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STABILITY OF THE Si LATTICE: ELECTRON-PHONON INTERACTIONS AND SUPERCONDUCTIVITY IN HEXAGONAL Si

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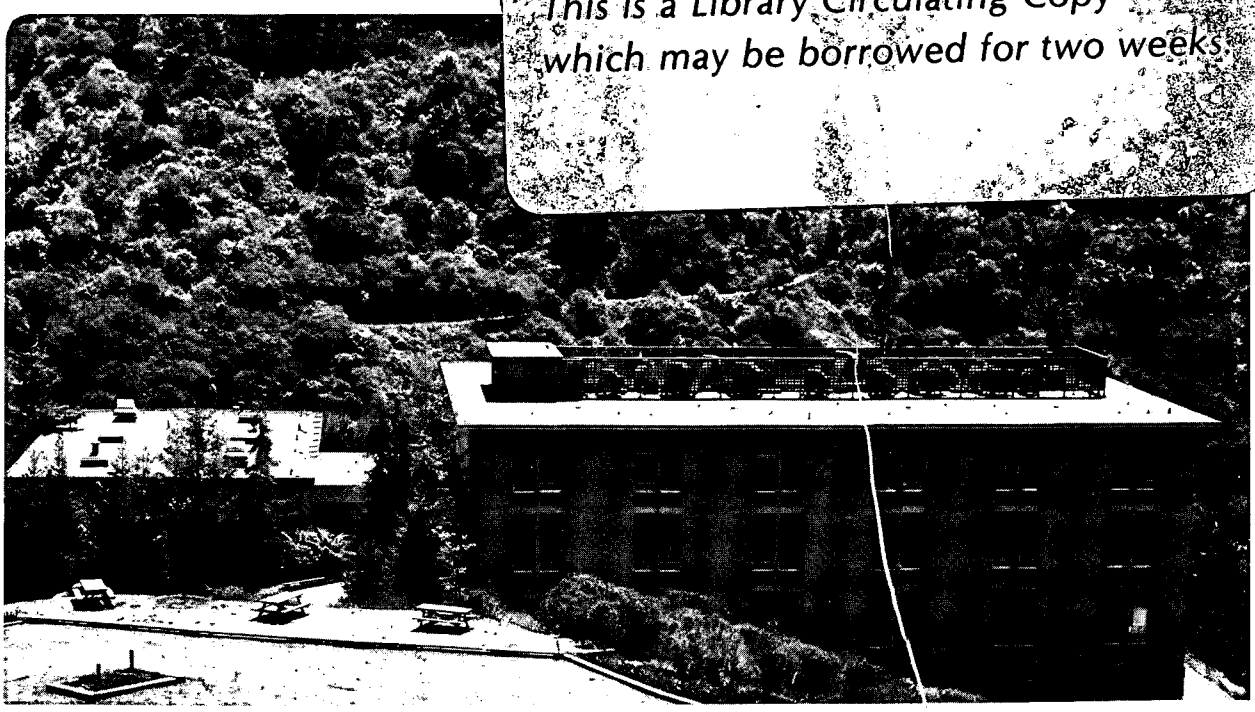

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DISCLAIMER

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ABSTRACT

Using ab initio pseudopotentials the properties of electrons, phonons and electron-phonon interactions in highly compressed Si have been computed. In particular, the hexagonal phases have been predicted to be superconducting by this calculation. The superconducting transition temperature for hexagonal Si has been measured for pressures between 16-45 GPa and compared to the theoretical value.

1. INTRODUCTION

The crystal structure of silicon undergoes a progression of transformations as it is compressed hydrostatically to pressures >10 GPa. From x-ray diffraction studies[1], this progression has been determined to be: diamond -> beta-tin -> primitive hexagonal(PH) -> hexagonal close packed(HCP), occuring approximately at the pressures 10, 16, and 40 GPa respectively (see Fig.1).

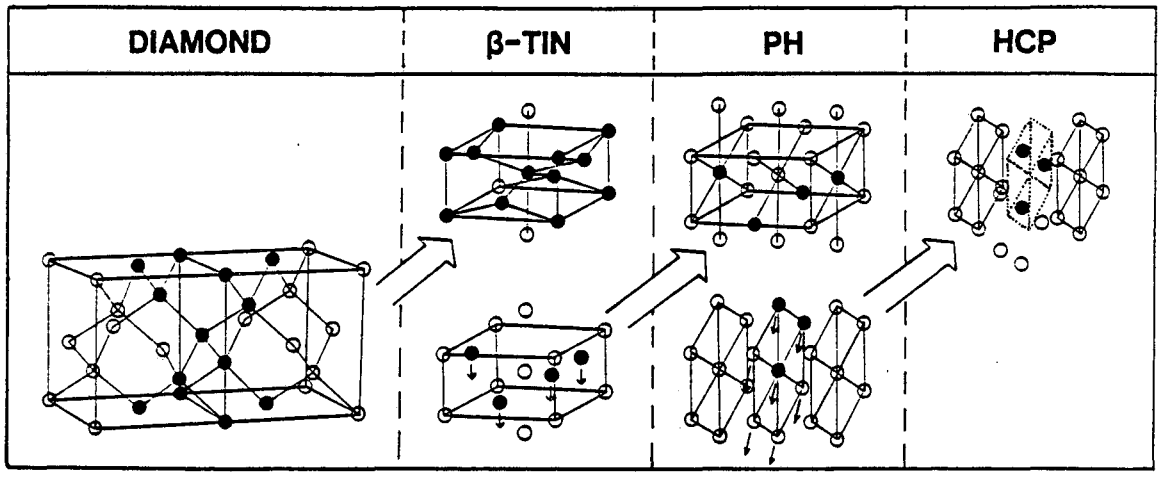

Fig.1. Progression of crystal structures for Si as it is compressed. Solid circles are meant to represent same atoms during transformation between structures.

The first of these phases is semiconducting, the latter ones are metallic. Furthermore, these metallic phases have been found to be superconducting. Because of the great sensitivity of the superconducting transition temperature (Tc) on the detailed properties of the electrons, phonons, and electron-phonon interaction, a study of the pressure (P) dependence of Tc leads to a better understanding of the Si lattice and its bonding.

2. THEORY

Using ab initio pseudopotentials and the frozen phonon approximation, the properties of electrons, phonons and the electron-phonon interaction λ in hexagonal (PH and HCP) Si have been calculated[2,3]. We found that unlike graphite, the bonding between the hexagonal planes of Si atoms is stronger than within the planes. Furthermore, pressure induces additional charge transfer from within the planes to the interplanar region. This charge transfer softens the transverse acoustic (TA) phonon propagating along the c-axis and enhances its electron-phonon interaction. This is shown more clearly from the P dependence of λ for phonons propagating along three directions. The softening of the TA phonon along the [001] direction is responsible for the increase in the corresponding λ near 40 GPa. Similarly, another soft phonon mode along the [011] direction is responsible for the large λ near 20GPa. Based on these theoretical values of λ we expect that Tc, which depends on λ averaged over the entire Brillion zone, to first decrease rapidly with P, reach a minimum above 30GPa and then increase before 40GPa.

3. EXPERIMENTAL

To verify these predictions we have measured the pressure dependence of Tc in Si between 16-46 GPa using a diamond anvil cell (DAC). A new technique has been developed to introduce wires into the DAC for four-probe resistance measurements under quasi-hydrostatic conditions. The sample was a single crystal of Si doped with 3×10^{14} cm^{-3} of phosphorus, 25x25x75 $(micron)^3$ in size, surrounded by a soft

powder and pressed against two half-loops of fine copper wire. A standard steel gasketing arrangement is used, except that Al_2O_3 powder is used to insulate the wires from the gasket. The sample pressure is determined in situ by measuring the shift in the fluorescence wavelength of tiny ruby chips placed adjacent to the sample.

The experimental results are shown in Fig.2a. In agreement with theory, Tc first decreases with pressure above 16 GPa and reaches a minimum value of 3.3 K around 37 GPa before increasing sharply to 4.9 K. The initial decrease agrees with the experimental results of Chang et al.[3]. Around 40 GPa Tc drops suddenly back to 3.5 K, presumably due to a transition into the HCP phase. In this phase Tc increases very slowly with pressure in agreement with the theoretical predictions.

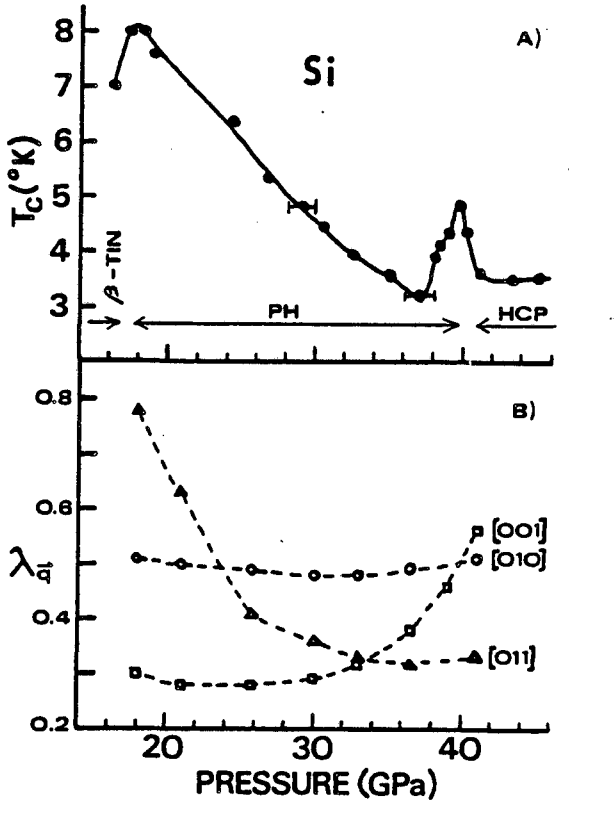

Fig.2 a) Measured superconducting transition temperature Tc for Si versus pressure. b) Calculated pressure dependent electron-phonon λ interaction strength for three phonon wavevectors.

4. DISCUSSION

Overall the experimental P dependence of Tc agrees with the theoretical prediction. A quantitative comparison between theory and experiment is not meaningful at present because λ has been calculated for phonons propagating along three directions only. Also in the present calculation λ is found to be divergent at the beta-tin -> PH transition because the frequency of the phonon in the [011] direction decreases to zero at the phase transition. Thus the present theory breaks down when a phonon mode becomes completely soft.

An additional phase transition into a new intermediated phase (Si VI) between PH and HCP has been observed by Olijnyk et al.[1] in Si between 36 and 40 GPa, (although this phase has not been seen by Spain and coworker[4]). The x-ray diffraction pattern of Si VI can be explained by a double HCP (DHCP) crystal structure, (stacking order ABACABAC...). However a total energy calculation performed for DH Si shows that this phase is metastable. If further x-ray diffraction studies confirm the existence of Si VI, then the increase in Tc between 37 and 40 GPa could be associated with this phase.

5. CONCLUSION

In conclusion, we have investigated both theoretically and experimentally superconductivity of highly compressed Si in its hexagonal phases. The measured pressure dependence of Tc can in large part be explained by the effect of soft phonon modes which accompany structural phase transitions.

6. REFERENCES

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7. ACKNOWLEDGEMENTS

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