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First-principles calculation of the elastic constants of AlAs

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We use an *ab initio* pseudopotential method within the local-density approximation to determine the full set of elastic moduli of AlAs which have not been directly established experimentally. We find $C_{11}=1.16$, $C_{12}=0.55$, and $C_{44}=0.57$ Mbar, close to those known for GaAs. Since similar calculations for GaAs and other materials produce results in close agreement with experiment, we conclude that our results for AlAs are also reliable.

I. INTRODUCTION

There is presently a great interest in superlattice structures made from $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ alloys. Although superlattice layers can be made containing Al-rich alloys and AlAs itself, bulk AlAs is not stable because of its hygroscopic nature. Among the properties of AlAs not well established experimentally are its elastic constants, which are relevant in the superlattices whenever strain is present. For example, analyses of experiments on superlattices under pressure[1] have depended crucially upon the magnitudes of the elastic constants as a function of Al composition in the $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ alloys.

Essentially all previous work has assumed that there are only small variations of elastic constants from GaAs to AlAs, which is supported by a number of studies of overall trends among the III-V materials.[2-4] In 1962 Keyes[2] showed that the dimensionless reduced bulk modulus and shear moduli (i.e., the elastic constants multiplied by the nearest-neighbor distance to the fourth power) exhibited smaller variations for different compounds of the same structure (diamond, zinc blende, or wurtzite) than did the original constants. Since AlAs and GaAs have essentially the same lattice constants at room temperature and pressure, this implies that the elastic properties of AlAs and GaAs should be extremely similar. Wiley[3] used this empirical result to tabulate elastic constants for AlAs which have been widely used. In a similar vein, Adachi[4] in 1985 used a linear relation between the elastic moduli and lattice constants of III-As compound materials to predict results for AlAs, which are virtually identical to GaAs.

Calculations of elastic constants for AlAs have been reported by Kagaya and Soma[5] who used the empirical pseudopotentials of the Ashcroft and variations of the local Heine-Abarenkov forms.[6] Because their calculations are not self-consistent, they found a range of results depending upon the approximations for the electronic screening (e.g., C_{44} ranges from 0.195 to 0.394 Mbar).

In this paper, we present *ab initio* calculations for the elastic moduli for AlAs using a norm-conserving nonlocal pseudopotential.[7] The methods of calculation have been developed by Nielsen and Martin[8,9] where they computed elastic constants for Si, Ge, and GaAs using the direct calculation of the macroscopic stress and force on the atoms in the solid. Based upon the previous successes of such calculations for related materials, especially GaAs, we believe that our results for AlAs are just as reliable and can be used to predict the elastic properties which are not yet experimentally established.

In previous work on AlAs, Froyen and Cohen[10] used *ab initio* methods to find the lattice constant and the bulk modulus, the latter derived from the equation of state. Similarly, Chang and Cohen[11] have calculated phonon frequencies and longitudinal effective charges with good agreement with experiment. de Gironcoli, Baroni, and Resta[12] introduced a way to calculate linear-response functions and they computed piezoelectric constants, zone-center phonon frequencies, internal strain parameters, effective charges, and static dielectric constants for AlAs and many other III-V crystals. In related work Barino, Giannozzi, and Testa[13,14] had earlier shown how to calculate linear elastic constants, however, they have given results only for Si.

In Sec. II we present a brief account of the method and in Sec. III we compare our work with other results. Finally, we draw some concluding remarks in Sec. IV.

II. METHOD

We have calculated the elastic properties of AlAs by computing the components of the stress tensor for small strains using the same methods as Nielsen and Martin.[9] The C_{11} and C_{12} elastic constants are derived from the harmonic relations $C_{11}=\sigma_1/\epsilon_1$ and $C_{12}=\sigma_2/\epsilon_1$ where σ_i and ϵ_i represent, respectively, the stress and the applied strain using the Voigt[15] notation. The macroscopic stress in the solid may be computed for a small strain by the use of the stress theorem, and the forces on the atoms derived from the Hellman-Feynman theorem.[8] There are internal displacements of the sublattices for an ϵ_4 strain (uniaxial strain in the [111] direction) and the atomic positions are not determined by the symmetry alone. Kleinman[16] defined an internal strain parameter ξ that describes the actual displacements of the atoms. Nielsen and Martin prescribe two independent calculations of stress and force which determine the three independent quantities C_{44}, ξ, and the optical Γ-phonon frequency ω_Γ, with one additional consistency check. The final stress-strain relation that they derive is

TABLE I. Lattice constant a_0, bulk modulus B, shear modulus C_s, and the elastic constants C_{ij} of AlAs. The "bare" elastic constant C^0_{44}, the optical Γ-phonon frequency ω_Γ, and the internal strain parameter ξ.

	a_0 (Å)	B (Mbar)	C_s (Mbar)	C_{11} (Mbar)	C_{12} (Mbar)	C^0_{44} (Mbar)	C_{44} (Mbar)	ω_Γ (THz)	ξ
Calc.	5.59±0.01	0.75±0.01	0.31±0.01	1.16±0.01	0.55±0.01	0.80±0.01	0.57±0.01	10.8±0.2	0.58±0.03
Wiley, Ref. 3		0.773	0.358	1.250	0.534		0.542		
Adachi, Ref. 4		0.779	0.318	1.202	0.567		0.589		
de Gironcoli *et al.*, Ref. 12	5.58								0.617
Expt., Ref. 22	5.660							10.89	

$$\sigma_4=[C^0_{44}-\Omega^{-1}\Phi(\xi a_0/4)^2]\epsilon_4=C_{44}\epsilon_4$$

where C^0_{44} is the elastic constant in the absence of internal displacements and Φ is the calculated force equal to $\mu\omega^2_\Gamma$ where μ is the reduced mass; a_0 is the equilibrium lattice constant and Ω the cell volume.

In the present work we use the Kerker[7] norm-conserving nonlocal pseudopotentials which were derived from all-electron local-density-approximation calculations of the free atoms employing the Ceperley-Alder[17] form for the exchange and correlation energies. Kinetic energy cutoffs of 18 and 24 Ry were applied to the Hamiltonian in a plane-wave basis and our results show good convergence for total energies, forces, and stresses at 18 Ry. We use an ellipsoidal cutoff in the strained lattices to include the same plane waves as in the undistorted crystal. Ten special points were used for the Brillouin-zone **k** integration[18,19] for the undistorted lattice. For the lower symmetry strained lattices there are 20 and 30 points for strains applied in the (100) and (111) directions, respectively. The calculations were done at the theoretical lattice constant of 5.59 Å determined at both 18 and 24 Ry by fitting the total energies to the empirical Murnaghan[20] equation of state.

III. RESULTS

In Table I we present our results at 24 Ry and 10 special **k** points which differ negligibly from the results at 18 Ry. The bulk modulus is defined as $B=(C_{11}+2C_{12})/3$ and the shear modulus as $C_s=(C_{11}-C_{12})/2$. We compare our results with references mentioned in Sec. I.

We use strains of $\epsilon_1=\pm 0.004$ to compute the C_{11} and C_{12} constants. A relative displacement of the atoms by u and a strain of the lattice by ϵ_4 along the (111) direction provide the two independent calculations that determine C_{44}. The change of the (111) bond length in the strained lattice by $(1-\xi)\epsilon_4 a\sqrt{3}/4$ defines the internal strain parameter ξ which is found to be in the range $0<\xi<1$. For a particular ϵ_4 strain of the lattice the effect of the relative displacement u is to produce zero force on the atoms. We performed our calculations with $u=\pm 0.002a$ and $\epsilon_4=\pm 0.004$.

We carried out similar calculations for GaAs using the Kerker pseudopotential at 18 and 24 Ry for which the equilibrium lattice constant was found to be 5.47 Å—much lower than the experimental value of 5.642 Å. Despite this difference, our elastic constants compare very well with experiment and with the results of Nielsen and Martin who used the potential of Bachelet, Hamann, and Schlüter.[21] The calculated (experimental) values in Mbar of C_{11}, C_{12}, and C_{44} were 1.38 (1.22), 0.55 (0.57), and 0.66 (0.60), respectively. We note that it is apparently very important to calculate elastic constants at the theoretical lattice constant since very different results are found if the experimental lattice constant is used.

IV. CONCLUSIONS

We have used an *ab initio* pseudopotential in a plane-wave basis with kinetic energy cutoffs of 18 and 24 Ry to compute highly converged total energies, forces, and stresses in AlAs and GaAs to arrive at the elastic moduli of these materials. Because all our calculations are *ab initio* with no adjustments, we believe that the results of Table I for AlAs should be as accurate as those for GaAs, and can be considered as a prediction for AlAs.

Our first-principles calculations give additional support for the scaling of the elastic moduli proposed by Keyes[2] and the use of this idea in the empirically predicted constants for AlAs given by Wiley.[3]

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