This work applies aperture-coupled resonators (ACRs) to realize a high-selectivity frequency-selective surface (FSS) at 60 GHz. In a generic ACR FSS, one or both of electrical and magnetic coupling paths can, theoretically, be constructed by appropriately designing coupling apertures. To investigate the operating principle of the ACR FSS, an equivalent-circuit model is first given and analyzed using the odd- and even-mode method. A novel ACR FSS structure with dominant electrical coupling is then proposed. This FSS consists of two crossed-dipole resonator arrays and one rectangular coupling aperture array in between. The constructed out-of-phase signal paths cause two transmission zeros (TZs) near the skirts of the narrow passband, thereby considerably enhancing the selectivity. Parametric study of the proposed ACR FSS was performed. Benefiting from the symmetric structure and low profile, the proposed FSS exhibits good angular stability and polarization stability. A prototype of the proposed ACR FSS at 60 GHz was fabricated and characterized experimentally. The measured results agree well with the full-wave and circuit simulated results, thus verifying the FSS design.