Giant orbital magnetic moments and paramagnetic shift in artificial relativistic atoms and molecules

Zhehao Ge1,7, Sergey Slizovskiy2,3,7, Peter Polizogopoulos1, Toyanath Joshi1, Takashi Taniguchi4, Kenji Watanabe5, David Lederman1, Vladimir I. Fal’ko2,3,6 & Jairo Velasco Jr1

Materials such as graphene and topological insulators host massless Dirac fermions that enable the study of relativistic quantum phenomena. Single quantum dots and coupled quantum dots formed with massless Dirac fermions can be viewed as artificial relativistic atoms and molecules, respectively. Such structures offer a unique testbed to study atomic and molecular physics in the ultrarelativistic regime (particle speed close to the speed of light). Here we use a scanning tunnelling microscope to create and probe single and coupled electrostatically defined graphene quantum dots to unravel the magnetic-field responses of artificial relativistic nanostructures. We observe a giant orbital Zeeman splitting and orbital magnetic moment up to ~70 meV T–1 and ~600 μB (μB, Bohr magneton) in single graphene quantum dots. For coupled graphene quantum dots, Aharonov–Bohm oscillations and a strong Van Vleck paramagnetic shift of ~20 meV T–2 are observed. Our findings provide fundamental insights into relativistic quantum dot states, which can be potentially leveraged for use in quantum information science.
constant and $g$ is the Landé factor) is invalid. This is because massless Dirac fermions disobey the non-relativistic relationship between velocity and momentum, namely, $\mathbf{p} = m\mathbf{v}$. Instead, $\mathbf{p}$ is given by the area of the atomic orbit (in $\alpha^2 r^2$) multiplied by the electrical current ($\alpha^2 2\pi r$), which results in $\mathbf{p} = -e\mathbf{v} \times \mathbf{r} / 2$. Because of this, the large and constant Dirac velocity $\mathbf{v}$, together with a sizable atomic orbital radius $r$ can produce extremely large $\mathbf{p}$ for artificial relativistic atoms. One direct consequence of this large $\mathbf{p}$ is a giant Zeeman splitting for artificial atomic orbital states in a magnetic field ($B$), which can potentially be useful for sensing. Such properties of artificial relativistic atoms, however, have not been experimentally demonstrated to date.

In this Article, we show $B$-dependent scanning tunnelling spectroscopy (STS) of gate-tunable single and coupled graphene quantum dots (GQDs) to unravel the unique $B$ responses of artificial relativistic atoms and molecules. For single GQD states, we observed large linear splitting in $B$, which evidenced the giant $\mathbf{p}$ of these states. Next, we studied the angular quantum number and gate dependence of GQD $\mathbf{p}$ and found good agreement with theory. For coupled GQD states, we found that the linear splitting in $B$ was quenched. Instead, a $B^2$-dependent energy shift towards lower energy was observed due to the existence of a strong paramagnetic shift in relativistic atomic systems. These phenomena underscore the uniqueness of GQDs compared with conventional semiconductor QDs.

**Observation of linear orbital Zeeman splitting**

We study GQDs defined by electrostatically induced circular p–n junctions with scanning tunnelling microscopy (STM) (Fig. 1a). Although Klein tunnelling makes it difficult to confine massless Dirac fermions, their oblique incidence onto the circular p–n junction boundary (Fig. 1b) avoids the 100% transmission occurring at normal incidence. This allows for the formation of quasi-bound states in GQDs, which has been confirmed in previous experiments. In zero $B$, the clockwise and counterclockwise quasi-bound states possessing the same radial quantum number ($n$) and angular quantum numbers ($\pm m$) are degenerate due to time-reversal symmetry. The directions of their $\mathbf{p}$, however, are opposite (Fig. 1c). Thus, by applying an external $B$, the degeneracy between the clockwise and counterclockwise quasi-bound states is lifted through an orbital Zeeman effect (Fig. 1d), leading to a splitting energy of $\Delta E = 2 |\mathbf{p} \cdot B|$. This linear orbital Zeeman splitting can be used to measure $\mathbf{p}$ of GQD states.

Importantly, the Berry-phase change in the GQD states\(^{19,24}\) in $B$ precludes the measurement of $\mathbf{p}$. To avoid this, we create GQDs with unprecedentedly sharp potential wells (Supplementary Section 2). Our method involves using a two-step tip-voltage-pulsing technique based on prior works\(^{13,22}\) (Supplementary Section 3) on samples with reduced hexagonal boron nitride (hBN) thickness. Figure 1e shows a typical $dV/dI$ spectrum along a line across the centre of a circular p–n junction created with this technique on a large-angle (14.1°) twisted bilayer graphene (tBLG)/hBN sample. By tracking the graphene charge density and gate dependence of GQD states with different $\mathbf{p}$, the splitting and merging of GQD states is evident as $\mathbf{p}$ increases. To visualize this behaviour more clearly, $dV/dI(V_G, B)$ with high $B$ resolution was acquired (Fig. 2d). These data were taken from the GQD (Fig. 2a,b) at $d = 40$ nm: here $d^2V/dV_G^2(V_G, B)$ are presented to enhance the visibility of the $dV/dI$ peaks (Extended Data Fig. 1 shows the raw $dV/dI(V_G, B)$ data). We observe a clear linear splitting for each QD state.

We attribute this behaviour to orbital Zeeman splitting and find that it is present at locations off the QD centre but absent near the centre where low-$m$ states concentrate (Supplementary Section 4). All these experimental findings are in good agreement with simulations based on a tight binding (TB) model for a QD (Supplementary Section 5) with a quadratic potential well (Fig. 2e–h), thus further supporting our qualitative understanding. The slight deviations visible between experiment and simulation at negative energies is probably due to the deviation in the experimental potential from the quadratic potential used in our simulation.

**Angular quantum number and gate dependence of orbital magnetic moment**

After confirming the existence of orbital Zeeman splitting, we now extract $\mathbf{p}$ of our artificial relativistic atom and study its angular quantum number and gate dependence. With spatially resolved $dV/dI(V_G, B)$ spectra (Fig. 2a,b), we can assign both radial and angular quantum numbers ($n, m$) for the GQD states in $d^2V/dV_G^2(V_G, B)$ plots\(^{13,22}\) (Supplementary Section 6): the assigned quantum numbers are shown in Fig. 2d.h. By using the simple consideration discussed in Fig. 1d ($(\Delta E = 2 |\mathbf{p} \cdot B|)$, the $\mathbf{p}$ value of the GQD states can then be extracted from the slopes of $\Delta E$ as a function of $B$. Figure 3a shows the extracted $\Delta E(B)$ and the corresponding linear fits for QD states with different $m$ values at $V_G = -16$ V (Supplementary Section 7), a clear increase in slope is seen for states with larger $m$. The magnitude of $\mathbf{p}$ ($\mu$) as a function of $m$ extracted from the slopes of the linear fits (Fig. 3a) is plotted in Fig. 3b. An increase from $\sim200 \mu_B$ to $\sim500 \mu_B$ for $\mu$ is seen when $m$ is increased from 2.5 to 10.5.

Next, we compare our experimentally extracted $\mu$ with theory. Approximately, the measured $\mu$ values are on the order of $\sim300 \mu_B$, which agrees well with the $\mu$ value of a current loop ($\mu = \gamma s$) for charge flowing with graphene’s Fermi velocity $v_F = 10^6$ m s\(^{-1}\) and with a loop radius of $r = 35$ nm. For a more formal comparison, we calculated the $m$-resolved local density of states LDOS($E, B$) from a continuum model for GQDs with quadratic potential wells (Supplementary Section 8); some results are shown in Fig. 3c–e. From such plots, we can extract $\mu$ for GQD states with different quantum numbers, and the extracted $\mu$ for states with $n = 0$ and $m = 2.5$–10.5 are plotted in Fig. 3b. Here we notice that the experimental results (red triangles) do not overlay any individual theory curve (empty circles). Additionally, the experimental results at larger $m$ appear closer to the theoretical curves calculated with a smaller $|\mathbf{p}|$. Akin to the discrepancy shown in Fig. 2, the discrepancy evident here is probably caused by the deviation in the experimental potential from a quadratic potential at negative $V_G$ (Extended Data Fig. 2). Nonetheless, for both experiment and theory, a clear increase in $\mu$ is seen with increasing $m$.

We now explore the gate dependence of $\mu$ for our artificial relativistic atoms. Figure 3f–h shows the $dV/dI(V_G, B, d)$ spectra measured at $V_G = -20$, $-10$ and $0$ V and $B = 0.2$ T for the same QD in Fig. 2a. The potential-well sharpness of the GQDs in our experiments can be tuned by $V_G$ (Supplementary Section 10). Apparently, as the potential-well sharpness increases with $V_G$, the splitting energy reduces. Figure 3i shows the experimentally extracted $\Delta E(B)$ for the $n = 0, m = 5.5$ QD state at various $V_G$ values; the $d^2V/dV_G^2(V_G, B)$ data at these $V_G$ values are shown in Extended Data Fig. 3. A clear increase in slope is seen as $V_G$ decreases, indicating an enhancement in $\mu$ as $V_G$ decreases. To see this more quantitatively, we plot the extracted $\mu$ values as a function of $m$ (Fig. 3j) for GQD states with different $m$ values measured at various $V_G$ values.
The extracted $\mu$ values are generally smaller at larger $V_C$ (sharper potential well) for all the QD states. The observed gate tuning of $\mu$ can be understood as a result of orbital size tuning of the QD states with $V_C$: sharper dots at larger $V_C$ have current loops with a smaller radius. In contrast to non-relativistic atoms, $\mu$ for relativistic atoms is governed by the orbital radius instead of angular momentum. Therefore, it is uniquely possible to tune $\mu$ of our artificial relativistic atom by $V_C$ and maintain the same quantum numbers. These results are also supported by the theoretically calculated $\mu(m)$ values for GQDs with different potential-well sharpness values (Fig. 3b).

We now compare the observed $\mu$ value in the GQDs with those of other systems. The value of $\mu$ observed in this work is orders of magnitude larger than those observed in natural atoms$^{28}$ and semiconductor QDs$^{29–31}$, and is also several times larger than that observed in Bernal-stacked bilayer graphene (BLG) QDs$^{32–34}$ (Table 1). Although similar $\mu$ values have been observed in Bernal-stacked trilayer graphene QDs$^{34}$, the findings in this work have several distinctions/advantages. Briefly, GQDs can achieve similar $\mu$ with a smaller QD size and maintain linear splitting within a larger $B$ range compared with trilayer graphene QDs (Supplementary Section 11). Thus, the extremely large orbital Zeeman splitting observed in our GQDs ($\sim 23–58$ meV T$^{-1}$) together with their nanometre-scale sizes offers a unique opportunity to fabricate magnetometer arrays with nanometre-scale spatial resolution (Supplementary Section 12). This is difficult to achieve for the current state of the art$^{35,36}$.

**Coupled double GQDs**

Having thoroughly investigated the $\mu$ values of single GQDs, below, we study the $B$ response of coupled double GQDs, which can be viewed as artificial relativistic molecules$^{37}$. These structures were created on a graphene/hBN sample by fabricating two circular p-doped regions with centres separated by 150 nm with our two-step tip-voltage-pulsing technique (Supplementary Section 3). Figure 4a shows a $d^2I/dV_s^2 (V_s, d)$ plot measured along a line across the centres of two dots for $B = 0$ T at $V_C = 0$ V: the red and blue patterns in the plot correspond to $d^2I/dV_s^2$ peaks and valleys, respectively. These three different regions can be identified (labelled as regions (i), (ii) and (iii) in Fig. 4a). In region (i), the energy spacing between the $d^2I/dV_s^2$ peaks is half those in regions (ii) and (iii): in region (iii), different nodal patterns appear compared with region (ii). These features are distinct from uncoupled double GQDs made with a similar fabrication technique (Supplementary Section 12).

Next, we map the $B$ response of our coupled GQDs. The plots of $d^2I/dV_s^2 (V_s, B)$ measured in the three distinct regions of the coupled dots are shown in Fig. 4b–e. First, in region (i), we notice that the QD states display a positive energy shift that is proportional to $B^2$; such...
behaviour is more evident in the zoomed-in view (Fig. 4c). The parabolic energy shift observed here is -20 meV T⁻². Second, in region (ii), we encounter a linear splitting resembling the observation in single GQDs (Fig. 4d). Finally, region (iii) reveals a distinct behaviour compared with the other regions, namely, a staggered pattern for $d_\ell/dV_s$ peaks (Fig. 4e). All these observations are qualitatively reproduced with a TB model for a coupled double GQD (Supplementary Section 13).

To attain an intuitive understanding of these observations, we consider semiclassical orbits within strongly coupled double GQDs. For QD states in a single GQD, their semiclassical orbits can be approximated as circular orbits. Once these two QD states strongly couple, the circular orbits of individual QD states merge into a figure-eight orbit. A schematic of such an orbit indicated by yellow rings embedded in a double GQD is shown in Fig. 4f. Here the arrows indicate the reversed current-flow directions of the two rings with the figure-eight orbit. Note that the single-dot states with the same direction of circulation on the dot couple only weakly (for example, by potential gradient) due to the counterpropagation of states at the touching dot edges. Closer to each individual QD centre, we expect the QD states to have a smaller radius and are decoupled, thus forming circular orbits (Fig. 4f, green rings). In our experiment, regions (i) and (iii) correspond to QD states with figure-eight orbits and region (ii) corresponds to QD states with circular orbits. With this understanding in hand, the half-energy spacing observed in region (i) compared with region (ii) is due to the length of the figure-eight orbit being twice that of the circular orbit. This is because according to the semiclassical quantization rule, the energy spacing between the QGD states is $\Delta E = \frac{\mu L^2}{2\hbar}$, where $\hbar$ is the Planck constant, $\mu$ is the graphene Fermi velocity and $L$ is the semiclassical orbit length. Consequently, the large $B$-induced linear splitting observed in region (ii) can be explained by the large $\mu$ value of the circular orbits akin to uncoupled GQDs.

**Van Vleck paramagnetic shift and Aharonov-Bohm effect**

We now discuss how the unique $B$ response observed in region (i) corresponds to the emergence of a Van Vleck paramagnetic shift due to the relativistic nature of our artificial molecule. Because each of the two rings of the figure-eight orbit have reversed current-flow directions, their $\mu$ are in opposite directions but with the same magnitude. Therefore, the net $\mu$ value of an entire figure-eight orbit will be zero, thereby explaining the disappearance of linear splitting in $B$ in regions (i) and (iii). Moreover, the positive parabolic energy shift for holes observed in region (i) is caused by a Van Vleck paramagnetic shift, which is a second-order perturbative $B$ response. Classically, this effect stems from a Lorentz force that expands (contracts) the orbits with $\mu$ aligned (anti-aligned) to $B$, resulting in an increase (decrease) in $\mu$. The quenching of the first-order $B$ effect in the figure-eight orbits helps the detection of this second-order effect. Importantly, for non-relativistic systems such as natural atoms and semiconductor QDs, a Larmor diamagnetic shift due to the change in electron orbital velocity in $B$ also exists and is usually stronger than the...
For works in which the \( \mu \) values are not directly given, we convert the observed Zeeman diamagnetism in non-relativistic systems (Supplementary Section 14). Such a \( B \)-induced response is thus unique to ultrarelativistic artificial atoms and molecules.

Finally, we discuss the origin of the staggered red stripe patterns in \( dI/dV \) (\( V_G, B \)) plot in region (iii) and attribute them to Aharonov–Bohm (AB) oscillations occurring in region (iii). When a particle returns to its original position after travelling along a closed path, constructive (destructive) interference leading to enhanced (reduced) LDOS occurs if the action along the closed path \( \Phi = \oint p \cdot dq \) equals to an even (odd) number of \( \pi \). Here \( p \) and \( q \) are the canonical momentum and position coordinates, respectively. As shown in Fig. 4g, two distinct eigenenergies exist for the figure-eight orbits with constructive (yellow dot) and destructive (black dot) interference at the figure-eight orbit centre. This is because changes returns to this location after travelling half of a figure-eight orbit. By applying an external \( B \), the AB effect causes the pickup of additional phases (\( \Delta \varphi = \frac{\Phi}{\pi} \)), where \( \Phi \) is the magnetic flux through each circular segment) with opposite signs for the two circular orbits flowing in opposite directions. As a result, the energy level of the figure-eight orbit does not change in \( B \) to the first order, but the interference type at the figure-eight orbit centre switches depending on the amount of \( \Delta \varphi \) picked up by each circular segment (Fig. 4g). This explains the staggered pattern observed in region (iii) (Fig. 4e), where the red and blue stripes (corresponding to constructive and destructive interference, respectively) alternate at a constant \( V_G \) in \( B \). In addition, the assigned \( \Delta \varphi \) values (Fig. 4e) are in good agreement with our Van Vleck paramagnetic shift. However, for QGDs, the Larmor diamagnetic shift is absent because of the constant velocity of massless Dirac fermions. Alternatively, this can be understood as resulting from the graphene Hamiltonian in \( B \) lacking a \( B^2 \) term that produces Larmor magnetic shift. However, this can be understood as resulting from the graphene Hamiltonian in \( B \) lacking a \( B^2 \) term that produces Larmor diamagnetism in non-relativistic systems (Supplementary Section 14). Such a \( B \)-induced response is thus unique to ultrarelativistic artificial atoms and molecules.

Finally, we discuss the origin of the staggered red stripe patterns in \( dI/dV \) (\( V_G, B \)) plot in region (iii) and attribute them to Aharonov–Bohm (AB) oscillations occurring in region (iii). When a particle returns to its original position after travelling along a closed path, constructive (destructive) interference leading to enhanced (reduced) LDOS occurs if the action along the closed path \( \Phi = \oint p \cdot dq \) equals to an even (odd) number of \( \pi \). Here \( p \) and \( q \) are the canonical momentum and position coordinates, respectively. As shown in Fig. 4g, two distinct eigenenergies exist for the figure-eight orbits with constructive (yellow dot) and destructive (black dot) interference at the figure-eight orbit centre. This is because changes returns to this location after travelling half of a figure-eight orbit. By applying an external \( B \), the AB effect causes the pickup of additional phases (\( \Delta \varphi = \frac{\Phi}{\pi} \)), where \( \Phi \) is the magnetic flux through each circular segment) with opposite signs for the two circular orbits flowing in opposite directions. As a result, the energy level of the figure-eight orbit does not change in \( B \) to the first order, but the interference type at the figure-eight orbit centre switches depending on the amount of \( \Delta \varphi \) picked up by each circular segment (Fig. 4g). This explains the staggered pattern observed in region (iii) (Fig. 4e), where the red and blue stripes (corresponding to constructive and destructive interference, respectively) alternate at a constant \( V_G \) in \( B \). In addition, the assigned \( \Delta \varphi \) values (Fig. 4e) are in good agreement with our Van Vleck paramagnetic shift. However, for QGDs, the Larmor diamagnetic shift is absent because of the constant velocity of massless Dirac fermions. Alternatively, this can be understood as resulting from the graphene Hamiltonian in \( B \) lacking a \( B^2 \) term that produces Larmor
A experimental double-dot geometry (Supplementary Section 15). This observed AB oscillation in the LDOS intensity of coupled GQDs can also be potentially used for B sensing.

Conclusions
We observed giant \( \mu \) and large orbital Zeeman splitting in artificial relativistic atoms formed with single GQDs. We also observed strong Van Vleck paramagnetic shifts and AB oscillations in artificial relativistic molecules formed with coupled double GQDs. These phenomena stem from the long-lived states near the GQD edges, thus resembling persistent currents in metallic rings\(^{30,31} \). Notably, our work demonstrates the potential for realizing giant and tunable persistent currents in GQDs. Such currents can potentially be used in quantum information processing\(^{32} \) and quantum simulation\(^{33} \).

**Online content**
Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41565-023-01327-0.

**References**
1. Novoselov, K. S. et al. Two-dimensional gas of massless Dirac fermions in graphene. Nature 438, 197–200 (2005).
2. Katsnelson, M., Novoselov, K. & Geim, A. Chiral tunnelling and the Klein paradox in graphene. Nat. Phys. 2, 620–625 (2006).
3. Young, A. F. & Kim, P. Quantum interference and Klein tunnelling in graphene heterojunctions. Nat. Phys. 5, 222–226 (2009).
4. Wang, Y. et al. Observing atomic collapse resonances in artificial nuclei on graphene. Science 340, 734–737 (2013).
5. Lu, J. et al. Frustrated supercritical collapse in tunable charge arrays on graphene. Nat. Commun. 10, 477 (2019).
6. Chen, S. et al. Electron optics with p-n junctions in ballistic graphene. Science 353, 1522–1525 (2016).
7. Cheianov, V. V., Fal’ko, V. & Atkinson, B. The focusing of electron flow and a Veselago lens in graphene p-n junctions. Science 315, 1252–1255 (2007).
8. Cheianov, V. V. & Fal’ko, V. I. Selective transmission of Dirac electrons and ballistic magnetoresistance of p–n junctions in graphene. Phys. Rev. B 74, 041403 (2006).
9. Liu, M.-H., Gorini, C. & Richter, K. Creating and steering highly directional electron beams in graphene. Phys. Rev. Lett. 118, 066801 (2017).
10. Chakraborty, T. Quantum Dots: A Survey of the Properties of Artificial Atoms (Elsevier, 1999).
11. Reimann, S. M. & Manninen, M. Electronic structure of quantum dots. Rev. Mod. Phys. 74, 1283 (2002).
12. Kouwenhoven, L. P., Austing, D. & Tarucha, S. Few-electron quantum dots. Rep. Prog. Phys. 64, 701 (2001).
13. Hanson, R., Kouwenhoven, L. P., Petta, J. R., Tarucha, S. & Vandersypen, L. M. Spins in few-electron quantum dots. Rev. Mod. Phys. 79, 1217 (2007).
14. Lodahl, P., Mahmoodian, S. & Stobbe, S. Interfacing single photons and single quantum dots with photonic nanostructures. Rev. Mod. Phys. 87, 347 (2015).
15. Zhao, Y. et al. Creating and probing electron whispering-gallery modes in graphene. Science 348, 672–675 (2015).
16. Freitag, N. M. et al. Electrostatically confined monolayer graphene quantum dots with orbital and valley splittings. Nano Lett. 16, 5798–5805 (2016).
17. Gutiérrez, C., Brown, L., Kim, C.-J., Park, J. & Pasupathy, A. N. Klein tunnelling and electron trapping in nanometre-scale graphene quantum dots. Nat. Phys. 12, 1069–1075 (2016).
18. Lee, J. et al. Imaging electrostatically confined Dirac fermions in graphene quantum dots. Nat. Phys. 12, 1032–1036 (2016).
19. Ghahari, F. et al. An on/off Berry phase switch in circular graphene resonators. Science 356, 845–849 (2017).
20. Jiang, Y. et al. Tuning a circular p–n junction in graphene from quantum confinement to optical guiding. Nat. Nanotechnol. 12, 1045–1049 (2017).
21. Bai, K.-K. et al. Generating atomically sharp p–n junctions in graphene and testing quantum electron optics on the nanoscale. Phys. Rev. B 97, 045413 (2018).
22. Freitag, N. M. et al. Large tunable valley splitting in edge-free graphene quantum dots on boron nitride. Nat. Nanotechnol. 13, 392–397 (2018).
23. Quezada-López, E. A. et al. Comprehensive electrostatic modeling of exposed quantum dots in graphene/hexagonal boron nitride heterostructures. Nanomaterials 10, 1154 (2020).
24. Behn, W. A. et al. Measuring and tuning the potential landscape of electrostatically defined quantum dots in graphene. Nano Lett. 21, 5013–5020 (2021).
25. Zhang, J., Jiang, Y.-P., Ma, X.-C. & Xue, Q.-K. Berry-phase switch in graphene quantum dots. Phys. Rev. Lett. 128, 126402 (2022).
26. Rodriguez-Nieva, J. F. & Levitov, L. S. Berry phase jumps and giant nonreciprocity in Dirac quantum dots. Phys. Rev. B 94, 235406 (2016).
27. Ge, Z. et al. Visualization and manipulation of bilayer graphene quantum dots with broken rotational symmetry and nontrivial topology. Nano Lett. 20, 8682–8688 (2020).
28. Bethe, H. A. & Salpeter, E. E. Quantum Mechanics of One- and Two-Electron Atoms (Springer Science & Business Media, 2012).
29. Rinaldi, R. et al. Zeeman effect in parabolic quantum dots. Phys. Rev. Lett. 77, 342 (1996).
30. Paskov, P. et al. Magnetoluminescence of highly excited InAs/GaAs self-assembled quantum dots. Phys. Rev. B 62, 7344 (2000).
31. Raymond, S. et al. Excitonic energy shell structure of self-assembled InGaAs/GaAs quantum dots. Phys. Rev. Lett. 92, 187402 (2004).
32. Ren, Y.-N., Cheng, Q., Sun, Q.-F. & He, L. Realizing valley-polarized energy spectra in bilayer graphene quantum dots via continuously tunable Berry phases. Phys. Rev. Lett. 128, 206805 (2022).
33. Tong, C. et al. Tunable valley splitting and bipolar operation in graphene quantum dots. Nano Lett. 21, 1068–1073 (2021).
34. Ge, Z. et al. Control of giant topological magnetic moment and valley splitting in trilayer graphene. Phys. Rev. Lett. 127, 136402 (2021).
35. Lenz, J. & Edelstein, S. Magnetic sensors and their applications. IEEE Sens. J. 6, 631–649 (2006).
36. Degen, C. L., Reinhard, F. & Cappellaro, P. Quantum sensing. Rev. Mod. Phys. 89, 035002 (2017).
37. Fu, Z.-Q. et al. Relativistic artificial molecules realized by two coupled graphene quantum dots. Nano Lett. 20, 6738–6743 (2020).
38. Ashcroft, N. W. & Mermin, N. D. Solid State Physics (Cengage Learning, 1976).
39. Arimondo, E., Ciampini, D. & Rizzo, C. Chapter one—spectroscopy of natural and artificial atoms in magnetic fields. In Advances In Atomic, Molecular, and Optical Physics 65, 1–66 (Elsevier, 2016).
40. Ambegaokar, V. & Eckern, U. Coherence and persistent currents in mesoscopic rings. Phys. Rev. Lett. 65, 381 (1990).
41. Bleszynski-Jayich, A. et al. Persistent currents in normal metal rings. Science 326, 272–275 (2009).
42. Mooij, J. et al. Josephson persistent-current qubit. Science 285, 1036–1039 (1999).
43. Nisoli, C., Moessner, R. & Schiffer, P. Colloquium: artificial spin ice: designing and imaging magnetic frustration. Rev. Mod. Phys. 85, 1473 (2013).

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations. Springer Nature or its licensor (e.g. a society or other partner) holds exclusive rights to this article under a publishing agreement with the author(s) or other rightsholder(s); author self-archiving of the accepted manuscript version of this article is solely governed by the terms of such publishing agreement and applicable law.

© The Author(s), under exclusive licence to Springer Nature Limited 2023
Methods
Sample fabrication
The graphene/hBN and tBLG/hBN samples were assembled with a standard polymer-based transfer method\textsuperscript{44}. For the graphene/hBN sample, a graphene flake exfoliated on a methyl methacrylate (MMA) substrate was mechanically placed on top of an ~20-nm-thick hBN flake that rests on a 285 nm SiO\textsubscript{2}/Si++ substrate. For the tBLG sample, we first use a monolayer graphene flake on MMA to pick up another monolayer graphene flake on the SiO\textsubscript{2}/Si++ substrate and then we place tBLG/MMA onto an ~20-nm-thick hBN flake that rests on a 285 nm SiO\textsubscript{2}/Si++ substrate. For both samples, the MMA scaffold was dissolved in a subsequent solvent bath. The assembled heterostructure is then annealed in forming gas (Ar/H\textsubscript{2}) for ~12 h at 400 °C to reduce the residual polymer after the heterostructure assembly procedure. Next, an electrical contact to the sample is made by thermally evaporating 7 nm Cr and 200 nm Au using a metallic stencil mask. To further improve the sample surface cleanliness, the heterostructure is then mechanically cleaned using an atomic force microscope\textsuperscript{45}, which is done in a glovebox filled with N\textsubscript{2} gas. We perform sequential scans in the contact mode (set point, 0.2 V; scanning speed, ~15 µm s\textsuperscript{-1}; resolution, 1,024 × 1,024 pixels) to sweep regions of ~30 × 30 µm\textsuperscript{2} by a Cypher S atomic force microscope with Econo-ESP-Au tips from Oxford Instruments. Finally, the heterostructure is annealed in an ultrahigh vacuum at 400 °C for 7 h before being introduced into the STM chamber.

STM/STS measurements
The STM/STS measurements were conducted in an ultrahigh vacuum with pressures higher than 1 × 10\textsuperscript{−10} mbar at 4.8 K in a Createc LT-STM instrument. Electrochemically etched tungsten tips calibrated on a Au(111) surface were used in the experiments. The lock-in a.c. signal frequency used for the STS measurements was 704 Hz.

Data availability
Source data are provided with this paper. Any additional material is available from the corresponding authors upon reasonable request.

Code availability
All the codes used in this Article are available from the corresponding authors upon request.

References
44. Zomer, P., Dash, S., Tombros, N. & Van Wees, B. A transfer technique for high mobility graphene devices on commercially available hexagonal boron nitride. Appl. Phys. Lett. 99, 232104 (2011).
45. Goossens, A. et al. Mechanical cleaning of graphene. Appl. Phys. Lett. 100, 073110 (2012).

Acknowledgements
We thank the Hummingbird Computational Cluster team at University of California Santa Cruz for providing computational resources for the numerical TB calculations performed in this work, and M. Hance for providing insight into the accelerator physics considerations related to our experimental findings. J.V.J. and Z.G. acknowledge support from the National Science Foundation under award DMR-1753367. J.V.J. acknowledges support from the Army Research Office under contract W911NF-17-1-0473. V.I.F. and S.S. acknowledge support from the European Graphene Flagship Core 3 Project. V.I.F. acknowledges support from the Lloyd Register Foundation Nanotechnology Grant and EPSRC grants EP/VO07033/1, EP/S030719/1 and EP/NO10345/1. K.W. and T.T. acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan, via grant no. JPMXP0112101001 and JSPS KAKENHI via grant no. JP20H00354.

Author contributions
Z.G. and J.V.J. conceived the work and designed the research strategy. Z.G. fabricated the samples and performed the data analysis under the supervision of J.V.J. K.W. and T.T. provided the hBN crystals. Z.G. carried out the tunnelling spectroscopy measurements with assistance from P.P. and T.J. under the supervision of D.L. and J.V.J. S.S. developed the interpretation for experimental findings and performed the continuum model calculations under the supervision of V.I.F. Z.G. performed the numerical TB calculations with input from S.S. under the supervision of V.I.F. and J.V.J. Z.G. and J.V.J. wrote the paper. All the authors discussed the paper and commented on the manuscript.

Competing interests
The authors declare no competing interests.

Additional information
Extended data is available for this paper at https://doi.org/10.1038/s41565-023-01327-0.

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41565-023-01327-0.

Correspondence and requests for materials should be addressed to Vladimir I. Fal’ko or Jairo Velasco.

Peer review information Nature Nanotechnology thanks the anonymous reviewers for their contribution to the peer review of this work.

Reprints and permissions information is available at www.nature.com/reprints.
Extended Data Fig. 1 | Raw $dI/dV_S(V_S, B)$ used to get $d^3I/dV_S^3(V_S, B)$ plot in Fig. 2d. $dI/dV_S(V_S, B)$ taken at $d = 40$ nm for the same GQD shown in Fig. 2a,b at $V_G = -16$ V. The set point used to acquire the tunneling spectra was $I = 1$ nA, $V_S = -60$ mV, with a 2 mV ac modulation.
Extended Data Fig. 2 | Deviation between experimental potential well and parabolic potential well. a, Experimentally measured $dI/dV_s(V_s, d)$ for the same QD shown in Fig. 2a at $V_G = -16$ V along a line across the center of a circular graphene pn junction. Colored lines are quadratic potential wells with different $\kappa$ values. The set point used to acquire the tunneling spectra was $I = 1$ nA, $V_s = -200$ mV, with a 2 mV ac modulation. b, Schematic of the deviation between experimental potential well and parabolic potential well at more negative energies. The experimental potential well deviates from parabolic potential well at more negative energies, and the actual QD radius will be larger than the parabolic potential well. This explains the faster increase of experimentally measured $\mu$ than the theoretical values for graphene QDs with a quadratic potential well.
Extended Data Fig. 3 | $d^3I/dV^3_s(V_s, B)$ plot at different $V_g$. a, $d^3I/dV^3_s(V_s, B)$ at $V_g = -24$ V and at $d = 36$ nm. b, $d^3I/dV^3_s(V_s, B)$ at $V_g = -20$ V and at $d = 40$ nm. c, $d^3I/dV^3_s(V_s, B)$ at $V_g = -10$ V and at $d = 36$ nm. d, $d^3I/dV^3_s(V_s, B)$ at $V_g = 0$ V and at $d = 25$ nm. The quantum number $(n,m)$ in (a–d) corresponds to radial and angular quantum number, respectively.