Nonlinear optical properties of GaAs quantum wires

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A nonlinear change in the absorption at discrete frequencies has been observed in chrysotile asbestos samples whose hollow channels, ≈6 nm in diameter, were filled with GaAs. The effect is attributed to saturation of optical transitions between quantum-well levels in GaAs quantum wires as they are excited by picosecond laser pulses. © *1995 American Institute of Physics.*

Researchers have recently been attracted to the unusual properties of semiconductor nanostructures in which the motion of the carriers is confined in two dimensions, i.e., quasi-1D structures or quantum wires. Quantum-size effects in the energy in quasi-1D systems become more prominent than those in quasi-2D systems. The energy gaps between the electron states are larger, and the density of states changes from a constant value for a 2D nanostructure to a discrete nature (with distinct peaks) for an ideal quasi-1D structure. Quasi-1D structures are promising for use as the active media of lasers which would have a low lasing threshold and which would operate at high temperatures, for the development of new types of transistors, for applications in optoelectronics, etc.

Several methods are available for fabricating semiconductor quantum wires from GaAs (Refs. 1–5). However, they generally require elaborate and expensive methods of molecular beam epitaxy or liquid phase epitaxy in combination with etching, the deposition of contacts, or cleavage of a resulting quasi-2D structure at a small angle or perpendicular to the surface. In the latter case, epitaxy is carried out a second time to deposit new layers of semiconductors on the cleaved surface (which is either smooth or terraced, with steps of atomic scale).

Unfortunately, these methods cannot produce samples containing quantum wires with dimensions sufficient for studies of optical and nonlinear-optics properties of quasi-1D structures.

A new method for producing quantum wires of GaAs in a transparent matrix was recently realized.[6] The idea is to use the method of Ref. 7 to fill hollow channels of nanometer radius in a chrysotile asbestos matrix with molten GaAs. The dimensions of the resulting samples make it possible to measure the linear absorption of GaAs quantum wires, to observe optical transitions between quantum-well levels, and to observe a substantial anisotropy of the absorption of light polarized either along or perpendicular to the quantum wires.

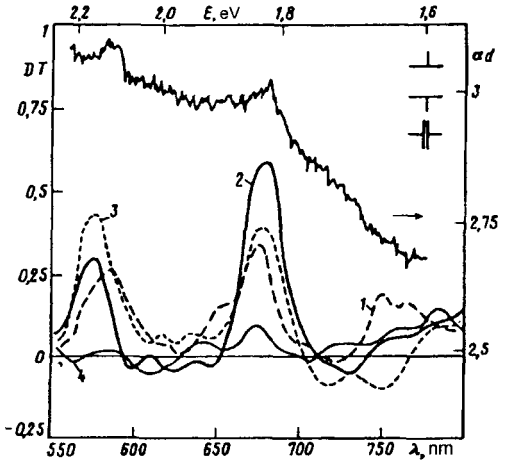

FIG. 1. Spectra of the linear absorption and differential transmission found for various time delays between the exciting and probe pulses, for GaAs quantum wires in chrysotile asbestos nanotubes. 1—15 ps (the probe pulse leads the exciting pulse); 2—0 ps; 3—20 ps; 4—50 ps.

In this letter we are reporting the observation of a nonlinear absorption by GaAs quantum conductors.

The test samples consisted of a chrysotile asbestos matrix (with the composition $Mg_3Si_2O_5(OH)_4$) in the form of a regular array of close-packed nanotubes filled with GaAs (see the inset in Fig. 2). The outside diameter of these nanotubes was about 30 nm. The inside diameter was measured by an electron microscope and found to be 6 nm (this is the diameter of the GaAs-filled channels). The typical length of the resulting GaAs wires was about 30 nm. To study the nonlinear absorption and its dynamic properties, we excited the GaAs quantum wires with ultrashort pulses of the second harmonic of a Nd:YAG laser (output pulse length of 30 ps, photon energy of 2.33 eV), and we probed them with ultrashort pulses of a picosecond continuum ("white light") at various times after the application of the pump pulse (we used an optical delay line). Differential transition spectra[8]

$$DT(\lambda)=\frac{T(\lambda)-T_0(\lambda)}{T_0(\lambda)}, \tag{1}$$

were measured with multichannel optical detectors. Here $T(\lambda)$ and $T_0(\lambda)$ are the transmission spectra of the excited and unexcited sample. The measurements were taken while the samples were at room temperature. Scattering of light was reduced by placing the samples in an immersion fluid.

Figure 1 shows a line absorption spectrum and a $DT(\lambda)$ spectrum of a 1D GaAs sample. In the line spectrum there are absorption bands peaking at 1.8 and 2.2 eV (corresponding absorption bands were detected in Ref. 6) and a knee at 1.7 eV. In the differential transmission spectrum we can clearly see brightening bands peaking at 1.8 and 2.2 eV (Fig. 1). For certain regions of the sample (the superimposed beams of the

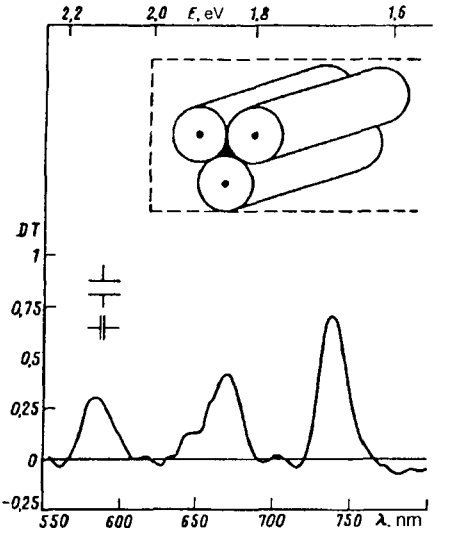

FIG. 2. Differential transmisssion spectrum for a region of a sample. There is a brightening band, peaking at about 1.7 eV. This spectrum was recorded as the exciting and probe pulses coincided in time. The inset shows the layout of the chrysotile asbestos matrix with the GaAs quantum wires.

probe light and the exciting light were moved over the sample), we observed an additional brightening band, peaking at 1.7 eV (Fig. 2). The brightening bands at 1.8 and 2.2 eV essentially disappeared from the differential transmission spectra after 50 ps (Fig. 1). The brightening band at 1.7 eV underwent relaxation more rapidly (its relaxation time could not be measured because of the limited time resolution of the apparatus).

The brightening bands at 1.8 and 2.2 eV can be attributed to saturation of optical transitions between low-lying quantum-well levels of electrons and holes corresponding to valence bands of the GaAs quantum wires (they correspond to absorption bands in the linear absorption spectrum of a sample found in both Ref. 6 and the present study). The energy of the first optical transition between the lower quantum-well level of the conduction band and the upper quantum-well level of the valence band can be estimated in the effective-mass approximation from[9]

$$E = E_g^i + X_{01}^2 \hbar^2 / 2\mu\rho^2, \tag{2}$$

where E_g^i is the width of the band gap (i is the corresponding valence band), $X_{01} \cong 2.4$, $\mu = m_e m_h^i / (m_e + m_h^i)$ is the reduced effective mass (m_e is the effective mass of an electron, m_h^i is the reduced effective mass of a hole in the corresponding valence band), and ρ is the radius of the quantum wire.

The brightening band at 1.8 eV in the differential transmission spectrum can then be identified with saturation of an optical transition between an upper quantum-well level of the valence band and a lower quantum-well level of electrons in GaAs quantum wires 6 nm in diameter. The band at 2.2 eV can be identified with saturation of a transition

between an upper quantum-well level for holes of the spin–orbit-split valence band and a lower quantum-well level of quantum wires of the same diameter.

One might suggest that the brightening band at 1.7 eV arises from saturation of a lower optical transmission between quantum-well levels of electrons and holes in the quantum wires, which form because the volume between the tubes of the chrysotile asbestos is filled with semiconductor (see the inset in Fig. 2). The decrease in the relaxation time of the brightening band at 1.7 eV in comparison with that of the bands at 1.8 and 2.2 eV can apparently be attributed to a greater effect of nonradiative surface recombination in quantum wires having a larger specific surface area.

The fast nonlinearity observed in the GaAs quantum wires, which is quite pronounced (an induced brightening was detected at excitation levels $\leqslant 100$ MW/cm^2), and the fast nonlinearity in the GaAs quantum wires support the conclusion that this material holds promise for effective optical switching.

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