

Comment on "Direct Measurement of the 'Giant' Adiabatic Temperature Change in $Gd_5Si_2Ge_2$ "

The authors of this paper [1] claim that the adiabatic temperature rise calculated from heat capacity data is much overestimated when compared to their direct measurements. Therefore, the validity of prior work [2], along with the use of Maxwell's relationship

$$(\partial M/\partial T)_H = (\partial S/\partial H)_T \quad (1)$$

for $Gd_5(Si_2Ge_2)$, was questioned by [1]. Unfortunately nonequilibrium direct ΔT_{ad} cannot be compared to the nearly equilibrium values obtained from the heat capacity measurements. It is well known that kinetic effects are intrinsic to any first-order transformation [3]. In the compound UNiAl displaying a first-order metamagnetic transition, we observed a cancellation of the intrinsic magnetocaloric effect due to kinetic effects [4]. Giguère *et al.* [1] measured ΔT_{ad} by moving the sample into a high magnetic field in a matter of a few seconds. However, when the magnetic field is ramped at a rate of about 2 T per min, the ΔT_{ad} is essentially identical to that calculated from the heat capacity (see Fig. 1). Figure 1 also illustrates that the results are sample dependent. Hence, ΔT_{ad} measured directly under near equilibrium conditions is in excellent agreement with the nearly equilibrium values calculated from heat capacity data collected on the same material.

Because of the discrepancy between their nonequilibrium measurements and the nearly equilibrium heat capacity data, the authors of Ref. [1] claim that Maxwell's relation does not apply but the Clausius-Clapeyron equation is perfectly valid. If their first point (i.e., a hidden parameter is responsible for the change in magnetization) holds, the Clausius-Clapeyron equation cannot be valid either. Therefore the only bona fide objection to the

applicability of Maxwell's relation at equilibrium would be nondefined derivatives in Eq. (1). This is the case only for an *ideal* first-order phase transition occurring infinitely fast at constant T , P , and H . All available experimental data, including Fig. 1 of Giguère *et al.* [1], point to the opposite: Near equilibrium, $(\partial M/\partial T)_H$ and $(\partial S/\partial H)_T$ are, indeed, well defined for $Gd_5(Si_2Ge_2)$. Thus there appears to be no need to use the Clausius-Clapeyron equation to account for the latent heat in the first-order phase transition. Additionally, the Clausius-Clapeyron relation would imply that the temperature step is independent of the field step, which is not observed in Fig. 3 of Giguère *et al.* [1].

Furthermore, the nature of the magnetic transition of $Gd_5Ge_2Si_2$ is quite different from the transition in FeRh. In $Gd_5Ge_2Si_2$ we deal with a transition from the paramagnetic to the ferromagnetic state in conjunction with a crystallographic transition which can be induced reversibly by a magnetic field, and the magnetization is a well-defined parameter. In FeRh, however, the transition is between the ferromagnetic and antiferromagnetic states, and in the antiferromagnetic state the staggered magnetization has to be considered as the relevant parameter. Finally, we note that $Gd_5(Si_2Ge_2)$ and the related $Gd_5(Si_{1-x}Ge_x)_4$ alloys are extremely complex and exciting materials which require much more detailed experimental and theoretical studies before their nature is fully understood.

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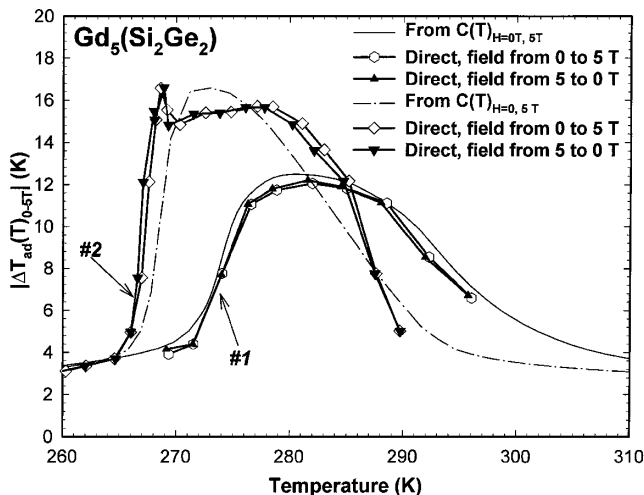


FIG. 1. The directly measured magnetocaloric effect in $Gd_5(Si_2Ge_2)$ for a magnetic field change from 0 to 5 T and 5 to 0 T and that calculated from heat capacity data measured in 0 and 5 T. The two samples (#1 and #2) were from different batches of the alloys with the same starting stoichiometry.

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