

The qubits for quantum information processing are encoded in two-level quantum systems $\{|0\rangle, |1\rangle\}$ (ref. 1), and can be realized, for example, by two spin states $\{|\uparrow\rangle, |\downarrow\rangle\}$ of an electron at the conduction band edge of a semiconductor²⁻⁴. Although Si enjoys a number of advantages over III-V semiconductors in this respect, including long spin coherence lifetime (associated with its weak spin-orbit coupling and small content of non-zero nuclear-spin isotopes)^{5,6}, as well as advanced fabrication know-how, its major drawback is the (sixfold) orbital degeneracy of its lowest conduction band (Fig. 1a) located close to the X point in the Brillouin zone. This is no longer a two-level system determined solely by its spin, leading to considerable leakage and decoherence driven by the energetic proximity among the degenerate orbitals^{6,7}. Whereas this six-fold valley degeneracy in the O_h -symmetric bulk Si can be partially removed by application of tensile biaxial strain⁸, thus, isolating the two lowest $|+z\rangle$ and $|-z\rangle$ components from the rest (Fig. 1b), the creation of a sufficiently large energy splitting within this Z-valley subspace (hereby called valley splitting (VS), see Fig. 1c) has proven to be a challenge for the experimental realization of spin-only qubits in Si⁶. This is clearly indicated by the very limited range of VS (of the order of 1 meV or less) attainable for Si quantum wells surrounded by Ge-Si alloy barriers in experiment⁹⁻¹⁵ and theory¹⁶⁻²³, which seriously hinders the further development of Si-based quantum computation.

The geometry of the basic physical system explored (Fig. 1d) includes a Si slab (well) interfaced by a material with higher conduction band (barrier). The VS of this system depends on a multitude of degrees of freedom present in the actual device growth. The Si well of thickness d clad by barrier materials of composition X_b is coherently strained on a substrate with the planar lattice constant a_s (determined by its composition

splittings of PbSe^{29} . In principle, the splitting within the Z-valleys

superlattice barriers with a shorter period of 16 MLs (see Supplementary Fig. S2a–c). Similar results are obtained for an Si well with the thickness of 47 MLs (located at an odd peak of Fig. 3a, see Supplementary Fig. S2d–f). To better understand this, we explore a simple case—the fixed 40 MLs Si well embedded in Ge_nSi_n superlattice barriers with $n = 1, 2, 4, 8, 16$, as shown in Fig. 5a. We see that the barrier of Ge_4Si_4 superlattice indeed exhibits the largest VS (>7 meV), whereas all other barriers (including pure Ge) show typically low VS (<2 meV). This indicates that the starting sublayer thinner or thicker than Ge_4 seems to equally suppress VS. We unravel the underlying origin within the EMA context. Briefly, the VS induced by an Si/Ge (ascending offset) interface has opposite sign to the Ge/Si (descending offset) interface with the same wave-function. Choosing the interface positions to match the maxima/minima of the VS at the ascending/descending interfaces would maximize the total VS. It is impossible to match the interface positions perfectly to the incommensurate oscillations of well-thickness-dependent VS (Fig. 3), but the Ge_4 sublayer is the closest we can get to this matching within the bilayer growth constraint we impose (better commensurability would be achieved if we chose to analyse any layer thickness, including odd numbers of MLs). Conversely, starting with a Ge_2 sublayer cladding the Si well, we find a destructive interference, in agreement with the suppressed VS for Ge_2Si_2 superlattice barrier in Fig. 5a. This engineering is analogous to that of a distributed Bragg reflector (see Supplementary Note 3 for detailed description). But the fact that the oscillations are incommensurate with the lattice and the strong dependence of VS on atomic ordering makes it impossible to analytically

nature of alloys induces disorder ranging from the geometry of the interface plane to the inhomogeneous strain fields³⁹. The leakage of electrons tunnelling through the superlattice barrier should also be suppressed as the electronic density inside the barrier is much reduced. The structure we proposed is accessible within the current experimental fabrication capabilities such as the molecular beam epitaxy^{8,38,40,41} and chemical vapour deposition^{8,40-44}. Particularly

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