Charge carrier relaxation processes in TbAs nanoinclusions in GaAs measured by optical-pump THz-probe transient absorption spectroscopy

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Rare-earth materials epitaxially codeposited with III-V semiconductors form small, spherical rare-earth-monopnictide nanoparticles embedded within the III-V host. The small size of these particles (approximately 1.5 nm diameter) suggests that interesting electronic properties might emerge as a result of both confinement and surface states. However, ErAs nanoparticles do not exhibit any signs of quantum confinement or an emergent band gap, and these experimental observations are understood theoretically. We use ultrafast pump-probe spectroscopy to investigate the electronic structure of TbAs nanoparticles embedded in a GaAs host, which were expected to be similar to ErAs. We study the dynamics of carrier relaxation into the TbAs states, which essentially act as traps, using optical-pump terahertz-probe transient absorption spectroscopy. By analyzing how the carrier relaxation rates depend on pump fluence and sample temperature, we conclude that the TbAs states are saturable. Saturable traps suggest the existence of a band gap for TbAs nanoparticles, in sharp contrast with the results for ErAs.

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I. INTRODUCTION

Carrier relaxation and transport dynamics in semiconductors are extremely sensitive to the electronic structure of traps, dopants, or nanoparticles. Control over these carrier dynamics is often essential for the development of new optoelectronic devices. For example, THz sources generating radiation via optoelectronic switches require materials with fast carrier relaxation times, but thermoelectrics require materials that minimize carrier relaxation. Rare-earth-monopnictide nanoinclusions embedded in III-V semiconductors have a substantial impact on carrier dynamics. Previous work on ErAs nanoinclusions demonstrates that it is possible to tune these dynamics with the size, composition, and density of the nanoinclusions [1–5].

TbAs nanoinclusions are in the same class of materials as ErAs and may provide another opportunity for tunable control of carrier dynamics. TbAs has a lattice constant of 5.815 Å while ErAs has a lattice constant of 5.734 Å [6]. Therefore the lattice mismatch between TbAs:InGaAs is smaller than for ErAs:InGaAs when they are lattice matched to InP (lattice constant 5.87 Å). It is thought that this improved epitaxial relationship will lead to more favorable electronic properties. For example, ErAs nanoparticles serve to reduce the thermal conductivity of GaAs through phonon scattering, increase the Seebeck coefficient through energy-dependent electron scattering, and increase electrical conductivity through nanoparticle donation of electrons [7,8]. TbAs may offer similar possibilities. Unlike with ErAs nanoparticles [8], the smaller lattice mismatch and therefore smaller strain between TbAs and InGaAs lattice matched to InP may pin the Fermi level below the matrix conduction band, which would improve the Seebeck coefficient without sacrificing good electrical conductivity [9].

Although TbAs nanoinclusions are predicted to be of interest for device applications, the electronic structure of the TbAs nanoinclusions is presently unknown. In this paper, we elucidate the electronic structure of TbAs nanoinclusions epitaxially embedded within GaAs. We pursue this objective with an analysis of the ultrafast relaxation processes of optically excited carriers. Femtosecond laser-generated optical-pump probe experiments have been used extensively to investigate carrier dynamics in semiconductors, particularly for materials with potential optoelectronic applications [10,11]. Trapping and relaxation processes, which typically occur on ps time scales, are of particular importance in devices where recombination via trap states may limit the device performance or the repetition rate at which the devices can operate. A high-fluence optical-pump optical probe experiment has been previously used to investigate trap saturation and Auger recombination in low temperature grown GaAs [12]. Optical-pump THz-probe techniques have also been used to measure short carrier relaxation times and high effective mobilities in radiation-damaged silicon on sapphire [13]. Pump probe studies of ErAs:GaAs have shown subpicosecond relaxation times, with full recovery times on the order of a few picoseconds; these recovery times vary with superlattice period and pump fluence [5]. Building on these techniques, we use a fluence-dependent optical-pump THz-probe method to probe the dynamics of carrier relaxation in TbAs:GaAs. Our results reveal that the states of TbAs nanoinclusions are saturable, which implies the existence of a band gap for TbAs nanoinclusions. This result is quite different than the results observed for ErAs nanoinclusions and cannot be explained by present theories of the electronic structure of TbAs nanoinclusions.

II. EXPERIMENTAL DESIGN

A. Sample fabrication

The TbAs:GaAs sample was grown on a (001) undoped GaAs substrate with a 50 nm thick GaAs buffer layer. The

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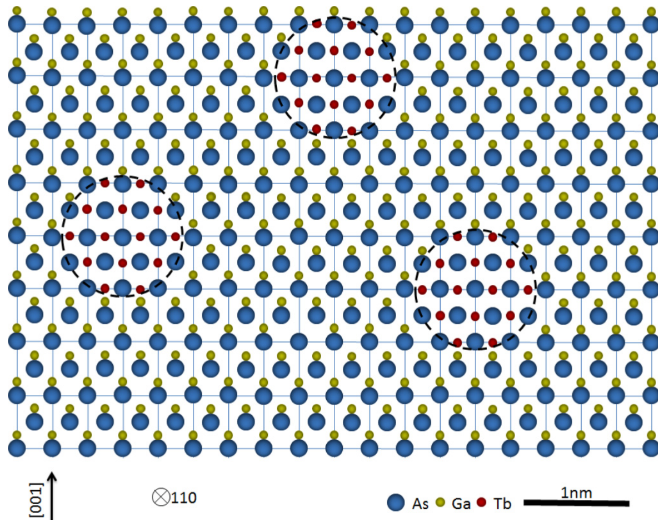

FIG. 1. (Color online) Schematic showing TbAs nanoparticles within a GaAs host matrix, formed by codepositing terbium, gallium, and arsenic.

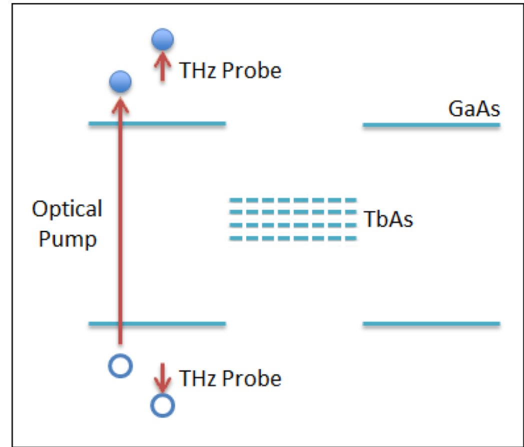

FIG. 2. (Color online) Optical pump creates electron-hole pairs. THz probe is absorbed only when electrons are in the conduction band or holes are in the valence band.

growth was completed using an OSEMI NextGEN solid-source molecular beam epitaxy (MBE) system, equipped with effusion cells for Ga and Tb, and a two-zone valved cracker source for As. During growth, using a substrate temperature of 490 °C measured by band edge thermometry, terbium was codeposited with gallium and arsenic such that the solid solubility limit of Tb in GaAs was exceeded and nanoparticles precipitated throughout the material (Fig. 1). No capping layer was grown on top of the TbAs:GaAs layer. Growth and thermal desorption of oxides were monitored with reflection high-energy electron diffraction (RHEED). A GaAs growth rate of 1 μm/h was used for an expected film thickness of 1 μm. An arsenic-to-gallium flux ratio of about 26:1 was used, measured by beam equivalent pressure (BEP) using an ionization gauge. Due to the low vapor pressure of Tb, it is very difficult to measure the flux of Tb from BEP measurements. Thus, the TbAs concentration was calibrated by matching the Rutherford backscattering spectroscopy (RBS) measured concentration to the Tb effusion cell temperature. For this sample, a Tb source temperature of 1345 °C was used, resulting in a TbAs concentration of 2.12%. Based on these growth conditions and using an average nanoparticle diameter of 1.1 nm, measured from samples of similar growths [14], a TbAs nanoinclusion density of 3.2×10^{19} cm^{-3} was calculated.

B. THz transient absorption spectroscopy

Optical-pump THz-probe experiments were performed using a regeneratively amplified Ti:sapphire laser system producing 3.2 mJ, 35 fs pulses centered on 800 nm at a 1 kHz repetition rate. A portion of the amplifier output is used to generate and detect the terahertz pulses through the electro-optic effect in ZnTe, and the rest of the power is used to optically pump the sample. The probe is a broadband THz pulse spanning 0.1–2.75 THz. The delay stage is capable of taking data up to an approximately 400 ps delay between pump and probe pulses [5]. Incident pump fluences of 2–200 μJ/cm^2 were used to excite the samples. The temperature dependence study was completed using a liquid helium cooled cryostat capable of reaching temperatures as low as 4.5 K.

In the absence of a pump beam, there are no carriers in the conduction or valence bands of the GaAs and the THz probe cannot be absorbed. The 800 nm pump beam excites carriers across the band gap, leading to a population of electrons in the conduction band and holes in the valence band. Because of the large number of available energy states in the conduction and valence bands, these electrons and holes are both able to absorb low energy photons from the THz-probe beam. The absorption of the optical pump and THz probe is depicted in the energy diagram of Fig. 2.

Electrons and holes generated by the optical pump will relax down to the GaAs band edge in less than 100 fs [15,16]. This initial relaxation due to thermalization is too fast to be resolved by this experiment, which is limited by the pulse duration of the probe (approximately 700 fs). Carriers at the band edge can relax by radiative or nonradiative recombination across the GaAs band gap or by relaxation into states associated with the TbAs nanoinclusions. The rate of decay of the carrier population, as measured by the decay of THz absorption, allows us to investigate these carrier relaxation mechanisms.

Transient absorption data is collected for delay times between 0 and 400 ps using a delay stage step size of 0.3333 ps/step. By averaging ten complete data runs for each pump fluence and temperature, we improve the signal-to-noise ratio enough to resolve the dynamics happening on time scales down to a few ps. We fit the data to an exponential decay using a linear least squares method with a Levenberg-Marquardt algorithm in order to determine the relaxation rates of the charge carriers.

C. Data analysis and physical model

Figures 3(a) and 3(b) show the average intensity of the THz-probe beam after it passes through the sample as a function of the delay between the optical pump and the THz probe. The data displayed is the average of ten data runs at room temperature for a pump fluence of 6 μW/cm^2. A single-exponential curve (solid blue curve) does not accurately

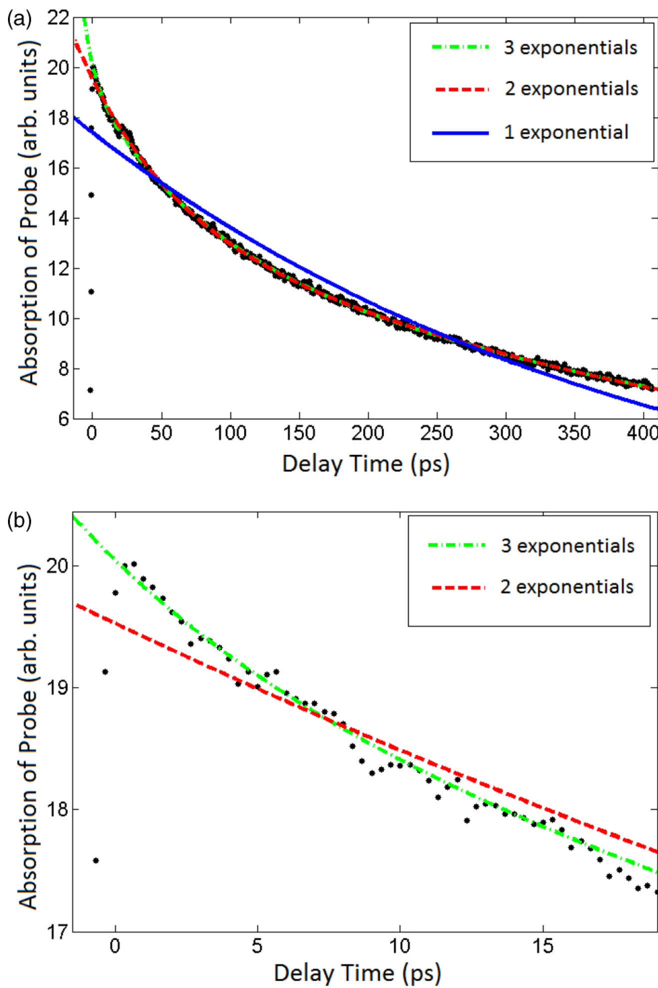

FIG. 3. (Color online) (a) For larger delay times (50–400 ps), both a biexponential (red dashed line) and a triexponential equation (green dashed-dotted line) fit the data with good agreement. (b) For shorter delay times (0–50 ps), a biexponential equation does not accurately reflect the relaxation dynamics.

describe the relaxation dynamics seen over the full 400 ps range of delay times [Fig. 3(a)].

We find that a biexponential curve, displayed as a red dashed line in Fig. 3(a), fits the data well on a long time scale, but does not accurately capture the relaxation dynamics happening during the first 10 ps after photoexcitation. In Fig. 3(b) we compare a biexponential curve (red dashed line) and a triexponential curve (green dashed-dotted curve) over the first 20 ps after photoexcitation. Figure 3(b) reveals the discrepancy of the biexponential fit for short delay times and confirms that a triexponential fit is necessary to capture these fast relaxation dynamics.

Using a four-exponential fit does not improve the adjusted R^2 value relative to the values obtained with a triexponential equation and the three- and four-exponential fits are completely indistinguishable. We therefore conclude that the relaxation dynamics in this sample are described by a triexponential model for absorption (I) of the THz probe as a function of the delay time (t) between the optical pump and

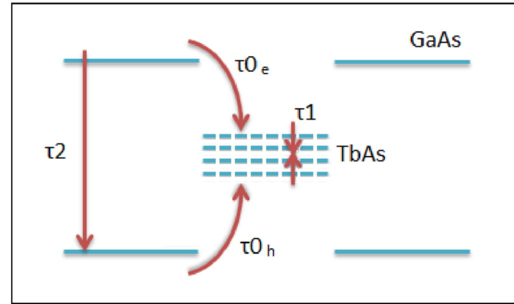

FIG. 4. (Color online) Charge carriers can relax via three processes: into the TbAs trap states ($\tau 0$), via trap states emptied by electron-hole annihilation within the traps ($\tau 1$), or radiative recombination across the bulk GaAs ($\tau 2$).

the THz probe:

$$I = A \exp\left(\frac{-t}{\tau 0}\right) + B \exp\left(\frac{-t}{\tau 1}\right) + C \exp\left(\frac{-t}{\tau 2}\right). \quad (1)$$

The three relaxation time constants returned by the triexponential fits are statistically distinct. $\tau 0$ has an average value of 5.4 ps for the various pump fluences and temperatures studied, $\tau 1$ ranges from 65 to 135 ps, and $\tau 2$ from 500 to 3700 ps. The amplitude coefficients A, B, and C provide information about the fraction of carriers participating in each relaxation process and will be discussed along with the relaxation rates.

Figure 4 schematically depicts our assignment of the three-exponential decay time constants to distinct physical relaxation processes. We will justify these assignments in the next sections with a systematic analysis of our experimental data and then use the results to draw conclusions about the electronic structure of the TbAs nanoinclusions. The shortest time constant, $\tau 0$, is assigned to the relaxation of carriers from the conduction and valence bands into the TbAs trap states [3]. The second shortest time constant, $\tau 1$, is assigned to the emptying of the trap states, which allows additional electrons to quickly relax from the conduction band into the newly available trap states. The longest time constant, $\tau 2$, is assigned to the relaxation of electrons across the bulk band gap. We will show that the observation of these three distinct relaxation processes, along with their observed dependence on laser fluence and temperature, suggests the existence of saturable trap states associated with the TbAs nanoinclusions, and thus the existence of a band gap for the TbAs nanoinclusions.

III. RESULTS AND DISCUSSION

A. Fluence dependence measurements

To understand the electronic structure of the TbAs nanoinclusions, we investigate the carrier decay as a function of both temperature and the intensity of the optical pump. The temperature was varied from 4.5 to 296 K at an optical-pump fluence of 2 μW/cm^2 and the optical-pump fluence was varied between 2 and 200 μW/cm^2 at room temperature. We consider both the relaxation time constants ($\tau 0$, $\tau 1$, and $\tau 2$) and the relative amplitudes of each decay process (A, B, and C) returned by the triexponential fit to each data set. As we show below, these amplitude coefficients provide insight into

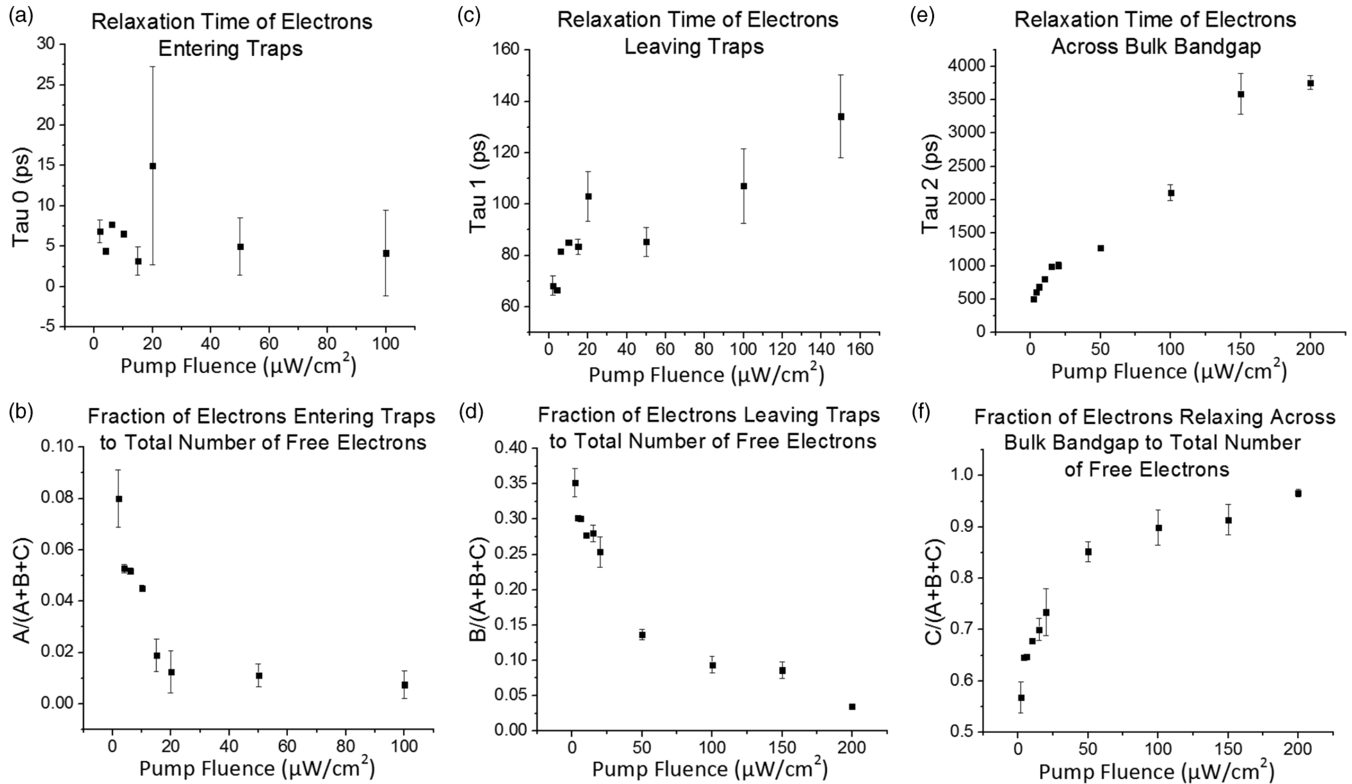

FIG. 5. The relaxation lifetime $\tau 0$ of carriers into the TbAs traps (a) shows no statistically significant fluence dependence. The fraction of electrons entering (b) and exiting (d) the TbAs traps decreases with increasing pump fluence, indicating trap saturation. The relaxation lifetime $\tau 1$ of carriers out of the TbAs traps (c) increases with fluence, which suggests that recombination between carriers in the conduction and valence bands of the GaAs matrix and those in the TbAs nanoinclusions is a significant nonradiative recombination pathway. The relaxation lifetime $\tau 2$ of carriers across the bulk band gap (e) increases with fluence, consistent with trap saturation. The fraction of electrons relaxing across the bulk band gap (f) increases with fluence, which indicates that at high pump fluences the carrier population exceeds the saturation threshold and all additional carriers must relax via bulk radiative decay.

the fraction of charge carriers participating in each relaxation process as well as evidence for trap state saturation when the intensity of the optical pump is increased.

We consider first the dependence of the relaxation rates and amplitude coefficients on optical-pump fluence. The parameters returned by our triexponential fit are plotted in Fig. 5 as a function of the pump fluence. As shown in Fig. 5(a), the relaxation time constant for carriers relaxing into the traps ($\tau 0$) does not show any statistically significant dependence on pump fluence and has an average value of 5.4 ps. As noted previously [16], the photon energy absorbed by electrons in the conduction band is similar to the photon energy absorbed by holes in the valence band. Because both electrons and holes can cause bleaching, it is not possible to distinguish which portion of the probe beam is absorbed by the electrons and which is absorbed by the holes. Therefore we only observe the overall effect of these two processes,

$$\tau 0 = \left(\frac{1}{\tau 0_e} + \frac{1}{\tau 0_h}\right)^{-1}, \tag{2}$$

where $\tau 0_e$ reflects the relaxation of electrons into trap states, and $\tau 0_h$ is the relaxation of holes. The difference in the rates of electrons and holes relaxing into the TbAs states is dependent on the location of the TbAs states within the band gap. Unfortunately the difference is not distinguishable by this experimental technique.

Although $\tau 0$ is independent of pump fluence, the fraction of carriers participating in this relaxation process, calculated by the amplitude ratio $A/(A + B + C)$, does exhibit a strong dependence on pump fluence. As shown in Fig. 5(b), the fraction of carriers participating in this fastest relaxation process decreases quickly with increasing pump fluence and drops to near zero when the pump fluence reaches 20 $\mu\text{W/cm}^2$. This evidence indicates that the rate of charge carrier capture does not change with pump fluence, but the number of carriers that can relax into the TbAs trap states is finite. As the pump fluence increases, the additional optically generated charge carriers are unable to relax into the TbAs trap states and must relax by a different physical process. The dependence on pump intensity of the amplitude fraction for the fastest decay process suggests that the TbAs trap states are saturable.

We propose that the second decay constant, $\tau 1$, which ranges from approximately 65 to 135 ps, is due to trap emptying. As the traps empty, carriers at the band edge can relax into the newly empty trap states, thus reducing the THz absorption. The rate of relaxation from the band edge into the trap states is fast, around 5.4 ps as we have just shown, and thus the rate limiting step for this carrier relaxation process is the trap emptying lifetime $\tau 1$. The fraction of charge carriers

whose relaxation dynamics are governed by trap emptying, $B/(A+B+C)$, decreases with increasing pump fluence, as shown in Fig. 5(d). This result is consistent with the observed change in the fraction of carriers initially relaxing into the traps $[A/(A+B+C)]$ because both processes are limited by the saturation of trap states.

There are two possible trap emptying pathways: radiative and nonradiative recombination [17]. We believe radiative relaxation is unlikely to be the dominant trap emptying pathway for two reasons. First, radiative lifetimes for nanostructures are typically on the order of 1 ns [18], which is significantly longer than the lifetimes observed for $\tau 1$. Second, spectrophotometry and Fourier transform infrared (FTIR) measurements out to 12 μm reveal only one absorption peak for TbAs:GaAs, at 750 meV [14]. However, we observe no photoluminescence associated with this absorption peak, suggesting that this peak could not be associated with a direct band gap. In the absence of a direct band gap, the probability of radiative recombination is negligible. The combination of these factors leads us to believe that nonradiative recombination is likely the dominant trap emptying process responsible for $\tau 1$.

Thermal relaxation through metalliclike states of the TbAs nanoinclusions would provide an efficient pathway for nonradiative recombination. However, rapid thermalization of electrons and holes through the continuous density of states of a metal would lead to extremely fast trap emptying. We observe that the fraction of carriers relaxing into the traps, $A/(A+B+C)$, decreases rapidly with increasing pump fluence, which indicates trap saturation. Trap state saturation is incompatible with rapid trap emptying via thermalization, which leads us to conclude that the TbAs states are not metalliclike. We believe nonradiative recombination of trapped electrons and holes is the likely relaxation pathway. A similar mechanism was proposed to explain the multiexponential recovery, including a 30 ps component, observed in transient absorption experiments that studied electron relaxation via trap states in aqueous CdS colloids [19] and quantum-size crystallites of ZnCdS in an aqueous colloidal solution [20].

The exact nature of the electron-hole nonradiative recombination process cannot be determined from these data. Possible mechanisms include (1) phonon-mediated nonradiative recombination across an indirect band gap, (2) Auger recombination involving excitation of carriers confined within the TbAs nanoinclusions, and (3) recombination of free electrons or holes in the conduction or valence band with oppositely charged carriers confined within the nanoinclusions. Phonon-mediated recombination across an indirect band gap is improbable due to the temperature-dependent measurements discussed below. Auger recombination typically results in increased relaxation rates (decreased lifetimes) as increasing fluence increases the number of carriers confined in a nanostructure and thus increases the probability that Auger recombination takes place [21]. We do not observe a decrease in the lifetime associated with $\tau 1$, the trap emptying rate, as the laser fluence is increased. We therefore believe Auger processes involving carriers confined within the TbAs nanoinclusions are not a dominant trap emptying process.

We observe that the relaxation time constant attributed to trap emptying ($\tau 1$) increases from 65 to 135 ps with increasing fluence, as shown in Fig. 5(c). Notably, the time constant continues to increase with increasing pump fluence well past 20 μW/cm^2, the point at which the data for $\tau 0$ suggest that the traps are saturated [Fig. 5(b)]. If the traps are saturated, increasing pump fluence can only result in additional carriers occupying the conduction and valence bands. The dependence of $\tau 1$ on increasing occupation of the conduction and valence bands suggests that recombination between carriers in the conduction and valence bands of the GaAs matrix and those in the conduction and valence bands of the TbAs nanoinclusions is a significant nonradiative recombination pathway. The existence of this mechanism is supported by the observation that the fraction of carriers participating in the trap emptying process decreases more slowly with increasing fluence [Fig. 5(d)] than the fraction of carriers participating in the direct trapping process [Fig. 5(b)].

The fraction of charge carriers relaxing across the bulk band gap, $C/(A+B+C)$, increases with increasing fluence and slowly approaches unity, as shown in Fig. 5(f). This result is consistent with the decrease in the fraction of charge carriers participating in the fastest relaxation pathways discussed above. If there are a finite number of saturable traps, there are a finite number of carriers that can participate in the fast relaxation processes enabled by the traps. At high pump fluences the carrier population exceeds the saturation threshold and all additional carriers must relax via bulk radiative decay. We observe that the relaxation time constant for charge carriers relaxing across the bulk band gap ($\tau 2$) increases with increasing pump fluence, as shown in Fig. 5(e). This result is consistent with our previous work on trap saturation: Higher pump fluences saturate the traps and lead to a bulk radiative lifetime that becomes longer and approaches the normal radiative lifetime of GaAs without any nanoinclusions or traps [22].

B. Temperature dependence measurements

We turn now to investigating the dependence of carrier relaxation dynamics on temperature. As shown in Fig. 6, the

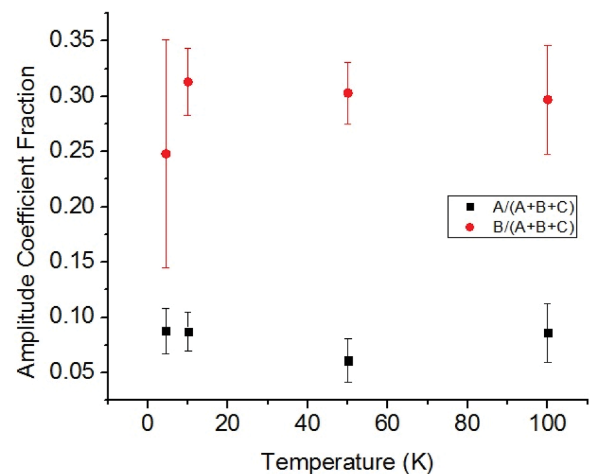

FIG. 6. (Color online) For a 2 μW/cm^2 pump fluence, the fraction of carriers entering the TbAs traps $[A/(A+B+C)$, black squares] and the fraction leaving the traps $[B/(A+B+C)$, red circles] do not show any statistically significant dependence on temperature.

fraction of charge carriers relaxing directly into trap states $[A/(A+B+C)]$ and the fraction whose relaxation dynamics are dominated by the trap emptying process $[B/(A+B+C)]$ are not dependent on temperature. Similarly, $\tau0$, $\tau1$, $\tau2$, and $C/(A+B+C)$ do not show a statistically significant temperature dependence over the temperature range 4.5–296 K (data not shown). If phonon-related recombination was a significant relaxation process, the trap states would display increasing lifetimes with lower temperatures. The absence of a temperature dependence for the relaxation times is consistent with previous optical-pump THz-probe experiments completed on ErAs [5] and supports two conclusions: (1) The trap states are relatively deep and (2) phonon-mediated nonradiative recombination across an indirect band gap is unlikely to be a dominant trap emptying pathway.

C. Comparison to theoretical calculations

Studies have shown the possibility of a semimetal-to-semiconductor transition when some materials are sufficiently spatially confined [23,24]. To evaluate this possibility in ErAs nanoinclusions, linear-muffin-tin-orbital calculations have been used to compute the energy band structure and the density of states of an ErAs:GaAs superlattice. Using this approach, Said found that the presence of interface states prevents the heterostructure from transitioning from a semimetal into a semiconductor when the number of ErAs monolayers was varied [25]. Furthermore, a Maxwell-Garnett formulation and semiclassical transport theory suggest that near-infrared absorption in ErAs nanoinclusions should be attributed to a plasmon resonance rather than the opening of a band gap [26]. A simple hard-walled finite-potential model predicts a gap opening for embedded ErAs nanoparticles with diameters of approximately 3 nm [27], but cross-sectional scanning tunneling spectroscopy reveals that the local density of states exhibits a finite minimum at the Fermi level, demonstrating that the nanoinclusions remain semimetallic [28]. The latter study proposed that the interface atomic structure results in electronic states that prevent the opening of a band gap.

Following the results of these studies of ErAs, it was expected that TbAs, which is in the same class of materials as ErAs, would also fail to exhibit a semimetal-to-semiconductor transition when spatially confined. When considering the effects of interface states [29] and strain [30] on the Fermi level, and subsequently the possible transition from a semimetallic material to a semiconductor, the *f*-shell electrons of ErAs can be dealt with by the frozen core approximation. Therefore it is reasonable to assume that the small differences in the number of *f*-shell electrons contained by ErAs and TbAs would cause minimal changes in their electronic properties. Our evidence of trap saturation in TbAs nanoinclusions indicates a surprising transition to semiconductor behavior and suggests a more sophisticated theoretical treatment may be necessary to understand the electronic structure of TbAs nanoinclusions.

IV. CONCLUSION

We have investigated carrier relaxation processes in GaAs samples containing embedded TbAs nanoinclusions on a GaAs substrate using an optical-pump THz-probe transient absorption spectroscopy experiment. Fitting the data to a triple exponential decay yields numerical values for the carrier relaxation lifetimes as well as the fraction of carriers participating in each relaxation process. We observe a pump fluence dependence of the fraction of carriers participating in fast relaxation processes, as well as the bulk lifetime. These dependences suggest that the TbAs states are saturable, ruling out the existence of metallic states that would allow fast thermalization of carriers and therefore prohibit saturation. Absorption spectra and the evidence for trap state saturation point to the existence of a band gap. The absence of photoluminescence associated with the only optical absorption peak allows us to rule out the existence of a direct band gap. The data therefore suggest that spatial confinement of TbAs likely results in nanoinclusions with the electronic structure of an indirect gap semiconductor. The trap filling dynamics at low pump fluence are not affected by temperature over the range of 4.5–293 K, which suggests that the trap states are deep in the band gap and that phonon-mediated recombination across an indirect band gap is not a dominant trap emptying process. The dependence of trap emptying rate on increasing population of carriers in the conduction and valence bands suggests the existence of important nonradiative recombination pathways involving both free carriers and those confined within the nanoinclusions.

The results presented here provide insight into the dynamics of carriers in GaAs containing TbAs nanoinclusions and will guide the future development of TbAs:GaAs materials for optoelectronic device applications. For example, it may be possible to simultaneously make use of the Fermi-level pinning of TbAs and the fast recovery time of ErAs by using a core-shell structure in order to develop new materials for THz device applications. Development of such materials will rely on the analysis of electronic properties and carrier dynamics developed here.

ACKNOWLEDGMENTS

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