

PROGRESS REPORT

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**Excitons in Semiconducting Superlattices, Quantum
Wells, and Ternary Alloys**

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Contents

Summary	3
Introduction	4
<u>Work done under this grant in the budget period</u>	
1. Type II short-period AlAs/GaAs superlattices	8
Exciton kinetics in indirect gap AlAs/GaAs superlattices	
Energy levels and uniaxial stress effects in very short	
period AlAs/GaAs superlattices	
Electron-hole plasma in indirect gap AlAs/GaAs superlattices	
2. Oxygen in gallium phosphide	13
3. Strain-confined excitons in microstructures	14
4. Resonant Raman Scattering in II-VI compound superlattices	15
5. Theoretical work	16
Publications based on work supported by this grant	17
References	19

Excitons in Semiconducting Superlattices, Quantum Wells, and Ternary Alloys

Summary

It is now possible to fabricate semiconducting layered structures with precisely defined layer thicknesses of a few atomic diameters. Examples are the "quantum well" and the "superlattice" structures, in which semiconductors with different band gaps are interleaved. "Microstructures" can be produced from this material by patterning and etching them on a small (~10nm) scale. Their electronic properties are quite different from those of the constituents and offer interesting new possibilities both in device design and in basic physics. This proposal aims to improve our understanding of optically excited states ("excitons" and "electron-hole plasmas") in these structures. Time-resolved tunable laser spectroscopy, with and without external perturbations such as magnetic field, electric field, and uniaxial stress, will be used principally to study the following phenomena.

1. Exciton states in very short period superlattices, where the familiar "effective mass model" of semiconductor states breaks down, and there is a wide variety of theoretical predictions.
2. The electron-hole plasma which forms when the excitation density is high. This plasma may be in a liquid state at low temperatures. Our efforts will concentrate on establishing if a liquid state in fact exists, and on those phenomena such as Alfvén waves which occur also in classical plasmas. In the short period superlattices which are our primary concern, electrons and holes are spatially separated, leading to internal electric fields which might be expected to have a pronounced effect on the plasma properties.

Work will also continue on ternary alloys, primarily to establish if the alloy disorder produces a mobility edge for excitons, and on II-VI compounds, where the principal interest at present is in the nature of the exciton-phonon coupling. A new class of structures, GaAs-based superlattices containing thin metallic layers of rare earth pnictides, will also be studied spectroscopically.

Introduction

The new crystal-growth techniques of molecular beam epitaxy (MBE), metal-organic chemical vapor deposition (MOCVD) and related techniques, permit the fabrication of structures made up of atomically flat layers of semiconductors such as GaAs and AlAs¹. The electronic properties of such structures are quite different in many respects from those of the parent bulk materials. Study of these structures can give new insight into basic problems of solid-state physics, particularly those associated with low dimensionality, and have led to unexpected and fundamental discoveries, such as the integral and fractional quantum Hall effect. Furthermore, these structures are of great technological interest, since they give the device designer new options in, and unprecedented control over, the properties of his working material.

Of the many physical techniques for the study of such structures, optical spectroscopy is unique in that it gives information about excited states as well as the ground state of the system². Such information is obviously useful to the designer of opto-electronic devices; it is also essential to the understanding of the basic physics of these structures. The energies, wave functions and dynamics of electronically excited states are more sensitive than is the ground state to the physical assumptions of any model used to describe the system. Furthermore, through such phenomena as the Raman effect and phonon-assisted transitions, the atomic vibrations of the structure can be probed; these too differ greatly from those of the parent materials.

The fundamental optical process in an semiconductor or insulator is the creation or destruction of an electron-hole pair with the corresponding absorption or emission of a photon. The electron and hole attract each other and form a neutral bound entity called the exciton. At low temperature and low excitation intensity, transitions involving the creation and destruction of excitons dominate the optical properties of reasonably pure crystals³. In a perfect system excitons move freely; in a real system the exciton dynamics give

valuable insight into deviations from perfection (such as impurities, defects, interface roughness, and composition fluctuations) as well as into the exciton-phonon interaction.

In bulk semiconductors of technological interest, the small binding energy of the exciton permits the device designer to ignore excitonic effects, and the chief practical use of the optical spectroscopy of excitons has been as an analytic tool. In the layered structures that we will consider here, on the other hand, excitonic effects dominate the optical spectra even at room temperature, and are responsible for the non-linear optical response which is essential to the new generation of optical devices, such as logic gates and optical switches based on optical bistability⁴. Thus the study of excitons in these structures has come to play an important technological role in the field of opto-electronics.

In this proposal we are concerned with two types of structure, the "quantum well" and the "superlattice". A "quantum well" consists of a thin (~100Å) layer of semiconducting material (e.g. GaAs) with a small bandgap, sandwiched between layers of a larger bandgap materials (e.g. AlAs)⁵. Electrons and holes are confined to the layer with the smaller bandgap, and perpendicular to the layer their motion is quantized. Within any one such quantum state they behave as particles free to move only in two dimensions. Optically created electrons and holes combine to form two dimensional excitons which would move freely in the layer if it were perfect. The principal deviation from perfection is usually interface roughness, to which the exciton dynamics are very sensitive.

Frequently, several identical quantum wells are grown in one "multi-quantum well" structure. The quantum wells are far enough apart that the electronic wave functions in neighboring wells do not overlap, and for many purposes the quantum wells can be regarded as independent. However, they are still coupled by the electromagnetic field, and the energy transfer processes due to this are an interesting and barely touched subject of study.

In a "superlattice" (SL), the barrier layers are sufficiently thin to permit penetration of the electron or hole wave functions through them. Particle motion is now three-dimensional, but highly anisotropic. We are interested in SLs which have periods less than about 60Å, since above this the superlattice differs little from a multi-quantum well structure. At short periods remarkable new phenomena, such as spatial separation of electron and hole states, have recently been observed⁶. In the past year we have paid particular attention to SLs with periods of 12Å or less. Until recently it was believed that such SLs would be indistinguishable, except perhaps for a slight anisotropy, from the bulk alloy, but this has been shown to be mistaken: as we shall see below, AlAs/GaAs SLs containing only a single monolayer of each constituent, if of good quality, have been shown to have an electron band structure fundamentally different from that of the bulk alloy. In these SLs the usual methods of calculating SL band structure based on the effective mass approximation, such as the envelope function method⁷ and the Kronig-Penney model⁸, break down, and have been replaced with more or less success by a plethora of so-called "first principles" calculations⁹. We have found that even the best of these are not altogether successful in explaining the experimental data.

In the GaAs/AlAs system the constituents have almost identical lattice constants and can therefore grow epitaxially without internal strain. It is also possible to grow structures in which the constituents have different lattice constants, and the lattice mismatch will be taken up by elastic strain rather than by the generation of dislocations at the interfaces if the layers are sufficiently thin¹⁰. The energy levels of such structures are profoundly affected by the internal strain.

The new crystal growth techniques can also be used to produce ternary and quaternary semiconducting alloys, such as $\text{Ga}_{1-x}\text{Al}_x\text{As}$ and $\text{Ga}_{1-x-y}\text{Al}_x\text{In}_y\text{As}$, of accurately controlled and uniform composition¹¹. In such material it should be possible to distinguish the effect on

the exciton dynamics of the intrinsic potential fluctuations due to the statistical distribution of the constituent atoms, from the effects of fluctuations which arise from non-uniform growth. The intrinsic fluctuations in an alloy are due to the differences in the pseudopotentials of the components and are much weaker (\sim meV) than those in an amorphous semiconductor, where they are so large (\sim eV) that excitons cannot exist. Excitons in a ternary alloy can provide a model for the effects of disorder in the more complex amorphous case.

SLs made of II-VI compounds and their alloys are also being fabricated. The chief practical interest of such compounds is in the wide range of direct bandgap that they offer: they are essential for the development of devices operating at wavelengths shorter than 600 nm or longer than a few μ m. Furthermore, they readily incorporate magnetic atoms, and SLs in which one or both of the layers is magnetic (ferro-, antiferro- or spin glass) have some remarkable properties¹². The physics of non-magnetic II-VI SLs is of interest because the electron-phonon interaction is considerably stronger than it is in III-Vs.

Work done under this grant during the budget period

1. Type II short-period AlAs/GaAs superlattices

In a type II AlAs/GaAs superlattice (SL) the lowest electron state is in the AlAs layer while the hole state is in the GaAs layer⁶. AlAs is an indirect gap semiconductor; i.e. its conduction band (electron) minimum is not at the Brillouin zone center Γ , as is the valence band (hole) maximum, but is at or near the X point on the zone boundary. If the GaAs layer is sufficiently thin, the confinement energy pushes the lowest Γ electron state above the X state of AlAs. Hence the lowest exciton consists of the AlAs X electron and the GaAs Γ hole, and is indirect in momentum space, so that its decay by photoluminescence (PL) is forbidden to zero'th order by momentum conservation. The transition is made weakly allowed by four possible mechanisms which mix the Γ and X electron states: by phonons, by the SL potential, by impurities, and by random scattering at the interface. The first mechanism cannot give a no-phonon line, and since a no-phonon line is in fact observed in PL, at least one of the other three must be present.

In a cubic crystal there are three degenerate X electron states, corresponding to electron momenta directed along the x, y or z directions. In the SL this degeneracy is lifted and we label the resulting states X_z and X_{xy} , where we have taken the growth direction to be the z axis. Only X_z is mixed with Γ by the SL potential, but any state can be mixed by interface disorder. Electrons relax rapidly and non-radiatively to the lowest X state. The low temperature PL is therefore predominantly from this state, though very weak PL is sometimes observed from the Γ state, for which the radiative transition probability is high enough to compete with the non-radiative $\Gamma \rightarrow X$ transfer.

The questions addressed during the period covered by this report were:

- (1) What is the relative importance of the different mechanisms which make the transition allowed? How large is the mixing of the Γ and X states?
- (2) What is the order and separation of the X_z and X_{xy} states, particularly in very short period SLs where the effective mass approximation might be expected to break down? Recent "first principles" calculations make predictions which need to be tested⁹.
- (3) How do the excitons associated with the different conduction band states couple to phonons?
- (4) Can the predicted⁹ change of symmetry of the states with SL period (odd or even) be observed?
- (5) (a) What new effects can we expect at high excitation? (b) Can an electron-hole liquid form in a Type II SL, as it does at low temperatures in other indirect gap semiconductors such as Si and Ge¹³?

The work done under this grant has established a firm answer to question (1) and, except for some quantitative details, (2) and (3). We are working on question (4) but we have not reached a clearcut conclusion, and our data may still be limited by sample quality. In regard to (5), intriguing results have been obtained in PL at high excitation which suggest that the answer to question (5b) may be yes, but more work is needed to confirm this.

Question (1) has been addressed in two studies: on the Stark effect and on the time decay of the PL.

Stark effect: In an X-exciton the electron and hole occupy different layers, while in a Γ -exciton they are both in the GaAs layer. Hence they respond differently to an applied electric field, and can in fact be made to cross. If Γ and X are mixed the states anticross. In a collaboration with M-H Meynadier and others this anticrossing has been observed (Publication # 1) and a mixing of 2 meV deduced, consistent with the observed lifetime of the X-exciton. This mixing is less than the inhomogeneous linewidth and it is difficult to extract an accurate

value from the data. We have attempted to improve on this by making time-resolved measurements. However, inhomogeneous broadening of the line greatly complicates the results and we are still working on the analysis.

Time decay: Differences between the time decay reported by different groups^{6,14} on nominally identical Type II SLs were sorted out and found to be due to different experimental conditions. By careful measurements over a range of intensities, we found that the non-exponential decay at low intensity is a consequence of interface disorder, and changes continuously to exponential decay as the intensity is increased. This phenomenon can be attributed to delocalization of the excitons at high density and consequent averaging out of the interface disorder. The non-exponential decay at a sample-independent rate is consistent with decay from localized X_z excitons¹⁵, and implies that the PL is not impurity-related.

(Publication #3,13)

The main thrust of our work in this area has been directed towards question (2): i.e. understanding the lowest conduction band structure of very short period $(\text{GaAs})_n(\text{AlAs})_n$ SLs, in which the layer thickness n is 4 monolayers or less. Well characterized SLs with such thin layers have recently become available from L. N. Pfeiffer of A.T.& T. Bell Labs. We have studied the photoluminescence (PL) and photoluminescence excitation (PLE) spectra of these SLs, both as grown and under uniaxial stress applied parallel to the growth direction. We have established that the PL of these SLs is intrinsic, apart from a relatively weak and easily identifiable line associated with defects. This defect line dominated previous PL measurements on nominally similar SLs^{16,17}. Comparison with an $\text{Al}_{0.5}\text{Ga}_{0.5}\text{As}$ alloy grown by the identical technique shows that the material is indeed a true superlattice even for $n=1$. We find that for $n \geq 4$ the spectrum is similar to that previously reported in wider SLs, whose non-exponential time decay is discussed above. The PLE spectrum, the time decay, the shift of the line with stress, and the weak phonon sidebands, are all evidence that the transition involves the X_z

exciton. This has been confirmed by ODMR¹⁶. For $n \leq 3$, on the other hand, the zero stress PL spectrum, with strong phonon sidebands, the PLE spectra, the slow quasi-exponential time decay, and the shift of the PL in low stress clearly indicate the X_{xy} exciton. The spectra are similar to those obtained on SLs with relatively wide AlAs layers¹⁸, in which mismatch strain makes the X_{xy} exciton lowest, as has also been proved by ODMR¹⁹. At a stress of 1-2 kbar the states cross and the X_z exciton dominates the PL. When allowance is made for the lattice mismatch strain, which tends to lower the X_{xy} state, we find that the $X_{xy} - X_z$ splitting in zero stress is much less than predicted by "first principles" calculations, and goes to zero within experimental accuracy for $n \leq 2$. Calculations using the effective mass approximation do predict this near degeneracy, but this approximation is not valid in such short period structures. Thus the question of how these very short period SLs are to be described theoretically is still open. (Publication #5, 7, 10, 12, 14). At present our values for the splitting are not as precise as they could be, primarily because the uniaxial stress data cannot be quantitatively interpreted without knowledge of the valence band shift. The recent discovery of $\Gamma \rightarrow \Gamma$ emission (with intensity $\sim 10^{-4}$ of the X emission) from these very short period SLs (Publication #8) makes it possible to determine this shift experimentally, and we expect to resolve this question before the end of the grant period.

On question (3), the first problem is to assign the phonon sidebands observed. There are some discrepancies between the results of different laboratories which need to be resolved, but there do appear to be consistent differences which depend on the SL period. We are working on this in collaboration with B.A. Wilson and R.C. Spitzer, both until recently of A.T. & T. Bell Labs. (Publication #18).

On question (4), in the last few months we have observed some of the predicted symmetry effects and our results were reported at international conferences (pubs #10, 12). However, the predicted effect of parity has not so far been detected, probably because of surface electric fields.

Question (5), on the effect of high excitation intensity and the possible observation of an electron-hole liquid, requires a little discussion of the background. When the exciton density becomes high in a semiconductor, the excitons may form excitonic molecules, or "bi-excitons"²⁰. At higher densities still there is a transition to a new state, the electron-hole plasma, in which each electron and hole moves freely in the average field of the other particles, rather than being bound to one other particle as in an exciton. Normally the plasma is gaseous and expands to fill any volume available to it, but at low temperatures it may be liquid with a well defined density, and forms "droplets" surrounded by a low density gas of excitons¹³. These droplets may be more stable than the biexciton. This phenomenon is well known in conventional indirect semiconductors such as silicon and germanium, but has not been unambiguously observed in direct gap semiconductors such as gallium arsenide. There are two reasons for this: electrons in direct gap semiconductors usually have small effective masses, destabilizing the liquid; and the short radiative lifetime of the direct exciton does not give time for the liquid to form even if it is the stable state. Since the Type II AlAs/GaAs SL has an indirect gap, it seems likely that an electron-hole liquid can form, but it is not known how the dimensionality of the electron states, which are two- rather than three-dimensional, affects the stability of the liquid. It has been argued by Hawrylak²¹ that low dimensionality enhances the stability, but it is not clear if his calculations are in fact applicable to this system. Because the electron and hole are spatially separated in a Type II superlattice, there is a strong internal field at high particle density, so that the band gap tends to increase with density rather than decrease as in the bulk .

It has been found at Bellcore and at Dartmouth that in SLs of a certain range of layer thickness a new optical transition, the "M line", appears in the PL at high excitation. It has a well-defined threshold, as a function of excitation, somewhat higher than the rather broad threshold for plasma formation, and its lineshape over a

certain range of excitation, is consistent with that expected for an electron-hole liquid. We have recently made time-resolved measurements which show that the plasma density remains almost constant while the integrated intensity falls by an order of magnitude, implying that the volume occupied by the plasma decreases rather than increases with time. This also seems to imply a liquid rather than a gaseous plasma. However, it has a number of unusual features, the most striking of which is that the M line is at a somewhat higher energy than the exciton energy. This is inconsistent with the usual picture of the liquid being the most stable (i.e. lowest energy) state. One hypothesis, on which we are working now, is that the lowest electron state does not form a liquid, but that when the electron density is sufficiently high a higher conduction band minimum becomes occupied, and this perhaps can form a liquid. (Publications #11,12,17).

2. Oxygen in gallium phosphide

There exists now a very powerful and apparently successful *a priori* theory of the effect of short-range (atomic scale) potentials on the electron states of semiconductors²². This theory has been applied principally to "deep impurity" states in semiconductors: i.e. states in which the electron or hole is localized by a very strong short-range impurity potential. It can also be applied to the electron states of quantum wells and SLs, since in these, too, strong short-range potentials are present at the interfaces⁹. It is, therefore, very important that the foundations of the theory be securely established by comparison with experiment. In 1978, work by Monemar and Samuelson²³ on the deep donor state of oxygen in GaP cast serious doubt on these foundations. They found that the ground state of O^+ , which according to the theory is simply the ground state of the unperturbed crystal and can have no structure, is split by about 0.4 meV. This result caused a sharp controversy to develop. It was argued, principally by P.J.Dean²⁴, that the interpretation of the experiments was faulty and that the effects observed were associated with an oxygen-acceptor pair rather than with isolated oxygen (to which the theory applies). T.N.Morgan²⁵, on the other hand, developed a

complicated new theory, very different from and apparently inconsistent with the "standard" theory mentioned above, which could account for the existence of the extra states. Morgan's theory has been criticised²⁵ both for its extreme complication (it predicts a very large number of states, very few of which are observed) and for its lack of *a priori* justification. The question was unresolved at the time of Dean's death in 1984 and has been largely ignored since.

Our measurements show that the Monemar and Samuelson result is strongly sample dependent. This appeared at first sight to support Dean's interpretation, but Morgan in a private communication pointed out that the new data were in fact also consistent with his theory. In an attempt to resolve this question, we have refined the excitation measurements, paying particular attention to the threshold region where the excitation is extremely weak, and have partially avoided the problems raised by Dean by comparing the spectrum with that obtained for two-step luminescence, in which an electron is first excited from the valence band to the 0° state and then further excited to the conduction band. Our results confirm that Monemar & Samuelson's result is indeed intrinsic to the oxygen, and is not associated with the presence of the acceptor, so that the theoretical dilemma remains.

(Publication #20).

3. Strain-confined excitons in microstructures.

In a "quantum well" carrier confinement is in one dimension, normal to the growth direction. It is now possible to fabricate microstructures with lateral confinement, i.e. confinement transverse to the growth direction, such as the "quantum wire" in which the carriers are confined in two dimensions, and the "quantum dot" in which the confinement is in all three²⁶. Up to now this has been achieved two main methods. One is by patterned etching of a quantum well layer²⁷: this has the disadvantage that there is rapid non-radiative recombination at the exposed surface. The other by is growth on specially structured substrates. While remarkable results have recently been obtained by different versions of these techniques²⁸,

the process is difficult to make reproducible, and the active regions necessarily form a rather small fraction of the total area. In collaboration with K.Kash and others at Bellcore we found that lateral confinement can be produced the strain in a quantum well layer due to changes in topography just above it (publication #2). Strain confinement on a macroscopic scale has been studied in detail in bulk semiconductors²⁹, but not previously applied to quantum well systems, or on a sub-micron scale. Because the strain prevents excitons from migrating to surfaces the quantum efficiency of the microstructure is high. The strain is proportional to the lattice mismatch between the barrier and quantum well material, which in the case of the AlAs overlayer used was extremely small, so that confinement was only obtained at low temperatures. The technique has now been extended to overlayers such as amorphous carbon which have a large mismatch³⁰, and confinement at room temperature is in principle possible, with important device implications. This is the substance of a separate proposal by one of us (M.D.Sturge) and K. Kash.

4. Resonant Raman Scattering in II-VI compound superlattices

In collaboration with Dr Finkman of the Technion, Israel, and the CNRS Laboratories in Grenoble, France, we have measured resonant LO phonon Raman scattering and selectively excited phonon-assisted luminescence in a compensated strain CdTe/Cd_{0.91}Zn_{0.09}Te superlattice. This system is of interest for two reasons: it provides a non-magnetic model for the II-VI magnetic semiconductor superlattice, and the light hole (LH) and heavy hole (HH) reside in different layers³¹. Strong Raman resonances are observed at the direct LH and HH excitons. The polarization of the HH scattering for (001) and (110) polarized light shows that it is at least 90% due to the Frohlich interaction (FI), while the LH scattering shows equal contributions from FI and deformation potential coupling (DP). The difference is not due to the mass difference, but is presumably related to the fact that the LH exciton is type II in this superlattice. We are working on the theory of this effect, which may give useful information on the electron and hole wave-functions in these superlattices. Strong unpolarized

resonances associated with impurity bound excitons are also observed: while these look like Raman scattering they are almost certainly due to selectively excited luminescence (Publication #14).

5.Theoretical work

Several theoretical studies peripheral to this work have been made. A new and rather simple result has been obtained, relating the observed energy relaxation rate of electrons and holes in a highly excited semiconductor to the electron-phonon coupling, and applied to data obtained at the Institute of Semiconductors, Beijing, China, on a multi-quantum well system (Publication #6). The non-linear pressure shift of the electronic energy levels of quantum wells has been analyzed (Publication #9). A model for the effect of the EL2 defect center on oxygen-related vibrations in GaAs has been developed (Publication #16). A new theoretical analysis explains quantitatively the binding energy of the hole in an exciton bound to an electron-attractive isoelectronic trap such as N in GaP (Publication #21).

Publications

The following publications are based wholly or in part on work supported by this grant.

Papers published or submitted for publication

1. Indirect-direct anticrossing in GaAs/AlAs superlattices induced by quantum confined Stark effect: evidence of Γ -X mixing. M-H Meynadier, R E Nahory, J.M.Worlock, M C Tamargo, J.L.de Miguel and M D Sturge, Phys. Rev. Lett. 60, 1338 (1988).
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3. Photoluminescence decay time studies of type II GaAs/AlAs quantum well structures. M.D.Sturge, Janet L. Mackay, Colette Maloney and J.K.Pribram. J. Appl. Phys. 66, 5639 (1989)
4. Introduction to the optical properties of quantum wells and superlattices. M.D.Sturge and M-H Meynadier. J. Lumin. 44, 199 (1989).
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6. Hot carrier relaxation processes and nonequilibrium phonon effect in multiple quantum well structures. Weikun Ge, Z Xu, Y Li, Z Xu, J Xu, B Zheng and W Zhuang, J. Lumin. 46, 137 (1990).
7. Energy levels of very short period $\text{GaAs}_n\text{AlAs}_n$ superlattices Weikun Ge, M.D.Sturge, W.D.Schmidt, L.N.Pfeiffer and K.W.West. Appl. Phys. Lett. 57, 55 (1990).
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14. Resonant Raman scattering and selectively excited luminescence in a CdTe/CdZnTe superlattice.
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16. EL2 ionization induced interconversion among three Ga-O-Ga LVM absorption bands in semi-insulating GaAs.
 Weikun Ge, Chung-ying Song and B. Pajot.
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17. Multiple quantum wells under intense optical excitation.
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Papers in preparation

18. Phonon assisted photoluminescence in AlAs/GaAs superlattices
 Weikun Ge, R.C. Spitzer, B.A. Wilson and M.D.Sturge

19. Survey of direct bandgaps in GaAs/AlAs superlattices
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20. Is the ground state of O^+ in GaP split?
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21. Binding energy of the hole in the exciton bound to an isoelectronic trap. Yong Zhang.

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