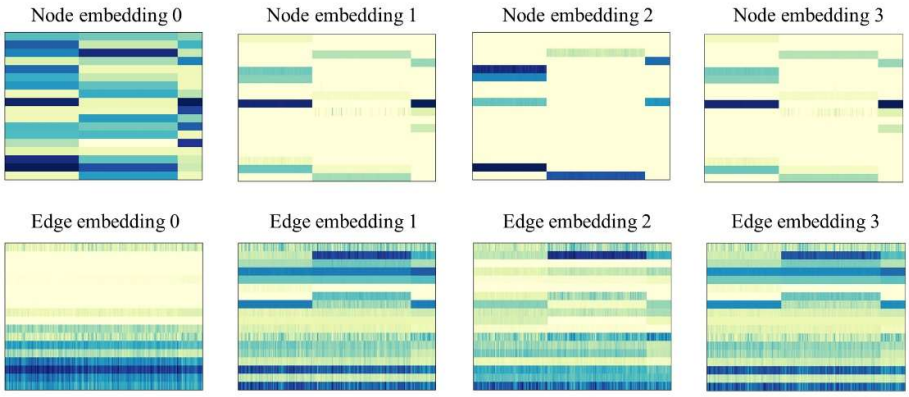
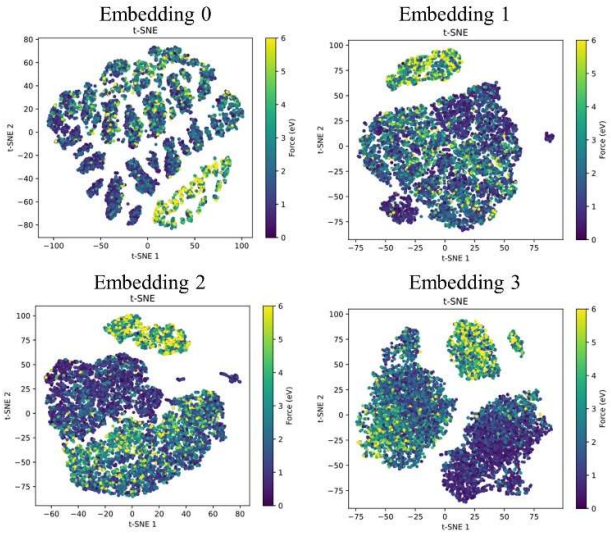
Efficient modelling of ionic and electronic interactions by resistive memory-based reservoir graph neural network

Content of Supplementary Figures

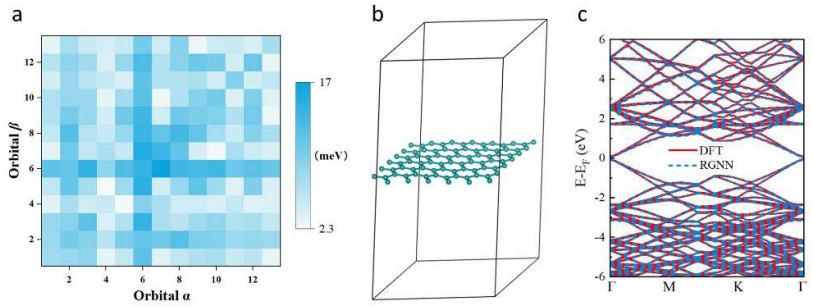


Supplementary Figure 1. Embedding update process for molecular dynamics simulations based on our co-design. In order, embedding 0 is the initial embedding, embedding 1 is updated by the first random MP layer, embedding 2 is updated by the second random MP layer, and embedding 3 is updated by the trainable embedding layer. The edge embedding is the average of all the edges for each atom. It indicates both the node and edge embeddings updated by the random MP layers (node embedding 2 and edge embedding 2) are similar to those updated by the trainable embedding layer (node embedding 3 and edge embedding 3). It therefore confirms that the random weights from resistive memory play a major role in the embedding update process of the RGNN.

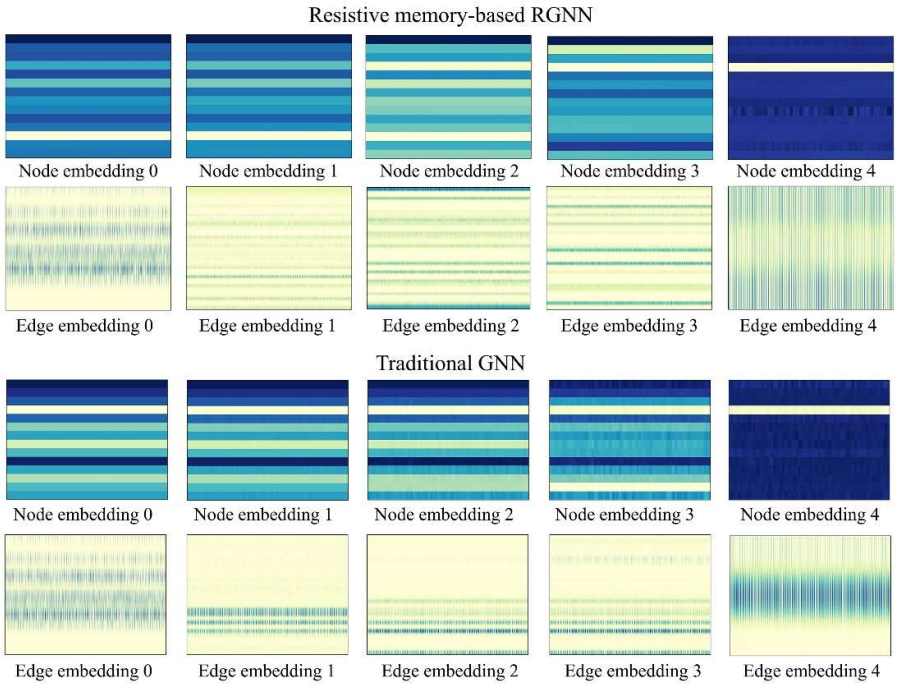


Supplementary Figure 2. Correlation between edge embeddings and atomic forces during embedding update. The average of all edge embeddings of each atom is calculated, and then the t-SNE method is used to reduce the dimension, and the color map is used to represent the atomic force. In order, embedding 0 is the initial edge embedding, embedding 1 is updated by the first random MP layer,

embedding 2 is updated by the second random MP layer, and embedding 3 is updated by the trainable embedding layer. As the embedding is updated, the correlation between edge embeddings and atomic forces gradually increases. The relevance of atomic forces and edge embeddings is already evident in embedding 2 without trainable embedding layers, thus proving the major contribution of random weights.

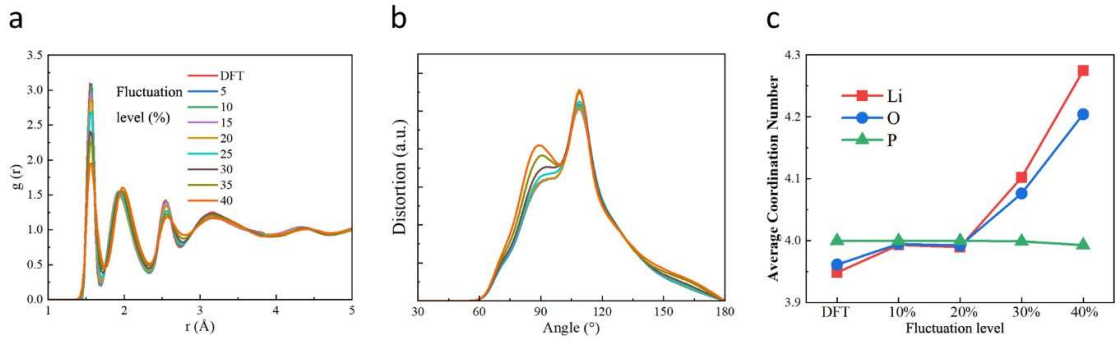


Supplementary Figure 3. Testing results of the trained RGNN model in electronic structure calculation experiment. **a**, MAEs of Hamiltonian of each orbital calculated on the testing dataset. **b**, Atomic structure of one graphene model sampled from the testing dataset. **c**, Comparison of band structures of graphene (b) based on software RGNN and DFT. It confirms that the RGNN model can accurately predict the Hamiltonian of graphene after sufficient training, and further calculate the energy band structure.

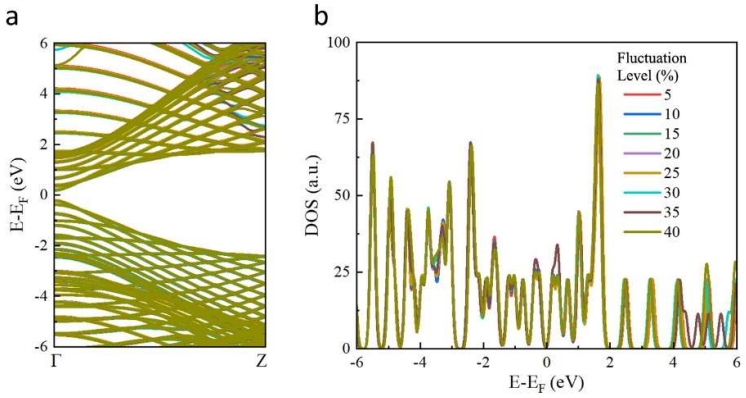


Supplementary Figure 4. Comparison of the embedding update process of our co-design and traditional GNN models. The initial node and edge embedding are denoted as 0, then the embeddings 1, 2 and 3

are updated using random MP layers, and the embedding 4 is update using trainable embedding layer. Comparing to traditional GNN, the resistive memory-based RGNN model has the similar node and edge embeddings during the update process. It explains why our co-design can accurately predict the Hamiltonian using the random weights.

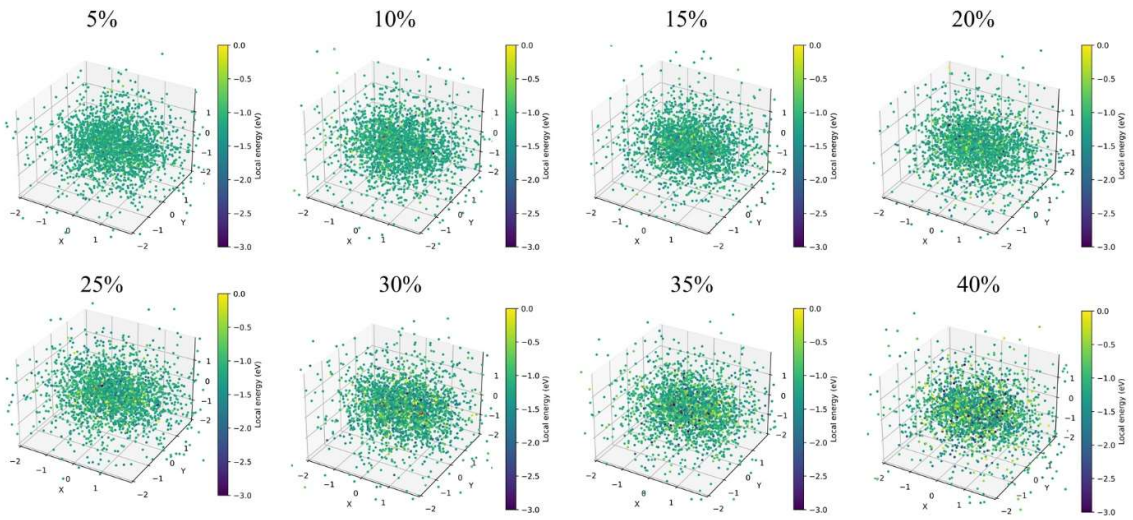


Supplementary Figure 5. Local structures are analyzed under various conductance fluctuation levels. Different levels of weight fluctuation (from 5% to 40%) are incorporated into the RGNN model predicting atomic force, followed by MD simulation. The local structures of the resulting amorphous model are computed, including the pair distribution function (a), bond angle distribution function (b), and coordination number distribution (c). As the fluctuation level increases, the main peak of the pair distribution function diminishes, the 90° peak of the bond angle distribution function intensifies, and the average coordination number of Li and O rises. This suggests that conductance fluctuations introduce structural errors in the MD simulation. However, when the fluctuation level is below 20%, the local structure error is minimal and does not significantly impact the understanding of the material's nature. This confirms that the MD simulation based on our co-design is resistant to conductance fluctuations in resistive memory.



Supplementary Figure 6. Electronic structures under various conductance fluctuation levels. Different levels of weight fluctuations (from 5% to 40%) are added to the RGNN model predicting Hamiltonian, and then to calculate the energy band and DOS of carbon nanotube (zigzag). As the fluctuation level

increases, both the energy band and DOS change only slightly. It confirms the conductance fluctuation of resistive memory could not affect the calculation of electronic structure.



Supplementary Figure 7. Electronic configurations and local energies under various conductance fluctuation levels. To investigate the impact of conductance fluctuation on the performance of our co-design, we simulate the various conductance fluctuations (5% to 40%). These conductance fluctuations are incorporated into the RGNN-based wavefunction of H_2, which is then used to perform MCMC steps to update the electronic configurations. After numerous MCMC steps, the potential electron positions and corresponding local energies are displayed. As the fluctuation level increases, the distribution of electron positions remains relatively unchanged, while the distribution range of local energy expands significantly. At a 40% fluctuation level, a substantial number of local energy values deviate from the original ground state energy (approximately -1.173 eV), which is the primary cause of the error in the calculated total energy. However, when the fluctuation level is below 20%, the local energy distribution is not significantly affected, enabling relatively high calculation accuracy.