Femtosecond response times and high optical nonlinearity in beryllium-doped low-temperature grown GaAs

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We have investigated the effect of beryllium doping on the optical nonlinearity and on the carrier dynamics in low-temperature (LT) grown GaAs for various growth temperatures and doping levels. Pump–probe experiments with 20 fs pulses and quantitative measurements of the nonlinear absorption show that in undoped LT GaAs, ultrafast response times are only obtained at the expense of low absorption modulation. In contrast, in Be-doped LT GaAs, high absorption modulation is maintained for response times as short as 100 fs. These results are qualitatively explained accounting for the point-defect-related optical transitions in LT-GaAs. © *1999 American Institute of Physics.* [S0003-6951(99)02509-7]

Ultrafast all-optical switching devices based on III–V semiconductor materials[1] have found many applications in high-bit-rate communication systems[2] and in femto- and picosecond laser pulse generation.[3,4] Ultrafast all-optical switching with resonant nonlinearities requires materials with a fast temporal response of the nonlinear absorption, high absorption modulation, and low absorption when the material is totally saturated, i.e., low nonsaturable losses. Furthermore, the bandwidth of the nonlinearity has to be large for applications on the sub-100 fs time scale, requiring above-band gap excitation of the semiconductor continuum transitions.[5] A standard approach for obtaining sub-ps response times in III–V semiconductors, such as GaAs, is low-temperature (LT) growth.[6] Although the time response of LT-GaAs has been extensively investigated,[6–8] not much is known about the strength of the nonlinearity. In this letter, we study both the nonlinearity and the time response of LT GaAs. We demonstrate that fast response times of undoped LT GaAs are only obtained at the expense of low absorption modulation and high nonsaturable losses. Most significantly, we show that Be doping of LT GaAs[9] yields a material with an ultrafast, 100 fs response time, high modulation, and low nonsaturable losses. Our results demonstrate that this material is superior to undoped LT GaAs for all-optical switching applications. We will discuss the physics responsible for these findings.

We have studied 500 nm thick as-grown molecular beam epitaxy (MBE) LT GaAs layers, either undoped (nominal growth temperature $T_g = 200-580\,^{\circ}\mathrm{C}$) or doped with $1\times10^{19}\,\mathrm{cm}^{-3}$ or $3\times10^{19}\,\mathrm{cm}^{-3}$ Be (nominal $T_g = 250, 300\,^{\circ}\mathrm{C}$). These layers were either grown on $Al_{0.15}Ga_{0.85}As$/AlAs Bragg mirrors or on GaAs substrates with etch stop layers. The substrates of the etch stop samples were removed by wet etching to allow for transmission experiments. All samples were antireflection coated on the front surface. We have verified by x-ray diffraction and transmission electron microscopy that the structural properties of the LT GaAs layers are independent of the substrate type and that all layers have good quality. The Bragg mirror samples were used for quantitative measurements of the nonlinearity in a reflection geometry, since a more accurate calibration can be obtained with these samples, and for pump–probe with 100 fs pulses. The etch stop samples were used for pump–probe experiments in a transmission geometry with 20 fs pulses, for which the bandwidth of the Bragg mirrors was too small. We note that all experiments measure absorption changes, independent of the type of sample and the geometry, and that the effective sample thickness in the reflection geometry is twice the physical thickness. All optical measurements were performed at room temperature.

First, we recall some basic properties of undoped LT GaAs. During LT growth, excess As is incorporated as As antisite point defects As_{Ga}, which are double donors close to the center of the band gap. Less than 10% of the As_{Ga} are ionized, As^{+}_{Ga}, while the majority is neutral, As^{0}_{Ga}.[10] The concentration of As_{Ga} increases with decreasing growth temperature T_g, reaching $10^{20}\,\mathrm{cm}^{-3}$ for $T_g = 200\,^{\circ}\mathrm{C}$ and a beam equivalent pressure (BEP) of about 20.[10] It is well established that the fast response time of LT GaAs is due to carrier trapping into defects, which becomes faster with decreasing T_g.[6–8,11] Electrons are trapped into ionized As^{+}_{Ga}.[7,11]

The temporal decay of the absorption modulation, measured in degenerate pump–probe experiments with 100 fs pulses centered at 830 nm, is shown in Fig. 1 for 300 °C grown samples, undoped or with different Be concentrations [Be]. A faster response is obtained with increasing Be concentration. We characterize the temporal response by the $1/e$

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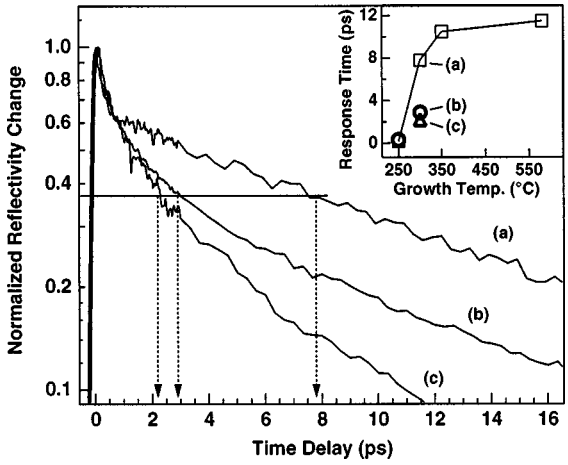

FIG. 1. Normalized reflectivity change of 500 nm thick LT GaAs layers grown at 300 °C for different doping levels: undoped (a), $[\mathrm{Be}]=1\times10^{19}\,\mathrm{cm}^{-3}$ (b), and $[\mathrm{Be}]=3\times10^{19}\,\mathrm{cm}^{-3}$ (c). The 100 fs excitation pulses were centered at 830 nm (carrier density$\approx5\times10^{18}\,\mathrm{cm}^{-3}$). Inset: 1/*e* decay times for a larger set of samples vs growth temperature: Undoped (□), $[\mathrm{Be}]=1\times10^{19}\,\mathrm{cm}^{-3}$ (○), $[\mathrm{Be}]=3\times10^{19}\,\mathrm{cm}^{-3}$ (△).

decay times, shown versus growth temperature in the inset for a larger set of samples. Be doping results in a much faster decay for $T_g=300\,^{\circ}\mathrm{C}$ while the decay for $T_g=250\,^{\circ}\mathrm{C}$ remains essentially unchanged. Considerably faster response times are obtained from pump–probe experiments with 20 fs pulses (shown in Fig. 2) for the same set of samples as in Fig. 1. Again, we find that Be doping results in a faster initial decay, as fast as 100 fs, which is clearly demonstrated by the 1/*e* decay times plotted versus growth temperature in the inset of Fig. 2.

We recall that the Fermi level is lowered upon acceptor doping, which gives rise to a larger fraction of $\mathrm{As}_{\mathrm{Ga}}^{+}$ and to the formation of doubly charged $\mathrm{As}_{\mathrm{Ga}}^{2+}$ for high Be concentration.[9] Consequently, we attribute the faster response for increased Be concentrations to enhanced electron trapping due to the larger concentration of ionized antisites, in agreement with earlier work.[9] Moreover, we recall that fast trapping is followed by slower recombination of trapped

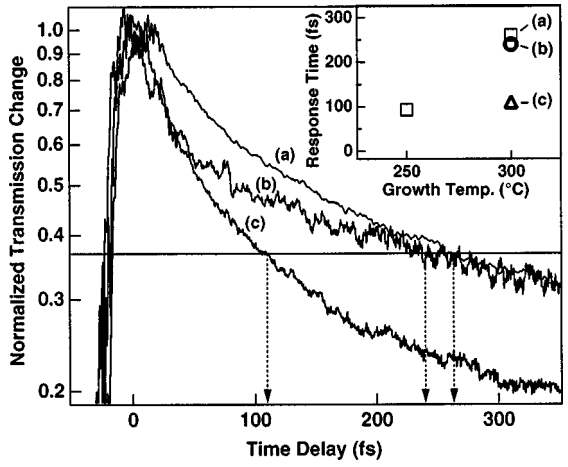

FIG. 2. Normalized transmission change of 500 nm thick LT GaAs layers grown at 300 °C for different doping levels: undoped (a), $[\mathrm{Be}]=1\times10^{19}\,\mathrm{cm}^{-3}$ (b), and $[\mathrm{Be}]=3\times10^{19}\,\mathrm{cm}^{-3}$ (c). The 20 fs excitation pulses were centered at 810 nm (carrier density$\approx5\times10^{17}\,\mathrm{cm}^{-3}$). Inset: 1/*e* decay times for a larger set of samples vs growth temperature: Undoped (□), $[\mathrm{Be}]=1\times10^{19}\,\mathrm{cm}^{-3}$ (○), and $[\mathrm{Be}]=3\times10^{19}\,\mathrm{cm}^{-3}$ (△).

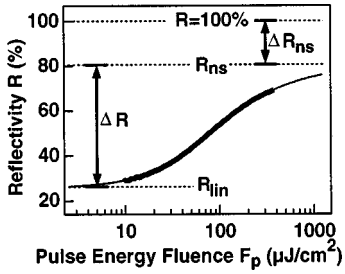

FIG. 3. Reflectivity R of a single pulse vs pulse energy fluence F_p: (thick line) measured data, (thin line) fit yielding the linear reflectivity R_{lin}, the saturated reflectivity R_{ns}, the absorption modulation ΔR, and the nonsaturable losses ΔR_{ns}.

carriers in LT GaAs[7,12] and that trapping is slowed down due to trap filling at higher carrier densities and longer times.[8] This is the reason why the time response measured at higher carrier densities ($5\times10^{18}\,\mathrm{cm}^{-3}$) with 100 fs pulses is slower than the one measured at $5\times10^{17}\,\mathrm{cm}^{-3}$ with the 20 fs pulses. Only measurements with pulse widths well below 100 fs can reveal the 100 fs time response of LT GaAs:Be.

The modulation and the nonsaturable losses are determined from quantitative measurements of the reflectivity as a function of the pulse energy fluence. The experimental data are extrapolated to the linear regime and to arbitrarily high fluences using the traveling wave rate equation model for a two-level absorber.[13,14] This yields the linear and the maximum reflectivity R_{lin} and R_{ns}. We define the modulation as $\Delta R=R_{\mathrm{ns}}-R_{\mathrm{lin}}$ and the nonsaturable losses as $\Delta R_{\mathrm{ns}}=1-R_{\mathrm{ns}}$, as illustrated in Fig. 3.

Figure 4 shows ΔR and ΔR_{ns} versus the response times from Fig. 1, measured with 100 fs pulses. In undoped LT GaAs, ΔR substantially decreases and ΔR_{ns} drastically increases for shorter response times corresponding to lower growth temperatures. A substantial improvement is obtained by Be doping. The data clearly demonstrate that for a given response time, LT GaAs:Be exhibits by far larger modulation and lower nonsaturable losses than undoped LT GaAs. Alternatively, for a given modulation a much faster response is obtained. This makes Be-doped LT GaAs a superior material

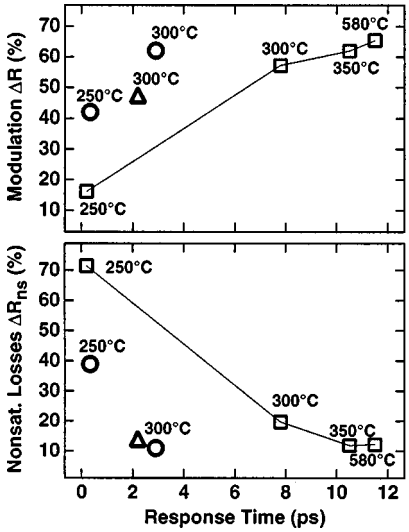

FIG. 4. Modulation ΔR and nonsaturable losses ΔR_{ns} vs 1/*e* decay times from Fig. 1. Undoped LT GaAs (□); LT GaAs:Be with $[\mathrm{Be}]=1\times10^{19}\,\mathrm{cm}^{-3}$ (○) and with $[\mathrm{Be}]=3\times10^{19}\,\mathrm{cm}^{-3}$ (△).

for applications in ultrafast all-optical switching.

We will now discuss the physics behind these results. It is well known from linear optics that the transition from neutral As^0_{Ga} to the conduction band (CB) substantially contributes to the above-band gap absorption[15] due to its high optical cross section.[16] The cross sections for the transition from the valence band (VB) to the ionized As^+_{Ga} and for the second optical ionization of As^+_{Ga} are small[16] and these transitions can be neglected. We expect that the As^0_{Ga}–CB transition is much more difficult to saturate than the VB–CB transition due to the large density of As^0_{Ga} and the high density of final states, 0.7 eV above the bottom of the CB. Therefore, we assume that the As^0_{Ga}–CB transition substantially contributes to the nonsaturable losses. This implies that an increase/decrease of the As^0_{Ga} concentration results in larger/smaller nonsaturable losses. In fact, we have experimentally found that the increased As^0_{Ga} concentration at lower growth temperatures in undoped LT GaAs results in larger nonsaturable losses (see Fig. 4). Moreover, we have verified by near-infrared absorption (NIRA)[10] that Be doping reduces the As^0_{Ga} concentration in our samples. As a consequence, reduced nonsaturable losses are observed in Be-doped LT GaAs for a given growth temperature.

In order to show how the As^0_{Ga}–CB transition affects the modulation, we make a very simple model to demonstrate the essence of the effect. The linear absorption coefficient α_{lin} is written as the sum of the VB–CB absorption α_s and the As^0_{Ga}–CB absorption α_{ns}. This yields $R_{\mathrm{lin}} = \exp[-2d(\alpha_{\mathrm{ns}}+\alpha_s)]$, for a sample thickness d. Assuming that the VB–CB absorption can be totally bleached and that the As^0_{Ga}–CB absorption does not depend on the pulse energy fluence, we obtain in the saturated regime $R_{\mathrm{ns}} = \exp(-2d\alpha_{\mathrm{ns}})$, and $\Delta R = R_{\mathrm{ns}} - R_{\mathrm{lin}} = \exp(-2d\alpha_{\mathrm{ns}})[1 - \exp(-2d\alpha_s)]$. This simple model illustrates how an increase of the As^0_{Ga}–CB absorption α_{ns} reduces both the linear reflectivity and the modulation ΔR. The decrease of ΔR with increasing α_{ns} is seen in Fig. 4 for decreasing growth temperatures in undoped LT GaAs. The decrease of the linear reflectivity with decreasing T_g was also observed (data not shown), in agreement with Ref. 15. We note that a careful analysis of our data on undoped LT GaAs shows that the As^0_{Ga}–CB absorption is responsible for the major fraction of the nonsaturable losses. Other sources for the nonsaturable losses are currently being investigated.

In summary, we have quantitatively studied the nonlinearity and the initial time response, resulting from carrier trapping, in undoped and Be-doped LT GaAs. We find that, in undoped LT GaAs, fast trapping is accompanied by weak absorption modulation. This is due to the presence of both ionized As^+_{Ga} electron traps and neutral As^0_{Ga}, giving rise to As^0_{Ga}–CB absorption which is difficult to saturate. Be doping reduces the concentration of As^0_{Ga} and increases the concentration of ionized As^+_{Ga}. Therefore, Be-doped LT GaAs combines a fast time response with high modulation, yielding a material with superior properties for applications in ultrafast nonlinear optics.

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