Polarization Dependent Excitonic Optical Nonlinearities in GaAs/AlGaAs Multiple Quantum Wells under Anisotropic In-Plane Strain

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The photoinduced bleaching and blueshift of the lowest heavy-hole-like exciton in an anisotropically strained multiple quantum well exhibits a large enhancement for femtosecond light pulses polarized parallel to the axis of maximum compressive strain relative to those measured using orthogonally polarized light. These results imply that the strain-induced anisotropy in k space leads to exciton-exciton interactions which possess a strong dependence on the orientation with respect to the strain axis, relative orientation, and spatial extent of the photogenerated excitonic dipoles.

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The introduction of an anisotropic homogeneous strain in a solid results in a lowering of symmetry with respect to that of the strain-free material [1]. This principle may be demonstrated in multiple quantum well (MQW) structures through the creation of such a strain in the plane of the layers, which leads to a reduction of the fourfold rotation symmetry about a growth axis normal to the (100)-oriented substrate, and produces a mixing of the heavy- and light-hole valence band wave functions near $k_{//} = 0$ [2,3]. Recently it has been shown that controllable, internal anisotropic strain can be readily achieved in MQW's [4], leading to the observation of a strong dependence of the optical constants in the spectral region near the exciton peaks on the orientation of linearly polarized light incident normal to the structure [5]. These results have been exploited in the tailoring of the valence band Bloch functions for the realization of a new class of polarization- and phase-sensitive semiconductor devices [4] which combine the elements of high contrast and high speed crucial for the next generation of optoelectronic applications. One of the most intriguing aspects of these devices is the ultrafast large-angle (42°) polarization rotation observed on a femtosecond time scale [6], which, when compared with the ~1° rotation associated with anisotropic k-space filling in unstrained GaAs [7], indicates that anisotropic strains as small as 0.2% produce dramatic changes in the polarization dependence of the nonlinear optical properties. Application of a perturbation such as controllable anisotropic strain in conjunction with polarization-sensitive nonlinear optical techniques allows for the first time the systematic investigation of the important effect of the reduction of symmetry on the fundamental nature of excitons and exciton-exciton interactions, an area possessing far-reaching technical ramifications.

In this Letter we present the first study of the nonlinear optical properties of excitons in MQW structures under anisotropic in-plane strain. In particular, we have used femtosecond pump-probe spectroscopy to observe a striking polarization dependence of these properties for the resonantly excited lowest heavy-hole-like exciton state of the structure. This dependence is manifested most dramatically at zero time delay for light polarized parallel to the axis of maximum compressive strain through an enhancement of the photoinduced bleaching by *greater than one order of magnitude* and of the blueshift of the exciton resonance by nearly a factor of 2 relative to the same quantities measured with orthogonally polarized light. These results imply that the strain-induced anisotropy in k space leads to exciton-exciton interactions which possess a strong dependence on the orientation with respect to the strain axis, relative orientation, and spatial extent of the nonequilibrium excitonic dipoles.

The sample employed in our investigations is a p-i-n structure in which the intrinsic region consists of a 100 period, nominally (150 Å GaAs)/(50 Å $Al_{0.1}Ga_{0.9}As$) MQW. This structure was removed from its (100) host GaAs substrate by epitaxial lift-off [8] and bonded to a lithium tantalate ($LiTaO_3$) substrate cut such that its room temperature linear thermal expansion coefficient in the y direction, a_y, matched that of GaAs while its orthogonal counterpart, a_x, was significantly larger [4]. Using this design a maximum compressive uniaxial strain of ~0.2% in the x direction was obtained at the measurement temperature of 30 K. Photoinduced transmission measurements utilizing the conventional pump-probe geometry were performed with the aid of 100 fs pulses derived from a self-mode-locked Ti:sapphire laser. The center frequency of the laser was chosen for the resonant generation of cold excitons corresponding to the lowest excited state of the strain-perturbed system, which is primarily heavy-hole excitonlike in character [5]. Polarization rotators enabled the independent orientation of the pump and probe polarization vectors with respect to the x and y axes of the sample.

Figure 1 shows the normalized photoinduced transmission $\Delta T/T_0$ as a function of time delay between the pump and probe pulses for the four possible permutations of pump and probe polarizations parallel and perpendicular to the x axis. A spectrometer placed before the probe detector was tuned to the heavy-hole exciton peak (1.5285 eV), and the transmission T_0 was measured in the

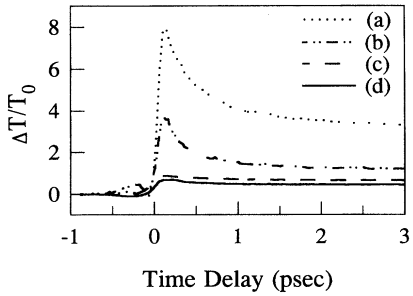

FIG. 1. Photoinduced bleaching $\Delta T/T_0$ as a function of time delay for pump (e) and probe (p) polarizations such that (a) $e//p//x$; (b) $e//y$, $p//x$; (c) $e//x$, $p//y$; and (d) $e//p//y$.

configuration for which the probe preceded the pump by 1 ps. Comparison of the four curves indicates that the $\Delta T/T_0$ data collected with the probe polarization parallel to the x axis [curves (a) and (b)] are strikingly different from those for which the probe polarization is orthogonal to this axis [curves (c) and (d)] in both magnitude and shape. The peak value of $\Delta T/T_0$ for the case in which the pump (e) and probe (p) polarizations are parallel to the x axis [curve (a)] is more than one order of magnitude greater than that measured with the polarizations of both beams perpendicular to this axis [curve (d)]. The maximum is strongly dependent on pump polarization for $p//x$, varying by more than a factor of 2 for $e \perp x$ [curve (b)], but is relatively insensitive to the orientation of the electric field vector of the exciting beam when the probe is perpendicular to the x axis, as seen from curve (c). Although all of the data exhibit decays with a time constant of ~0.5 ps, curves (a) and (b) show a sharp onset in photoinduced bleaching at $t = 0^+$, which decays to a plateau of less than half the maximum value, while curves (c) and (d) are characterized by a more step-function-like response possessing a smaller decay.

Further information about the anisotropy in the nonlinear optical properties can be obtained from the spectra shown in Fig. 2. While all of the data for the four experimental configurations indicate a reduction in the peak absorption and a concomitant line broadening [9] at $t = 0^+$ which are commensurate with the bleaching maxima of Fig. 1, they also exhibit blueshifts of the exciton resonance which possess subpicosecond decays similar to those shown in that figure. The largest measured blueshift occurs at $t = 0^+$, and a maximum value of $\Delta E = 1.5$ meV is attained for the $e//p//x$ case. This shift is nearly a factor of 2 greater than its counterparts for the $e//p//y$ and $e//y, p//x$ configurations (0.8 meV), and about 3 times larger than that found for the $e//x, p//y$ case (0.6 meV). These numbers are striking in that the maximum blueshift under similar experimental conditions for an unstrained MQW with similar well and barrier widths but with 30% aluminum content is only 0.5 meV [10]. The persistence of this blueshift for time delays significantly longer than the pulse autocorrelation width indicates that this phenomenon is primarily

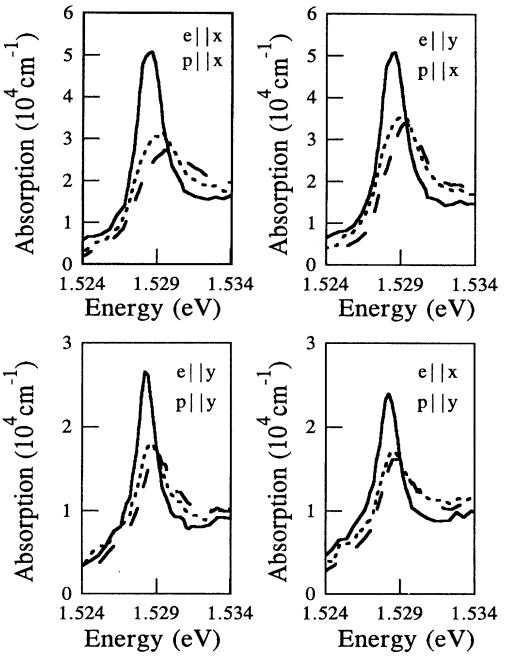

FIG. 2. Time-resolved absorption spectra for delays of −1 ps (lines), 0 ps (long dashes), and 3 ps (short dashes). The orientations of the pump (e) and probe (p) polarizations with respect to the sample axes are displayed in the figure.

associated with the creation of real excitations in the wells [11] rather than with the ac Stark effect [12].

Although the data of Fig. 2 show that in the absence of the pump the absorption of probe light polarized parallel to the x axis is nearly twice as large as that measured for the orthogonal polarization, it is evident that this anisotropic absorption cannot alone account for the anisotropy in the optical nonlinearity. In pump-probe experiments on materials not possessing anisotropic strain, the magnitude of $\Delta T/T_0$ is primarily dependent upon pump-induced carrier density effects. Viewed in this way, one would expect from the polarization dependence of the absorption data that $\Delta T/T_0$ in our measurements for the $e//x$ configurations would be, at most, twice as large as that obtained for the $e//y$ cases, irrespective of the probe polarization. Examination of Fig. 1 indicates that the results are strikingly different from this expectation. Moreover, for a given pump polarization corresponding to the generation of a fixed carrier density, the bleaching signal at all time delays is significantly greater for $p//x$ than for $p \perp x$. This counterintuitive behavior suggests that a strain-induced modification of the exciton relative-motion orbital wave function occurs, which is manifested most dramatically through the dominant role of the orientation of the probe polarization with respect to the strain axis in the determination of the nonlinear optical properties of the anisotropically strained MQW.

In MQW structures which retain in-plane fourfold rotation symmetry, there are three mechanisms which at low temperature and intermediate carrier densities ($10^{10} < n <$

10^{11} cm^{-2}) provide major contributions to the bleaching at the photon energy corresponding to the exciton peak in the dark: line broadening due to exciton-exciton collisions [13], which diminishes the peak absorption while the integrated absorption remains relatively constant [9]; reduction of oscillator strength attributed to both the filling of the phase space associated with the single-particle fermion states which comprise the excitons and the related consequences of the short-range exchange effects [14,15]; and a correction of the electron and hole self-energies resulting from these short-range exchange effects which manifests itself in a blueshift of the exciton resonance corresponding to the energies that parallel-spin particles gain by avoiding each other [15]. In the composite-particle picture more appropriate at lower carrier densities, this shift may be viewed as emanating from a short-range hard-core repulsion of the excitons. None of these existing theories, however, is capable of accounting for the anisotropy in either the photoinduced bleaching or blueshifts observed in our experiments. In order to do so we have heuristically incorporated the strain-induced anisotropy in k space into these models as a guide for future theoretical calculations.

Recent calculations [16] of the effects of in-plane uniaxial stress on the dispersion of the heavy- and light-hole valence bands in a MQW show that for a 0.2% strain in the x direction similar to that employed in our experiments, the constant-energy surfaces for the lowest heavy-hole valence band approach ellipsoids in k space whose major axis is along k_y and is significantly longer than the minor axis along k_x. This implies that excitons in equilibrium are composed of a linear combination of single particle states which have larger k values in the k_y direction than in the k_x direction, with the associated real space charge distribution exhibiting a peak position r_x and extent along the x direction greater than that of the counterparts of these quantities along the y axis. The absorption of linearly polarized light results in the creation of nonequilibrium excitons with a coherent macroscopic polarization characterized by dipoles aligned along the direction of the electric field vector of the light with spatial extent determined by that of the major and minor axes of the real space charge distribution. Excitonic dipoles generated by x-polarized light are of greatest extent, and therefore possess the highest scattering probability [9,13], the largest efficiency for single particle k-space filling [14,15], and the greatest exchange self-energy corrections due to repulsion effects [15], while those dipoles excited along the y direction are of least extent, with a concomitant smaller scattering probability, phase space filling efficiency, and composite particle hard-core repulsion. These factors indicate that the broadening, blueshift, and reduction in oscillator strength of the exciton peak for a given pump polarization should be greater and therefore yield larger bleaching for $p//x$ than for $p\perp x$ at all delay times, in agreement with the experimental results.

It is also evident from the data of Figs. 1 and 2 that the relative orientation of pump- and probe-created excitons plays an important role in the determination of the bleaching near $t = 0^+$. Polarization selection rules for the optical matrix elements [5] indicate that the interband transition rate for light polarized parallel to the x axis is significantly greater than that for its orthogonal counterpart, leading to a larger density ($\sim 3 \times 10^{10}$ cm^{-2}) of excitonic dipoles aligned along the x direction for $e//x$ than that observed along the y axis ($\sim 2 \times 10^{10}$ cm^{-2}) for $e\perp x$. This factor combined with the smaller density of states along k_x and the higher scattering probability and phase space filling efficiency for dipoles created by x-polarized light leads to a reduction in absorption at the exciton peak corresponding to a bleaching $\Delta T/T_0$ for the $e//p//x$ configuration nearly 10 times greater than the $\Delta T/T_0$ measured for the $e//p//y$ case. The added effect of a blueshift which is nearly twice as large due to the enhancement of the exchange self-energy correction associated with the hard-core repulsion of pump- and probe-generated dipoles aligned along the x direction yields a maximum total bleaching at the measurement wavelength more than one order of magnitude greater for $e//p//x$ than for $e//p//y$, as shown in Fig. 1. The bleaching peaks corresponding to the two cases for cross-polarized pump and probe beams lie between the two extremes, with the $\Delta T/T_0$ for $p//x$ significantly greater than that for $p\perp x$, in accordance with our previous discussion. Comparison of the bleaching maxima at $t = 0^+$ as a function of pump polarization for a given probe orientation offers further insight into the data. Although the density of photogenerated excitons is 1.5 times larger for $e//x$ than for $e\perp x$, the maximum bleaching at the exciton peak for $e//p//x$ is ~ 2.7 times greater than that for $e//y, p//x$, while it is only ~ 1.1 times greater for $e//x, p//y$ relative to that observed for $e//p//y$. These results show that the mechanisms which produce bleaching are more effective when the pump-created excitonic dipoles are parallel rather than perpendicular to those generated by the probe beam, thus illustrating the importance of the initial degree of phase space filling orientational overlap in the determination of the magnitude of the absorption reduction at $t = 0^+$. This alignment effect is also evident in the blueshift data, which exhibits a greater initial shift for $e//p$ than for $e\perp p$ for a given orientation of the probe polarization.

At longer delay times, the data of Fig. 2 indicate that both peak absorption recovery and blueshift decay play important roles in the bleaching decays of Fig. 1. The influence of the former is most dramatically illustrated in the $e//p//x$ data, which possesses an absorption recovery by 3 ps corresponding to a decay of $\Delta T/T_0$ to approximately half of its maximum value, in contrast to the smaller absorption recovery found for the other three configurations. The effect of the temporal decline of the blueshift in conjunction with the absorption recovery and broadening is most clearly seen in the decay of $\Delta T/T_0$ to less than half of its $t = 0^+$ value within 3 ps for the $e//y, p//x$ data.

The subpicosecond bleaching decays of Fig. 1 are not observed in similar low temperature experiments on unstrained MQW structures, for which a decay time of ~10 ps has been reported [14]. Although such rapid decays in shallow MQW's associated with the spin relaxation due to valence band mixing at $k_{//} \neq 0$ have been measured using nonlinear circular dichroism techniques [17], it is important to note that the anisotropic strain employed in our experiments mixes heavy- and light-hole valence bands which near $k_{//} = 0$ couple to circularly polarized light of the same helicity [1,2,5], thus rendering our measurements with linearly polarized light insensitive to this spin relaxation. We therefore attribute our bleaching decays to the orientational relaxation of the excitonic dipoles produced by the exciton-exciton interactions described above, which aid in the development of a real space charge distribution approaching that of the equilibrium excitons. The directional anisotropy in the density of states suggests that the equilibrium charge is distributed more heavily along the y direction, thus indicating that a greater absorption recovery due to this orientational relaxation is expected for the $e//p//x$ data than for the other three cases, in agreement with experiment. Further insight may be gained by comparison of the $e//x$ and $e\perp x$ data at a time delay $t = 3$ ps such that the orientational relaxation should be mostly completed, with the ratio of the bleaching magnitudes for a given probe polarization corresponding to that of the carrier densities, $n(e//x)/n(e\perp x)$. The fact that this ratio is smaller (1.2) for $p\perp x$ and larger (1.8) for $p//x$ than $n(e//x)/n(e\perp x) \sim 1.5$ implies that at higher excitation densities the orientational relaxation may be impeded by the greater inability of nonequilibrium excitons to scatter into increasingly occupied final states which characterize the equilibrium charge distribution. Although the dephasing times may possess both a polarization and carrier density dependence, the added effect of this orientational relaxation "bottleneck" suggests a bleaching decay which becomes insensitive to increasing carrier density, as observed in polarization rotation experiments [6], for which the rotation decays with a time constant of ~0.5 ps at carrier densities in excess of 10^{11} cm^{-2}.

In conclusion, we have reported the first measurement of the polarization dependence of excitonic optical nonlinearities in MQW's under anisotropic in-plane strain. A large anisotropy in the photoinduced bleaching at the measurement energy has been observed which exhibits a strong dependence on the orientation of the probe polarization with respect to the axis of maximum compressive strain (x axis). This result is linked to the strain-induced anisotropy in k space, which for the absorption of linearly polarized light leads to the generation of excitonic dipoles aligned along the electric field vector of the light with spatial extent and therefore scattering rate, phase space filling efficiency, and exchange self-energy corrections dictated by the orientation of the light polarization. The polarization dependence is most dramatically illustrated for the experimental configuration in which both pump and probe polarizations are parallel to the x axis, for which case excitonic dipoles of greatest extent are created, with a concomitant enhancement of the photoinduced bleaching by greater than one order of magnitude and of the blueshift by nearly a factor of 2 relative to the same quantities measured with both polarizations parallel to the y axis, for which dipoles of least extent are generated. The subpicosecond bleaching decays are attributed to an orientational relaxation of the excitonic dipoles toward an equilibrium charge distribution determined by the k-space anisotropy.

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