Terahertz Radiation Induced by Subband-Gap Femtosecond Optical Excitation of GaAs

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We have observed terahertz radiation induced by subband-gap femtosecond optical excitation from GaAs. We believe this is due to screening of the surface depletion field by virtual photocarriers (the inverse Franz-Keldysh effect).

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Recently, Yamanishi [1] and Chemla, Miller, and Schmitt-Rink [2] proposed independently that screening of the static field in a quantum well structure by virtual carriers induced by a short laser pulse tuned at the transparent region can generate a short electrical transient. Since no real carriers have been generated, this electrical transient is induced by a reactive current, and is free of the carriers' scattering processes. It is almost as fast as the optical pulse. Later, Yablonovitch and co-workers extended this idea to bulk material [3]. Basically, this is an inverse Franz-Keldysh effect.

In this Letter, we report observation of terahertz electromagnetic radiation induced by subband-gap femtosecond optical excitation. The experimental method we used is the newly developed optically induced terahertz radiation technique [4]. We illuminate a bulk GaAs surface with a femtosecond optical pulse from a tunable laser to inject real or virtual carriers depending on the exciting photon energy. This induces a fast electrical transient due to polarization of real or virtual photocarriers in response to the surface depletion field. This fast electrical transient radiates a short terahertz electromagnetic pulse.

A pump and probe technique is used for the measurement. The femtosecond laser beam, which is unfocused, illuminates the sample to induce subpicosecond terahertz radiation. The radiation is detected by a photoconducting dipole antenna [5,6] which is optically gated by a fraction of the laser beam. The temporal wave form of the radiated signal is measured by recording the gated current from the detector as a function of the relative time delay between two beams.

The detector has a time resolution of approximately 0.6 ps and a corresponding frequency bandwidth of 0.5 THz. An important property of our detector is that it is coherent, i.e., it measures the amplitude and the phase instead of the intensity of the radiated electric field. Since the radiation can come from either real or virtual carriers depending on whether the photon energy is above or below the band gap, we would expect to see changes in both the amplitude and shape of the radiated wave form as we tune the photon energy through the band edge. This approach serves as a unique probing method to study carrier dynamics on a subpicosecond time scale [7].

The femtosecond pulse laser we used was a tunable dual-jet hybrid mode-locked Styrl 9 dye laser synchronously pumped at 76 MHz by a frequency-doubled Nd-doped yttrium-lithium-fluoride laser. It produced 100-fs pulses with a total average power of about 80 mW. The laser wavelength was tunable from 815 to 870 nm. The spectrum had a full width at half maximum of about 10 meV. The experimental setup is similar to our previous one [4]. The GaAs sample was placed in a cryogenic Dewar. The dipole detector was located at the specular angle, about 2 cm away from the sample, which insured detection of the far-field electromagnetic radiation. For simplicity, we use $\delta\varepsilon$ to denote the difference between the center photon energy and band edge $E_g(T)$ of the semiconductor at temperature T.

Figure 1 shows part of the radiated wave forms at different $\delta\varepsilon$ from a (100)-oriented undoped semi-insulating GaAs sample. Figure 2 is the amplitude of the radiated signal versus $\delta\varepsilon$. The temperature of the sample is always kept below 130 K. However, to achieve a large detuning range, both temperature tuning and wavelength

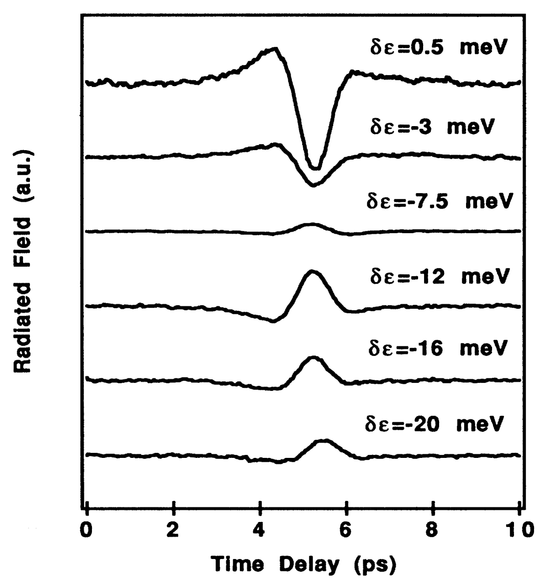

FIG. 1. Part of the experimental results from an undoped semi-insulating GaAs sample. The sample temperature is kept below 130 K. Both temperature and laser wavelength tuning are adopted to achieve the largest detuning.

tuning are adopted. When the laser center photon energy is well above the band edge, the radiation has a large amplitude, and the main peak is negative. As we decrease photon energy below the band edge, the amplitude of the radiated signal drops quickly. Finally, as we keep decreasing photon energy well below the band edge, the radiated wave form reverses its sign, with the main peak changing from negative to positive as $\delta\varepsilon$ is tuned only 3 meV. Furthermore, as shown in Fig. 2, after the sign reversal, the signal amplitude recovers somewhat and then decays at a much slower rate. Clearly, a different physical mechanism is governing the radiation process when we use subband-gap optical excitation.

To gain more insight from the experimental results, first, consider the radiation from a real photocurrent. In the far-field region, the temporal wave form should follow the negative first time derivative of the transient photocurrent. It can be roughly estimated with an assumption of the time dependence of the transient current. In our case, the depletion width is roughly several μm, and the static depletion field is estimated to be about a few kV/cm. Wysin, Smith, and Redondo made a detailed Monte Carlo calculation of the transient photocurrent in GaAs [8]. Their results suggest that in GaAs, under a biasing field around 5 kV/cm, and illuminated by a femtosecond laser pulse with a center photon energy of 1.5 eV, the photocurrent will rise very quickly, in about 200 or 300 fs, reach an overshoot value, then gradually decrease, and relax down to the normal steady-state value on a picosecond time scale. The first time derivative of the transient current will be a sharp positive peak followed by a small slow negative lobe. Therefore, the main

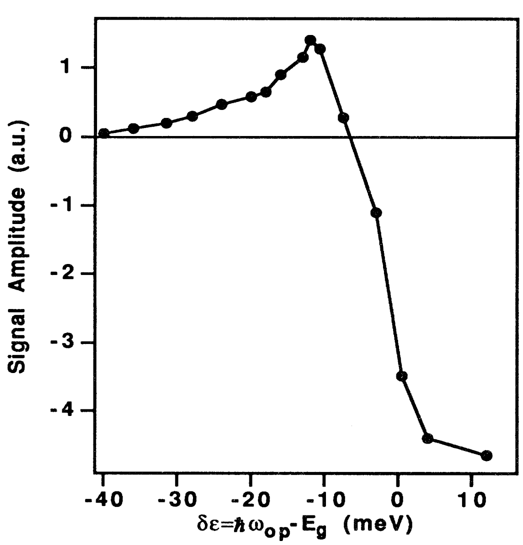

FIG. 2. Amplitude of radiation vs the detuning energy. Radiation caused by the virtual-carrier effect has a slower decay dependence on the detuning energy than that caused by real absorption.

peak of the radiated signal will be negative. Using the numerical values from their paper, we estimate that the amplitude of the detected radiation would be about a few V/cm for excitation above the band gap; this is close to what we detected in the experiment. For excitation near or just below the band edge, the signal should drop very quickly, since the optical absorption exponentially decreases [9]. This is consistent with the fast decay of the signal as $\delta\varepsilon$ is tuned through the band edge as shown in Fig. 2.

When the photon energy is tuned well below the band gap, the real photocurrent diminishes. However, as described by Yamanishi [1], Chemla, Miller, and Schmitt-Rink [2], and Yablonovitch and co-workers [3], there is a reactive photocurrent caused by the polarization of the virtual photocarriers, which can be described by a third-order nonlinear polarization:

$$\mathbf{P}^{\mathrm{NL}}=\chi^{(3)}(0,0,-\omega_{\mathrm{op}},\omega_{\mathrm{op}})\mathbf{E}_{\mathrm{op}}\mathbf{E}^{*}_{\mathrm{op}}\mathbf{E}_{\mathrm{dc}}\,, \tag{1}$$

where $\chi^{(3)}(0,0,-\omega_{\mathrm{op}},\omega_{\mathrm{op}})$ is the nonlinear susceptibility due to the inverse Franz-Keldysh effect [1-3]. For a bulk GaAs sample assuming an abrupt band edge, Yablonovitch and co-workers obtained the following simplified expression:

$$\chi^{(3)}(0,0,-\omega_{\mathrm{op}},\omega_{\mathrm{op}})=\frac{1}{24\pi^2}\frac{e^2acn}{E_{cv}m_{\mathrm{eff}}}\left(\frac{\hbar}{\Delta}\right)^3 \approx\frac{1.46\times10^{-3}}{\Delta^3}\ \mathrm{esu}\,, \tag{2}$$

where Δ is the detuning energy, $\Delta=E_g-\hbar\omega_{\mathrm{op}}$ in meV [3].

This nonlinear polarization has the spatial and temporal dependence of the optical excitation pulse. Clearly it is enhanced by the near-resonance excitation. The far-field wave form induced by $\mathbf{P}^{\mathrm{NL}}$ should follow the negative second time derivative of $\mathbf{P}^{\mathrm{NL}}$. Our optical pulse is very close to sech^2. The radiated wave form would have a large main peak with two small lobes of the opposite polarity. The polarity of the main peak is the same as that of $\chi^{(3)}$, which is positive. The magnitude of the radiation induced by the inverse Franz-Keldysh effect can also be roughly estimated. From Refs. [3,10,11], for our experimental conditions, $\chi^{(3)}(0,0,-\omega_{\mathrm{op}},\omega_{\mathrm{op}})$ ranges from 10^{-8} to 10^{-6} esu depending on how close the photon energy is to the band edge. Using these numbers, from basic radiation theory, we estimate that the detected radiated field due to the virtual current would have a magnitude in the range of a few mV/cm to nearly 1 V/cm, which is within our detectability.

Because of the finite spectral width of the femtosecond laser pulse and the band-tail states, a large number of real photocarriers are generated even when $\delta\varepsilon$ is negative. Therefore, when the optical excitation is just below the band edge, radiation comes from both the real and the virtual photocurrents. When the real photocurrent dominates, the detected radiation has a negative main peak. As we decrease the photon energy, the real photocurrent exponentially decreases, while the reactive current drops at a much slower rate. Eventually, the radiation due to virtual photocarriers should start to dominate and override the effect due to real absorption. Since the radiation induced by the reactive current has a positive main peak, the main peak of the detected wave form should change sign as observed. This sign reversal is very dramatic depending on the sharpness of the band edge and the spectral width of the laser pulse. From the literature value of the optical-absorption coefficient [9], the sign reversal occurs at a point where the absorption coefficient is more than an order of magnitude smaller than its above-band-gap value so that fewer real carriers are generated. The amplitude of the radiation due to the virtual photocarriers is consistent with our rough estimation.

When we decrease the photon energy further, the signal eventually dies out. Because of the finite spectral width of the optical pulse, the decay dependence on detuning energy would not be a simple inverse cubic dependence as indicated in Eq. (2), which is based on monochromatic optical excitation and an assumption of the abrupt band edge. However, the signal decays much more slowly than that caused by the real photocurrent as shown in Fig. 2.

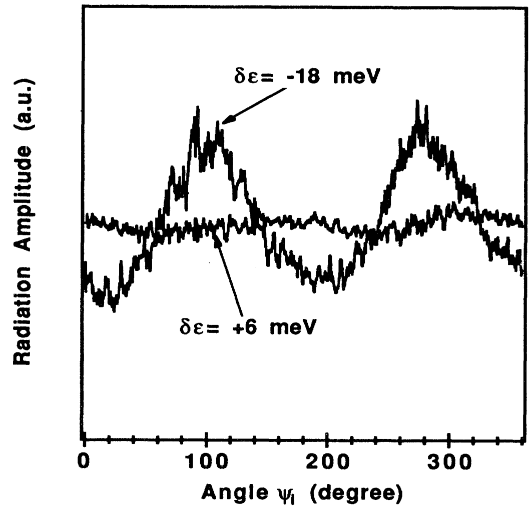

FIG. 3. Amplitude dependence on the optical polarization. Fresnel loss has been taken into account. Radiation caused by the virtual-carrier effect shows strong polarization dependence while that caused by real absorption shows little dependence.

Another property of the inverse Franz-Keldysh effect is the crystal symmetry dependence. For GaAs, $\chi^{(3)}$ has nonzero elements: $\chi^{(3)}_{xxxx}$, $\chi^{(3)}_{xyxy}$, and $\chi^{(3)}_{xxyy}$. The detected signal should be proportional to $\mathbf{d}\cdot\mathbf{P}^{\rm NL}$, where $\mathbf{d}$ is the unit vector of the polarization of the dipole detector. Therefore, when we rotate the polarization angle ψ_i of the pumping beam with respect to the incident plane, the amplitude of the signal due to the virtual current will have a ψ_i dependence in addition to the optical Fresnel loss. In our experimental setup, the detected amplitude variation with respect to the optical polarization angle would be

$$S_{\rm virtual}=F(\psi_i,\theta_i,\theta_r)\left[(\chi^{(3)}_{xxyy}+\chi^{(3)}_{xyxy})+\left(3\chi^{(3)}_{xxxx}\sin^2\theta_r-\frac{\chi^{(3)}_{xxyy}+\chi^{(3)}_{xyxy}}{2n}[\cos(\theta_i+\theta_r)+3\cos(\theta_i-\theta_r)]\right)\cos^2\psi_r\right], \qquad (3)$$

where F is the optical Fresnel loss, n is the optical index of refraction, θ_i,θ_r are optical incident and refracted angles, and ψ_i,ψ_r are polarization angles with respect to the plane of incidence, for incident and refracted beams, respectively, i.e., $\tan\psi_r=\tan\psi_i\cos(\theta_i-\theta_r)$. On the other hand, for the optical excitation *just above* the band edge of GaAs, we would not expect any polarization dependence for the radiated signal except the optical Fresnel loss. This offers another criterion to separate the virtual-carrier effect from the real-carrier effect. This is confirmed in the experiment. Figure 3 shows the radiation amplitude dependence on the optical polarization angle for $\delta\varepsilon=+6$ meV and $\delta\varepsilon=-18$ meV, where the optical Fresnel loss has been taken into account.

Several aspects of our experiment merit some discussion. First, in our experiment, the laser beam is unfocused, therefore the optical flux is below 10^9 cm^{-2} per pulse. Therefore, multiphoton absorption, which usually occurs in transmission-line measurements [12], is absent here. Second, our technique preserves all the phase information of the terahertz radiation. Therefore, we can easily separate the effect due to the reactive current from that of the real photocurrent, even if there is a small amount of real absorption due to tail states or impurities. Third, since the electromagnetic wave propagates in free space, there is no material dispersion present. Theoretically, the radiation caused by the inverse Franz-Keldysh effect should be faster than that caused by the real photocurrent, but, we are unable to confirm this at this time, since the detected signal from both the real and the reactive currents is limited by the frequency response of our detector.

In conclusion, we have observed terahertz radiation induced by the inverse Franz-Keldysh effect. Since its speed is not limited by the real-carrier relaxation processes, it may have applications for ultrafast devices with extremely large bandwidths.

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