinearity appearing in Eq. (2).

As the most direct test of the nonlinear transport hypothesis for the nonlocal photorefractive response, we simultaneously measured the two-wave mixing and the current-field curve of several GaAs samples (with the largest Γ-L separation) prepared with gold contacts evaporated onto the sample shortly after a HF acid etch. In Fig. 3 we show the current-field curve and the photorefractive phase shift as functions of applied field for a GaAs sample with excellent low-field Ohmic behavior. The deviation from Ohmic behavior above 3 kV/cm is unmistakable, and has the standard signature of velocity saturation due to intervalley scattering [13]. The photorefractive phase shift in this sample turns on near the same applied field as the onset of intervalley scattering. Although the experimentally measured velocity saturation
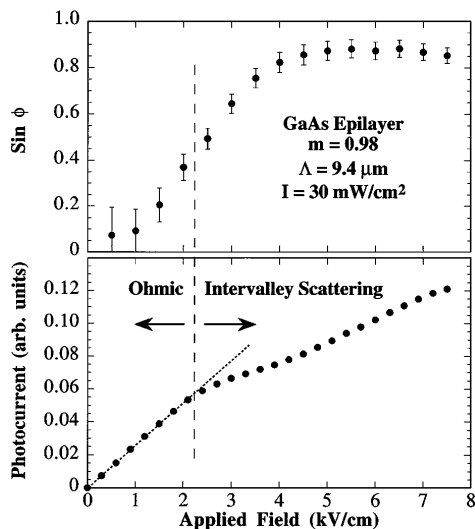
FIG. 3. Sine of the photorefractive phase shift compared with the current-field curve as a function of applied field. The strong onset of the photorefractive phase shift coincides with the onset of intervalley scattering, seen as a marked deviation from Ohmic behavior for fields above 3 kV/cm.

in the lower part of Fig. 3 is broadened by the inhomogeneous distribution of the electric field between the coplanar contacts (which is an intrinsic property of the two-dimensional geometry of the transverse-field devices [20]), the agreement between the onset of the phase shift and the onset of intervalley scattering is excellent. Similar agreement was observed for the other samples.

In conclusion, these experiments clearly and quantitatively establish the direct role of velocity saturation in generating the nonlocal photorefractive phase shift in thin trap-dominated semiconductor films. Because of the strong similarities between photorefractive phenomena and more general optically induced space-charge phenomena in trap-dominated semiconductors, the results described here should have general importance for a broad range of related problems in photoconductive and other optoelectronic devices.

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