A revision of the value for the Cu nuclear quadrupole moment (NQM) is reported based on high-accuracy ab initio calculations on the Cu electric field gradients in the CuF and CuCl molecules. Electron-correlation effects have systematically been taken into account using a hierarchy of coupled-cluster methods including up to quadruple excitations. It is shown that the CCSD(T)$_A$ method provides a more reliable treatment of triples corrections for Cu electric-field gradients than the ubiquitously applied CCSD(T) method, which is tentatively attributed to the importance of the wavefunction relaxation in the calculations of a core property. Augmenting large-basis-set CCSD(T)$_A$ results with the remaining corrections obtained using additive schemes, including full triples contributions, quadruples contributions, zero-point vibrational corrections, spin-orbit corrections, as well as the correction from the Gaunt term, a new value of 209.7(50) mbarn for the Cu NQM has been obtained. The new value substantially reduces the uncertainty of this parameter in comparison to the standard value of 220(15) mbarn obtained from a previous muonic experiment.