The synthesis of position and momentum density matrices is a representation of the phase-space density. The density matrix contains the position density of the electrons. The bonding is reflected by strong off-diagonal features of the off-diagonal part of the first-order matrix $Y(r,r')$. In terms of their Fourier transform, the reciprocal form factor $B(q)$, the momentum densities $p(p)$ are related to the off-diagonal part of the first-order matrix $Y(r,r')$ by a projection. Coulson’s charge and bond-order matrix is an example of such a matrix, and generally chemical bonding is reflected by strong off-diagonal features of the density matrix of the electrons. The power of the interpretation of momentum densities by this concept can be demonstrated for the electronic structure of a number of molecules.

The density matrix contains the position density of the particles as its diagonal as well, and in fact the matrix is a representation of the phase-space density. Therefore, the challenging goal in electron density work, first formulated by Pauli in 1933 for the wave function, is the determination of the density matrix from experimental data. While previous work by Clinton, Massa, Frishberg, Pecora et al. is restricted to Hartree-Fock representable systems and is handicapped by slow iteration, a new approach combines full generality with favourable convergence properties. It is shown that thus the synthesis of position and momentum density data to a quantum mechanically correct unified description of systems has become feasible.