Materials and device concepts for electrocaloric refrigeration

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Electrocaloric (EC) basic research is mostly focused on materials with a first-order phase transition providing a large EC temperature change ΔT_{EC}. However, near the Curie temperature T_C in the ferroelectric phase, the spontaneous and induced polarization contributions to the EC effect partially cancel each other out. Additionally, there is a peak of dielectric losses in this temperature region. Dielectric losses affect not only the heat balance, but they also make the temperature change of adiabatic heating larger than that of adiabatic cooling. This is not suitable for devices subjected to repeated thermodynamic cycles.

This work considers EC device operation above the temperature T_m of the dielectric permittivity peak $\varepsilon(T_m)$. The complex physics of ferroelectrics and relaxors close to T_m is replaced by considering a modified Curie-Weiss law predicting $\partial\varepsilon/\partial T$ as a function of four parameters: $\varepsilon(T_m)$, the intercept of the $\varepsilon(T)^{-1}$ vs T line with the T-axis, T_0, the parameter Ω characterizing the distribution of local Curie temperatures of materials which exhibit a diffuse phase transition (DPT), and the DPT diffuseness γ. The requirements to electrocaloric materials derived on this base ($\varepsilon(T_m) > 3000$, Ω = 20-50 K, γ = 1.2...2), are fulfilled by relaxor single crystals, relaxor ceramics and ferroelectric or relaxor thin films deposited by advanced technologies. Refrigerators using reverse Brayton, Ericsson and Stirling cycles are compared.

The field dependence of ΔT_{EC} is shown to be determined by the field dependence of ε. Perovskites with a nearly ideal close-packed structure possess field induced paralectric-ferroelectric (above T_C) or antiferroelectric-ferroelectric phase transitions. This results in a Clausius-Clapeyron contribution to ΔT_{EC} due to the latent heat of the field induced first-order phase transition. The critical field of field-induced phase transitions is in the order of a few V/µm. Therefore, the Clausius-Clapeyron contribution is of relevance only for bulk ceramic-based EC refrigerators.

Since the cooling power density dq/dt characterizes a thermal flux determined by the ratio of the temperature span to the total thermal resistance of the device, a comparable to bulk ceramics dq/dt may be realized also using thin film devices which are operated with higher cycle frequency employing the lower thin film thermal relaxation times.