Temperature dependence of the absorption saturation relaxation time in light- and heavy-ion-irradiated bulk GaAs

J. Mangeney,[a] N. Stelmakh, F. Aniel, P. Boucaud, and J.-M. Lourtioz
Institut d'Électronique Fondamentale, UMR CNRS 8622, Université Paris XI, 91405 Orsay Cedex, France

(Received 23 January 2002; accepted for publication 6 May 2002)

The absorption saturation relaxation time in light- and heavy-ion-irradiated GaAs saturable absorbers has been measured as a function of the temperature in the range from 7 to 300 K. For both types of samples, the relaxation time is shorter than 4 ps at 7 K. A regular increase of this time with temperature is observed for light-ion-irradiated samples, a value of 9.5 ps being reached at room temperature. In contrast, an almost temperature-independent relaxation time is found for heavy-ion-irradiated samples. The results are interpreted on the basis of a simplified relaxation model accounting for capture and emission from defect levels. We suggest that light-ion irradiation creates shallow centers whereas heavy-ion irradiation creates deep centers. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1489086]

Fast saturable absorbers are crucial components for high-bit-rate optical data processing[1] and ultra-short pulse generation.[2] The device response time (i.e., the absorption saturation relaxation time) is usually shortened by introducing capture and recombination centers during[3] or after[1,2,4] the crystal growth. The ion irradiation is one of the most efficient techniques to obtain relaxation times as short as few picoseconds in a large choice of semiconductor materials either based on GaAs[1,2] or InP.[4] Previous studies[5,6] have shown that the ion mass has an influence on the spatial distribution of the atomic displacements induced by irradiation. Heavy-ion irradiation produces clusters of point defects, while light-ion irradiation produces isolated point defects. The defect morphology is in turn expected to affect the two main mechanisms involved in the absorption saturation relaxation: the free-carrier capture by defect levels and the carrier recombination. Direct electrical measurements of the electronic capture cross section have previously been carried out to study the capture process.[5] This approach is however not effective at short capture times.

In this letter, we propose an all-optical approach to study the ion irradiation effect. The all-optical approach consists in measuring the absorption saturation relaxation time dependence with temperature. Pump-probe experiments are carried out over a large range of temperatures from low temperature (7 K) to room temperature (300 K). Both proton- and Au^+-irradiated GaAs samples are studied. The capture time value and its dependence versus temperature are shown to provide information on the nature of defects created by ion irradiation. From theoretical considerations, we suggest that isolated point defects act as shallow centers whereas clusters of point defects behave as deep neutral centers. Experimental results also provide useful information on the device stability against temperature fluctuations. This point is crucial for applications.

The samples used in the experiments were grown by molecular beam epitaxy on a GaAs substrate. They consist of 10-μm-thick GaAs deposited on a 1-μm-thick $Al_{0.3}Ga_{0.7}As$ etch-stop layer. Windows of 300 μm diameter were opened into the substrate by chemical etching. Irradiation with H^+ and Au^+ were performed with the TANDEM accelerator located in Orsay. The ion energies used for bombardment were high enough (1.5 and 200 MeV for H^+ and Au^+, respectively) to ensure a uniform defect distribution in the first micron depth, which is the light probed depth. According to TRIM[6] simulations, the ion penetration depth was predicted to be much longer than the 10 μm GaAs thickness. In other words, the ions go through the absorber layer without being implanted. The irradiation doses were 10^{15} and 10^{12} cm^{-2} for the H^+ and Au^+-irradiated samples, respectively.

Degenerate pump-probe experiments were performed using an optical setup that consists of a Ti:sapphire mode-locked oscillator pumped by an Nd:Yag laser. The pulse repetition rate was 80 MHz. The pump and probe pulse duration was about 150 fs. Pump and probe were cross-polarized and focused onto the sample with an estimated spot diameter of 100 μm. The pump and probe pulse energies were 80 and 8 pJ, respectively. At each temperature, the pump and probe wavelengths were adjusted near the GaAs band edge to obtain the maximum differential transmission between the unsaturated and saturated absorption states. The samples were placed in a helium-flow cryostat. Figure 1 shows the differential transmission of H^+-irradiated (a) and Au^+-irradiated (b) samples measured at 7 and 300 K as a function of the pump-probe delay time. At 7 K, the time-resolved differential transmission signal rapidly decreases for both samples and reaches a negative value at long delays. This offset is more important for Au^+-irradiated sample. This effect is not observed at room temperature. This negative offset likely results from pump-induced absorption.[7] Indeed, a similar effect has previously been reported for low-temperature-grown GaAs[8] and was attributed to an electronic excitation from trap levels to the conduction band. The relaxation time is presently obtained by fitting the initial decay to a single decreasing exponential. Near liquid helium temperature, both samples exhibit a relaxation time close to 3 ps. The relaxation time of the H^+-irradiated sample increases up to ~9.5

[a]Author to whom correspondence should be addressed; electronic mail: mangeney@ief.u-psud.fr

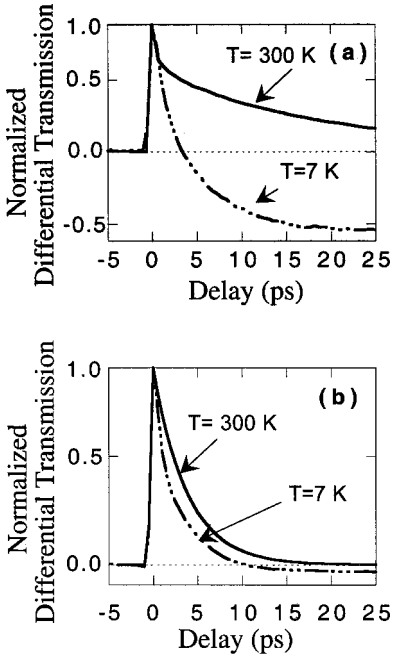

FIG. 1. Normalized differential transmission of proton- and Au^+-irradiated samples measured vs the pump-probe delay at two sample temperatures (7 and 300 K). The top figure (a) is for proton-irradiated samples, while the bottom figure (b) is for Au^+-irradiated samples.

ps at 300 K, while that of the Au^+-irradiated sample remains essentially unchanged.

Figure 2 shows the dependence of the relaxation time as a function of the temperature. The relaxation time invariance of Au^+-irradiated GaAs with temperature is clearly seen. In contrast, a monotonic increase of the relaxation time is observed for proton-irradiated GaAs, thus indicating a slower capture process. In both cases, the first decay of the differential transmission follows a single decreasing exponential, that corresponds to the carrier trapping. Free carriers move in the crystal with an average kinetic energy E_0 of the order of kT. When they approach a defect region (defined by a volume V_d), they can be trapped. In order to be captured, the carrier must lose kinetic energy that is carried away either by optical or acoustic phonons. Radiative recombination has presently a negligible influence at picosecond time scales. Auger recombination can also be disregarded due to the small average carrier density. However, according to the type of defect, the number of phonons involved in the transition from the conduction band edge to the trap level is variable and different capture probabilities must be considered. In the

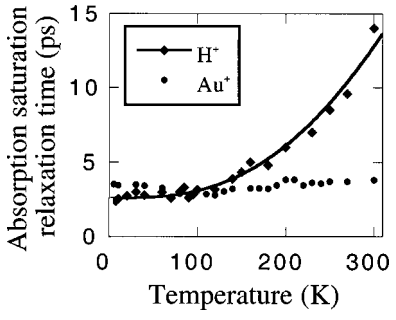

FIG. 2. Temperature dependences of the absorption relaxation time measured for proton- and Au^+-irradiated samples.

case of shallow centers, mainly one-phonon transitions are involved. In turn, carrier re-emission from defect levels can not be neglected. The competition between capture and emission thus leads to an apparent larger value of the capture time. As the emission process is more likely at high temperatures, the apparent capture time also increases with temperature. In contrast, for deep centers, the carrier capture involves nonradiative transitions with a multiphonon (MP) emission. The temperature dependence of the capture time is then controlled by the deep level charge.[9] For both attractive and repulsive centers, the capture time is expected to decrease with temperature. For neutral defect centers and moderate temperatures, the capture time is temperature independent, as shown by R. Pässler.[10] The present experimental results thus suggest that proton irradiation induces shallow defects in GaAs, while Au^+ irradiation induces deep neutral defects. To confirm the assumption for proton irradiation, we propose the following simplified model for the free carrier capture by shallow centers.

Shallow defects act as Coulomb attractive centers with a series of closely spaced excited states allowing one-phonon transitions. After interacting with a phonon, an electron can either escape with an energy gain or move closer to the ground state with an energy loss. The probability for the carrier to reach a lower energy level without being re-emitted is near unity for a carrier binding energy $U \geqslant 4\,kT$.[11] Including the free carrier energy E_0, the largest orbits that have to be considered correspond to state energies $E_0+U=5\,kT$. For an attractive Coulomb center of energy $V(r) = -Ze^2/4\pi\epsilon r$, the corresponding radius r of the largest orbits is $r=Ze^2/20\pi\epsilon\,kT$. The defect active volume $V_d = \frac{4}{3}\pi r^3 = \frac{4}{3}\pi(Ze^2/20\pi\epsilon\,kT)^3$ is then the volume within which the electron can reach the lower energy level without being re-emitted. Since only transitions with one-phonon emission are considered, the transition rate is proportional to $\langle n_q\rangle+1$, $\langle n_q\rangle=(e^{\hbar\omega_{\text{phonon}}/kT}-1)^{-1}$ being the average phonon number. It is also proportional to the defect filling factor V_dN_d, where N_d is the defect concentration. Finally, the last step limiting the rate at which electrons leave the conduction band is a multiphonon transition from the defect excited level to the ground state. The capture time can then be expressed as

$$\tau=\frac{A(1-e^{-\hbar\omega/kT})}{\frac{4}{3}\pi\left(\frac{Ze^2}{20\pi\epsilon\,kT}\right)^3 N_d}+\tau_{\text{multiphonon}}, \tag{1}$$

where A is a constant that links the one-phonon transition time to the average phonon number. A defect concentration $N_d \approx 4\times10^{17}\ \text{cm}^{-3}$ is presently estimated from φ/d, where φ is the irradiation dose and d is the average distance between elementary displacement calculated by TRIM. The optical phonon energy is $\hbar\omega=36$ meV. The capture time in formula (1) has thus two adjustable parameters: τ_{MP} and A/Z^3. Actually, τ_{MP} can be identified to the experimental relaxation time of H^+-irradiated GaAs measured at low temperature. Indeed, the one-phonon transition is very fast at 7 K. Following this, a τ_{MP} value of 2.8 ps is found. The second parameter, A/Z^3, must be adjusted to 1.58×10^{-14} s to fit the theoretical predictions to experimental data. As seen in Fig. 2, an excellent agreement is obtained for this parameter

value. From the fitted A/Z^3 value, one can approximate the collision time with one-phonon emission to 0.016 ps at room temperature. This time value leads to electron mobility ($\mu = e\tau/m^*$) equals to 400 $cm^2/V/s$, that is of the order of magnitude typically obtained for irradiated GaAs materials.[12]

Let us now consider deep centers. For temperatures in the range defined by $kT < \hbar\omega_{\text{phonon}}$, theoretical works predict[8] a deep center capture coefficient that varies as $T^{-1/2}$; T^0; $T^{-3/2}e^{[-3(\theta/T)^{1/3}]}$ in the cases of attraction, neutrality, and repulsion, respectively. The parameter θ depends on the center charge. Our experimental observations reveal the dominance of deep neutral centers in the case of heavy-ion irradiation. In other words, the condensation of point defects leads to a temperature-independent relaxation time even for high dissipated power densities. However, the previous formulas only take into account the carrier trapping process, while the re-excitation of trapped carriers to the conduction band is not considered. Actually, the negative offset of differential transmission experimentally observed at long pump-probe delays (Fig. 1) can be attributed to this defect-to-band reabsorption. The effect, more pronounced in H^+-irradiated samples, is observed only at low temperatures. This can be explained by a faster carrier capture leading to an excess population of trapped carriers in the defect states. The re-excitation of trapped carriers to the conduction band is then very efficient. Additional investigations are required for a more complete description of the carrier recombination process.

In conclusion, we have performed degenerate pump-probe experiments on ion-irradiated saturable GaAs absorbers over a temperature range from 7 to 300 K. The absorption relaxation time of heavy-ion-irradiated samples is found to be almost independent of the temperature. In contrast, the relaxation time of proton-irradiated samples significantly increases with temperature. A preliminary interpretation of the data suggests that light-ion irradiation creates shallow centers, whereas heavy-ion irradiation creates deep neutral centers. Incidentally, the experiments have also revealed the thermal stability of heavy-ion-irradiated semiconductor materials. This characteristic is crucial for high bit rate applications.

The authors greatly thank B. Sermage, J. Lopez, and J.-L. Oudar for many fruitful discussions. They are also indebted to S. Cabaret for his technical assistance and D. Gardes for sample irradiation.

[1] A. Hirano, H. Kobayashi, H. Tsuda, R. Takahashi, M. Asobe, K. Sato, and K. Hagimoto, Electron. Lett. **34**, 198 (1998).
[2] I. E. Adams, E. S. Kintzer, M. Ramaswamy, J. G. Fujimoto, U. Keller, and M. T. Asom, Opt. Lett. **18**, 1940 (1993).
[3] S. Benjamin, H. Loka, A. Othonos, and P. W. Smith, Appl. Phys. Lett. **68**, 2544 (1996).
[4] E. Lugagne Delpon, J. L. Oudar, N. Bouché, R. Raj, A. Shen, N. Stemakh, and J. M. Lourtioz, Appl. Phys. Lett. **72**, 759 (1998).
[5] D. Pons and J. C. Bourgoin, J. Phys. C **18**, 3839 (1985).
[6] J. P. Biersack and L. G. Haggmark, Nucl. Instrum. Methods **174**, 257 (1980).
[7] U. Seigner, R. Fluck, G. Zhang, and U. Keller, Appl. Phys. Lett. **69**, 2566 (1996).
[8] S. D. Benjamin, H. S. Loka, A. Othonos, and P. W. E. Smith, Appl. Phys. Lett. **68**, 2544 (1996).
[9] R. Pässler, Phys. Status Solidi B **103**, 673 (1981).
[10] R. Pässler, Phys. Status Solidi B **85**, 203 (1978).
[11] M. Lax, Phys. Rev. **119**, 1502 (1960).
[12] M. Mikou, R. Carin, P. Bogdanski, and P. Marie, J. Phys. III, 1661 (1997).