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Electronic Structure of GaAs†

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Using a nonlocal pseudopotential, including spin-orbit interactions, we have calculated a band structure for GaAs which is in excellent agreement with electroreflectance, wavelength-modulation, and photoemission experimental results. As in a calculation by Pandey and Phillips, an order of magnitude in accuracy has been gained over existing band structures; the largest discrepancy with Schottky-barrier electroreflectance measurements is less than 0.08 eV. The E_0' conflict is resolved in favor of Aspnes's interpretation.

The nonlocal pseudopotential approach has yielded what we believe to be the most accurate band structure for GaAs to date.[1] In fact, as a result of the extensive experimental studies on this compound, this band structure may be the most accurate band structure, over a large energy range, available for any material. The largest discrepancy between the experimental critical-point energies as determined by the Schottky-barrier electroreflectance of Aspnes and Studna[2] and as calculated by our nonlocal pseudopotential scheme is less than 0.08 eV. Further, we are able to achieve comparable agreement with the derivative reflectivity spectrum from wavelength-modulation measurements, and the electronic density of states as measured by photoemission experiments. This represents nearly an order-of-magnitude improvement over previous band-structure calculations. We are also able to confirm the existence of the critical points of Δ symmetry appearing in the E_0' structure, which lie about 10% of the way from Γ to X, in agreement

with the proposed assignment of Aspnes and Studna.[2] Spin-orbit interactions are found to be of prime importance in the formation of these critical points.

The band structure was calculated using the empirical pseudopotential method (EPM) which has been discussed extensively elsewhere.[3] Since this method requires experimental information as input to fix the form factors, the recent experimental advances in electroreflectance techniques, in particular those developed by Aspnes,[2,4] and high-resolution photoemission spectroscopy[5] have proven to be quite valuable. According to Aspnes and Studna their electroreflectance measurements on GaAs have resulted in an order-of-magnitude improvement in the resolution and accuracy in the determination of critical-point energies as compared to previous spectroscopic work.[2] In addition, the advent of high-resolution photoemission spectroscopy has supplied us with detailed information on the lowest-lying valence bands in semiconductors. This information, in

turn, has indicated the necessity of nonlocal corrections to the usual local-pseudopotential calculations.[6,7] It is these two experimental areas of advancement, then, which have made an extremely accurate band structure possible.

In our calculation we have taken an atomic pseudopotential of the form

$$V_{NL}(\vec{r}) = V_L(r) + A_2 \exp(-r^2/R^2)\mathcal{P}_2, \qquad (1)$$

where $V_L(r)$ is the usual local pseudopotential, and $\mathcal{P}_2$ projects out the $l = 2$ angular-momentum component. This nonlocal pseudopotential is quite similar to the one used recently by us for Ge.[8]

The crystal potential is then taken to be a sum

of these atomic pseudopotentials, and the local part can be expressed in terms of symmetric and antisymmetric pseudopotential form factors.[3] For this calculation the symmetric form factors were given by $V^S(3) = -0.214$, $V^S(8) = 0.014$, $V^S(11) = 0.067$ Ry, and the antisymmetric form factors by $V^A(3) = 0.055$, $V^A(4) = 0.038$, $V^A(11) = 0.001$ Ry. The nonlocal well depths were taken to be $A_2(\text{Ga}) = 0.25$ and $A_2(\text{As}) = 1.25$ Ry. The parameter R in (1) is not of crucial importance in determining the band structure,[7] and since it is computationally simpler, we have constrained it to be equal to the radius used in Ge[8] for both the Ga and As contributions. Spin-orbit interactions were included in our calculation in a method similar to the one used by Saravia and Brust in Ge.[9]

In Table I we compare our calculated critical-point energies to the experimental results of Schottky-barrier electroreflectance, wavelength modulation, and photoemission. We also compare our results to two of the most accurate recent band calculations which have also included spin-orbit interactions. As can be noted, a vast improvement over the previous calculations has been obtained. The local EPM gives topologically incorrect results[1,8] for critical points, and it yields significant errors when compared to photoemission data. The orthogonalized–plane-wave (OPW) calculation results in better valence energies, but it does not yield accurate optical-transition energies. Since a typical discrepancy between the local EPM or OPW theoretical results and the experimental values is on the order of 0.5 eV, and a typical nonlocal EPM error is on the order 0.05 eV, we have obtained an order-of-magnitude improvement.

Once the band structure has been obtained, the imaginary part of the dielectric function can be calculated, and by using the Kramers-Kronig dispersion relations, the reflectivity can be obtained. In Fig. 1 the experimental and theoretical modulated reflectivity is given for GaAs. It is impressive that third-derivative electroreflectance, which gives accurate critical-point energies, yields an accurate first-derivative spectrum via band-structure analysis.

By examining the energy gradients and dipole matrix elements throughout the Brillouin zone, it is possible to determine the origin of structure in the imaginary part of the dielectric function. In such a manner we have analyzed the contributions to the E_0' reflectivity structure. This structure has been the subject of some discussion.[2,10] Rehn and Kyser, using transverse electroreflec-

TABLE I. Comparison of theoretical and experimental critical-point energies (in eV) for GaAs.

Transition	Experiment		Theory		
	Reflectivity Structure	Critical Point [d]	Non-local EPM	Local EPM [c]	OPW [g]
$E_0 \quad \Gamma_8^v - \Gamma_6^c$	1.52[a]	1.52	1.51	1.52	1.34
$E_0 + \Delta_0 \quad \Gamma_7^v - \Gamma_6^c$	1.86[b]	1.86	1.86	1.87	1.66
$E_1 \quad L_{4,5}^v - L_6^c$	3.02[c]	3.04	3.03	2.92	2.62
$E_1 + \Delta_1 \quad L_6^v - L_6^c$	3.25[c]	3.25	3.25	3.05	2.82
$E_0'(\Gamma) \quad \Gamma_8^v - \Gamma_7^c$	--	4.49	4.54	4.80	4.12
$E_0' + \Delta_0' \quad \Gamma_8^v - \Gamma_8^c$	--	4.66	4.71	4.93	4.30
$E_0' + \Delta_0 + \Delta_0' \quad \Gamma_7^v - \Gamma_8^c$	--	5.01	5.05	5.28	4.62
$E_0'(\Delta) \quad \Delta_5^v - \Delta_5^c$	4.44[c]	4.53	4.54	4.38	--
$E_0' + \Delta_0'(\Delta) \quad \Delta_5^v - \Delta_5^c$	4.64[c]	4.71	4.70	4.55	--
Σ	5.11[c]	5.14	5.07	4.88	--
$X_7^v - X_6^c$	--	4.94	4.92	4.40	4.33
$E_2 \quad X_6^v - X_6^c$	--	5.01	5.01	4.49	4.52
$X_7^v - X_7^c$	5.64[c]	5.34	5.28	4.67	4.58
$X_6^v - X_7^c$		5.42	5.38	4.76	4.67
	Photoemission				
	UPS[e]	XPS[f]			
$L_3^v - \Gamma_{15}^v$	0.8±0.2	1.4±0.3	1.31	0.85	1.06
$\Sigma_1^{min} - \Gamma_{15}^v$	4.1±0.2	4.4±0.2	4.23	3.35	--
$X_3^v - \Gamma_{15}^v$	6.9±0.2	7.1±0.2	6.98	6.23	6.43
$X_1^v - \Gamma_{15}^v$	10.0±0.2	10.7±0.3	9.37	10.00	10.24
$\Gamma_1^v - \Gamma_{15}^v$	12.9±0.5	13.8±0.4	12.55	12.10	12.44

[a] See Ref. 13.   [e] See Ref. 15.
[b] See Ref. 14.   [f] See Ref. 16.
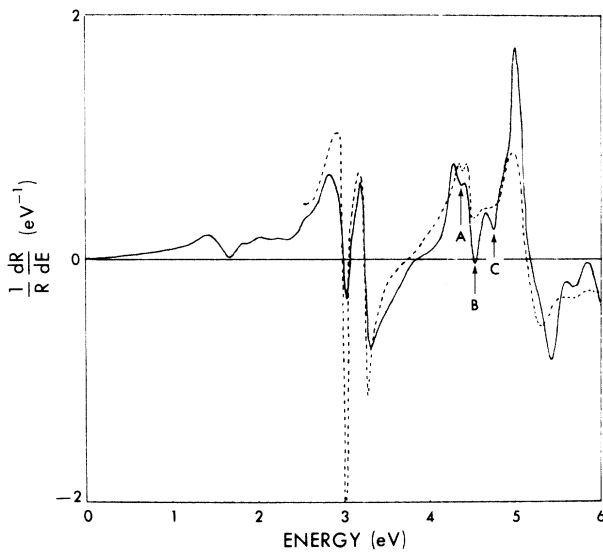[c] See Ref. 11.   [g] See Ref. 12.
[d] See Ref. 2.

FIG. 1. Comparison of theoretical (solid line) and experimental (dashed line) modulated reflectivity for GaAs. The experimental results are from Ref. 11. For A, B, C, see text.
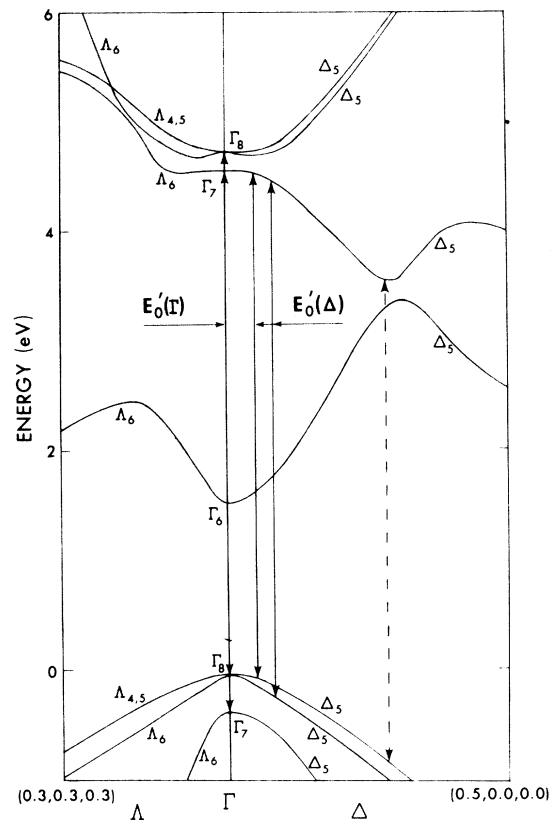

FIG. 2. Calculated band structure for GaAs near Γ showing the critical-point location for the E_0' structure. Also indicated (dashed line) is an M_0 critical point resulting from the pseudocrossing of the Δ_5 conduction bands.

tance, observed only a Δ symmetry for this structure.[10] They attributed the structure to derive from the pseudocrossing of the Δ_5 conduction bands. However, Aspnes and Studna have pointed out that this interpretation conflicts with band-structure calculations where some Γ symmetry structure is predicted.[2] Further, they proposed that the Δ symmetry structure arises from a pair of M_1 critical points approximately $\frac{1}{10}$ of the way from Γ to X. Our calculations agree with the Aspnes-Studna interpretation. We, indeed, find two M_1 critical points along Δ at between 5 and 10% of the way from Γ to X, as indicated in Fig. 2. It is these points, along with contributions from Γ, which cause the structure at 4.5 eV (B) and 4.7 eV (C) in our derivative spectrum (Fig. 1). We have also found, by calculating the band structure with and without spin-orbit interactions, that these interactions are crucial in altering the band shape near Γ and producing the critical points.

Aspnes and Studna also noted the possibility of the pseudocrossing producing some very weak structure at 4.4 eV in the electroreflectance data.[2] This also agrees with our results. The dashed line in Fig. 2 indicates an M_0 critical-point position near the pseudocrossing. This M_0 critical point produces the weak structure near 4.4 eV (A) in Fig. 1. It should be noted, however, that there exists a companion M_0 critical point

due to the spin-orbit splitting of the Δ_5 valence band. Since this companion occurs at about 0.1 eV higher energy, it is nearly degenerate with the E_0' structure from Γ and Δ at 4.5 eV. In our calculated derivative spectrum it is masked by the stronger M_1 critical points, and this may also be the case in the electroreflectance measurements.

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Measurements of Directional Correlation from Oriented Nuclei (DCO) and the Band Structure in ^{102}Pd

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A new technique for determining spin sequences and mixing ratios has been used to investigate the ground state band of ^{102}Pd. This new method is based on the observation of coincidences at only one angle rather than measuring a complete angular correlation. The results show that the unusual spin sequence previously proposed for the ground state band of ^{102}Pd is not correct.

In a recent Letter,[1] "forking" in the ground-state band of ^{102}Pd was reported. The proposed spin sequence, which is fundamental to the description of the band, was established on the basis of angular-*distribution* measurements of the γ rays following (HI, xn) reactions. In the present work the results of directional *correlation* observations on the ^{102}Pd γ rays using a new and simple method of data analysis are presented showing that the proposed spin assignments are incorrect.

The interpretation of γ-angular-distribution measurements following (HI, xn) reactions is often difficult because of γ transitions unresolved from the one of interest. This difficulty may be avoided by observing the directional correlation of two γ rays that are emitted in coincidence from oriented nuclear states populated in (HI, xn) reactions (directional correlation from oriented nuclei, DCO). The measurement of a complete directional correlation is unattractive in view of the large accelerator time required. However,

the DCO-ratio method proposed by Krane, Steffen, and Wheeler[2] is a practical and powerful method for the determination of multipole-mixing ratios of γ transitions and spin sequences.

The DCO-ratio method uses the coincidence rates $W(A(\gamma_1), B(\gamma_2))$ of two γ rays γ_1 and γ_2 that are emitted from an oriented ensemble of nuclei and are observed by two detectors A and B, respectively. The detectors are fixed at *asymmetric* directions with respect to the orientation axis of the ensemble (e.g., beam direction). If the role of the two detectors is reversed the coincidence rate $W(A(\gamma_2), B(\gamma_1))$ is observed and the ratio $R(A, B) = W(A(\gamma_1), B(\gamma_2))/W(A(\gamma_2), B(\gamma_1))$ of the two coincidence rates provides an observable that is very sensitive to the multipole mixing of, and to the spin changes involved in, the γ transitions. Many DCO ratios $R(A, B)$ can be simultaneously and accurately measured with modern computer-based electronics.

For "monotonic stretched" cascades (i.e., cascades with ΔI = const and $L = |\Delta I|$) the DCO ratio