Gallium arsenide photoluminescence under picosecond-laser-driven shock compression

X. Z. Lu, R. Garuthara, S. Lee, and R. R. Alfano
Institute for Ultrafast Spectroscopy and Lasers, The City College of New York, New York, New York 10031

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A pump-and-probe technique was used to investigate shock effects on the photoluminescence spectra ($\sim$833 nm) at $T = 80$ K due to the direct transition E_0 from the Γ_6 conduction band to the Γ_8 fourfold degenerate top valence band in GaAs. Under the shock loading condition, the photoluminescence peak was observed to blue shift and split into two components, corresponding to the transitions from the Γ_6 conduction band to the valence heavy- and light-hole subbands, because of symmetry breaking by the uniaxial shock compression along the [001] direction. From the blue shift of the photoluminescence peaks, we deduced our picosecond-laser-driven shock pressure of $\sim$10 kbar.

Significant changes in the optical and transport properties of semiconductors occur with the application of a uniaxial stress due to changes in symmetry and lattice parameters.[1–4] Knowledge of stress effects on band structure is important for operation of GaAs in high-speed ultrasmall devices and switches. In this letter we report on a new observation of the photoluminescence spectra from gallium arsenide (GaAs) under picosecond-laser-driven shock wave loading. The shock waves were generated by focusing intense picosecond laser pulses onto an aluminum (Al) foil[5–8] attached to the sample.

GaAs has a zinc-blende structure[9] of the space group T_d^2. The lowest conduction band is s-like and twofold degenerate, having a Γ_6 symmetry at the Brillouin zone center. The top valence band is a sixfold degenerate p-like band that splits into a fourfold $P_{3/2}$ multiplet ($J = 3/2$, $M_J = \pm 3/2$, $\pm 1/2$) with a Γ_8 symmetry and $P_{1/2}$ doublet ($J = 1/2$, $M_J = \pm 1/2$) with a Γ_7 symmetry due to the spin-orbit interaction. The band structure is shown in Fig. 1 (the left-hand side). The lowest energy band-to-band direct transition (labeled E_0) occurs at the center of the Brillouin zone. The total Hamiltonian matrix for the valence band in the presence of a uniaxial compression P along the [001] direction can be written[9]

$$H = \begin{pmatrix} v_2 & v_1 & v_3 \\ -\delta E_H - \tfrac{1}{2}\delta E_S & 0 & 0 \\ 0 & -\delta E_H + \tfrac{1}{2}\delta E_S & 2^{-1/2}\delta E_{S'} \\ 0 & 2^{-1/2}\delta E_{S'} & -\Delta_0 - \delta E_{H'} \end{pmatrix}, \tag{1}$$

where $\delta E_H = -a(S_{11} + 2S_{12})P$, $\delta E_{H'} = -a'(S_{11} + 2S_{12})P$, $\delta E_S = -2b(S_{11} - S_{12})P$, $\delta E_{S'} = -2b'(S_{11} - S_{12})P$, and Δ_0 is the spin-orbit splitting, S_{11} and S_{12} are components of the elastic compliance constants, a and a' are the deformation potentials of the Γ_8 and Γ_7 valence bands for hydrostatic effects, and b and b' are the deformation potentials of the Γ_8 and Γ_7 bands for shear effects. If the pressure-induced shift and splitting are much smaller than Δ_0, the eigenvalues of Eq. (1) can be written

$$\delta E_{v_2} = -\delta E_H - \tfrac{1}{2}\delta E_S, \tag{2a}$$

$$\delta E_{v_1} = -\delta E_H + \tfrac{1}{2}\delta E_S + \frac{(\delta E_{S'})^2}{2\Delta_0}, \tag{2b}$$

$$\delta E_{v_3} = -\Delta_0 - \delta E_{H'} - \frac{(\delta E_{S'})^2}{2\Delta_0}. \tag{2c}$$

Including the effect of the conduction-band shift, the pressure dependence of the energy gap changes is given by

$$\delta(E_c - E_{v_2}) = \Delta E_H + \tfrac{1}{2}\delta E_S, \tag{3a}$$

$$\delta(E_c - E_{v_1}) = \Delta E_H - \tfrac{1}{2}\delta E_S - \frac{(\delta E_{S'})^2}{2\Delta_0}, \tag{3b}$$

$$\delta(E_c - E_{v_3}) = \Delta_0 + \Delta E_{H'} + \frac{(\delta E_{S'})^2}{2\Delta_0}, \tag{3c}$$

where $\Delta E_H = -(c_1 + a)(S_{11} + 2S_{12})P$, $\Delta E_{H'} = -(c_1 + a')(S_{11} + 2S_{12})P$, and c_1 is the deformation potential of the conduction band. For light polarized perpendicularly to the stress axis c, the transitions between the s-like Γ_6 conduction band and all three valence bands, v_1, v_2, and v_3, are allowed.[9] From Eqs. (3a) and (3b), we deduce that under shock loading condition, the photoluminescence peak corresponding to the E_0 transition will be subject to a blue shift because of the hydrostatic component of the pressure [represented by the term ΔE_H in Eqs. (3a) and (3b)]. It will also split into two components corresponding to the transitions $E_0(1)$ and $E_0(2)$ (see Fig. 1, the right-hand side) because of the reduced symmetry by the uniaxial nature of the shock pressure [represented by the term δE_S in Eqs. (3a) and (3b)]. Since the heavy-hole band v_2 has a lower energy under uniaxial compression,[10] the corresponding transition

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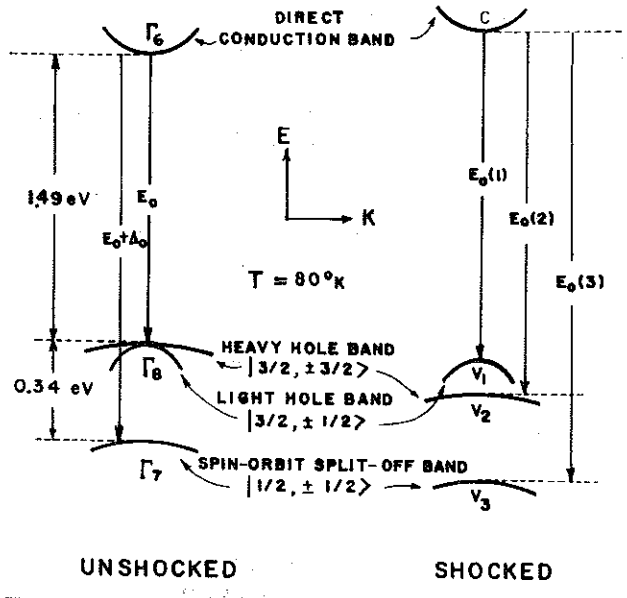

FIG. 1. Band structure of GaAs at $T = 80$ K for unshocked (left) and shocked (right) crystals.

$E_0(2)$ will have a larger blue shift. In addition, the blue shift of the emission peak of the electron-light-hole recombination [the transition $E_0(1)$] will increase sublinearly with the increasing pressure because of the pressure-induced coupling between the sets of $M_J = \pm\frac{1}{2}$ bands (v_1 and v_3).

A schematic diagram of the experimental geometry is shown in Fig. 2. The experimental setup was described in Ref. 11. A Quantel Nd:YAG laser of 30 ps pulse width and 25 mJ pulse energy at 1.064 μm was utilized as the pump beam to generate shock waves in the aluminum foil. The shock waves were launched into the GaAs sample located behind the aluminum foil. The probe beam of 27 ps at 532 nm was delayed through a white cell approximately by 46 ns relative to the arrival of the pump beam at the aluminum foil to ensure that the probe pulses reach the sample surface area right after the arrival of the shock front. This was necessary to observe the maximum effect of shock pressure.

The GaAs sample was n type with a carrier concentration of $\sim 10^{18}$ cm^{-3}. The thickness of the sample was about 50 μm with the cubic [001] axis perpendicular to the sample surface. The aluminum foil had a thickness of 20 μm. The sample and aluminum foil were loaded between two quartz plates and placed in the center of a liquid-nitrogen Dewar.

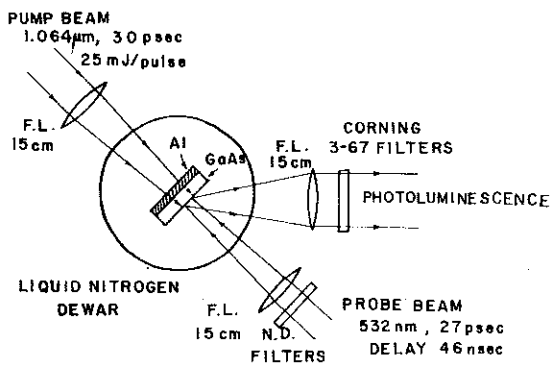

FIG. 2. Schematic diagram of the experimental geometry.

APIEZON N-grease was added to the interface of the sample and aluminum foil to ensure efficient mechanical coupling at $T = 80$ K. The pump beam was focused by a 15-cm focal length lens to a 500-μm-diam spot on the aluminum foil, which ensured planarity of the shock wave and a peak power density of $\sim 4.2\times 10^{11}$ W/cm^2. The probe pulse was focused to a 400-μm-diam spot by a 15-cm focal length lens on the sample, giving a peak power density of $\sim 1.6\times 10^7$ W/cm^2.

The emitted light was dispersed by a 1/4 m spectrometer, detected by a silicon-intensified target coupled to an optical multichannel analyzer and stored in a PDP 11/23 + host computer. The resolution of the detection system was about 2 nm. All spectra were recorded at $T = 80$ K by single shots of the laser.

The observed photoluminescence spectrum of GaAs without shock loading is dominated by a single peak due to the band-to-band recombination at the lowest energy gap E_0, as shown in Fig. 3 (left). No emission lines due to impurity transitions were observed. The effect of shock waves was most pronounced for the delay time between the pump and probe beam, $\tau = 40\sim 60$ ns. At $\tau \leqslant 35$ ns, no shock effect was observed. A typical spectrum showing the shock effects on the photoluminescence from GaAs is displayed in Fig. 3 for $\tau = 46$ ns and probe intensity 1.6×10^7 W/cm^2. Under shock loading, the emission peak was split into two, with blue shifts of 20 and 85 meV, respectively.

The lowest energy band gap in GaAs in the absence of the shock pressure can be written[12]

$$E_g(T) = 1.530\ \text{eV} - (5.0\times 10^{-4}\ \text{eV K}^{-1})T. \qquad (4)$$

The peak energy of the emission due to the band-to-band transition E_0 at $T = 80$ K is $E_p = E_g(80\ \text{K}) = 1.490$ eV, i.e., $\lambda_p = 832$ nm, which agrees with our observed unshocked emission line at 833 nm (Fig. 3) within our experimental accuracy. Since the energy gap $E_c - E_{v_2}$ expands linearly with the increasing pressure P, the blue shift of the transition energy $E_0(2)$ was chosen to calibrate the shock

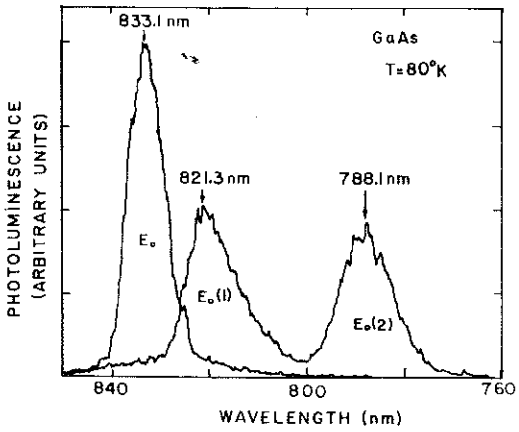

FIG. 3. Unshocked (left) and shocked (right) photoluminescence spectra at a delay time $\tau = 46$ ns and probe intensity 1.6×10^7 W/cm^2. The shocked spectrum consists of two peaks, corresponding to the transitions $E_0(1)$ and $E_0(2)$.

pressure. From Eq. (3a) the pressure coefficient of this transition energy is given by

$$\frac{dE_0(2)}{dP} = -(c_1 + a)(S_{11} + 2S_{12}) - b(S_{11} - S_{12}). \tag{5}$$

With the reported values,[13] $c_1 + a = -13.0$ eV, $b = -1.66$ eV, and[14] $S_{11} = 1.16 \times 10^{-3}$ kbar^{-1}, $S_{12} = -3.67 \times 10^{-4}$ kbar^{-1}, we obtain $dE_0(2)/dP = 8.5$ meV/kbar. Therefore, the experimentally measured blue shift of 85 meV (Fig. 3) for the electron-heavy-hole recombination emission line gives a magnitude of the laser-driven shock pressure in GaAs of ~ 10 kbar.

From Eq. (3b) and the reported values,[9] $b' = -2.47$ eV and $\Delta_0 = 0.34$ eV, we obtain $\delta(E_c - E_{v_1}) = 21.5$ meV for a pressure of 10 kbar, which consistently agrees with our observed blue shift of ~ 20 meV under the same shock compression condition. Therefore, the shocked system is in quasiequilibrium state.

In conclusion, the picosecond-laser-drive shock waves offer a new technique to investigate properties of materials at very high pressures with a table-top laser facility. Using this technique, we have observed blue shift and line splitting in the photoluminescence spectrum of GaAs due to band-gap expansion and symmetry breaking by the uniaxial shock compression along the [001] direction. The pressure range of the shock waves generated in our experiment by a 25-mJ picosecond laser pulse is ~ 10 kbar.

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[13]See Table II of Ref. 9. For $c_1 + a$ (i.e., $c_1 + a_1 + a_2$), we took an intermediate value -13.0 eV between the "present work" -8.38 eV and "theoretical calculation" -15.2 eV.
[14]The elastic constants C_{11} and C_{12} of GaAs at $T = 77$ K are $C_{11} = 1221$ kbar and $C_{12} = 566$ kbar (see p. 345 of Ref. 12). In cubic crystals the elastic compliance constants are related to the elastic constants by $S_{11} = (C_{11} + C_{12})/[(C_{11} - C_{12})\ (C_{11} + 2C_{12})]$, $S_{12} = -C_{12}/[(C_{11} - C_{12})\ (C_{11} + 2C_{12})]$, and $S_{44} = 1/C_{44}$ [see O. Madelung, in *Landolt-Bornstein Numerical Data and Functional Relationships in Science and Technology*, edited by K. H. Hellwege (Springer, Berlin, 1980), New Series, group III, Vol. 17a, p. 25].