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High-resolution alternating-field technique to determine the magnetocaloric effect of metals down to very low temperatures

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The magnetocaloric effect or “magnetic Grüneisen ratio” $\Gamma_H = T^{-1}(dT/dH)_S$ quantifies the cooling or heating of a material when an applied magnetic field is changed under adiabatic conditions. Recently, this property has attracted considerable interest in the field of quantum criticality. Here, we report the development of a low-frequency alternating-field technique for measurements of the magnetocaloric effect down to very low temperatures, which is an important property for the study of quantum critical points. We focus, in particular, on highly conducting metallic samples and discuss the influence of eddy current heating. By comparison with magnetization and specific heat measurements, we demonstrate that our fast and accurate technique gives quantitatively correct values for the magnetocaloric effect under truly adiabatic conditions. © 2011 American Institute of Physics. [doi:10.1063/1.3529433]

I. INTRODUCTION

Understanding the phenomena close to quantum critical points (QCPs) is one of the major challenges in condensed matter physics. A QCP emerges when the characteristic temperature of a system, e.g., the Néel temperature, is suppressed to zero by variation of a nonthermal parameter r like pressure, magnetic field or doping. Upon pressure-tuning a system toward a QCP, the Grüneisen ratio Γ is expected to display singular behavior, namely a divergence toward zero temperature, while by tuning a system by magnetic field toward quantum criticality Γ_H , as defined below, diverges.^{1,2} Both Grüneisen parameters consist of ratios of the control-parameter and temperature derivatives of the entropy, which is accumulated close to the QCP (S , α , C , and M denote the entropy, volume thermal expansion, specific heat, and magnetization, respectively):

$$\Gamma = -\frac{1}{V_m T} \frac{(\partial S / \partial p)_T}{(\partial S / \partial T)_p} = \frac{\alpha}{C_p}, \quad (1)$$

$$\Gamma_H = -\frac{1}{T} \frac{(\partial S / \partial H)_T}{(\partial S / \partial T)_H} = -\frac{(\partial M / \partial T)}{C_H} = \frac{1}{T} \frac{dT}{dH} \Big|_S.$$

Indeed, both Γ and Γ_H have been experimentally shown to diverge in materials near QCPs.^{3–6} Since the magnetic field, in contrast to pressure or doping, can easily be tuned continuously *in situ*, the magnetic Grüneisen ratio is ideally suited to investigate the nature of QCPs. So far, the magnetic Grüneisen ratio close to QCPs has been determined only indirectly by calculation from magnetization and specific heat data.⁶ Note, that Γ_H can be determined by a single measurement of the magnetocaloric effect (MCE), $(dT/dH)_S$, as shown above. Very recently, the field dependence of the entropy close to a QCP has been calculated from highly nonadiabatic (quasi isothermal) $T(H)$ traces, separating the effect of heat flow from/to the bath and the magnetocaloric effect of the sam-

ple, using independent measurements of the thermal conductance between sample and bath.⁷ However, a more direct measurement of the MCE under truly adiabatic conditions is highly desired for comparison with theoretical scenarios. Below, we show that the alternating-field technique is suitable for this purpose. Previously, the spin-flip transition in insulating GdVO₄ has been investigated using such technique down to 0.5 K.⁸ In their study, not much care has been taken on the absolute values of the MCE and eddy current heating plays no role, since the material is an electrical insulator. Below, we report the application of the low-frequency alternating-field technique to clean metals and down to mK temperatures. We carefully investigate the effect of eddy current heating. Furthermore, we prove, by comparison with magnetization and specific heat measurements, that accurate absolute values of the adiabatic MCE are obtained.

II. EXPERIMENTAL SETUP

The definition of the MCE implies that a change of the sample temperature ΔT caused by a small change of the magnetic field ΔH under adiabatic conditions needs to be detected. Below, we demonstrate how the MCE could be detected directly by applying a low-frequency alternating field, $H_0 \sin(2\pi f t)$, and detecting the temperature oscillation of the sample with the same frequency, $T_0 \sin[2\pi f(t + \delta t)]$ (δt is a small delay due to the thermal resistance between the sample and thermometer).

Our setup for generating the alternating field is shown in Fig. 1. A SR830 Lock-In amplifier (Stanford Research Systems) is used to produce a current with low frequency (typically 0.01–0.1 Hz, the choice of the frequency will be discussed later). The current is amplified by a Kepco bipolar power amplifier up to 8 A and sent through a modulation coil (8 A current corresponds to 25 mT) which is inserted to the main superconducting magnet and cooled within the liquid 4-He. The current is determined by measuring the voltage across a shunt resistance (4 m Ω) with a Keithley 195A

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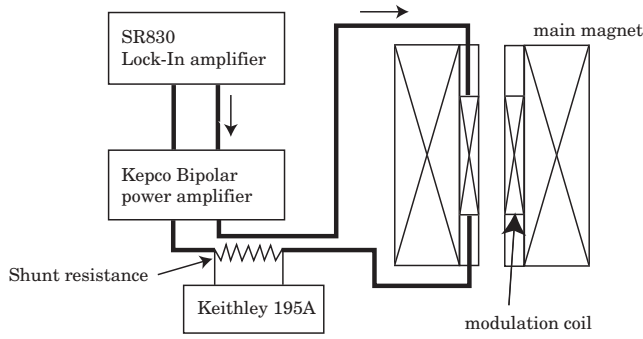


FIG. 1. Generation and control of the alternating magnetic field superposed to the large magnetic field of the main superconducting magnet.

multimeter. The modulation field is superimposed to the desired static magnetic field of interest, which is generated by a large superconducting magnet. Note that a finite static magnetic field is required since the MCE depends on the magnetization of the system [cf. Eq. (1)].

Figure 2 shows the setup to detect the sample temperature under quasiadiabatic conditions. The sample is glued on a thin sapphire plate and is cooled or heated through the weak thermal link (thin CuNi wire). If τ denotes the thermal relaxation through the link, the frequency of the field modulation must be adjusted such that $1/f \ll \tau$ in order to obtain effective adiabatic conditions. Such adjustment can easily be performed at several different temperatures by taking temperature traces with differing frequency and calculation of the MCE (see below). If the MCE is frequency independent, the measurements are performed under adiabatic conditions. The RuO_2 resistive thermometer is glued on the sample and its leads are made from superconducting filaments to avoid an additional heat leak. The sample temperature is measured using a LS370 resistance bridge (Lake Shore). The heat sink is weakly coupled to the mixing chamber and its temperature is proportional integral differential (PID) controlled with a heater within the field-compensated region of the large superconducting magnet. In a typical measurement of the temperature dependence of the MCE, we slowly sweep the temperature of the heat sink leading to a slow sweep of the average sample temperature. Due to the alternating magnetic field, the sample temperature oscillates through its average value. The amplitude of

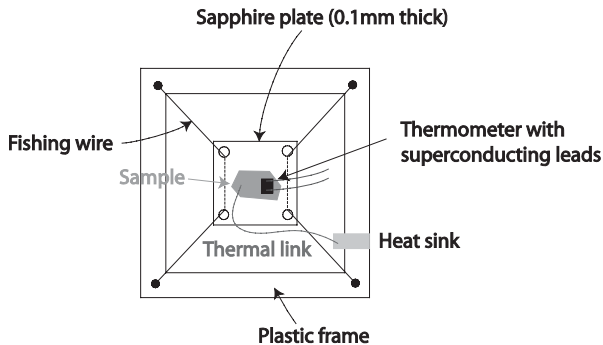


FIG. 2. Schematic view of the platform used for alternating field magnetocaloric effect measurements under quasiadiabatic conditions.

this oscillation gives the MCE, while the average value determines T .

III. APPLICATION TO STRONGLY CORRELATED MATERIALS

Figure 3 shows a typical example of a temperature sweep during an alternating field MCE measurement. We have chosen YbRh_2Si_2 as this is a system close to a QCP, at which a pronounced divergence of the magnetic Grüneisen ratio has been observed.⁶ The data discussed here are obtained on a slightly Fe-doped single crystal of 20.1 mg mass in a shape of plate with a thickness of 0.25 mm (residual resistivity ratio ~ 12).⁹ A relatively large piece of single crystal was chosen to determine precisely its magnetization and specific heat, which we will discuss later. The field was applied within the easy magnetic plane of the system (parallel to the plane of the sample plate), which is perpendicular to the tetragonal c -axis. The static field is set to 50 mT which is the critical field for undoped YbRh_2Si_2 and the alternating field has an amplitude of 4.5 mT with frequency of 0.1 Hz. A clear oscillation of the sample temperature due to the MCE is resolved. The inset of Fig. 3(a) displays the temperature variation on a larger time scale such that individual oscillations could not be resolved. Here the thickness of the line is a measure of the oscillation amplitude. Indeed the thickness of the $T(t)$ trace increases as temperature is decreased, indicating a larger temperature oscillation dT/dH with decreasing temperature. This is a clear evidence of quantum criticality in this material, since dT/dH is expected to decrease linearly with decreasing temperature for a noncritical material with constant magnetic Grüneisen ratio $\Gamma_H = \text{const}$.

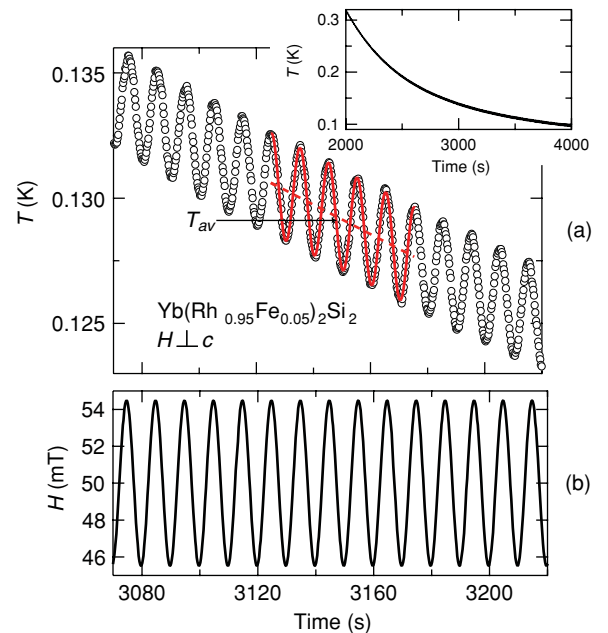


FIG. 3. (Color online) Sample temperature (a) and magnetic field (b) vs time for a measurement on doped YbRh_2Si_2 . The red solid and dotted lines show a fit to $T(t) = a_0 + a_1 t + a_2 t^2 + T_0 \sin[2\pi f(t - \delta t)]$ and its background term, $a_0 + a_1 t + a_2 t^2$, respectively. The inset displays the temperature variation of the sample over a longer time scale.

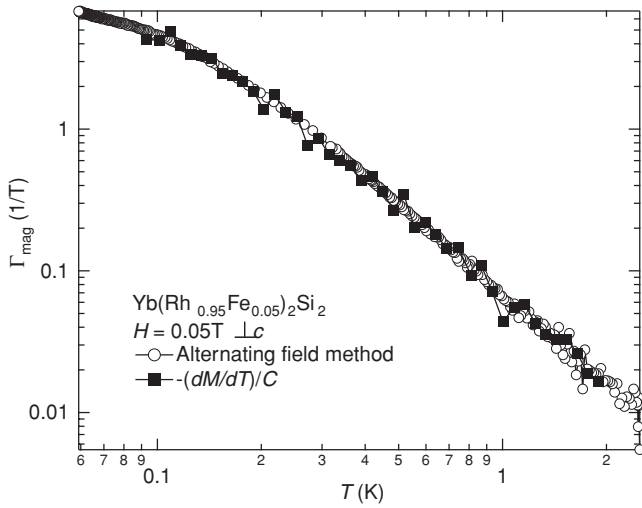


FIG. 4. Magnetic Grüneisen ratio for $\text{Yb}(\text{Rh}_{0.95}\text{Fe}_{0.05})_2\text{Si}_2$ as a function of temperature. The open circles are determined by the alternating field technique while the solid squares are calculated from magnetization and specific heat data using $-(dM/dT)/C$.

In order to quantify the MCE at a given average temperature T_{av} , we chose time intervals of five periods ($5 \times 1/f$ s in time) and determine the amplitude of the temperature oscillation by fitting the temperature trace to $T(t) = a_0 + a_1t + a_2t^2 + T_0 \sin[2\pi f(t - \delta t)]$ [red solid line in Fig. 3(a)], where f is the frequency of the field modulation $H(t) = H_{\text{av}} + H_0 \sin(2\pi ft)$. The second order polynomial in time [$a_0 + a_1t + a_2t^2$, cf. red dotted line in Fig. 3(a)] is added to the fitting function to take into account the background temperature drift due to the slow sweep of the heat sink temperature. The amplitude of the temperature oscillation T_0 divided by the field modulation amplitude H_0 gives the MCE. This value is associated to the average temperature T_{av} given by the background temperature at the center of the time interval, $T_{\text{av}} = a_0 + a_1(t_0 + 2.5/f) + a_2(t_0 + 2.5/f)^2$, where t_0 is the time of the first data point for fitting. The magnetic Grüneisen ratio is then calculated by $\Gamma_H(T_{\text{av}}) = 1/T_{\text{av}} \times T_0/H_0$. By shifting the fitting interval across the temperature trace, a dense and continuous curve of Γ_H is obtained, as shown in Fig. 4. In this measurement from 3 K down to 0.3 K, the temperature of the heat sink was slowly decreased, using PID control, and below 0.3 K the heater for the heat sink was turned off, since the thermal conductance of the thermal link is very small at low temperatures, and thus, the decrease of the average sample temperature is very slow. We have also performed a measurement with half value of the frequency ($f = 0.05$ Hz) and the same H_0 in the entire temperature range from 3 to 60 mK, using the same cooling rate of the heat sink, and confirmed that the result is exactly the same. This proves that the measurement is taken under effective adiabatic conditions ($1/f \ll \tau$; i.e., heat flow between sample and heat sink within one period is negligible.). The data are compared with $\Gamma_H(T)$ calculated using Eq. (1) from magnetization and specific heat data⁹ taken at the same field of 0.05 T and measured using the same piece of single crystal. The excellent agreement between the two independent ways to obtain the magnetic Grüneisen ratio prove that this tech-

nique is best suited to quantitatively obtain the true adiabatic MCE. Furthermore, the comparison shows the extraordinarily high resolution of the alternating field technique.

At last we discuss the problem of Joule heating caused by the field modulation. If the sample is very clean and the mean free path of the electrons is rather high, the frequency and the amplitude of alternating field have to be low to avoid Joule heating due to eddy currents. Figure 5 shows data obtained in the normal state of a high-quality single crystal of the clean heavy-fermion superconductor CeCoIn_5 in magnetic field applied parallel to c -axis. Note here that the material has a very large electronic mean free path of 810 Å (Ref. 10) and alternating field is applied perpendicular to the plane of a platelet sample ($1.3 \times 1.1 \times 0.33$ mm³) with a twice larger frequency of 0.2 Hz than the one used for Fe-doped YbRh_2Si_2 . This condition causes a severe problem with eddy current heating. As clearly seen in Fig. 5, the temperature of the sample oscillates with twice the frequency of the field modulation which is due to Joule heating. Eddy currents induced by the variation of a magnetic field are proportional to the time derivative of the field, $dH/dt = 2\pi f H_0 \cos(2\pi ft)$. Since eddy current heating is proportional to the square of the current, $(dH/dt)^2 \sim f^2 H_0^2 [1 + \cos(2\pi f t)]$, it results in an oscillation with $2f$ frequency. Note that the heating effect has two components; constant and $2f$ -oscillating ones. Figure 5 also shows that as soon as the alternating field is turned on, the average sample temperature raises immediately due to the constant heating component, followed by the $2f$ oscillation. We have also verified that the amplitude of $2f$ component increases rapidly with amplitude H_0 and frequency of the alternating field, whereas the $1f$ component due to the intrinsic MCE remains constant. The eddy current heating can very effectively be suppressed by a reduction of the cross section of the sample perpendicular to the field axis and as well as by reducing the frequency and/or amplitude of the field modulation.

At an elevated temperature, where the electrical conductivity is smaller, and therefore, the eddy current heating is less severe, a mixture of intrinsic $1f$ and extrinsic $2f$

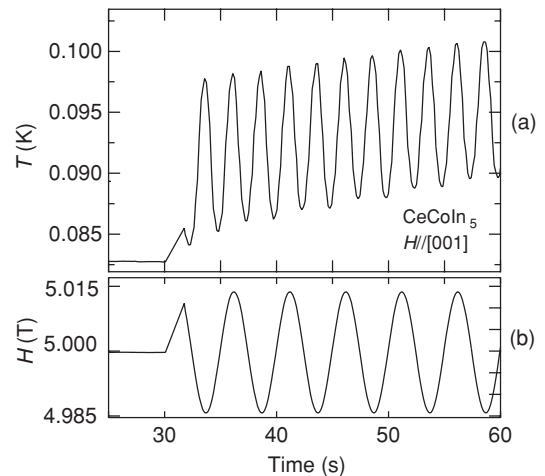


FIG. 5. Sample temperature (a) and magnetic field (b) vs time for a high-quality single crystal of CeCoIn_5 for $H \parallel [001]$. Note that the temperature oscillation has twice the frequency of the field modulation.

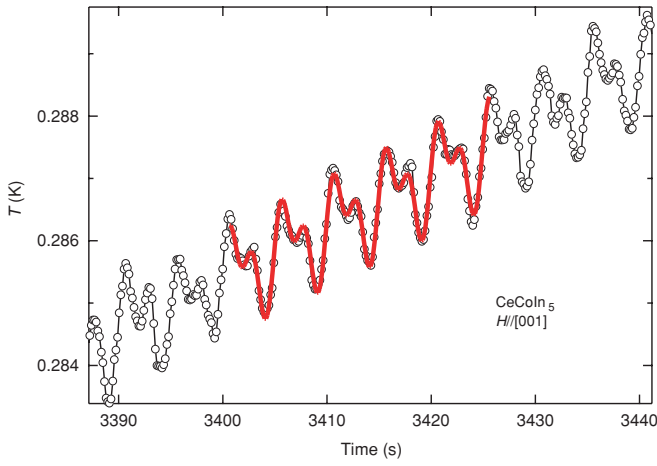


FIG. 6. (Color online) Mixture of $1f$ - and $2f$ -oscillations of sample temperature of CeCoIn_5 under the same condition as Fig. 5. The red line is a fit to $T(t) = a_0 + a_1 t + a_2 t^2 + T_0 \sin[2\pi f(t - \delta t)] + T_0^{2f} \cos[2\pi 2f(t - \delta t_{2f})]$.

oscillations can be found, as shown in Fig. 6. The temperature as a function of time is well fitted by a function with two oscillating components, $T(t) = a_0 + a_1 t + a_2 t^2 + T_0 \sin[2\pi f(t - \delta t)] + T_0^{2f} \cos[2\pi 2f(t - \delta t_{2f})]$ (red line in Fig. 6). We deduced T_0 , using several different frequencies but the same H_0 and found a constant T_0 . This indicates that the obtained T_0 is still valid under the influence of eddy current heating.

IV. SUMMARY

We developed a high-resolution low-temperature alternating-field technique for quasiadiabatic measurements of the MCE and magnetic Grüneisen ratio for clean metallic samples down to mK temperatures and in large magnetic fields. The performance check has revealed excellent

agreement with the magnetic Grüneisen ratio calculated by magnetization and specific heat measurements. The main advantages of the new technique compared to the latter are that it is much faster and easier and that its resolution is much higher. The capability of continuous temperature sweeps makes possible thorough investigations of the magnetic Grüneisen ratio in the entire H - T phase space, which is of particular interest for systems close to magnetic-field induced QCPs.

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