Photorefractive Gunn Effect

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We predict a new optically nonlinear effect in semiconductors in which a traveling interference pattern generated by two optical waves excites multiple high-field Gunn domains which move phase locked with the interference fringes. The optical waves simultaneously diffract off the refractive index change generated by this oscillating space-charge field. [S0031-9007(96)00145-7]

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In 1963 Gunn showed [1] that, if a dc electric field applied to a GaAs crystal exceeded a critical value, spontaneous current oscillations appeared in the external circuit. This Gunn effect arises from negative differential resistance which occurs when the applied field induces intervalley electron transitions from a high mobility Γ minimum into higher minima of the conduction band, where the effective mass is larger and the scattering rate is higher [2]. Thus, electron velocity decreases with increasing electric field when the field exceeds the critical value. This effect has been extensively studied [3] and is now used for high frequency oscillators. Later studies have shown that formation of high-field (Gunn) domains can be triggered by light [4] and that traveling high-field domains in a biased GaAs crystal can modulate a beam of light incident upon it [5].

The photorefractive effect in semiconductors is known for more than a decade. Wave mixing experiments in semi-insulating Cr doped GaAs and Fe doped InP [6] and in undoped GaAs [7] show moderate coupling between the interacting waves. However, the high mobility in these materials reduces their dielectric relaxation time, and makes them attractive for image processing [8] and sensitive optical detection [9]. Recent advances [10] in understanding the interaction between waves of different frequencies in a photorefractive crystal with an applied ac electric field have lead to optical phase-locked detection [11] and detection of microwave signals [12]. The photorefractive effect was also used to detect low frequency spontaneous current oscillations [13] which occur in biased InP and are *not* related to the Gunn effect.

In this Letter, we predict a new optically nonlinear effect in semiconductors. Two optical waves of slightly different frequencies are incident upon a biased semiconductor crystal doped with deep impurity centers (as in Refs. [6–13]). Their moving interference pattern excites multiple high-field Gunn domains which move *phase locked* with the interference fringes. The space-charge (high-field) domains modify the refractive index via Pockel's effect, creating a periodic index modulation. Finally, the optical waves that generated the Gunn domains diffract off the index modulation and couple to each other via cross-phase modulation. This is a new nonlinear optical effect that couples optical waves through optically triggered Gunn effect.

We start with the expression [2] for the drift velocity v of electrons in a material with negative differential resistance as a function of electric field E

$$v(E) = v_s\left[1 + \frac{E/E_s - 1}{1 + A(E/E_s)^\beta}\right], \tag{1}$$

where v_s is the saturation drift velocity, E_s the saturation field, and A and β are dimensionless constants which depend explicitly [2] on the mobility $\mu = v_s/E_s$. For example, for GaAs with $\mu = 0.5\ \mathrm{m^2\,V^{-1}\,sec^{-1}}$, $v_s = 8.5 \times 10^4$ m/sec, $E_s = 1.7 \times 10^5$ V/m, $A = 0.04$, and $\beta = 4$. Consider such a bulk crystal illuminated by two quasimonochromatic plane waves of slightly different frequencies ω and $\omega + \Omega$ ($\Omega \ll \omega$) and slightly different angles of incidence (Fig. 1). The light forms a moving interference pattern of intensity

$$I(z,t) = I_0[1 + m\cos(Kz + \Omega t)], \tag{2}$$

where $K = 2\pi/\Lambda$ is the interference wave number, m the modulation depth of the interference grating, and I_0 the average (total) intensity, equal to the modulus squared of the sum of the optical field amplitudes, averaged over time much longer than $2\pi/\omega$, but much shorter than $2\pi/\Omega$. As in the case of any photorefractive material, electrons are photoexcited from deep donor sites and are trapped either in acceptors or in ionized donors (no intrinsic excitation). We use the standard set of rate and continuity equations and Gauss' law that describe the photorefractive effect in a medium in which electrons are the sole charge carriers [14]

$$\partial N_D^i/\partial t = sI(N_D - N_D^i) - \gamma n N_D^i\,, \tag{3}$$

$$\partial n/\partial t - \partial N_D^i/\partial t = (1/q)\,\partial J/\partial z\,, \tag{4}$$

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$$\partial E/\partial z = (-q/\varepsilon_s)(n + N_A - N_D^i). \quad (5)$$

The current J includes diffusion, drift, and displacement terms [15]

$$J = qn\upsilon(E) + q\,\partial[D(E)n]/\partial z - \varepsilon_s\,\partial E/\partial t. \quad (6)$$

The independent variables are the lateral axis along which the current flows z and the time t. The dependent variables are $I(z,t)$, $n(z,t)$, the electron number density, $N_D^i(z,t)$, the number density of ionized donors, $J(z,t)$, the current density, $E(z,t)$, the space-charge field inside the crystal, $\upsilon(E)$, the electron drift velocity, and $D(E)$, the diffusion coefficient that depends on the electric field E. Note that for $E \ll E_s$ one can approximate $\upsilon(E) \cong \mu E$ and obtain through the Einstein relation [15] $D = \mu k_B T/q$, where k_B is Boltzmann's constant and T is the absolute temperature. Relevant crystal parameters are N_D, the total donor density, N_A, the density of negatively charged acceptors, s, the photoionization cross section, γ, the recombination rate coefficient, ε_s, the low frequency dielectric constant, and $-q$, the charge on the electron. The boundary condition is the dc voltage V applied between electrodes spaced by ℓ ($\ell \gg \Lambda$). The goal of this calculation is to find an explicit relation between the space-charge field $E(z,t)$ and the interference grating $I(z,t)$ of Eq. (2).

Solving Eqs. (3)–(6) for $E(I)$ is complicated. As the first step, we can neglect the diffusion term in Eq. (6) based on the fact that separate multiple domains exist only if the separation between them is larger than some critical distance, evaluated from Kroemer's criterion [2]. The domains are excited by $I(z,t)$, thus they will form at the maxima of the intensity pattern (separated by Λ). Using the above GaAs parameters and $\varepsilon_s = 13.2$, $E \approx 3E_s$, and $n \approx 10^{14}\ \mathrm{cm}^{-3}$, we find $qn\upsilon(E) \approx 3 \times 10^6\ \mathrm{F\,V\,m^{-2}\,sec^{-1}}$. Thus, if $|q\,\partial[D(E)n]/\partial z| \ll 10^6\ \mathrm{F\,V\,m^{-2}\,sec^{-1}}$, the diffusion term is negligible with respect to the drift term. To estimate when this occurs, we assume that the magnitude of D does not deviate much from the Einstein relation, thus $D \approx \mu k_B T/q$. This gives the condition $|\mu k_B T n K| \ll 10^6$, which is satisfied for $\Lambda = 2\pi/K \gg 1\ \mu\mathrm{m}$. We will use grating periods on the order of tens of microns, thus we neglect the diffusion term in Eq. (6).

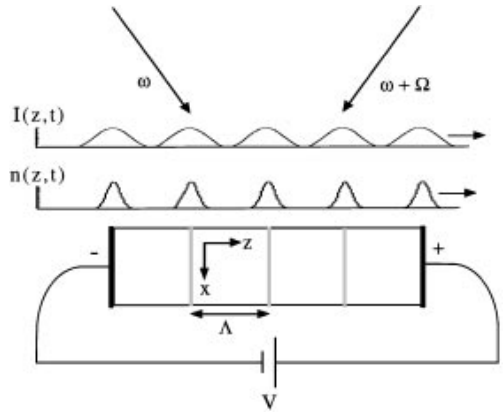

FIG. 1. Proposed experimental setup for the observation of the photorefractive Gunn effect.

We solve Eqs. (3)–(6) using an asymptotic expansion for sinusoidal steady state

$$E(z,t) = E_0 + mE_1\cos(Kz + \Omega t) + m^2E_2\cos(2Kz - 2\Omega t) + \cdots, \quad (7)$$

$$n(z,t) = n_0 + mn_1\cos(Kz + \Omega t) + m^2n_2\cos(2Kz - 2\Omega t) + \cdots, \quad (8)$$

$$N_D^i(z,t) = N_{D0}^i + mN_{D1}^i\cos(Kz + \Omega t) + m^2N_{D2}^i\cos(2Kz - 2\Omega t) + \cdots, \quad (9)$$

which converges when $m \ll 1$ and all the coefficients (E_i, n_i, N_{Dj}^i; $j = 0, 1, 2, \ldots$) are of the same order in each of the expansions of Eqs. (7)–(9). The sinusoidal steady state situation exists whenever the formation time of the Gunn domains is much shorter than $2\pi/\Omega$. Typical values are 3 psec domain formation time [2] and $2\pi/\Omega = 1$ nsec or longer.

The zero-order solutions are the usual lowest order solutions for biased photorefractive materials [14]: $N_{D0}^i = N_A$, $n_0 = sI_0(N_D - N_A)/\gamma N_A$ (for $n_0 \ll N_A$), and $E_0 = V/\ell$.

The first-order terms are found using a second approximation. Gunn domains evolve most easily when the negative differential resistance is the largest, and, at this point, they require the smallest applied field. For the GaAs example, this occurs for $E \approx 3E_s$. In a relatively large region around this point, υ is roughly a linear function of E. We approximate Eq. (1) for the above GaAs example as

$$\upsilon \cong \upsilon_s[2.21 - 0.245(E/E_s)] = 2.21\upsilon_s - 0.245\mu E \quad (10)$$

for the range $2.5 \le E/E_s \le 3.5$ (which implies that $|mE_1| \le E_s/2$). This approximation is shown in Fig. 2. Using Eq. (10) and the expansions of Eqs. (7)–(9) into Eqs. (3)–(6) give the equations for the first-order terms

$$i\Omega N_{D1}^i = sI_0(N_D - N_A - N_{D1}^i) - \gamma(n_0N_{D1}^i + n_1N_A), \quad (11)$$

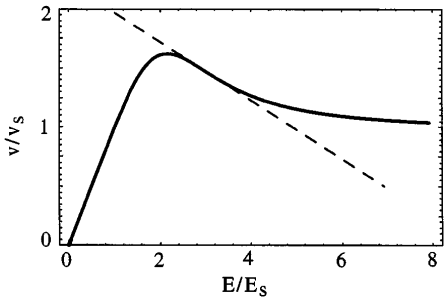

FIG. 2. Drift velocity of electrons as a function of electric field (normalized to the saturation field E_s) for GaAs with mobility $\mu = 0.5\ \mathrm{m^2\,V^{-1}\,sec^{-1}}$. The dashed line shows the linear approximation for $2.5 \le E/E_s \le 3.5$.

$$[\Omega - 2.21v_sK + 0.245\mu KE_0]n_1 - \Omega N_{D1}^i = -[i\Omega\varepsilon_s/q + 0.245\mu n_0]KE_1\,, \quad (12)$$

$$-iKE_1 = (q/\varepsilon_s)[n_1 - N_{D1}^i]\,. \quad (13)$$

We seek synchronous solutions, i.e., solutions in which the movement of all the unknowns is *locked* together for all t and z. This is equivalent to requiring $\Omega = \alpha K$ which leads to $\cos(Kz + \Omega t) = \cos[K(z + \alpha t)]$, where α is the phase velocity (a constant). Using this, and substituting Eq. (13) into Eqs. (11) and (12), produces

$$[i\alpha K + sI_0N_D/N_A + \gamma N_A]n_1 = s\,I_0(N_D - N_A) + [\alpha(\varepsilon_s/q)K^2 - i(\varepsilon_s/q)sI_0(N_D/N_A)K]E_1\,, \quad (14)$$

$$[-2.21v_s + 0.245\mu E_0]n_1 = -0.245\mu n_0E_1\,. \quad (15)$$

The last equation yields $E_1 = 6E_s(n_1/n_0)$ for $E_0 \approx 3E_s$. From Eq. (14) we get

$$\left[\left(sI_0\frac{N_D}{N_A} + \gamma N_A - 6E_s\frac{\alpha\varepsilon_s}{qn_0}K^2\right) + i\left(\alpha - 6E_s\frac{\varepsilon_s}{qn_0}sI_0\frac{N_D}{N_A}\right)K\right]n_1 = sI_0(N_D - N_A)\,. \quad (16)$$

From Eq. (16) it is obvious that n_1 can have both a component that is in phase with the interference grating [of Eq. (2)] and a component that is phase shifted by $\pi/2$ [the first and second terms on the left hand side of Eq. (16), respectively]. While the in-phase component can be eliminated only for some special values of K, the $\pi/2$ phase-shifted component can be eliminated (for *all* K) by a proper choice of setting $\alpha = 6E_s(\varepsilon_s/qn_0)sI_0N_D/N_A = 6E_s\varepsilon_s/qP_d\tau_R$, where $P_d = (N_D - N_A)N_A/N_D$ is the effective trap density and $\tau_R = (\gamma N_A)^{-1}$ is the recombination time. For most photorefractive media [14], $P_d \approx 10^{16}$ cm^{-3} and $\tau_R \approx 10^{-10}$ sec. Tuning the value of α can be performed by varying the voltage V or by adjusting the frequency difference between the optical waves Ω. This choice of α leads to

$$n_1 = \frac{sI_0(N_D - N_A)}{sI_0N_D/N_A + \gamma N_A - \alpha^2K^2N_A/sI_0N_D}\,. \quad (17)$$

The interference grating is now in phase with both n_1 and E_1, as observed from Eq. (15). As shown below, E_1 modulates the refractive index in phase with the interference pattern, leading to a nonlinear phase-coupling process (at *all* z and t) between the plane waves which form $I(z, t)$.

The last term in the denominator of Eq. (17) is proportional to $1/I_0$. Thus, two regimes of intensity exist, one where I_0 is sufficiently large so that this term is negligible with respect to sI_0N_A/N_D and to γN_A, and one where I_0 is sufficiently small so that this term is dominant.

Consider first the *high intensity regime.* The solutions are now simple:

$$n_1 = sI_0(N_D - N_A)/(sI_0N_D/N_A + \gamma N_A),$$
$$E_1 = 6E_sn_1/n_0 = 6E_s\gamma N_A/(sI_0N_D/N_A + \gamma N_A)\,. \quad (18)$$

Since E_1 decreases with increasing I_0, one can find the *lowest* intensity that still allows us to be in this regime. The approximation which simplifies Eq. (17) into (18) is justified for $sI_0 \gg (N_A/N_D)\tau_R(\alpha K)^2$. For GaAs parameters (above, and in Ref. [14]) $N_D = 10^{18}$ cm^{-3}, $N_A = 10^{16}$ cm^{-3}, $\alpha \approx 750$ m/sec, $s \approx 10^{-3}$ m^2 sec^{-1} W^{-1}, and for $\Lambda = 30$ μm (which implies $\Omega \approx 10^7$ sec^{-1}), the minimal intensity requirement is $I_0 \gg 2.5$ kW/cm^2.

So far we have shown that a moving sinusoidal intensity pattern incident upon a semiconductor biased at negative differential resistance gives rise to a periodic free carrier distribution of the same period that moves *phase locked* to the optical grating. The Gunn effect, however, creates *high-field* domains, i.e., regions where the space-charge field is *larger* than the bias field. This space-charge distribution will indeed result in a sequence of high-field (Gunn) domains if Kroemer's criterion

$$n_1L > 3\varepsilon_sv_s/q|\mu_d| \quad (20)$$

is satisfied [where L is the length of a Gunn device (in the case of a single domain) or the distance between adjacent domains (for multiple domain) and $\mu_d = dv/dE$]. In our case of $L = \Lambda = 30$ μm and $\mu_d = -0.245\mu$, we find that n_1 must be larger than 5×10^{13} cm^{-3}. From Eq. (18), this requires a minimum intensity of $I_0 = 50$ kW/cm^2. Since this minimum intensity is inversely proportional to Λ, a longer grating period reduces this requirement accordingly. It is, however, within reasonable experimental possibilities since two wave mixing experiments in GaAs have been performed with psec [16] and nsec [17] light pulses at more than 10 MW/cm^2 intensities. Under these conditions, an optically triggered sequence of high-field domains will form. The space-charge field for $\Lambda = 30$ μm and $\alpha \approx 750$ m/sec, with $I_0 = 100$ kW/cm^2 ($n_1 = 10^{14}$ cm^{-3}), is (to the first order in m)

$$E(z, t) = E_0 + mE_1\cos(Kz + \Omega t) \approx 3E_s\{1 + 2m\cos[K(z + \alpha t)]\}, \quad (21)$$

where the frequency difference between the optical waves is $\Omega = \alpha K = 2\pi \times 25$ MHz. For self-consistency with our expansion, we require $m \ll 1$. Thus, for $m = 0.1$, we expect the periodic part of E to be roughly $0.6E_s = 900$ V/cm. However, with I_0 that satisfies Kroemer's criterion we expect to see high-field domain nucleation that can even *exceed the value of* E_0. This can be found,

for example, by solving Eqs. (3)–(6) to additional orders of m and finding the high-field domains explicitly. The expansion converges, but additional terms do contribute to a high-field domain that moves with the first-order term of Eq. (21). Other ways to describe the domain nucleation process is through the transient solution of these equations or via stability analysis [18].

In the *low intensity regime,* the last term in the denominator of Eq. (17) is dominant. The solutions are

$$n_1 = -(sI_0)^2 (N_D - N_A) N_D / \alpha^2 N_A K^2 ,$$
$$E_1 = -6E_s \left(\frac{sI_0 N_D}{\alpha^2 \tau_R N_A} \right) \frac{1}{K^2} \cong \left(\frac{-q^2 sI_0 N_D N_A \tau_R}{6E_s \varepsilon_s^2} \right) \frac{1}{K^2} . \quad (22)$$

To be in this regime, $sI_0 \ll (N_A/N_D)\tau_R(\alpha K)^2$ must be satisfied. On the other hand, Kroemer's criterion implies that for $\Lambda \geq 30$ μm intensities higher than 50 kW/cm^2 are required for the formation of high-field domains. For $\Lambda \geq 30$ μm and the above GaAs parameters, we find that the intensity must be *much smaller* than 2.5 kW/cm^2. Thus, *high-field (Gunn) domains cannot form in this regime;* i.e., the space-charge field is sinusoidal (for $m \ll 1$) *despite* the negative differential resistance.

The space-charge field $E(z, t)$ gives rise to a periodic change in the refractive index $\Delta n(z, t)$ that moves in phase with it [19] [and hence with $I(z, t)$] via Pockel's effect $\Delta n(E) = -\frac{1}{2} n^3 r_{41} E$, where n is the refractive index and r_{41} the electro-optic coefficient (3.3×10^{-12} and 1.43×10^{-12} m/V for GaAs at $\lambda = 1$ μm, respectively). Here, we are interested in the high intensity regime that will give high-field domains. Even if we ignore the expected (large) contribution to the field from the high-field (Gunn) domains, we have found $mE_1 =$ 900 V/cm for the GaAs example. Thus, we expect to find periodic Δn with *at least* an amplitude of 2.3×10^{-6}. This refractive index modulation (which is a real-time polarization hologram) results in nonlinear phase coupling between the optical waves [19]. The magnitude of the nonlinear coupling can be estimated given the interaction length in the crystal. For a thickness of $X = 1$ cm we expect the total nonlinear phase to be $2\pi \Delta n X/\lambda \approx$ 0.15 rad. Thus, even if we do not include the high-field enhancement of E_1, we can still get an appreciable nonlinear effect. High-field enhancement and illumination with an interference of a large modulation depth ($m \sim 1$) can enhance the nonlinear refractive index change to $\Delta n \sim 10^{-4}$ and the nonlinear phase coupling around 15 rad, which is a large effect.

The main importance of this photorefractive Gunn effect is in being a new optically nonlinear effect which results from a highly nonlinear electric effect (the Gunn effect) and a second-order optical nonlinearity (Pockel's effect). When triggered by phase-locked interference fringes, this "natural" high-field behavior of the semiconductor medium can also drive a large nonlinear optical effect by greatly enhancing the local field and creating a moving real-time polarization hologram. Potential applications include optical switching, high efficiency wave mixing, fast and sensitive detection of temporal optical signals embedded in noise, and the ability to convert this optical information to electric signals directly.

In conclusion, we predict a new optically nonlinear effect in semiconductors: the photorefractive Gunn effect, which stems from an electric nonlinearity (Gunn effect), and a second-order optical nonlinearity (Pockel's effect).

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