NONLINEAR OPTICS IN QUANTUM-CONFINED STRUCTURES

Experimenters and theorists have combined two new and productive fields of physics—nonlinear optics and quantum confinement—to reveal a host of unusual and useful properties in semiconductor heterostructures.

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Nonlinear optical effects arise when one applies to a material system optical fields that are on the order of or larger than the atomic fields that exist within the system. Quantum size effects appear when the dimensions of a system become comparable to or even smaller than the natural length scale governing its quantum mechanics. Much recent progress has resulted from the combination of these two very productive areas of physics. Because of advances in semiconductor and laser technologies, it is now possible to apply very intense, ultrashort pulses of light to nanometer-scale semiconductor heterostructures. These conditions produce many new and exciting effects. This article aims to give the reader a flavor of the status of this quickly evolving field of research, with a special emphasis on the most novel and unexpected results.[1]

Nonlinear optical effects were first observed in and described for independent and localized systems such as dilute atomic vapors and molecular solutions. Researchers found that the concepts developed to describe these systems could be extended to ionic solids and molecular crystals, providing that some simple precautions (such as the use of local field correction) were taken. Thus until recently the description of nonlinear optical processes in solids paralleled that for atomic systems. However, in another field of condensed matter physics, superconductivity, collective effects are known to play a dominant role. These effects originate from the unique properties of interacting many-body systems with extended and delocalized elementary excitations (Cooper pairs). It is this collective behavior that provides superconductors with their most interesting and surprising properties.

The physics that governs the properties of optically excited semiconductors is special because it is based on two types of concepts: those developed to describe localized behavior and those developed to describe collective behavior. The near-bandgap interactions induced in semiconductors by incident light occur via interband transitions that originate from the atomic levels of the constituent atoms. However, the electron–hole pairs that participate in absorption and emission processes are delocalized, and electrons and holes interact strongly through the long-range Coulomb force. This unique combination of atomic-like and collective behavior results in a rich and complex array of properties. Electron–hole pairs can form neutral hydrogenic bound states, known as Wannier excitons, whose Bohr radii a_0 determine the natural length scales for optical processes in the semiconductor. Because the Bohr radii of these loosely bound electron–hole pairs are large on the atomic scale—a_0 ranges between roughly 50 and 500 Å—quantum size effects are important and rather easy to observe in the high-quality heterostructures currently available. Thus to some extent one has the ability to tailor the optical responses of the structures for specific purposes. (See the article by Yoshihisa Yamamoto and Richart Slusher on page 66.) The current interest in this field of research stems both from the exciting fundamental science and the potential applications.

Dimensionality and linear optics

Before discussing nonlinear optical properties, it is necessary and instructive to see how confinement in low-dimensional structures affects linear optical properties. Figure 1 shows the low-temperature absorption spectra $\alpha(\omega)$ of a series of high-quality indium gallium arsenide layers grown by Alfred Y. Cho at AT&T Bell Laboratories. The Bohr radius for this material is rather large, about 300 Å. The bottom spectrum corresponds to a single "three-dimensional" layer 6000 Å (about 20 a_0) thick. The three other spectra are from samples containing a total of 6000 Å of InGaAs in the form of ultrathin layers, or "quantum wells," separated by layers of the large-bandgap compound indium aluminum arsenide. These quantum well structures are made up of 30, 60 and 80 bilayers of thicknesses $L = 200$ Å ($a_0/2$), 100 Å ($a_0/3$) and 75 Å ($a_0/4$), respectively. In the 80-bilayer sample the InGaAs quantum well is thin enough to be considered quasi-two-dimensional.

This series of spectra clearly illustrates the effects of

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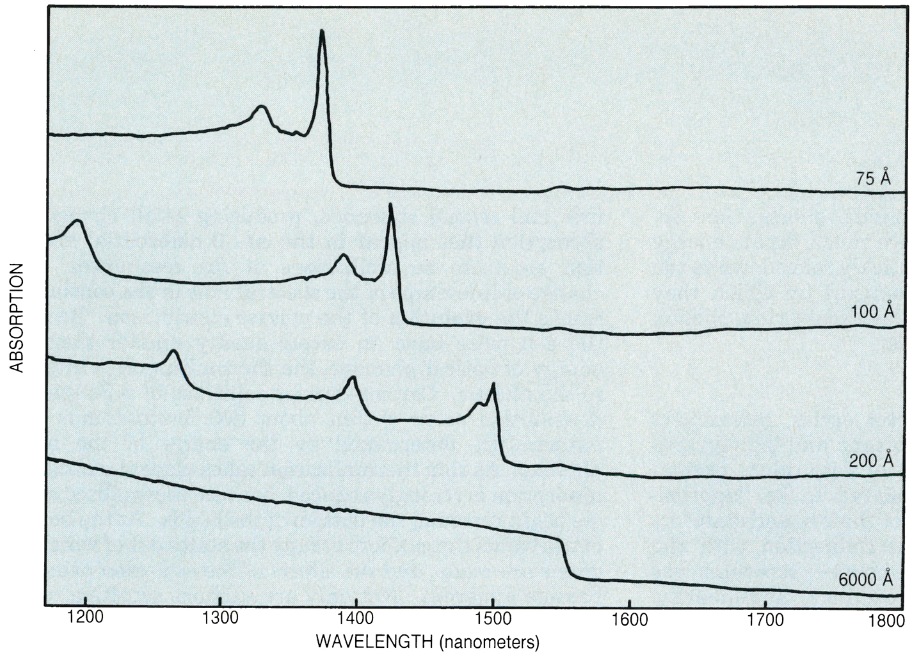


Transition from quasi-three-dimensional to quasi-two-dimensional behavior can be seen in these linear absorption spectra of InGaAs–InAlAs heterostructures. All the samples contain 6000 Å of active material (InGaAs). The bottom spectrum corresponds to a single "three-dimensional" layer 6000 Å thick (20 times the exciton Bohr radius a_0). The other three spectra are of ultrathin layers, or quantum wells, separated by barrier layers of large-bandgap material (InAlAs). Those samples consist of 30, 60 and 80 bilayers of thickness $L = 200$ Å, 100 Å and 75 Å, respectively. The bilayer thickness in the 80-bilayer sample, $L \sim a_0/4$, is thin enough to correspond to the quasi-two-dimensional limit. **Figure 1**

quantum confinement. The spectrum of the thick, 6000-Å sample consists of a step with no distinct exciton resonance (because of alloy disorder). The other spectra exhibit clear resonances followed by flat continua, indicative of the two-dimensional density of states. In addition, one can see several steps due to transitions that originate from the discrete quantization of the electron–hole wavefunctions in the direction normal to the layers.

By applying a magnetic field perpendicular to the plane of the quantum well, one can further confine e–h pairs. The crystal potential then confines the e–h pairs along the normal to the layers, and in the plane of the layers the magnetic field confines the pairs within the cyclotron radius $r_c \sim 1/\sqrt{B}$. Figure 2 shows the evolution of $\alpha(\omega)$ in the two-dimensional to zero-dimensional transition for a gallium arsenide quantum well structure 84 Å (roughly 0.6 a_0) thick.[2] At the highest field shown ($B =$ 12 tesla, $r_c \sim 70$ Å), one can say, very crudely, that e–h pairs are confined in very small "cylinders" of height L and radius r_c. The spectra show how the flat continuum in the $B = 0$ two-dimensional density of states evolves into a series of isolated peaks as the applied magnetic field increases, reaching a quasi-zero-dimensional density of states at $B = 12$ T. Furthermore, the details of evolution of these peaks and how they originate from the unresolved bound states near the gap give direct information on the band structure of the material and on the very special (parabolic) nature of the magnetic confinement.

This type of confinement enhances excitonic effects. In fact, for optoelectronic applications the most important property of quantum-confined heterostructures is that excitonic enhancement of optical transitions persists even above room temperature and in the presence of e–h plasmas of significant density.[1]

Nonlinear optical effects

In typical nonlinear optical experiments one applies to the sample an intense laser pulse, called the pump, which changes the sample's optical properties. The experimenter measures these changes by applying a second, very weak laser pulse, called the probe. Accurate and sensitive data acquisition techniques measure the optical transmission "seen" by the probe either when the pump is applied or in its absence; the difference between the two spectra measured by the probe pulse is known as the differential absorption spectrum. (See figure 3.) In this experimental configuration, however, the two laser pulses interfere within the sample, and photons can be scattered in directions compatible with the conservation of energy and momentum. These scattered photons are measured in coherent wave-mixing experiments, which give information on the way coherent polarization waves are generated in the sample and emit light.

When the pump pulse is applied, the state of a semiconductor can be described as a superposition of excited states. In other words, its condition is characterized by the populations and transition amplitudes of excitons induced by the pump. One can choose from among three energy regions to excite, or "pump," the semiconductor: the transparency region below the lowest exciton peak; coincident with an exciton resonance; or well above the resonances, in the continua of unbound states.

The nature of the nonlinear response is different in each case. If the sample is pumped in the transparency region there is, strictly speaking, no absorption; the populations in the exciton states are "virtual," lasting only as long as the pump is applied to the sample, and they are in phase with the pump field.

When the pump is resonant with an exciton state, its population dominates. Very early in the interaction the field of the pump laser drives populations back and forth between the ground state and the exciton, inducing oscillating transition amplitudes. Initially these oscillations are in phase with the driving field, so that the populations and amplitudes are "coherent." Eventually, though, scattering (by phonons, for example) produces dephasing. The populations remain in the exciton state, they become "real," photons from the pump disappear from the pump pulse, and what we call absorption occurs. Over a much longer time scale these excitons will recombine, emitting photons with a phase unrelated to that of the pump field.

For excitation in the continuum, the initial part of the process is similar to that in the resonant case. However, this early part of the process lasts a much shorter time, because the e–h pairs generated have much excess energy and are almost free. Hence they quickly relax down to the lowest exciton states. The mechanisms by which they reach this state include carrier–carrier scattering, phonon emission and bound-state formation.

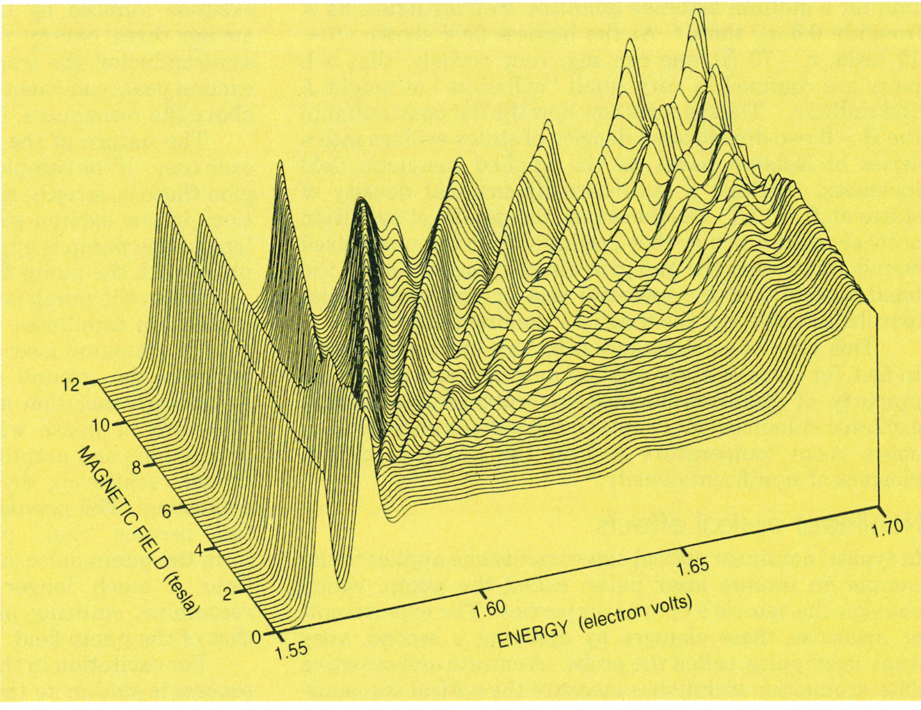
Relaxed populations

In the early days of semiconductor optics, researchers considered only linear effects important, and they investigated theoretically only the limit of very dilute exciton population. Excitons were considered to be "approximate" bosons, and the unbinding of their bound states by other e–h pairs was analyzed in connection with the metal–insulator transition. In this context, screening was considered the major source of excitonic nonlinearity. With the development of ultrashort-pulse lasers it became possible to separate experimentally the effects of screening from those due to the Pauli exclusion principle.[3]

Figure 3 shows the results of one experiment of this type: time-resolved differential absorption spectra for a series of time delays Δt between the pump and the probe before and just after excitation of a GaAs quantum well structure about 20 meV above the exciton resonance. The spectrum of the pump is shown at the bottom of the figure. Near $\Delta t = 0$ a nonthermal carrier distribution appears in the flat continuum between the two exciton series. Because of the Pauli principle, the absorption around the central energy of the pump photons is reduced, as evidenced by the spectral hole (shaded area in figure 3). The small shift of the spectral hole with respect to the laser spectrum results from many-body effects.[4] Furthermore, the e–h pairs immediately screen the excitons of the first and second subbands, producing small changes in absorption that appear in the $\Delta t \sim 0$ differential absorption spectrum as oscillations at the resonances. The change of line shape of the spectral hole in the continuum tracks the evolution of the carrier distribution. Because the e–h pairs have an excess energy smaller than the energy of optical phonons, the thermalization is internal to the plasma. Carrier–carrier collisions in a Boltzmann distribution occur within about 200 femtoseconds at a temperature determined by the energy of the pump photons. As this thermalization takes place the excitonic absorption is strongly reduced, because thermalized carriers begin reaching the bottom of the bands. At the bottom of the bands the carriers occupy the states out of which excitons are made, and the effects of the exclusion principle become apparent. Not only are excitons sensitive to the underlying Fermi statistics, but in quasi-two-dimensional systems the effects of Pauli exclusion dominate over the effect of screening.

From this type of experiment one can also extract information on the dynamics of the thermalization of "isolated" plasmas, an old problem of thermodynamics related to the Boltzmann H-theorem.[3] Carriers at the bottom of the bands will eventually form bound states. In this last phase of their evolution before recombination the exciton resonance shows a further blue shift,[5] which can be interpreted as a hard-core repulsion due to the exchange interaction. In the quasi-two-dimensional case the effects of exchange become visible because they are no longer compensated by the weakened screening.[6]

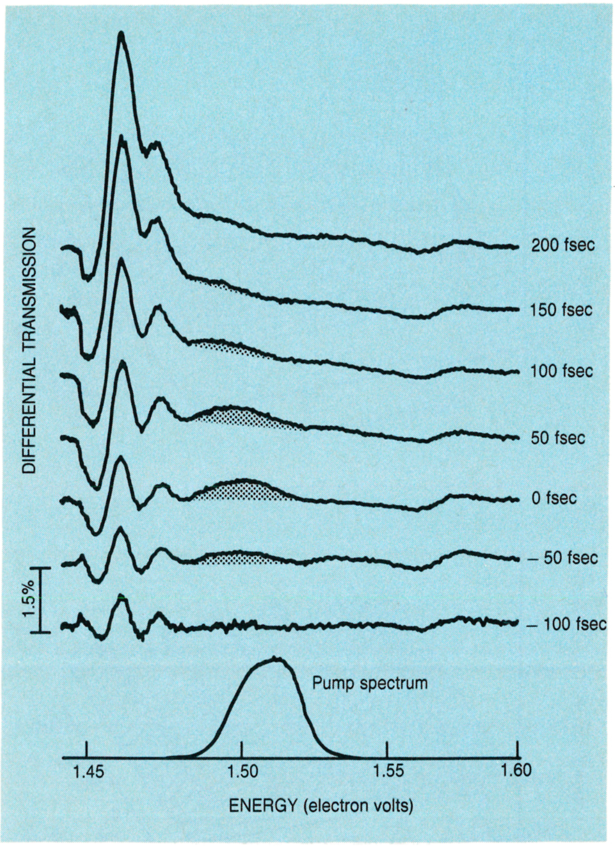
One can further confine the e–h pairs by applying a magnetic field, as described above. When no magnetic field is present the e–h pairs form the usual quasi-two-



Progressive confinement of electron–hole pairs from the quasi-two-dimensional regime to the quasi-zero-dimensional regime is evident in these linear absorption spectra of a GaAs quantum well structure subjected to increasingly strong perpendicular magnetic fields. (Adapted from ref. 2.) **Figure 2**

dimensional excitons; as the field strength approaches infinity the pairs tend toward electrons and holes in Landau levels. For intermediate field strengths they form magnetoexcitons. According to exact many-body results, in the presence of a strong magnetic field and at thermodynamic equilibrium, an ensemble of spin-polarized magnetoexcitons behaves like a gas of noninteracting, nonpolarizable point-like bosons.[7] Figures 4a and 4b show differential absorption spectra obtained at $B = 12$ T with a circularly polarized probe for the two polarizations of the pump that are resonant with the lowest magnetoexciton.[2] One observes strong responses up to high photon energy.

The difference between these two differential absorption spectra, shown in figure 4c, distinguishes the spin-dependent nonlinearities from the spin-independent ones. In these difference spectra the high-energy response disappears, indicating that the high-energy magnetoexcitons are sensitive only to charge-density effects such as screening and collisional broadening. Conversely, the lowest-energy excitons generated by the probe are sensitive to the spins of those created by the pump, as expected from the Pauli exclusion principle. In addition, one finds no trace of the blueshift that is seen at zero field and is indicative of the exciton–exciton interaction. Experiments performed with magnetic fields of different strengths have demonstrated that the hard-core repulsion blueshift does indeed decrease very strongly as field strength increases.[2] This occurs because as the field strength increases, magnetic confinement begins to dominate over the Coulomb interaction. Since the cyclotron radius is independent of mass, electrons and holes are forced into identical, overlapping and shrinking wavefunctions. Thus the magnetoexcitons become less polarizable and locally neutral, their interaction is reduced, and eventually, as the field strength approaches infinity, it vanishes. This is the essence of the exact many-body result, supported by experimental evidence.[2,7]



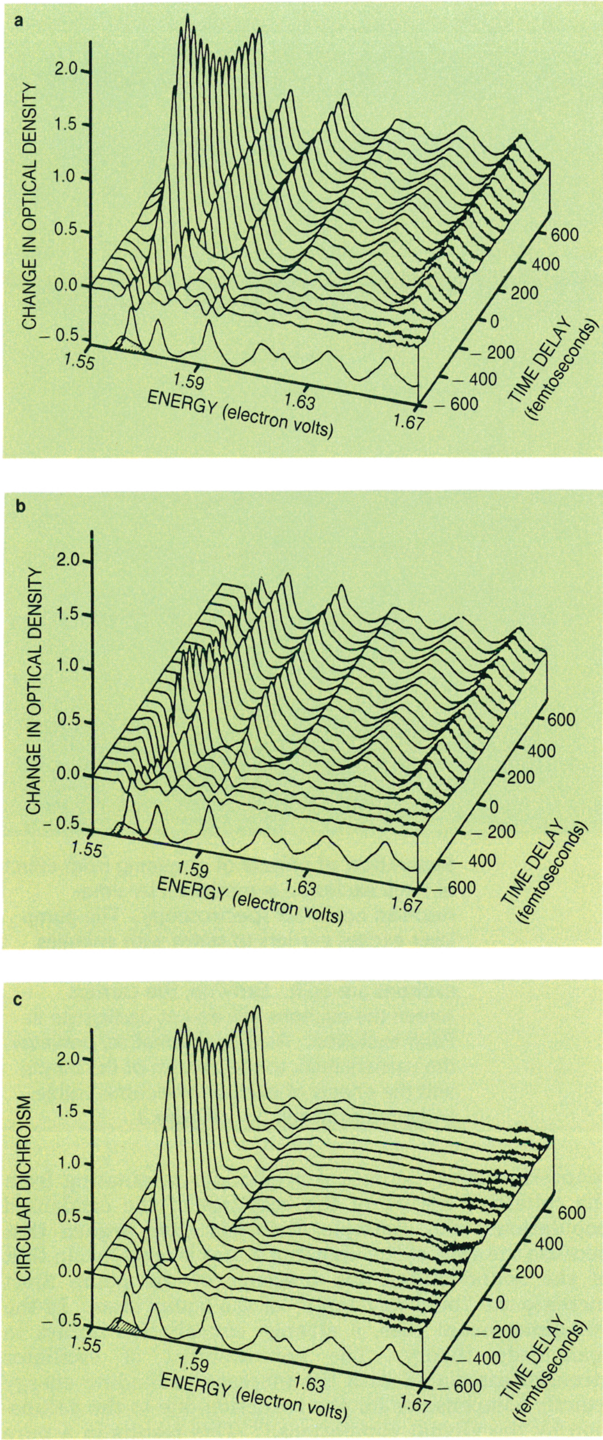
Separation of effects of screening from effect of Pauli exclusion is measured by time-resolved nonlinear spectroscopy. The pump laser excites carriers to states with energies above the energies of the states from which excitons are built. Early on, the carriers screen the excitons but do not participate in Pauli exclusion. As they thermalize, however, the carriers relax to the bottom of the bands, and the effects of exclusion become visible. (Adapted from ref. 3.) **Figure 3**

Coherent nonlinear optical effects

Coherent nonlinear optical effects occur before relaxation processes are able to destroy the phases of the populations and polarizations. The most striking situation involves excitation well below the absorption edge. In this case the virtual populations generated by the pump are all in phase. The weak probe then sees a blueshift of the exciton resonances, which recover their original positions immediately after the pump pulse has left the sample. An example of this effect, known as the excitonic ac Stark effect,[8,9] is shown in figure 5. This effect has attracted a lot of attention, because it can be viewed as a Bose condensation of populations in all the excitonic states stimulated by the "symmetry breaking" pump field.[9]

The theory of the ac Stark effect parallels the BCS theory of superconductors, with two changes that reflect the fact that the condensation is not spontaneous but stimulated: The chemical potential of the condensate is the energy of the pump photons, and as compared with the famous BCS equation, the "gap equation" contains an extra term (the pump Rabi energy $\mu E_{\rm p}$) that describes the coupling to this field. The excitonic ac Stark effect has been extensively investigated both experimentally and theoretically. Experiments have verified some rather counterintuitive theoretical predictions originating from the collective nature of the response of the condensed populations. For example, theorists have argued that because the shift of the unbound states is larger than that of the bound states, the presence of the pump must increase the "binding energy" of the bound state. In the two-dimensional case, a strange cancellation occurs in many-body theory: The enhancement of oscillator strength associated with the increase in binding energy exactly compensates for the reduction due to the occupation by the virtual populations.[10] This results in a pure shift of the resonances, without a change of magnitude, which indeed has been observed experimentally.[11]

Using ultrashort pulses one can generate real populations and investigate their behavior at early times, before and during dephasing. Time-resolved studies of the dynamics of coherent light scattering have revealed qualitative differences between the way semiconductors and independent atomic systems emit coherent light.[12,13] For atoms, the only nonlinearity that contributes originates from the exclusion principle and is therefore "instantaneous." In this case nonlinear wave-mixing occurs because of the interaction of the optical pulses with polarization waves. In semiconductors, because of the

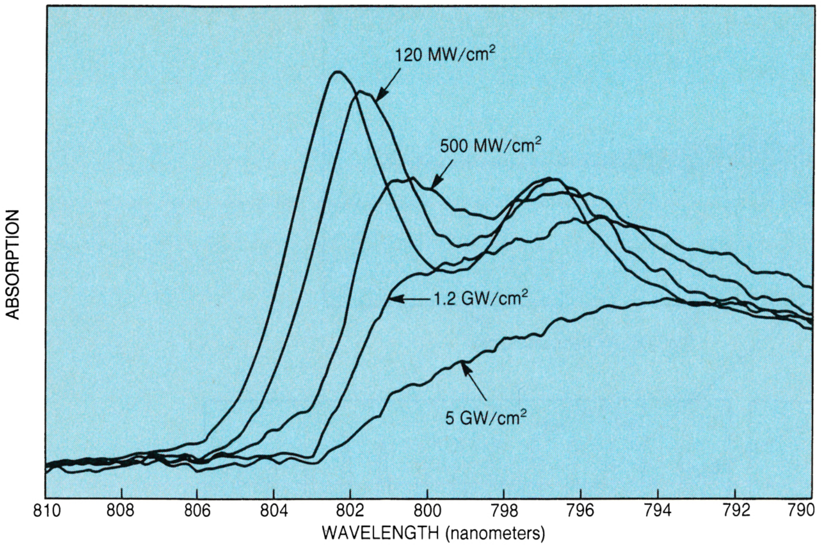


Time-resolved differential absorption spectra of quasi-zero-dimensional magnetoexcitons. Depending on its polarization the resonant pump creates populations of spin-polarized magnetoexcitons that affect the transmission of the broadband probe differently. **a,b:** Spectra obtained at a magnetic field of 12 tesla for the two polarizations of the pump that are resonant with the lowest magnetoexciton. **c:** The difference between the two spectra separates the effects of Pauli exclusion and exchange that are spin dependent from the spin-independent charge-density effects. (Adapted from ref. 2.) **Figure 4**

exciton–exciton Coulomb coupling, coherent polarization waves can interact with one another, and the corresponding nonlinearity also contributes to wave-mixing. The exciton–exciton interaction coupling bears a striking resemblance to the Ginzburg–Landau mechanism that, near the superconducting transition, drives the Cooper pairs toward a coherent state. Light scattering through polarization-wave coupling is not instantaneous and takes time to build up in the sample. Because the atom-like nonlinearity and the exciton–exciton interaction nonlinearity have different dynamics, they can be distinguished by experiments that resolve the temporal profile of the emission. Surprisingly, such measurements—performed recently on quantum well samples—demonstrated that emission through polarization-wave interaction is the dominant mechanism.[13,14] Because the Coulomb interaction is weakened by screening, the magnitude of the collective response can be altered by generating e–h pairs in the sample. Studies have demonstrated that, indeed, the collective effect is reduced by the presence of e–h pairs and that at high densities, where the Coulomb interaction is completely screened, it disappears altogether, so that only the contribution of the Pauli exclusion remains.[13]

So far we have limited our discussion to the measurement of the amplitude of the field emitted by or transmitted through the samples. The field's phase also contains important information about the mechanisms responsible for the establishment and decay of polarization waves. Unambiguous phase retrieval raises serious difficulties, for both fundamental and practical reasons. If the amplitude is a well-behaved and smooth function of time, however, a combination of interferometric correlation and power spectrum measurements gives a good indication of the phase changes during an ultrashort pulse.[15] In this case one can relate the magnitude of the phase shift to the fringe spacing of the signal autocorrelation as compared with that of the autocorrelation of a reference laser pulse. One can independently determine the sign of the phase shift by cross-correlation with the reference laser. The experiments are nevertheless extremely delicate, especially when the pulses contain very little energy (in the range of femtojoules), as is the case in spectroscopic experiments. Recently experimenters found that the coherent-wave-mixing signal emitted when a quantum well sample is excited close to the exciton resonances is actually frequency modulated.[15] When a single resonance is excited by a slightly detuned, ultrashort pulse, the emission starts at the exciton's natural frequency (like the sound waves emitted from a tuning fork after a brief chock), but within a couple of hundred femtoseconds it experiences nonlinear shifts that modulate the instantaneous frequency toward that of the laser. This behavior is due again to collective interaction of the delocalized excitons and cannot occur in independent atom-like systems.

An even more spectacular effect occurs when two resonances excited simultaneously, for example, the heavy-hole and light-hole peaks (see figure 2), contribute equally to the emission. In this case quantum beats are expected, and indeed they have been observed in amplitude measurements.[16] The interferometric measurement beautifully demonstrates the modulation of the instantaneous frequency.[15] (See figure 6.) The interferences are



Excitation of virtual e–h pairs well below the resonance energies gives these absorption spectra of quasi-two-dimensionally confined excitons. The instantaneous blueshift due to the ac Stark effect is clearly seen at low excitation intensity. As the pump intensity increases, the absorption line shape is altered. (Adapted from ref. 8.) **Figure 5**

clearly illustrated by the autocorrelation trace (figure 6a). The plot of the relative fringe spacing (figure 6b) shows that at the beginning of the pulse the frequency of the signal coincides with the laser frequency, but after about 120 fsec the signal frequency suddenly experiences a downward shift of exactly π before the modulation resumes at a slower rate. Figure 6c details the occurrence of this π shift by comparing a dozen of the fringes around $\Delta t = 0$ and around the first node of the autocorrelation trace. One sees very clearly how over a period of about ten fringes, the fringe pattern close to the node shifts by exactly one fringe in relation to the fringe pattern around the center. The accuracy of the measurement is approximately 0.13 fsec. Measurements of the dynamics of the instantaneous frequency of an ultrashort-pulse coherent-wave-mixing signal raise fundamental questions related to the time–energy representations of quantum mechanics and of signals, as considered by Eugene Wigner and Jacques Ville. Since experimental techniques are now sophisticated enough to perform such measurements with an accuracy close to the fundamental limit imposed by the uncertainty relations, it is likely that much needed theoretical investigations will now be done.

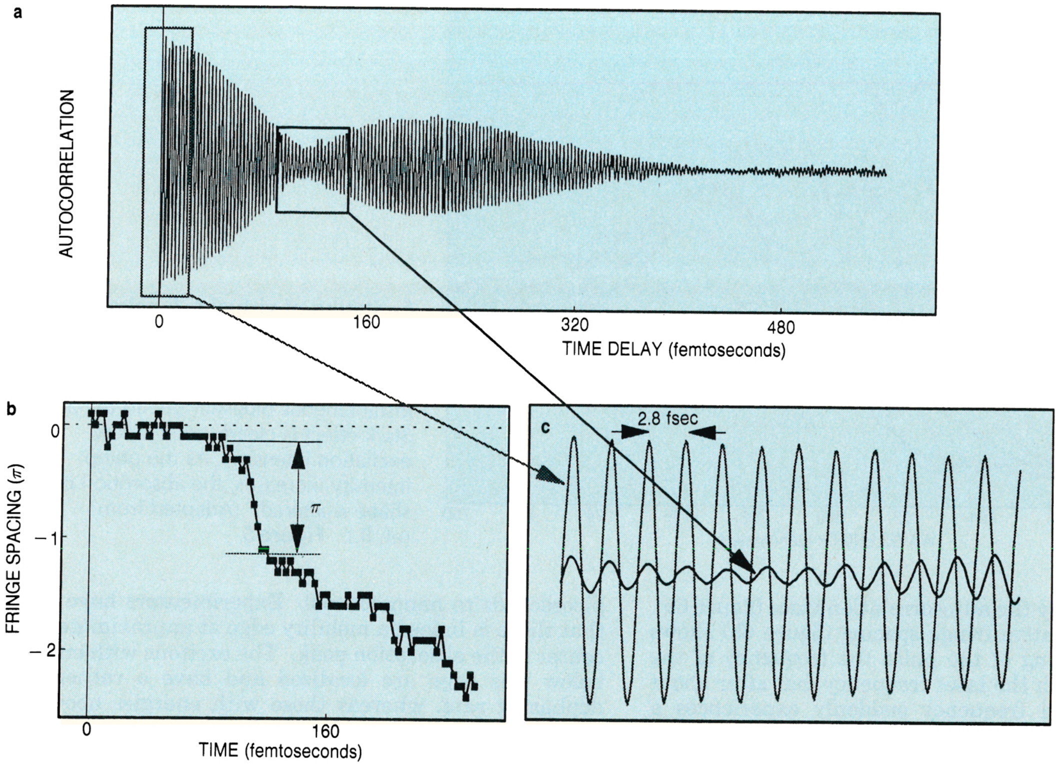
Coherent nonlinear effects can also be used to study the opposite regime: extremely slow processes.[17] Even under the best conditions for crystal growth, the limit on the definition of heterostructure interfaces is a single atomic layer. The effect of interface roughness on the energy of excitons in ultrathin layers is significant (a few meV). As a result, excitons can migrate within a layer by emission or absorption of acoustic phonons and experience scattering by the random potential associated with interface fluctuations. The very slow dynamics associated with this migration in a disordered system exhibit thermal activation and a mobility edge. At high temperature there are enough thermal phonons to delocalize the excitons, whereas at low temperature localization in the lowest energy states occurs. Wave-mixing experiments provide an elegant way to study these processes. Two coherent-wave lasers whose frequencies differ slightly ($\Delta\omega = \omega_1 - \omega_2$ ranges between zero and the gigahertz regime) produce both spatial and temporal modulation of the exciton populations probed by a third beam. One can view this modulation as a slowly moving interference pattern from which a third beam is scattered. A plot of the wave-mixing signal versus detuning $\Delta\omega$ provides information on the dynamics over time scales $\Delta t \sim \Delta\omega^{-1}$ (tens of picoseconds to nanoseconds). Experimenters have found that there is indeed a mobility edge at approximately the center of the absorption peak. The excitons with energies below this edge are localized and have a rather slow dephasing rate, whereas those with energies above the edge are delocalized and have a fast dephasing rate.[17]

This review, limited to general trends and focused on the most fundamental aspects of nonlinear optics in quantum-confined heterostructures, has concentrated on new results that reveal specific behaviors well differentiated from those found in other areas of nonlinear optics in condensed matter. For completeness, I should mention that investigators have used ultrashort-pulse time-resolved techniques extensively as spectroscopic tools to investigate other processes in heterostructures. For example, researchers have applied them to the study of gain dynamics in semiconductor lasers[18] and to the development of a comprehensive theory of these inverted media.[19] These methods also have been used to temporally resolve Bloch oscillations in superlattices under static electric fields[20] and polarization interferences from independent resonances.[21]

The many exciting results obtained in this field over the last decade are just an indication of what is still to come. Many challenges, experimental and theoretical, remain. For example, the structures that we can fabricate are at present very simple and are based on a limited number of materials. Clearly we need new growth or processing techniques able to produce one-dimensional and zero-dimensional structures that are defect and fluctuation free. And we expect that heterostructures involving more complex and diversified combinations of materials, especially materials with very different dielectric constants, will display new properties. We need to develop sensitive and versatile detection techniques able to probe a single nanostructure. Theories of relaxation going beyond mean-field approximation and able to describe the very-short-time dynamics, including realistic boundary conditions, are still missing. This is good news: There is much exciting research yet to be done.

* * *

A number of friends and collaborators have worked with me during the last ten years and I would like to thank them for their help and support. I wish to mention especially Stefan Schmitt-Rink. Stefan made an outstanding contribution to the theory of this new field before his tragic death last year. (See his obituary in PHYSICS TODAY*, March, page 104.) We were planning to write this*



Interferometric autocorrelation of the coherent-wave-mixing signal observed when two quasi-two-dimensional exciton resonances are excited by an ultrashort laser pulse. **a:** The autocorrelation trace exhibits beautiful interference patterns. **b:** The fringe spacing shows how the signal frequency, which coincides with that of the laser at the beginning of the pulse, suddenly experiences a shift of π after about 120 fsec. **c:** Over about ten fringes, the fringe pattern close to the first node shifts by exactly one fringe in relation to the fringe pattern around $\Delta t = 0$. The accuracy of the measurement is approximately 0.13 fsec. **Figure 6**

article together. He was my friend. I would like to dedicate this paper to his memory.

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