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In the last decade, convolution operators of matrix functions have received unusual attention due to their diverse applications. This monograph presents some new developments in the spectral theory of these operators. The setting is the L_p spaces of matrix-valued functions on locally compact groups. The focus is on the spectra and eigenspaces of convolution operators on these spaces, defined by matrix-valued measures. Among various spectral results, the L_2 -spectrum of such an operator is completely determined and as an application, the spectrum of a discrete Laplacian on a homogeneous graph is computed using this result. The contractivity properties of matrix convolution semigroups are studied and applications to harmonic functions on Lie groups and Riemannian symmetric spaces are discussed. An interesting feature is the presence of Jordan algebraic structures in matrix-harmonic functions.