

Supplementary data

How does relativity affect magnetically induced currents?

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All calculations have been calculated with the program ReSpect,¹ calculating both the magnetically induced currents and nuclear magnetic shielding constants at the relativistic four-component Kohn-Sham density functional level of theory using the Dirac-Coulomb Hamiltonian (DKS). For the shielding constant and current calculations, we apply the restricted magnetic balance condition and Gauge-Including Atomic Orbitals (GIAO) to improve basis set convergence and ensure gauge origin-independent results.^{2,3} We have used the uncontracted all-electron valence triple zeta basis set of Dyall for gold, mercury,⁴ and hydrogen⁵ in combination with the Perdew–Becke–Ernzerhof (PBE) functional.⁶ In the response calculations the non-collinear exchange-correlation kernel for a generalized gradient approximation functionals was employed.⁷ Automatic differentiation technique was employed in analytical calculation of exchange-correlation potential and kernel, as implemented in the XCFun library.⁸ The structure parameters (metal-hydrogen distances $r(\text{Au-H}) = 1.5351$ Å and $r(\text{Hg-H}) = 1.6565$ Å) have been determined with total energy scans, the molecular symmetry of HgH₂ was fixed at D_{∞h}.

For the numerical integration of the currents the magnetic field was set parallel to the z-axis and the atoms were aligned along the x-axis. Then perpendicular components of currents with respect to the integration plane were calculated on a rectangular grid (in the xz plane) starting 3 Å below the hydrogen atom in z direction and extending to 3 Å above the hydrogen atom. The grid extends as well in x direction by 6 Å (pointing away from the metal atom) and 120 x 120 points were calculated and summed up (no numerically significant differences upon extension as well in size as in number of points were found). This was done for AuH and HgH₂ both at full 4-component level of theory as well as at non-SOC level of theory. To remove SO effects from four-

component calculations we have done the following: omitting SO integrals in the four-component perturbation-free calculations and keeping all four-component operators in the response calculations. This procedure will result in a) removing “Passive spin-orbit effects” b) keeping “FC/SZ–KE” scalar but spin dependent terms c) partially retain “active spin-orbit effects” (for definition of quoted phrases see Ref. 9 and 10). Since “active spin-orbit effects” are negligible for light elements⁹ their otherwise undesirable partial presence in non-SO calculations is justified. The integral of the differences in currents (full 4-c and without SOC) were calculated as the difference of the integrals resulting from both procedures.

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