Steady-State Photovoltaic Effect in Asymmetrical Graded Superlattices

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In a graded InAlGaAs superlattice grown by molecular-beam epitaxy at 500 °C, we have observed a novel steady-state photovoltaic effect which arises from an internal polarization field under above-band-gap ($h\nu > E_g$) illumination. The saturation value is ~0.1 V and the response time to the illumination is limited by the oscilloscope resolution to ~2 ns. The observations are in quantitative agreement with a theoretical model by which we fit our data and extract a minority-carrier lifetime τ_e of ~30 ps. This effect is substantially reduced in a second wafer grown at 550 °C, in which τ_e is expected to be long.

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In a sawtooth superlattice structure, as shown in Fig. 1(a), a *transient* electrical polarization phenomenon was previously observed [1]. The structure was heavily doped in p type. In thermal equilibrium, the Fermi energy E_F pins the valence band at a constant level, and the built-in electric field exists in the conduction band. Upon above-band-gap ($h\nu > E_g$) illumination, electron-hole pairs are generated in the whole structure. The built-in field sweeps the electrons toward the lower-band-gap end and separates them from the holes. Thus, a polarization field is created. The polarization field unbalances the constant E_F, and is a nonequilibrium phenomenon. In the experiment, it decayed rapidly at a picosecond rate, and no steady-state polarization field was measured even in the presence of cw illumination.

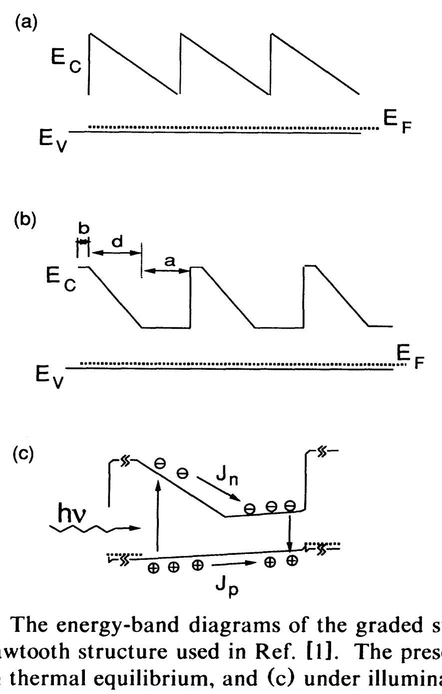

FIG. 1. The energy-band diagrams of the graded structures. (a) The sawtooth structure used in Ref. [1]. The present structure (b) in thermal equilibrium, and (c) under illumination. E_C is the conduction band edge, and E_V is the valence band edge.

Recently, a theoretical model [2] has been proposed in which a steady-state polarization field can be created if certain parameters of the graded structure are modified. Most importantly, a reduction of the minority-carrier lifetime τ_e is required, which can be achieved by, e.g., growing the structure at low temperatures [3]. τ_e determines the saturation value of the steady-state V_p, while interestingly τ_e is irrelevant to the transient effect [1]. The model can be briefly described with the aid of Figs. 1(b) and 1(c), where the layer thicknesses d and a are ~50 nm. Under cw illumination of photon energies above the band gap of layer d, electron-hole pairs are generated in both the d and a layers. The built-in field sweeps electrons from layer d to layer a instantaneously within a time period of less than 1 ps. Consequently, the electron density is higher in layer a, and electron-hole recombination occurs predominantly in this layer. A constant supply of holes from layer d to layer a must therefore be provided to satisfy the recombination. The steady state is established when the hole current density J_p equals that of the electrons J_n. As seen from Fig. 1(c), J_n can be approximated as the total generation rate of electrons in layer d, and be expressed as

$$J_n = e\alpha(I_\nu/h\nu)d\,, \tag{1}$$

where α is the absorption coefficient in layer d, I_ν is the light intensity, $h\nu$ is the photon energy, and $I_\nu/h\nu$ is the photon flux. Since no built-in electric field exists in the valence band, a steady-state polarization field E arises, which drags the holes and supports J_p. Thus, J_p is given by $ep\mu_h E$, where p and μ_h are, respectively, the concentration and mobility of holes. Because of the asymmetry of the superlattice, J_n is directed along the conduction-band gradient. This ensures the unidirectional flow of holes. As a result, E in each period of the superlattice adds up to a finite V_p between the top and the bottom of the superlattice:

$$V_p \sim NE\frac{d+a}{2} = N\alpha\frac{I_v}{h\nu}d\frac{1}{\mu_h(p_0+\tau_e\alpha I_v/h\nu)}\frac{d+a}{2}, \quad (2)$$

where N is the number of superlattice periods and p_0 is the doping concentration in the superlattice. From Eq. (2), the saturation value of V_p under intense illumination, i.e., when I_v becomes high enough, is limited by the condition $\tau_e\alpha I_v/h\nu \gg p_0$. Substituting the experimental values [1] into Eq. (2) and using $\mu_h \gtrsim 100$ cm^2/V s [4] and $\tau_e \sim 10^{-8}$ s [5] for their AlGaAs material grown by molecular-beam epitaxy (MBE) under normal conditions, one finds that V_p is limited to $\lesssim 10^{-4}$ V, too small to have been observed.

In this Letter, we report experiments on a graded InAlGaAs structure which was grown by MBE at 500 °C. We have observed a steady-state polarization field which gives rise to a photovoltage V_p of as high as 0.1 V. The intrinsic response time of this novel steady-state photovoltaic effect is the dielectric relaxation time, which is estimated to be about 2 ps in our samples. Experimentally, we measured an impulse response of 2 ns, limited by the time resolution of the oscilloscope. The observations are in quantitative agreement with the model [2]. By fitting the data by the model, we extract a τ_e of $\sim$30 ps in this low-temperature-grown structure. In a second wafer grown at 550 °C, where τ_e is expected to be long, the observed photovoltage was substantially reduced, confirming our explanations.

The graded-gap superlattice was grown by MBE on a (100)-oriented heavily p-type–doped InP substrate. All layers are Be doped. Ten periods of the Fig. 1(b) structure were grown between two 1-μm $In_{0.52}Al_{0.48}As$ window layers which were both doped to $\gtrsim 10^{19}$/cm^3. The a layer in each period is 50-nm $In_{0.53}Ga_{0.47}As$ doped to 5×10^{16}/cm^3, the d layer is 50-nm graded $In_{0.53}(Al_xGa_{1-x})_{0.47}As$ doped to 5×10^{16}/cm^3 with x ramped from 0 to 1, and the b layer is 5-nm $In_{0.52}Al_{0.48}As$ doped to 10^{18}/cm^3. The top window layer is followed by a 70-nm $In_{0.53}Ga_{0.47}As$ contact layer doped to $\gtrsim 10^{19}$/cm^3. For the purpose of reducing τ_e in the active regions, the superlattice was grown at 500 °C, while the window layers and the contact layer were grown at 550 °C. The unidirectional grading of the d layers and the abrupt interface between layers b and a were achieved by controlling the openings of the Al and Ga cells of the MBE machine.

The devices are 500 μm×500 μm squares defined by mesa etching 2.5 μm from the surface down to the middle of the bottom window layer. Each device has a top contact of 50 μm in diameter located 25 μm from one edge of the device and a contact to the bottom window layer outside the mesa area. The contact metal is composed of 80-nm Be, 30-nm Ti, and 120-nm Au, and the contacts were formed by a 15-s alloy at 390 °C. Each device has a linear I-V characteristic of $\sim$8-Ω slope between the contacts, indicative of good Ohmic contacts. To measure μ_h, we have also fabricated small devices which have top contacts 12.5 μm in diameter, and, after the mesa etching, the superlattice region was $\sim$9 μm in diameter. Such devices result in a linear I-V of $\sim$400-Ω resistance between the contacts. Since only the superlattice region is doped to 5×10^{16}/cm^3 and the other layers of the whole structure are heavily doped to $\gtrsim 10^{19}$/cm^3, the measured resistance is from the superlattice region. Thus, from the geometric parameters and the doping level, μ_h along the growth direction in the superlattice region is deduced to be $\sim$50 cm^2/V s. This mobility is much lower than would be expected from a similar material grown at ordinary MBE temperatures. According to Ref. [4], μ_h in $In_{0.53}Ga_{0.47}As$ doped to 5×10^{16}/cm^3 is of the order of 1000 cm^2/V s.

A Quantronix 114 Nd-doped yttrium-aluminum-garnet laser equipped with repetitive Q-switching and mode-locking capabilities was used in our experiment. The laser wavelength at 1.064 μm corresponds to a photon energy $h\nu = 1.165$ eV. As a consequence, only $\sim$60% of the d layer was absorbing and the $In_{0.52}Al_{0.48}As$ window layers were transparent to the laser light. The Q-switched pulse duration can be varied from 200 ns to 2 μs by varying the Q-switch repetition rate from 0.1 to 20 kHz. In contrast, the mode-locked pulses have a fixed pulse width of 100 ps at a 100-MHz repetition rate. The average laser power P_{av} illuminating the device was controlled by rotating the optical axis of a half-wave plate with respect to a polarizing beam splitter. The laser intensity I_v is further determined by the laser spot size on the device surface, which was varied by accurately moving the device with respect to a focusing microscope objective lens. The radius ρ of the $1/e$ laser intensity profile on the device surface was measured *in situ* with an optical fiber of 6-μm core diameter. During the experiment, a photoconductive Si p-i-n diode with a response time shorter than 5 ns was used to monitor the Q-switched pulses as well as the mode-locked pulse train. The mode-locked pulse width was independently measured with an autocorrelator. The photovoltage V_p of the device generated under the illumination of the laser light was measured between the top and the bottom contacts. No bias was applied to the device.

In contrast to the previous experiment by Capasso *et al.* [1], a steady-state V_p was observed under cw illumination in our experiment. For the first time, this is achieved in a graded superlattice. To increase the signal level and the range of measurement while maintaining P_{av} at a low level, we used the Q-switched pulses. We note here that the signal generated by a Q-switched pulse represents the steady-state response of the device for the following reasons: (1) As will be discussed later, the response time of the device was faster than $\sim$2 ns, which was 2–3 orders of magnitude shorter than the Q-switched pulse duration; (2) the peak value of V_p depended only on the peak intensity but not on the duration or the repetition rate of the Q-switched pulses; and (3) in the range of linear response at low light intensities, the signal V_p followed the Q-switched pulse shape exactly while the pulse duration was varied from 200 ns to 2 μs. Figure 2 displays the wave form of V_p together with that of a 300-ns laser pulse used to generate the photovoltage. These traces were obtained on a Tektronix 11302 oscilloscope with a 50-Ω channel input impedance which had a time resolution of $\sim$2 ns. At a low peak laser-pulse intensity, the device response is linear and V_p coincides exactly with the shape of the laser pulse, which is demonstrated in Fig. 2(a). However, as is shown in Fig. 2(b), V_p saturates at a high peak laser intensity. The maximum saturation value of V_p is in qualitative agreement with Eq. (2).

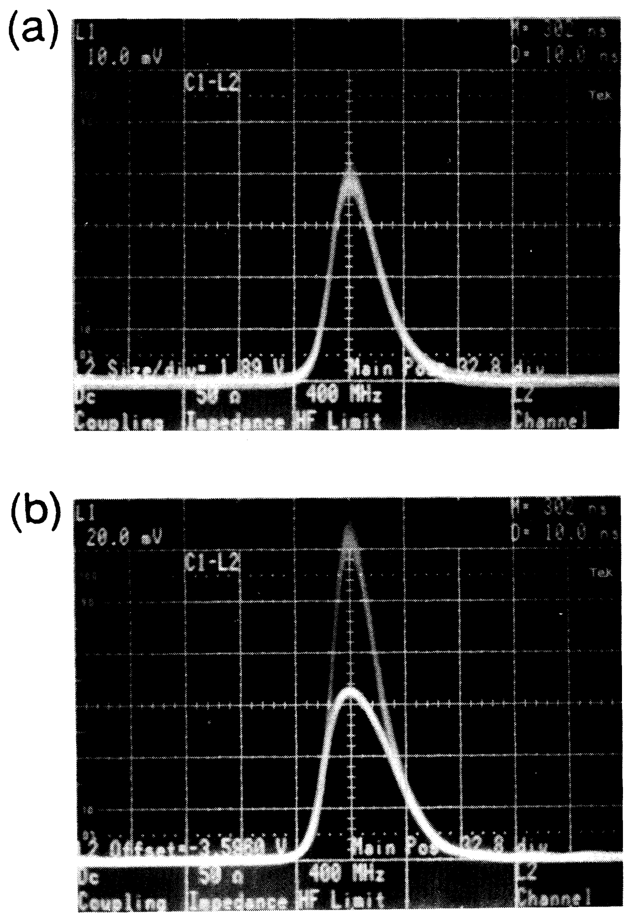

FIG. 2. The photovoltage signal (lower traces) generated by a 300-ns laser pulse (upper traces) which is monitored by a Si p-i-n detector. (a) When the laser intensity is low, the photovoltage signal follows linearly and coincides with the laser pulse signal. (b) When the laser intensity becomes high, the photovoltage saturates. The horizontal axis is 302 ns per division. The vertical axis is 10 and 20 mV per division in (a) and (b), respectively.

Before presenting quantitative results, we discuss further evidence for V_p being a photovoltage generated in the graded superlattice. First of all, both the signal V_p from the device and that from the monitoring Si p-i-n diode were displayed on a Tektronix 2246 oscilloscope with a 1-MΩ channel input impedance. In comparison to the wave forms obtained through the 11302 oscilloscope of 50-Ω input impedance and displayed in Fig. 2, both the peak value and the shape of the signal V_p remained almost unchanged, while the peak value of the signal from the photoconductive Si p-i-n diode was increased by orders of magnitude with a long decay time. As the laser beam was focused to a spot of 0.1 mm in radius on a portion of the device surface away from the top Ohmic contact, V_p existed as well and depended only slightly on the location of laser illumination, indicating that V_p did not arise from the contact. We further etched away the superlattice layers, made the same Ohmic contacts to the bottom window layer and to the back of the substrate, and repeated the same measurements under similar conditions. No V_p could be observed between these contacts, confirming that the observed photovoltage was generated in the superlattice layers.

Figure 3 shows the measured V_p as a function of I_ν in a log-log plot for two different laser spot sizes with radii $\rho=0.81$ and 0.1 mm, respectively, on the device surface. The data points were obtained at four different values of P_{av} with Q-switched pulses varying in duration from 200 ns to 2 μs. V_p is the peak value of the device signal and I_ν is the peak laser intensity. With $\rho=0.81$ mm, the laser spot covered the entire device quite uniformly, while only $\sim$12% of the device surface was illuminated with $\rho=0.1$

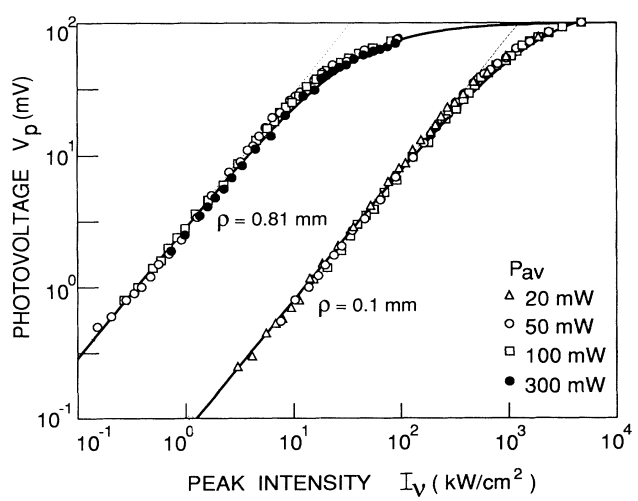

FIG. 3. Photovoltage as a function of the laser intensity. The solid curves are the best fit of the data points by Eq. (2), and the dotted lines of slope 1 are the extension of the linear region of the solid curves.

mm. Consequently, V_p was substantially reduced for the same peak laser intensity as ρ was changed from 0.81 to 0.1 mm. However, in both cases, V_p first increases linearly with I_ν, as is indicated by the dotted lines of slope 1 in Fig. 3, and saturates at a value of ~ 0.1 V at high intensities.

The data shown in Fig. 3 also agree quantitatively with Eq. (2). From Eq. (2), the linear-response region is when $\tau_e \alpha I_\nu / h\nu \ll p_0$. In this region, V_p is completely determined by I_ν, μ_h, α, p_0, and device dimensions. We note that the effective d layer is only ~ 30 nm for $h\nu = 1.165$ eV. As discussed earlier, μ_h has been independently determined to be ~ 50 cm^2/V s. Therefore, only α is left as a free parameter for fitting the tens of data points in the linear region. For $\rho = 0.81$ mm, the linear region is best fitted with $\alpha = 10^4$/cm with 10% accuracy. In reality α is not uniform in the active d and a layers. Nevertheless, the fitting value for α is very close to the average absorption coefficient of the absorbing active regions [6]. Using μ_h and α, we can further fit the entire set of data points including the saturation region to extract τ_e. The solid curves in Fig. 3 are the fit to the data points. For $\rho = 0.81$ mm, the extracted τ_e is 30 ps. For $\rho = 0.1$ mm, the data points can still be fitted. However, since the device was only partially illuminated by the laser beam, the fitting parameters are difficult to interpret.

The extracted value of τ_e is of physical importance. It is much lower than that expected for a normally grown material [5]. We have grown a second wafer of the same layered structure at 550 °C, in particular, including the superlattice regions. Devices were fabricated with the same procedures. However, the observed V_p was substantially reduced under the same experimental conditions. This confirms the importance of the low τ_e and is consistent with the theory [2].

The response speed of the device was studied with the 100-ps mode-locked pulses. Although the signal generated by a cw mode-locked pulse train can be observed, it was at the noise level of a sampling oscilloscope and could not be studied in detail. In order to generate clear signals of significant magnitude, the Q-switched mode-locked pulse train was used. This signal can be clearly displayed on the 11302 oscilloscope, but the time resolution is limited to 2 ns by the oscilloscope. The upper trace of Fig. 4 shows the central portion of a train of signal V_p pulses generated by the Q-switched mode-locked pulses. The lower trace is the signal from the monitoring Si p-i-n diode. The signal V_p has a full width at half maximum (FWHM) of ~ 2 ns, equivalent to the oscilloscope resolution, while the signal from the Si p-i-n diode has a FWHM of ~ 3.6 ns. Therefore, the device is faster than the Si p-i-n diode and the 2-ns oscilloscope response time. The ultimate speed is theoretically limited only by the intrinsic dielectric relaxation time, which is estimated to be ~ 2 ps in our devices. Further experiments along this line are currently in progress.

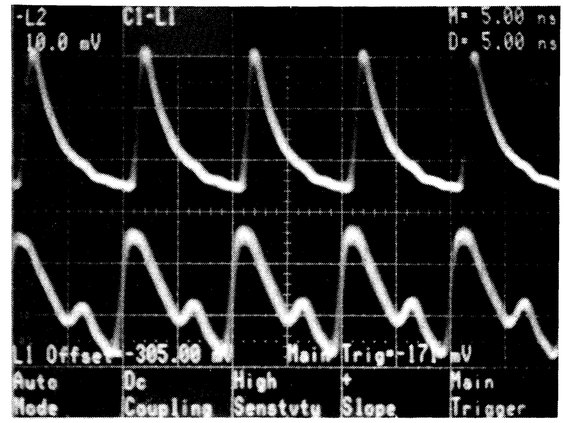

FIG. 4. The photovoltage (upper trace) in response to the Q-switched mode-locked laser pulses (lower trace) which have a width of 100 ps and a separation of 10 ns. The Q-switched envelope is 300 ns. The time resolution of the oscilloscope is 2 ns. The horizontal axis is 5 ns per division, and the vertical axis for the photovoltage is 10 mV per division.

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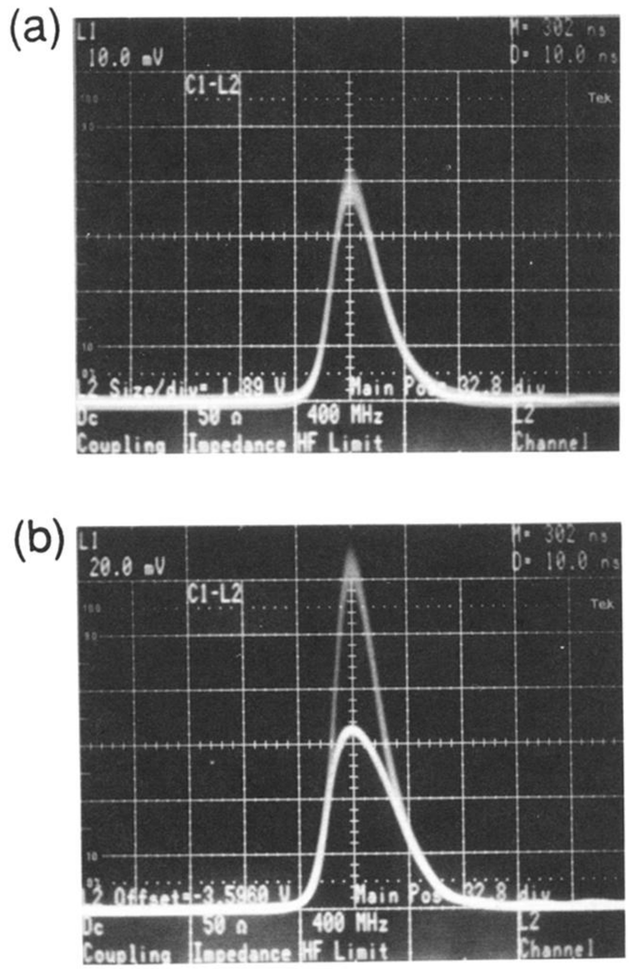

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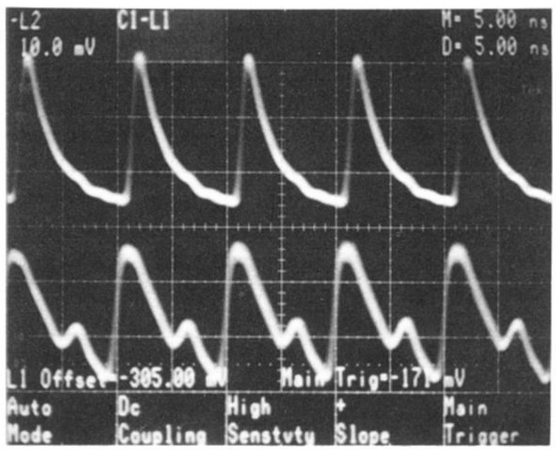

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