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Spatial modulation of the Fermi level by coherent illumination of undoped GaAs

D. D. Nolte,* D. H. Olson, and A. M. Glass
AT&T Bell Laboratories, Holmdel, New Jersey 07733
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The Fermi level in undoped GaAs has been modulated spatially by optically quenching $EL2$ defects. The spatial gradient of the Fermi level produces internal electric fields that are much larger than fields generated by thermal diffusion alone. The resulting band structure is equivalent to a periodic modulation-doped *p-i-p* structure of alternating insulating and *p*-type layers. The internal fields are detected via the electro-optic effect by the diffraction of a probe laser in a four-wave mixing geometry. The direct control of the Fermi level distinguishes this phenomenon from normal photorefractive behavior and introduces a novel nonlinear optical process.

Undoped, semi-insulating GaAs is produced through the compensation of shallow acceptors by the deep defect level known as $EL2$. This defect can be optically transformed from a deep donor into a metastable state that is electrically inactive, thereby transforming the crystal from a semi-insulating state into a metastable conducting state. The common signature of this optical transformation is photocapacitance quenching,[1,2] measured by monitoring the capacitance of a space-charge region of a conducting sample as the region is illuminated by photons with an energy near 1.2 eV. In this Rapid Communication, we describe the use of coherent laser beams to spatially modulate this quenching of $EL2$ at low temperatures in bulk semi-insulating GaAs. The quenching produces a direct modulation of the Fermi level, which drives a transfer of charge from the illuminated to the dark regions of the crystal. The large internal electric fields that result are much greater than fields produced by thermal diffusion alone. The spatially modulated electric field produces a diffraction grating through the electro-optic effect. The diffraction grating is probed with a second laser, and the presence of the internal electric fields is measured as a dramatic enhancement of the four-wave mixing efficiency.

The pump laser used was a cw 1.06 μm YAG:Nd laser (YAG denotes yttrium aluminum garnet). This source is optimal for quenching of $EL2$. The typical exposure received to quench $EL2$ is 100 mJ/cm^2. The pump laser is split into two beams that intersect inside the sample and interfere coherently, producing fringes of constructive and destructive interference. The spatial frequency of the fringes is $K=2\pi/\Lambda$, where $\Lambda=\lambda/2\sin\theta=3.4$ μm, $\lambda=1.06$ μm is the wavelength of the pump laser, and $\theta=9°$ is the half angle between the two arms of the pump laser. For temperatures above 140 K, semi-insulating GaAs behaves as a normal photorefractive semiconductor.[3,4] Photocarriers generated from $EL2$ diffuse thermally from the regions of high photocarrier density to the dark fringes and are retrapped on defect sites, producing an internally modulated space-charge field $E_{sc}(Kx)$. The value of the diffusion-generated space-charge field is[5]

$$E_{sc}=Kk_BT/e\,. \tag{1}$$

Below 140 K, on the other hand, much larger values of $E_{sc}(Kx)$ are created by $EL2$ quenching. The magnitude of the electric field can be measured from the concomitant change in the refractive index through the electro-optic effect $\Delta n(Kx)=-\frac{1}{2}n^3r_{41}E_{sc}(Kx)$, where r_{41} is the electro-optic coefficient. In a nondegenerate four-wave mixing geometry, the modulated index of refraction acts as a diffraction grating that diffracts a probe beam. Our probe beam is from a 1.32 μm YAG:Nd laser. The signal is measured by chopping the probe beam and detecting the diffracted beam with an $In_xGa_{1-x}As$ photodiode. The diffraction efficiency is given by

$$\eta=\sin^2\left(\frac{\pi\Delta n_1 d}{\lambda\cos\theta}\right),$$

where d is the thickness of the sample and Δn_1 is the first Fourier amplitude of the modulated index. The temperature dependence of the steady-state diffracted signal in GaAs is shown in Fig. 1. There is a gradual decrease of the diffraction efficiency below room temperature because of the decreasing diffusion field. Above room temperature there is a rapid decrease of the diffraction efficiency

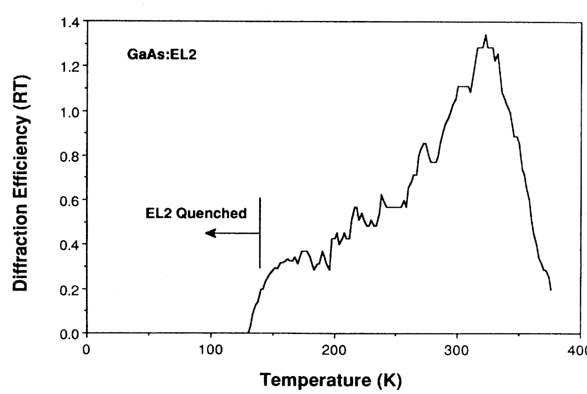

FIG. 1. Temperature dependence of the four-wave mixing diffracted signal of a GaAs:$EL2$ sample cooled down under pump illumination. The sample exhibits normal photorefractive behavior above 140 K. Below this temperature, the diffraction drops as $EL2$ is quenched and the crystal is no longer semi-insulating.

caused by dielectric relaxation from carriers thermally excited from $EL2$.

The diffraction efficiency behaves qualitatively differently below 140 K. When the sample is cooled with the pump illumination on, the diffraction efficiency is extinguished for temperatures below 140 K. This is seen as the knee near 140 K in Fig. 1. However, when the sample is cooled below 140 K without illumination, and the pump laser is then turned on, there is a rapid increase of the diffracted signal as $EL2$ centers are quenched in the regions of greatest intensity. For long exposures, the diffraction efficiency decreases as $EL2$ centers are slowly quenched even in the region of lowest intensity. This diffraction transient is shown in Fig. 2 at 60 K for a pump power of 20 mW/cm^2 in each arm. The diffracted signal reaches a maximum 2 sec after the pump is turned on. The maximum signal is almost three times greater than the diffraction efficiency at room temperature. If the pump beams are kept on, the signal decreases with a time constant of around 3 sec. However, if the pump beams are turned off as the signal reaches its maximum, then the Fermi-level modulation persists for the duration of the experiments, exceeding many hours. The gratings can be erased by warming the sample briefly to 140 K.

The maximum diffraction efficiency of the quenched-in gratings can be translated into a value for the first Fourier component of the internal electric field. Using the room-temperature diffraction efficiency for calibration, the largest diffraction efficiency achieved by $EL2$ quenching was nearly three times the room-temperature value. Given that the internal diffusion field from Eq. (1) at $T=295$ K is 470 V/cm, the maximum electric field amplitude is $E_t=\sqrt{3}E_{\mathrm{RT}}=812$ V/cm. This value is more than eight times larger than the diffusion field at 60 K. The charge density N_t transferred from the illuminated to the dark regions of the crystal to build in this field is derived from the expression

$$E_t=\frac{eN_t}{K\epsilon\epsilon_0}$$

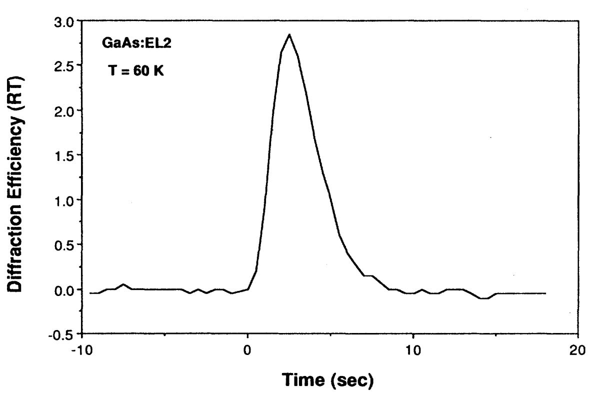

FIG. 2. Diffraction transient at $T=60$ K, after the sample was cooled in the dark, measured relative to the room-temperature diffraction efficiency. The YAG pump beam was turned on at $t=0$ sec. The maximum diffracted signal at $t=2.5$ sec was three times greater than the value at room temperature.

to be $N_t=1\times10^{14}$ cm^{-3}. Such a large transfer of charge and corresponding electric field can only be driven by a direct spatial modulation of the Fermi level. This is clearly possible in the case of $EL2$ because the quenching radiation changes the compensation of the material. In the illuminated regions, the Fermi level is driven towards the valence band, while in the dark regions the Fermi level remains pinned near the $EL2$ level. It is important to note the distinction between the Fermi level and quasi-Fermi levels. Quasi-Fermi levels are created by the redistribution of occupancies. In the standard photorefractive effect,[5] the quasi-Fermi levels define carrier densities that drive the transfer of charge through normal thermal diffusion. In sharp contrast, in the present case of quenching $EL2$, defect concentrations, not just occupancies, are changing with illumination. Therefore this case directly involves the Fermi level. This is the important distinction between normal photorefractive gratings and the quenched-in gratings described here. The gradient of the Fermi level defines an effective electric field that drives charge from the illuminated to the dark fringes, working in parallel with normal diffusion. The quenched-in grating is formally equivalent to a periodic modulation-doped *p-i-p* structure that alternates insulating and conducting *p*-type layers.

The maximum charge density $N_{\max}$ that can be transferred from the light to the dark fringes is limited by the maximum potential difference between the $EL2$ level and the valence band, $V_{\max}=(E_{EL2}-E_{\mathrm{VB}})=0.7$ eV. The potential difference caused by the transferred charge is

$$V_b=\frac{2eN_t}{K^2\epsilon\epsilon_0}\,.$$

The transferred charge density for maximum modulation of the Fermi level is found to be $N_{\max}=8\times10^{14}$ cm^{-3}. This limit is nearly an order of magnitude larger than the value of $N_t=1\times10^{14}$ cm^{-3} derived from the experiment. To examine the specific relationship between optical exposure and carrier transfer, two issues will be discussed in more detail: (i) a spatial-modulation amplitude less than unity, and (ii) the time evolution of the first Fourier amplitude of the Fermi level.

The quenching rate as a function of position is given by

$$\beta(x)=\beta_0[1+m\sin(Kx)]\,,$$

with[2] $\beta_0=s_p s^* I_0(s_n+s_p)$ where s_p and s_n are the optical cross sections for electron and hole emission, s^* is the cross section for transforming $EL2$ into its metastable state, and I_0 is the pump intensity. The modulation index m is the ratio of the beam intensities in the two arms of the pump laser. It determines the depth of the illumination modulation. In practice, the modulation index is less than unity because of small inbalance of beam intensities and because of internal reflections and scattering within the crystal. This leads to a finite quenching rate in the dark fringe.

The amount of $EL2$ quenched at position x and time t is

$$N_{EL2}(x,t)=N_{EL2}(t=0)\{1-\exp[-t\beta(x)]\}\,,$$

where $N_{EL2}(t=0)$ is the initial defect concentration. The quenched $EL2$ fraction as a function of time and position is shown in Fig. 3 for a pump intensity of $I_0 = 20$ mW/cm^2 and $m = 0.5$. When the density of unquenched $EL2$ decreases below $N_A - N_D$, the Fermi level drops relatively abruptly to the valence band. In Fig. 3 an initial compensation ratio of 0.1 is denoted by the horizontal line. For quenched fractions above this line, the Fermi level is pinned near the valence-band edge. The time evolution of the diffracted signal shown in Fig. 2 follows the time evolution of the square of the first-Fourier amplitude of the Fermi level. The diffraction efficiency increases rapidly at short times as an approximately sinusoidal variation of the Fermi level is formed by the modulated quenching rate. Maximum diffraction is reached when the Fermi level is modulated symmetrically between the light and dark fringes. This corresponds to $t = 2.5$ sec in Fig. 3. If the modulation index m is small, then the transferred charge will be reduced from the maximum allowed charge transfer, N_{max}. For longer times, the width of the central dark region is progressively pinched off and the Fourier amplitude decreases until the entire crystal is quenched. In our experiments, the reduction of charge transfer from the maximum allowed value corresponds to a modulation index of $m = 0.5$ which is a reasonable value in view of the high internal reflectivity of GaAs surfaces. Antireflection coatings should significantly increase the charge transfer.

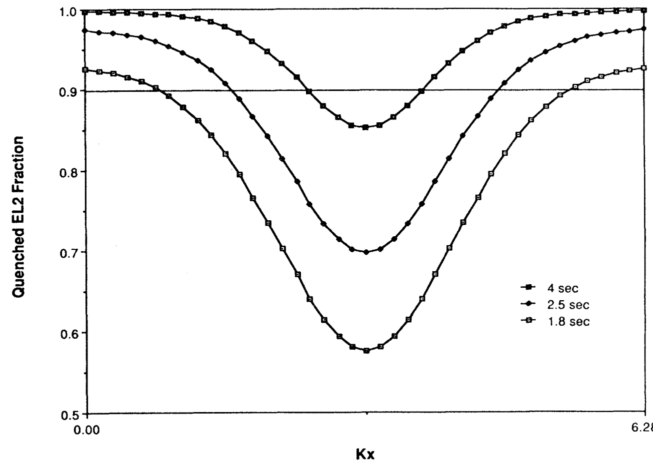

FIG. 3. Density of quenched $EL2$ as a function of position and time. The pump intensity is $I_0 = 20$ mW/cm^2 with $m = 0.5$, $s_n = 3\times10^{-16}$ cm^2, $s_p = 1.8\times10^{-16}$ cm^2, and $s^* = 2.4\times10^{-17}$ cm^2. The Fermi level drops to the valence band when the unquenched density drops below the shallow acceptor density. The horizontal line denotes this transition for an initial compensation ratio of 0.1. The maximum diffraction occurs at $t = 2.5$ sec.

In conclusion, we have demonstrated that by combining coherent illumination with the quenching of $EL2$ into its metastable state, large spatial modulation of the Fermi level is possible in undoped semi-insulating GaAs. The gradient of the Fermi level defines an effective electric field that drives charge transport from the illuminated to the dark fringes. The resulting internal electric fields are much greater than diffusion fields and can be retained for many hours. The electric field modulates the index of refraction, producing an index grating. Such gratings are used to diffract a probe laser, and are potentially useful for spatial light modulation or optical image processing. The ability to modulate the Fermi level directly distinguishes this phenomenon as a novel nonlinear optical mechanism, distinct from normal diffusive photorefractive effects.

*Present address: Department of Physics, Purdue University, West Lafayette, IN 47907.

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