| | $\langle\mathcal{H}\rangle_{ex}$ | $\langle\mathcal{H}\rangle_{th}$ | $Tangle$ | $Overlap$ | $\theta_1$ | $\theta_2$ | $\theta_3$ | $\theta_4$ | $\theta_5$ | $\theta_6$ |
|---|---|---|---|---|---|---|---|---|---|---|
| 1 | -2.5078 | -2.7073 | 0.0179 | 0.8923 | 3.3001 | 2.0815 | 2.7118 | 4.5107 | 5.7567 | 5.5922 |
| 2 | -2.0926 | -2.1451 | 0.2272 | 0.4888 | 2.2041 | 3.1980 | 3.8283 | 5.6272 | 0.5901 | 0.4256 |
| 3 | -2.0373 | -1.9210 | 0.0776 | 0.2396 | 4.4167 | 0.9855 | 3.8283 | 5.6272 | 0.5901 | 0.4256 |
| 4 | -1.8814 | -1.8707 | 0.2204 | 0.0331 | 4.4167 | 3.1980 | 1.6158 | 5.6272 | 0.5901 | 0.4256 |
| 5 | -1.8263 | -1.8630 | 0.2831 | 0.1849 | 4.4167 | 3.1980 | 3.8283 | 3.4147 | 0.5901 | 0.4256 |
| 6 | -0.9813 | -0.9516 | 0.1783 | 0.2804 | 4.4167 | 3.1980 | 3.8283 | 5.6272 | 4.6607 | 0.4256 |
| 7 | -1.6469 | -1.6768 | 0.0838 | 0.2184 | 4.4167 | 3.1980 | 3.8283 | 5.6272 | 0.5901 | 4.4962 |
| 8 | -1.6073 | -1.7091 | 0.5067 | 0.1037 | 5.4014 | 4.1827 | 4.8130 | 0.3288 | 2.4305 | 1.4103 |
| 9 | -0.8575 | -0.9881 | 0.0138 | 0.0945 | 5.1552 | 3.9365 | 4.5668 | 0.0826 | 4.5588 | 1.1641 |
| 10 | -2.6995 | -2.7383 | 0.0067 | 0.9205 | 5.8937 | 2.6397 | 3.2701 | 5.0690 | 0.0318 | 6.1505 |
| 11 | -2.2993 | -2.5159 | 0.0133 | 0.7448 | 3.8584 | 4.6751 | 3.2701 | 5.0690 | 0.0318 | 6.1505 |
| 12 | -1.9179 | -2.1183 | 0.6881 | 0.4342 | 3.8584 | 2.6397 | 5.3054 | 5.0690 | 0.0318 | 6.1505 |
| 13 | -2.3298 | -2.4948 | 0.0125 | 0.7347 | 3.8584 | 2.6397 | 3.2701 | 0.8211 | 0.0318 | 6.1505 |
| 14 | -2.3581 | -2.4798 | 0.0250 | 0.7361 | 3.8584 | 2.6397 | 3.2701 | 5.0690 | 0.0318 | 1.9026 |
| 15 | -1.6101 | -1.9230 | 0.7991 | 0.7196 | 3.8584 | 2.6397 | 3.2701 | 5.0690 | 2.0671 | 6.1505 |
| 16 | -1.4786 | -1.5846 | 0.1501 | 0.3406 | 4.3508 | 3.1321 | 3.7624 | 5.5613 | 4.0936 | 0.3596 |
| 17 | -1.8377 | -1.9169 | 0.5750 | 0.6002 | 3.9815 | 2.7628 | 3.3931 | 5.1921 | 1.0029 | 6.2736 |
| 18 | -1.9379 | -1.8873 | 0.0024 | 0.1088 | 4.5969 | 2.3606 | 2.9909 | 4.7898 | 6.0359 | 5.8713 |
| 19 | -1.9937 | -2.0772 | 0.0022 | 0.1768 | 4.8761 | 2.6397 | 3.2701 | 5.0690 | 0.0318 | 0.8850 |
| 20 | -2.1023 | -2.0829 | 0.0010 | 0.1764 | 4.8761 | 2.6397 | 3.2701 | 6.0866 | 0.0318 | 6.1505 |
| 21 | -2.0475 | -2.0774 | 0.0014 | 0.1767 | 4.8761 | 3.6574 | 3.2701 | 5.0690 | 0.0318 | 6.1505 |
| 22 | -2.0776 | -2.0871 | 0.0846 | 0.0817 | 4.8761 | 2.6397 | 4.2877 | 5.0690 | 0.0318 | 6.1505 |
| 23 | -1.3951 | -1.3407 | 0.0716 | 0.2161 | 4.8761 | 2.6397 | 3.2701 | 5.0690 | 1.0495 | 6.1505 |
| 24 | -1.1719 | -1.1289 | 0.4467 | 0.3230 | 5.1222 | 2.8859 | 3.5162 | 5.3152 | 5.2043 | 0.1135 |
| 25 | -1.9891 | -1.9942 | 0.0356 | 0.2589 | 4.9376 | 2.7013 | 3.3316 | 5.1305 | 0.5174 | 6.2120 |
| 26 | -2.2089 | -2.2807 | 0.2986 | 0.5363 | 5.5149 | 3.2786 | 3.9089 | 5.7078 | 0.4728 | 0.5062 |
| 27 | -2.0589 | -2.1332 | 0.4612 | 0.4875 | 5.3667 | 3.1304 | 3.7607 | 5.5596 | 5.9764 | 0.3579 |
| 28 | -2.2991 | -2.3281 | 0.4780 | 0.5147 | 5.5918 | 3.3555 | 3.9858 | 5.7847 | 0.0660 | 5.5093 |
| 29 | -2.1885 | -2.2572 | 0.4431 | 0.4142 | 5.8304 | 2.2372 | 4.2243 | 6.0233 | 0.0773 | 0.0301 |
| 30 | -2.1160 | -2.2005 | 0.0407 | 0.5254 | 5.4943 | 2.4664 | 3.8883 | 5.6872 | 0.5439 | 5.7133 |
| 31 | -2.8815 | -2.8495 | 0.0028 | 0.9916 | 6.1910 | 2.8993 | 3.2281 | 0.1007 | 0.3761 | 6.0583 |
| 32 | -2.3104 | -2.4283 | 0.4365 | 0.7411 | 0.5653 | 3.0291 | 2.6983 | 0.7582 | 0.5482 | 6.0122 |
| 33 | -2.2211 | -2.1728 | 0.1510 | 0.4109 | 0.3461 | 2.9858 | 4.2318 | 5.4653 | 0.4908 | 6.0275 |
| 34 | -2.5510 | -2.5875 | 0.0148 | 0.8172 | 0.1062 | 3.3323 | 3.7280 | 5.7908 | 6.2442 | 0.2862 |
| 35 | -2.8393 | -2.8501 | 0.0008 | 0.9936 | 6.2397 | 3.9266 | 3.2265 | 5.3772 | 0.3888 | 6.0549 |
| 36 | -2.4879 | -2.6233 | 0.0308 | 0.8471 | 0.5135 | 3.1011 | 3.3145 | 5.5825 | 6.2485 | 5.3339 |
| 37 | -2.4566 | -2.3986 | 0.0401 | 0.7715 | 5.7381 | 3.4323 | 2.6859 | 5.8640 | 6.0554 | 5.8646 |
| 38 | -2.2352 | -2.4707 | 0.3266 | 0.6791 | 0.5412 | 3.0883 | 2.4986 | 5.5711 | 0.0991 | 0.2180 |
| 39 | -2.5119 | -2.6311 | 0.0723 | 0.8258 | 5.8999 | 3.2887 | 3.6140 | 5.7313 | 0.0742 | 5.7573 |
| 40 | -2.1806 | -2.2142 | 0.1772 | 0.4847 | 0.4488 | 2.9636 | 4.1078 | 5.4475 | 0.4936 | 6.1102 |
| 41 | -2.7794 | -2.7658 | 0.0009 | 0.9542 | 5.9866 | 3.3151 | 3.0414 | 5.7599 | 6.2357 | 5.9260 |
| 42 | -2.3013 | -2.3962 | 0.0664 | 0.6364 | 5.4034 | 3.3661 | 3.3882 | 5.7882 | 0.2961 | 0.5550 |
| 43 | -2.8255 | -2.7967 | 0.0022 | 0.9607 | 0.1652 | 3.1674 | 3.3329 | 5.6339 | 0.0481 | 5.7100 |
| 44 | -2.7888 | -2.8022 | 0.0132 | 0.9637 | 0.1998 | 3.1381 | 2.9950 | 5.6070 | 0.1785 | 0.1158 |
| 45 | -2.4054 | -2.6302 | 0.1425 | 0.8569 | 6.0247 | 3.0298 | 2.6366 | 5.4862 | 0.3643 | 5.5301 |
| 46 | -2.7119 | -2.7756 | 0.0001 | 0.9445 | 0.0150 | 3.2567 | 3.4552 | 5.7146 | 0.0618 | 0.0264 |
| 47 | -2.5890 | -2.7247 | 0.0375 | 0.9190 | 0.3720 | 3.9280 | 3.1563 | 0.1400 | 0.3035 | 6.0021 |
| 48 | -2.8318 | -2.8236 | 0.0007 | 0.9763 | 6.0841 | 2.9618 | 3.2416 | 5.4075 | 0.0997 | 6.1134 |
| 49 | -2.8154 | -2.8045 | 0.0108 | 0.9698 | 6.1793 | 3.2128 | 2.9000 | 5.6752 | 0.2861 | 5.7776 |
| 50 | -2.7204 | -2.7443 | 0.0407 | 0.9399 | 0.2720 | 3.1202 | 3.2667 | 5.6017 | 0.5066 | 6.1117 |
| 51 | -2.8434 | -2.8365 | 0.0016 | 0.9887 | 6.1288 | 3.2664 | 3.0977 | 5.7203 | 0.0910 | 5.9724 |
| 52 | -2.7332 | -2.7545 | 0.0025 | 0.9268 | 5.9408 | 3.3400 | 3.3473 | 5.7924 | 0.2514 | 5.4965 |
| 53 | -2.8295 | -2.8453 | 0.0010 | 0.9928 | 0.0642 | 3.1886 | 3.0831 | 5.6533 | 0.1967 | 6.1734 |
| 54 | -2.7891 | -2.7508 | 0.0001 | 0.9278 | 0.0172 | 3.2572 | 3.5033 | 5.7169 | 0.1141 | 6.2498 |
| 55 | -2.7689 | -2.8469 | 0.0023 | 0.9942 | 6.2096 | 3.2239 | 3.0508 | 5.6856 | 0.2431 | 5.8956 |
| 56 | -2.7714 | -2.8492 | 0.0026 | 0.9931 | 6.1599 | 3.0829 | 3.1629 | 6.0521 | 0.2336 | 6.0153 |
| 57 | -2.8427 | -2.8511 | 0.0016 | 0.9930 | 6.2153 | 3.4129 | 3.2273 | 5.8806 | 0.3824 | 6.0566 |
| 58 | -2.7948 | -2.8407 | 0.0018 | 0.9856 | 6.1375 | 2.9306 | 3.2349 | 5.8957 | 0.2379 | 6.0858 |
| 59 | -2.7675 | -2.8547 | 0.0001 | 0.9977 | 6.2692 | 3.0439 | 3.1556 | 6.0186 | 0.2864 | 6.1158 |
| 60 | -2.7752 | -2.8349 | 0.0000 | 0.9842 | 0.0365 | 3.0333 | 3.2805 | 6.0089 | 0.2121 | 5.8841 |
| 61 | -2.8024 | -2.8481 | 0.0060 | 0.9943 | 6.1852 | 3.0561 | 3.0641 | 6.0295 | 6.6143 | 5.9179 |
| 62 | -2.8322 | -2.8400 | 0.0036 | 0.9847 | 6.1337 | 3.0944 | 3.2437 | 6.0650 | 6.5878 | 5.8902 |
| 63 | -2.8791 | -2.8449 | 0.0006 | 0.9890 | 6.2342 | 3.0593 | 3.2633 | 6.0357 | 6.6644 | 5.9490 |
| 64 | -2.7867 | -2.8311 | 0.0413 | 0.9797 | 6.0460 | 3.1175 | 3.1399 | 6.0879 | 6.7422 | 6.1018 |
| 65 | -2.8534 | -2.8457 | 0.0139 | 0.9889 | 6.1144 | 3.0965 | 3.1751 | 6.0682 | 6.6805 | 6.0474 |
| 66 | -2.8242 | -2.8574 | 0.0031 | 0.9977 | 6.2204 | 3.2756 | 3.1656 | 6.2586 | 6.7695 | 5.8873 |
| 67 | -2.8221 | -2.8157 | 0.0019 | 0.9675 | 6.1845 | 3.2233 | 3.3703 | 6.2011 | 6.7281 | 6.0449 |
| 68 | -2.7665 | -2.8380 | 0.0027 | 0.9829 | 6.1847 | 3.1815 | 3.2937 | 6.1582 | 6.6996 | 6.0132 |
| 69 | -2.7585 | -2.8479 | 0.0015 | 0.9907 | 6.2126 | 2.9793 | 3.2457 | 6.2098 | 6.6618 | 6.0036 |
| 70 | -2.8464 | -2.8494 | 0.0069 | 0.9913 | 6.1527 | 2.9979 | 3.2016 | 6.2260 | 6.6699 | 6.0528 |
| 71 | -2.7616 | -2.8507 | 0.0021 | 0.9925 | 6.2032 | 3.1561 | 3.2277 | 6.1322 | 6.6624 | 6.0574 |
| 72 | -2.8526 | -2.8456 | 0.0035 | 0.9886 | 6.1623 | 2.9969 | 3.2359 | 6.2244 | 6.6235 | 5.9742 |
| 73 | -2.8756 | -2.8545 | 0.0033 | 0.9953 | 6.2057 | 3.0875 | 3.1969 | 6.3212 | 6.7144 | 5.9728 |
| 74 | -2.8543 | -2.8543 | 0.0042 | 0.9963 | 6.1881 | 2.9777 | 3.1461 | 6.2067 | 6.6368 | 5.9881 |
| 75 | -2.7703 | -2.8517 | 0.0077 | 0.9935 | 6.1550 | 3.0591 | 3.1664 | 6.2884 | 6.6603 | 6.0309 |
| 76 | -2.7991 | -2.8493 | 0.0086 | 0.9925 | 6.1484 | 2.8500 | 3.1639 | 6.4180 | 6.6589 | 5.9683 |
| 77 | -2.8093 | -2.8498 | 0.0027 | 0.9920 | 6.1944 | 2.8773 | 3.2244 | 6.3051 | 6.6606 | 5.9739 |
| 78 | -2.8871 | -2.8480 | 0.0013 | 0.9907 | 6.2163 | 3.0955 | 3.2470 | 6.1378 | 6.6626 | 6.0384 |
| 79 | -2.8141 | -2.8392 | 0.0002 | 0.9854 | 6.2502 | 3.2183 | 3.2886 | 5.9977 | 6.6644 | 6.0735 |
| 80 | -2.8701 | -2.8528 | 0.0045 | 0.9939 | 6.1776 | 3.1410 | 3.1941 | 6.1950 | 6.6615 | 6.0543 |
| 81 | -2.8481 | -2.8524 | 0.0011 | 0.9943 | 6.2276 | 3.0680 | 3.2144 | 6.2637 | 6.6495 | 5.9759 |
| 82 | -2.8878 | -2.8525 | 0.0020 | 0.9941 | 6.2089 | 3.0505 | 3.2112 | 6.2542 | 6.6546 | 5.9951 |
| 83 | -2.8744 | -2.8564 | 0.0014 | 0.9974 | 6.2335 | 3.0870 | 3.1719 | 6.2752 | 6.7061 | 6.0614 |
| 84 | -2.8794 | -2.8444 | 0.0013 | 0.9880 | 6.2229 | 3.1426 | 3.2703 | 6.3157 | 6.7160 | 6.0720 |
| 85 | -2.8618 | -2.8475 | 0.0005 | 0.9911 | 6.2485 | 2.9798 | 3.2477 | 6.3677 | 6.7095 | 6.0117 |
| 86 | -2.7606 | -2.8528 | 0.0031 | 0.9939 | 6.1953 | 3.1007 | 3.2075 | 6.2382 | 6.6735 | 6.0436 |
| 87 | -2.7711 | -2.8506 | 0.0016 | 0.9926 | 6.2126 | 3.0730 | 3.2291 | 6.1960 | 6.6586 | 6.0168 |
| 88 | -2.8412 | -2.8512 | 0.0024 | 0.9929 | 6.1999 | 2.9749 | 3.2197 | 6.3191 | 6.6569 | 6.0267 |
| 89 | -2.8097 | -2.8493 | 0.0016 | 0.9915 | 6.2159 | 3.0965 | 3.2408 | 6.2850 | 6.6853 | 6.0335 |
| 90 | -2.7855 | -2.8535 | 0.0026 | 0.9947 | 6.2073 | 3.0690 | 3.2040 | 6.2877 | 6.6845 | 5.9839 |
| 91 | -2.8392 | -2.8549 | 0.0018 | 0.9960 | 6.2212 | 3.0687 | 3.1916 | 6.2647 | 6.6804 | 6.0283 |
| 92 | -2.8207 | -2.8529 | 0.0031 | 0.9941 | 6.1933 | 3.0957 | 3.2027 | 6.2246 | 6.6580 | 6.0247 |
| 93 | -2.7630 | -2.8542 | 0.0030 | 0.9953 | 6.2029 | 3.0455 | 3.1942 | 6.3491 | 6.6813 | 6.0140 |
| 94 | -2.7949 | -2.8517 | 0.0019 | 0.9934 | 6.2102 | 3.0661 | 3.2204 | 6.2343 | 6.6643 | 6.0161 |
| 95 | -2.8657 | -2.8510 | 0.0017 | 0.9929 | 6.2092 | 3.0485 | 3.2247 | 6.2396 | 6.6487 | 6.0575 |
| 96 | -2.7915 | -2.8528 | 0.0023 | 0.9941 | 6.2060 | 3.0455 | 3.2098 | 6.2948 | 6.6637 | 6.0392 |
| 97 | -2.8469 | -2.8520 | 0.0020 | 0.9936 | 6.2091 | 3.0610 | 3.2177 | 6.2494 | 6.6641 | 6.0218 |
| 98 | -2.8688 | -2.8542 | 0.0027 | 0.9956 | 6.1980 | 3.0033 | 3.1817 | 6.2322 | 6.6356 | 6.0178 |
| 99 | -2.7382 | -2.8525 | 0.0013 | 0.9943 | 6.2222 | 2.9732 | 3.2129 | 6.2951 | 6.6554 | 6.0244 |
| 100 | -2.8243 | -2.8530 | 0.0026 | 0.9943 | 6.2005 | 3.0651 | 3.2052 | 6.2422 | 6.6574 | 6.0246 |

**Supplementary Table** 1: **Experimental results for the optimisation run shown in Figure 3.** Experimental results for the first 60 steps of the optimization run for $R = 90$ pm. $\langle\mathcal{H}\rangle_{ex}$ is the experimentally computed energy, $\langle\mathcal{H}\rangle_{th}$ is the energy calculated analytically at each experimental step. The arbitrary normalised two-bit states $|\psi\rangle = \alpha\,|00\rangle + \beta\,|01\rangle + \gamma\,|10\rangle + \delta\,|11\rangle$ are represented by six real numbers, $\{\theta_1, \ldots, \theta_6\}$. The first state is randomly generated.

| $R$ | $II$ | $IX$ | $IZ$ | $XI$ | $XX$ | $XZ$ | $ZI$ | $ZX$ | $ZZ$ |
|---|---|---|---|---|---|---|---|---|---|
| 0.05 | 33.9557 | -0.1515 | -2.4784 | -0.1515 | 0.1412 | 0.1515 | -2.4784 | 0.1515 | 0.2746 |
| 0.1 | 13.3605 | -0.1626 | -2.4368 | -0.1626 | 0.2097 | 0.1626 | -2.4368 | 0.1626 | 0.2081 |
| 0.15 | 6.8232 | -0.1537 | -2.3801 | -0.1537 | 0.2680 | 0.1537 | -2.3801 | 0.1537 | 0.1512 |
| 0.2 | 3.6330 | -0.1405 | -2.2899 | -0.1405 | 0.3027 | 0.1405 | -2.2899 | 0.1405 | 0.1176 |
| 0.25 | 1.7012 | -0.1324 | -2.1683 | -0.1324 | 0.3211 | 0.1324 | -2.1683 | 0.1324 | 0.1010 |
| 0.3 | 0.3821 | -0.1306 | -2.0305 | -0.1306 | 0.3303 | 0.1306 | -2.0305 | 0.1306 | 0.0943 |
| 0.35 | -0.5810 | -0.1335 | -1.8905 | -0.1335 | 0.3344 | 0.1335 | -1.8905 | 0.1335 | 0.0936 |
| 0.4 | -1.3119 | -0.1396 | -1.7568 | -0.1396 | 0.3352 | 0.1396 | -1.7568 | 0.1396 | 0.0969 |
| 0.45 | -1.8796 | -0.1477 | -1.6339 | -0.1477 | 0.3339 | 0.1477 | -1.6339 | 0.1477 | 0.1030 |
| 0.5 | -2.3275 | -0.1570 | -1.5236 | -0.1570 | 0.3309 | 0.1570 | -1.5236 | 0.1570 | 0.1115 |
| 0.55 | -2.6844 | -0.1669 | -1.4264 | -0.1669 | 0.3264 | 0.1669 | -1.4264 | 0.1669 | 0.1218 |
| 0.6 | -2.9708 | -0.1770 | -1.3418 | -0.1770 | 0.3206 | 0.1770 | -1.3418 | 0.1770 | 0.1339 |
| 0.65 | -3.2020 | -0.1871 | -1.2691 | -0.1871 | 0.3135 | 0.1871 | -1.2691 | 0.1871 | 0.1475 |
| 0.7 | -3.3893 | -0.1968 | -1.2073 | -0.1968 | 0.3052 | 0.1968 | -1.2073 | 0.1968 | 0.1626 |
| 0.75 | -3.5417 | -0.2060 | -1.1552 | -0.2060 | 0.2958 | 0.2060 | -1.1552 | 0.2060 | 0.1791 |
| 0.8 | -3.6660 | -0.2145 | -1.1117 | -0.2145 | 0.2853 | 0.2145 | -1.1117 | 0.2145 | 0.1968 |
| 0.85 | -3.7675 | -0.2222 | -1.0758 | -0.2222 | 0.2738 | 0.2222 | -1.0758 | 0.2222 | 0.2157 |
| 0.9 | -3.8505 | -0.2288 | -1.0466 | -0.2288 | 0.2613 | 0.2288 | -1.0466 | 0.2288 | 0.2356 |
| 0.95 | -3.9183 | -0.2343 | -1.0233 | -0.2343 | 0.2481 | 0.2343 | -1.0233 | 0.2343 | 0.2564 |
| 1 | -3.9734 | -0.2385 | -1.0052 | -0.2385 | 0.2343 | 0.2385 | -1.0052 | 0.2385 | 0.2779 |
| 1.05 | -4.0180 | -0.2414 | -0.9916 | -0.2414 | 0.2199 | 0.2414 | -0.9916 | 0.2414 | 0.3000 |
| 1.1 | -4.0539 | -0.2430 | -0.9820 | -0.2430 | 0.2053 | 0.2430 | -0.9820 | 0.2430 | 0.3225 |
| 1.15 | -4.0825 | -0.2431 | -0.9758 | -0.2431 | 0.1904 | 0.2431 | -0.9758 | 0.2431 | 0.3451 |
| 1.2 | -4.1050 | -0.2418 | -0.9725 | -0.2418 | 0.1756 | 0.2418 | -0.9725 | 0.2418 | 0.3678 |
| 1.25 | -4.1224 | -0.2392 | -0.9716 | -0.2392 | 0.1610 | 0.2392 | -0.9716 | 0.2392 | 0.3902 |
| 1.3 | -4.1356 | -0.2353 | -0.9728 | -0.2353 | 0.1466 | 0.2353 | -0.9728 | 0.2353 | 0.4123 |
| 1.35 | -4.1454 | -0.2301 | -0.9757 | -0.2301 | 0.1327 | 0.2301 | -0.9757 | 0.2301 | 0.4339 |
| 1.4 | -4.1523 | -0.2239 | -0.9798 | -0.2239 | 0.1194 | 0.2239 | -0.9798 | 0.2239 | 0.4549 |
| 1.45 | -4.1568 | -0.2167 | -0.9850 | -0.2167 | 0.1068 | 0.2167 | -0.9850 | 0.2167 | 0.4751 |
| 1.5 | -4.1594 | -0.2086 | -0.9910 | -0.2086 | 0.0948 | 0.2086 | -0.9910 | 0.2086 | 0.4945 |
| 1.55 | -4.1605 | -0.1998 | -0.9975 | -0.1998 | 0.0837 | 0.1998 | -0.9975 | 0.1998 | 0.5129 |
| 1.6 | -4.1602 | -0.1905 | -1.0045 | -0.1905 | 0.0734 | 0.1905 | -1.0045 | 0.1905 | 0.5304 |
| 1.65 | -4.1589 | -0.1807 | -1.0116 | -0.1807 | 0.0640 | 0.1807 | -1.0116 | 0.1807 | 0.5468 |
| 1.7 | -4.1568 | -0.1707 | -1.0189 | -0.1707 | 0.0555 | 0.1707 | -1.0189 | 0.1707 | 0.5622 |
| 1.75 | -4.1540 | -0.1605 | -1.0262 | -0.1605 | 0.0479 | 0.1605 | -1.0262 | 0.1605 | 0.5766 |
| 1.8 | -4.1508 | -0.1503 | -1.0334 | -0.1503 | 0.0410 | 0.1503 | -1.0334 | 0.1503 | 0.5899 |
| 1.85 | -4.1471 | -0.1403 | -1.0404 | -0.1403 | 0.0350 | 0.1403 | -1.0404 | 0.1403 | 0.6023 |
| 1.9 | -4.1431 | -0.1305 | -1.0473 | -0.1305 | 0.0297 | 0.1305 | -1.0473 | 0.1305 | 0.6138 |
| 1.95 | -4.1390 | -0.1210 | -1.0540 | -0.1210 | 0.0251 | 0.1210 | -1.0540 | 0.1210 | 0.6244 |
| 2 | -4.1347 | -0.1119 | -1.0605 | -0.1119 | 0.0212 | 0.1119 | -1.0605 | 0.1119 | 0.6342 |
| 2.05 | -4.1303 | -0.1031 | -1.0667 | -0.1031 | 0.0178 | 0.1031 | -1.0667 | 0.1031 | 0.6432 |
| 2.1 | -4.1258 | -0.0949 | -1.0727 | -0.0949 | 0.0148 | 0.0949 | -1.0727 | 0.0949 | 0.6516 |
| 2.15 | -4.1214 | -0.0871 | -1.0785 | -0.0871 | 0.0124 | 0.0871 | -1.0785 | 0.0871 | 0.6594 |
| 2.2 | -4.1169 | -0.0797 | -1.0840 | -0.0797 | 0.0103 | 0.0797 | -1.0840 | 0.0797 | 0.6666 |
| 2.25 | -4.1125 | -0.0729 | -1.0893 | -0.0729 | 0.0085 | 0.0729 | -1.0893 | 0.0729 | 0.6733 |
| 2.3 | -4.1082 | -0.0665 | -1.0944 | -0.0665 | 0.0070 | 0.0665 | -1.0944 | 0.0665 | 0.6796 |
| 2.35 | -4.1040 | -0.0606 | -1.0993 | -0.0606 | 0.0058 | 0.0606 | -1.0993 | 0.0606 | 0.6854 |
| 2.4 | -4.0998 | -0.0551 | -1.1040 | -0.0551 | 0.0047 | 0.0551 | -1.1040 | 0.0551 | 0.6909 |
| 2.45 | -4.0957 | -0.0500 | -1.1085 | -0.0500 | 0.0039 | 0.0500 | -1.1085 | 0.0500 | 0.6961 |
| 2.5 | -4.0918 | -0.0454 | -1.1128 | -0.0454 | 0.0032 | 0.0454 | -1.1128 | 0.0454 | 0.7010 |
| 2.55 | -4.0879 | -0.0411 | -1.1170 | -0.0411 | 0.0026 | 0.0411 | -1.1170 | 0.0411 | 0.7056 |
| 2.6 | -4.0841 | -0.0371 | -1.1210 | -0.0371 | 0.0021 | 0.0371 | -1.1210 | 0.0371 | 0.7099 |
| 2.65 | -4.0805 | -0.0335 | -1.1248 | -0.0335 | 0.0017 | 0.0335 | -1.1248 | 0.0335 | 0.7141 |
| 2.7 | -4.0769 | -0.0303 | -1.1285 | -0.0303 | 0.0014 | 0.0303 | -1.1285 | 0.0303 | 0.7181 |
| 2.75 | -4.0735 | -0.0273 | -1.1321 | -0.0273 | 0.0011 | 0.0273 | -1.1321 | 0.0273 | 0.7218 |
| 2.8 | -4.0701 | -0.0245 | -1.1356 | -0.0245 | 0.0009 | 0.0245 | -1.1356 | 0.0245 | 0.7254 |
| 2.85 | -4.0669 | -0.0221 | -1.1389 | -0.0221 | 0.0007 | 0.0221 | -1.1389 | 0.0221 | 0.7289 |
| 2.9 | -4.0638 | -0.0198 | -1.1421 | -0.0198 | 0.0006 | 0.0198 | -1.1421 | 0.0198 | 0.7322 |
| 2.95 | -4.0607 | -0.0178 | -1.1452 | -0.0178 | 0.0005 | 0.0178 | -1.1452 | 0.0178 | 0.7354 |
| 3 | -4.0578 | -0.0159 | -1.1482 | -0.0159 | 0.0004 | 0.0159 | -1.1482 | 0.0159 | 0.7385 |
| 3.05 | -4.0549 | -0.0142 | -1.1511 | -0.0142 | 0.0003 | 0.0142 | -1.1511 | 0.0142 | 0.7414 |
| 3.1 | -4.0521 | -0.0127 | -1.1539 | -0.0127 | 0.0002 | 0.0127 | -1.1539 | 0.0127 | 0.7443 |
| 3.15 | -4.0494 | -0.0114 | -1.1566 | -0.0114 | 0.0002 | 0.0114 | -1.1566 | 0.0114 | 0.7470 |
| 3.2 | -4.0468 | -0.0101 | -1.1592 | -0.0101 | 0.0001 | 0.0101 | -1.1592 | 0.0101 | 0.7497 |
| 3.25 | -4.0443 | -0.0090 | -1.1618 | -0.0090 | 0.0001 | 0.0090 | -1.1618 | 0.0090 | 0.7522 |
| 3.3 | -4.0418 | -0.0081 | -1.1643 | -0.0081 | 0.0001 | 0.0081 | -1.1643 | 0.0081 | 0.7547 |
| 3.35 | -4.0394 | -0.0072 | -1.1666 | -0.0072 | 0.0001 | 0.0072 | -1.1666 | 0.0072 | 0.7571 |
| 3.4 | -4.0371 | -0.0064 | -1.1690 | -0.0064 | 0.0001 | 0.0064 | -1.1690 | 0.0064 | 0.7595 |
| 3.45 | -4.0349 | -0.0056 | -1.1712 | -0.0056 | 0.0000 | 0.0056 | -1.1712 | 0.0056 | 0.7617 |
| 3.5 | -4.0327 | -0.0050 | -1.1734 | -0.0050 | 0.0000 | 0.0050 | -1.1734 | 0.0050 | 0.7639 |
| 3.55 | -4.0306 | -0.0044 | -1.1756 | -0.0044 | 0.0000 | 0.0044 | -1.1756 | 0.0044 | 0.7661 |
| 3.6 | -4.0285 | -0.0039 | -1.1776 | -0.0039 | 0.0000 | 0.0039 | -1.1776 | 0.0039 | 0.7681 |
| 3.65 | -4.0265 | -0.0035 | -1.1796 | -0.0035 | 0.0000 | 0.0035 | -1.1796 | 0.0035 | 0.7702 |
| 3.7 | -4.0245 | -0.0030 | -1.1816 | -0.0030 | 0.0000 | 0.0030 | -1.1816 | 0.0030 | 0.7721 |
| 3.75 | -4.0226 | -0.0027 | -1.1835 | -0.0027 | 0.0000 | 0.0027 | -1.1835 | 0.0027 | 0.7740 |
| 3.8 | -4.0208 | -0.0024 | -1.1854 | -0.0024 | 0.0000 | 0.0024 | -1.1854 | 0.0024 | 0.7759 |
| 3.85 | -4.0190 | -0.0021 | -1.1872 | -0.0021 | 0.0000 | 0.0021 | -1.1872 | 0.0021 | 0.7777 |
| 3.9 | -4.0172 | -0.0018 | -1.1889 | -0.0018 | 0.0000 | 0.0018 | -1.1889 | 0.0018 | 0.7795 |
| 3.95 | -4.0155 | -0.0016 | -1.1906 | -0.0016 | 0.0000 | 0.0016 | -1.1906 | 0.0016 | 0.7812 |

**Supplementary Table 2: The coefficients $h^i_\alpha(R)$ and $h^{ij}_{\alpha\beta}(R)$, determined using the PSI3 computational package [1]**

## Supplementary Methods

**Quantum eigenvector preparation algorithm**
Below we detail the steps involved in implementing the quantum variational eigensolver (QVE) algorithm.

1. Design a quantum circuit, controlled by a set of experimental parameters $\{\theta_i\}$, which can prepare a class of states. Using this device, prepare the initial state $|\psi^0\rangle$ and define the objective function $f(\{\theta_i^n\}) = \langle\psi(\{\theta_i^n\})|\,\mathcal{H}\,|\psi(\{\theta_i^n\})\rangle$, which efficiently maps the set of experimental parameters to the expectation value of the Hamiltonian and is computed in this work by QEE. $n$ denotes the current iteration of the algorithm.
2. Let $n = 0$
3. Repeat until optimization is completed
   (a) Call QEE with $\{\theta_i\}$ as input:
      i. Using the QPU, compute $\langle\sigma_\alpha^i\rangle$, $\langle\sigma_\alpha^i\sigma_\beta^j\rangle$, $\langle\sigma_\alpha^i\sigma_\beta^j\sigma_\gamma^k\rangle$, ..., on $|\psi^n\rangle$ for all terms of $\mathcal{H}$.
      ii. Classically sum on CPU the values from the QPU with their appropriate weights, $h$, to obtain $f(\{\theta_i^n\})$
   (b) Feed $f(\{\theta_i^n\})$ to the classical minimization algorithm (e.g. gradient descent or Nelder-Mead Simplex method), and allow it to determine $\{\theta_i^{n+1}\}$.

**Second Quantized Hamiltonian**
When taken with the Born-Oppenheimer approximation, the Hamiltonian of an electronic system can be generally written [2] as

$$\mathcal{H}(R) = \sum_{pq} h_{pq}(R)\hat{a}_p^\dagger\hat{a}_q + \sum_{pqrs} h_{pqrs}(R)\hat{a}_p^\dagger\hat{a}_q^\dagger\hat{a}_r\hat{a}_s \quad (1)$$

where $\hat{a}_i^\dagger$ and $\hat{a}_j$ are the fermionic creation and annihilation operators that act on the single particle basis functions chosen to represent the electronic system and obey the canonical anti-commutation relations $\{\hat{a}_i^\dagger, \hat{a}_j\} = \delta_{ij}$ and $\{\hat{a}_i, \hat{a}_j\} = \{\hat{a}_i^\dagger, \hat{a}_j^\dagger\} = 0$. $R$ is a vector representing the positions of the Nuclei in the system, and is fixed for any given geometry. The constants $h_{pq}(R)$ and $h_{pqrs}(R)$ are evaluated using an initial Hartree-Fock calculation and relate the second quantized Hamiltonian to the first quantized Hamiltonian. They are calculated as

$$h_{pq} = \int dr\ \chi_p(r)^* \left(-\frac{1}{2}\nabla^2 - \sum_\alpha \frac{Z_\alpha}{|r_\alpha - r|}\right)\chi_q(r) \quad (2)$$

$$h_{pqrs} = \int dr_1\ dr_2\ \frac{\chi_p(r_1)^*\chi_q(r_2)^*\chi_r(r_1)\chi_s(r_2)}{|r_1 - r_2|} \quad (3)$$

where $\chi_p(r)$ are single particle spin orbitals, $Z_\alpha$ is the nuclear charge, and $r_\alpha$ is the nuclear position. From the definition of the Hamiltonian, it is clear that the number of terms in the Hamiltonian is $O(N^4)$ in general, where $N$ is the number of single particle basis functions used. The map from the Fermionic algebra of the second quantized Hamiltonian to the distinguishable spin algebra of qubits is given by the Jordan-Wigner transformation [3], which for our purposes can be concisely written as

$$\hat{a}_j \to I^{\otimes j-1} \otimes \sigma_+ \otimes \sigma_z^{\otimes N-j} \quad (4)$$

$$\hat{a}_j^\dagger \to I^{\otimes j-1} \otimes \sigma_- \otimes \sigma_z^{\otimes N-j} \quad (5)$$

where $\sigma_+$ and $\sigma_-$ are the Pauli spin raising and lowering operators respectively. It is clear that this transformation does not increase the number of terms present in the Hamiltonian, it merely changes their form and the spaces on which they act. Thus the requirement that the Hamiltonian is a sum of polynomially many products of Pauli operators is satisfied. As a result, the expectation value of any second quantized chemistry Hamiltonian can be efficiently measured with our scheme.

For the specific case of He-H$^+$ in a minimal, STO-3G basis [2], it turns out that full configuration interaction (FCI) Hamiltonian has dimension four, thus a more compact representation is possible than in the general case. In this case, the FCI Hamiltonian can be written down for each geometry expanded in terms of the tensor products of two Pauli operators. Thus the Hamiltonian is given explicitly by an FCI calculation in the PSI3 computational package [1] and can be written as

$$\mathcal{H}(R) = \sum_{i\alpha} h_\alpha^i(R)\sigma_\alpha^i + \sum_{ij\alpha\beta} h_{\alpha\beta}^{ij}(R)\sigma_\alpha^i\sigma_\beta^j \quad (6)$$

The coefficients $h_\alpha^i(R)$ and $h_{\alpha\beta}^{ij}(R)$ are tabulated in Supplementary Table 2.

**Finding excited states**
Frequently, one may be interested in eigenvectors and

eigenvalues related to excited states (interior eigenvalues). Fortunately our scheme can be used with only minor modification to find these excited states by repeating the procedure on $\mathcal{H}_\lambda = (\mathcal{H} - \lambda)^2$. The folded spectrum method [4, 5] allows a variational method to converge to the eigenvector closest to the shift parameter $\lambda$. By scanning through a range of $\lambda$ values, one can recover the eigenvectors and eigenvalues of interest. Although this operation incurs a small polynomial overhead —the number of terms in the Hamiltonian is quadratic with respect to the original Hamiltonian— this extra cost is marginal compared to the cost of solving the problem classically.

**Application to $k-$sparse Hamiltonians**

The method described in the main body of this work may be applied to general $k-$sparse Hamiltonian matrices which are row-computable even when no efficient tensor decomposition is evident with only minor modification. A Hamiltonian $\mathcal{H}$ is referred to as $k-$sparse if there are at most $k$ non-zero elements in each row and column of the matrix and row computable if there is an efficient algorthim for finding the locations and values of the non-zero matrix elements in each row of $\mathcal{H}$.

Let $\mathcal{H}$ be a $2^n \times 2^n$ $k-$sparse row-computable Hamiltonian. A result by Berry et al. [6] shows that $\mathcal{H}$ may be decomposed as $\mathcal{H} = \sum_{j=1}^{m} \mathcal{H}_j$ with $m = 6k^2$, $\mathcal{H}_j$ being a $1-$sparse matrix and each $\mathcal{H}_j$ may be efficiently simulated ($e^{-i\mathcal{H}_j t}$ may be acted on the qubits) by making only $O(\log^* n)$ queries to the Hamiltonian $\mathcal{H}$. Alternatively, a more recent result by Childs et al. [7] shows that it is possible to use a star decomposition of the Hamiltonian such that $m = 6k$ and each $\mathcal{H}_j$ is now a galaxy which can be efficiently simulated using $O(k + \log^* N)$ queries to the Hamiltonian. Either of these schemes may be used to implement our algorithm efficiently for a general $k-$sparse matrix, and the choice may be allowed to depend on the particular setup available. Following a prescription by Knill et al. [8], the ability to simulate $\mathcal{H}_j$ is sufficient for efficient measurement of the expectation value $\langle \mathcal{H}_j \rangle$. After determining these values, one may proceed as before in the algorithm as outlined in the main text and use them to determine new parameters for the classical minimization.

**Computational Scaling**

In this section, we demonstrate the polynomial scaling of each iteration of our algorithm with respect to system size, and contrast that with the exponential scaling of the current best-known classical algorithm for the same task. Suppose that the algorithm has progressed to an iteration $j$ in which we have prepared a state vector $|\psi^j\rangle$ which is stored in $n$ qubits and parameterized by the set of parameters $\{\theta_i^j\}$.

We wish to find the average value of the Hamiltonian, $\langle \mathcal{H} \rangle$ on this state. We will assume that there are $M$ terms comprising the Hamiltonian, and assume that $M$ is polynomial in the size of the physical system of interest. Without loss of generality, we select a single term from the Hamiltonian, $\mathcal{H}_i$ that acts on $k$ bits of the state, and denote the average of this term by $\langle \mathcal{H}_i \rangle = h \langle \tilde{\sigma} \rangle$ where $h$ is a constant and $\tilde{\sigma}$ is the $k-$fold tensor product of Pauli operators acting on the system. As the expectation value of a tensor product of an arbitrary number of Pauli operators can be measured in constant time and the spectrum of each of these operators is bounded, if the desired precision on the value is given by $p$, we expect the cost of this estimation to be $O(|h|^2\ p^{-2})$ repetitions of the preparation and measurement procedure. Thus we estimate the cost of each function evaluation to be $O(|h_{\max}|^2 M\ p^{-2})$. For most modern classical minimization algorithms (including the Nelder-Mead simplex method [9]), the cost of a single update step, scales linearly or at worst polynomially in the number of parameters included in the minimization [10]. By assumption, the number of parameters in the set $\{\theta_i^j\}$, is polynomial in the system size. Thus the total cost per iteration is roughly given by $O(n^r |h_{\max}|^2\ M\ p^{-2})$ for some small constant $r$ which is determined by the encoding of the quantum state and the classical minimization method used.

Contrasting this to the situation where the entire algorithm is performed classically, a much different scaling results. Storage of the quantum state vector $|\psi^j\rangle$ using currently known exact encodings of quantum states, requires knowing $2^n$ complex numbers. Moreover, given this quantum state, the computation of the expectation value $\langle \tilde{\sigma} \rangle = \langle \psi^j | \tilde{\sigma} | \psi^j \rangle$ using modern methods requires $O(2^n)$ floating point operations. Thus a single function evaluation is expected to require exponential resources in both storage and computation when performed on a classical computer. Moreover, the number of parameters which a classical minimization algorithm must manipulate to represent this state exactly is $2^n$. Thus performing even a single minimization step to determine $|\psi^{j+1}\rangle$ requires an exponential number of function evaluations, each of which carries an exponential cost. One can roughly estimate the scaling of this procedure as $O(M\ 2^{n(r+1)})$

From this coarse analysis, we conclude that our algorithm attains an exponential advantage in the cost

of a single iteration over the best known classical algorithms, provided the assumptions on the Hamiltonian and quantum state are satisfied. While convergence to the final ground state must still respect the known complexity QMA-Complete complexity of this task [11], we believe this still demonstrates the value of our algorithm, especially in light of the limited quantum resource requirements.

**Computing the phase shifters' value to prepare an arbitrary 2-qubit pure state**

An arbitrary 2-qubit pure state can be parametrized in the following form

$$\begin{aligned}|\psi\rangle =& \cos(\tfrac{\theta_0}{2})\cos(\tfrac{\theta_1}{2})\,|0_1 0_2\rangle \\ &+ \cos(\tfrac{\theta_0}{2})\sin(\tfrac{\theta_1}{2})e^{i\omega_1}\,|0_1 1_2\rangle \\ &+ \sin(\tfrac{\theta_0}{2})e^{i\omega_0}\cos(\tfrac{\theta_2}{2})\,|1_1 0_2\rangle \\ &+ \sin(\tfrac{\theta_0}{2})e^{i\omega_0}\sin(\tfrac{\theta_2}{2})e^{i\omega_2}\,|1_1 1_2\rangle\,, \qquad (7)\end{aligned}$$

$$\theta_i \in [0,\pi],\ \ \omega_i \in [0,2\pi].$$

This parameterization is normalized, has the global phase removed and can be factorized as follows:

$$\begin{aligned}|\psi\rangle &= \cos\left(\tfrac{\theta_0}{2}\right)|0_1\rangle\,|T_0\rangle + e^{i\omega_0}\sin\left(\tfrac{\theta_0}{2}\right)|1_1\rangle\,|T_1\rangle\,, \qquad (8)\\ |T_0\rangle &= \cos\left(\tfrac{\theta_1}{2}\right)|0_2\rangle + e^{i\omega_1}\sin\left(\tfrac{\theta_1}{2}\right)|1_2\rangle\,,\\ |T_1\rangle &= \cos\left(\tfrac{\theta_2}{2}\right)|0_2\rangle + e^{i\omega_2}\sin\left(\tfrac{\theta_2}{2}\right)|1_2\rangle\,.\end{aligned}$$

First, we find the midpoint $|T_{mid}\rangle$ between the qubit states $|T_0\rangle$ and $|T_1\rangle$.

At this point it is possible to find the angles of the phase shifters $\phi_7$ and $\phi_8$:

$$\phi_7 = \frac{\pi}{2} + \arcsin\left(\sin(\theta_m)\sin(\omega_m)\right) \qquad (9)$$

$$\phi_8 = \pi - \arctan\left(\frac{\cos(\theta_m)}{\sin(\theta_m)\cos(\omega_m)}\right) \qquad (10)$$

The phase shifters $\phi_5$ and $\phi_6$ are not required for the state preparation. For computing the phase shifters' value, we set them to $\pi$ and $\pi$ respectively to implement the identity operation.

Next, we compute the transformation of the state $|\psi\rangle$ launched into the end of the chip and travelling backwards throughout the Mach-Zehnder interferometers identified by phase shifters $\phi_{5,6,7,8}$ and the CNOT gate. We obtain the state

$$\begin{aligned}|\psi'\rangle &= \cos\left(\tfrac{\theta_0}{2}\right)|0_1\rangle\,|T_0'\rangle + e^{i\omega_0}\sin\left(\tfrac{\theta_0}{2}\right)|1_1\rangle\,|T_1'\rangle\,, \qquad (11)\\ |T_0'\rangle &= e^{i\tau_0}\cos(\tfrac{\theta_p}{2})\,|0_2\rangle + e^{i(\omega_p+\tau_0)}\sin(\tfrac{\theta_p}{2})\,|1_2\rangle\,,\\ |T_1'\rangle &= e^{i\tau_1}\cos(\tfrac{\theta_p}{2})\,|0_2\rangle + e^{i(\omega_p+\tau_1)}\sin(\tfrac{\theta_p}{2})\,|1_2\rangle\,.\end{aligned}$$

Note that the correct description of our linear optical CNOT gate is

$$\begin{pmatrix} 0 & -i & 0 & 0 \\ -i & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix} \qquad (12)$$

From the parameterization of these states, the final four phase shifters ($\phi_{1-4}$) can be found:

$$\phi_1 = \theta_0 + \pi \qquad (13)$$

$$\phi_2 = \omega_0 + \tau_1 - \tau_0 - \pi \qquad (14)$$

$$\phi_3 = \theta_p + \pi, \qquad (15)$$

$$\phi_4 = \omega_p. \qquad (16)$$

# Supplementary References


[1] Crawford, T. D. *et al.* Psi3: An open-source ab initio electronic structure package. *J. Comp. Chem.* **28**, 1610–1616 (2007).

[2] Helgaker, T., Jorgensen, P. & Olsen, J. *Molecular Electronic Structure Theory* (Wiley, Sussex, 2002).

[3] Jordan, P. & Wigner, E. über das paulische äquivalenzverbot. *Zeitschrift für Physik* **47**, 631–651 (1928).

[4] MacDonald, J. K. L. On the modified ritz variation method. *Phys. Rev.* **46**, 828–828 (1934).

[5] Wang, L.-W. & Zunger, A. Solving schr[oumlaut]dinger's equation around a desired energy: Application to silicon quantum dots. *J. Chem. Phys.* **100**, 2394–2397 (1994).

[6] Berry, D., Ahokas, G., Cleve, R. & Sanders, B. Efficient quantum algorithms for simulating sparse hamiltonians. *Comm. Math. Phys.* **270**, 359–371 (2007).

[7] Childs, A. & Kothari, R. Simulating sparse hamiltonians with star decompositions. In Dam, W., Kendon, V. & Severini, S. (eds.) *Theory of Quantum Computation, Communication, and Cryptography*, vol. 6519 of *Lecture Notes in Computer Science*, 94–103 (Springer Berlin Heidelberg, 2011).

[8] Knill, E., Ortiz, G. & Somma, R. D. Optimal quantum measurements of expectation values of observables. *Phys. Rev. A* **75**, 012328 (2007).

[9] Nelder, J. A. & Mead, R. A simplex method for function minimization. *The Computer Journal* **7**, 308–313 (1965).

[10] Fletcher, R. *Practical methods of optimization* (Wiley-Interscience, New York, NY, USA, 1987), 2nd edn.

[11] Kempe, J., Kitaev, A. & Regev, O. The complexity of the local hamiltonian problem. *SIAM J. Comput.* **35**, 1070–1097 (2006).