Attojoule Calorimetry of Mesoscopic Superconducting Loops

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We report the first experimental evidence of nontrivial thermal behavior of the simplest mesoscopic system—a superconducting loop. By measuring the specific heat C of an array of 450 000 noninteracting aluminum loops with very high accuracy of $\sim$20 fJ/K, we show that the loops go through a periodic sequence of phase transitions (with a period of an integer number of magnetic flux quanta) as the magnetic flux threading each loop is increased. The transitions are well described by the Ginzburg-Landau theory and are accompanied by discontinuities of C of only several thousands of Boltzmann constants k_B.

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Halfway between microscopic and macroscopic worlds, mesoscopic systems are known to exhibit a series of unique phenomena which disappear on smaller as well as on larger scales (see Refs. [1–3] for review): oscillations of transition temperature in thin superconducting cylinders [4], magnetic flux quantization [5], magnetoresistance oscillations [6], persistent currents [7], etc. The study of these phenomena is becoming increasingly important because mesoscopic, several nanometers in size, elements are the base of this century's electronics and are likely to revolutionize many areas of human activity, such as medicine, biotechnology, and information processing [2,3,8]. Although the electric transport in mesoscopic systems has been mainly studied during the past 20 years, the thermal transport has also attracted some attention very recently [9,10]. Yet, very little is known about the thermodynamic and thermal behavior of mesoscopic systems (i.e., behavior of their entropy, specific heat, etc.). Meanwhile, thermodynamics of nanostructured systems, new phase transitions intrinsic for them [11,12], the energies needed for their heating, the heat released when the system changes its state, will certainly be important in numerous future applications of nanoelectronic devices [8].

In the present Letter we report highly sensitive specific heat measurements performed on the simplest mesoscopic system—a superconducting loop of size comparable to the superconducting coherence length $\xi(T)$ in a magnetic field. We show that even this simple system exhibits behavior that differs substantially from that observed in macroscopic superconductors. More precisely, we observe multiple phase transitions between states with different numbers of magnetic vortices in the increasing magnetic field, accompanied by discontinuities of the specific heat as small as only a few thousands of k_B. These mesoscopic phase transitions are due to the entrance of superconducting vortices into the sample and have recently been observed in superconducting loops similar to ours by susceptibility measurements [13] and by Hall magnetometry [14,15]. Similar phenomena have been demonstrated to exist in mesoscopic disks [16,17].

Our sample is composed of 450 thousand nominally identical, noninteracting [18] aluminum square loops (size $= 2\ \mu$m, $w = 230$ nm arm width, $d = 40$ nm thickness, separation of neighboring loops $= 2\ \mu$m, total mass of the sample $m = 80$ ng), patterned by electron beam lithography on a suspended sensor composed of a very thin (4 μm thick) silicon membrane and two integrated

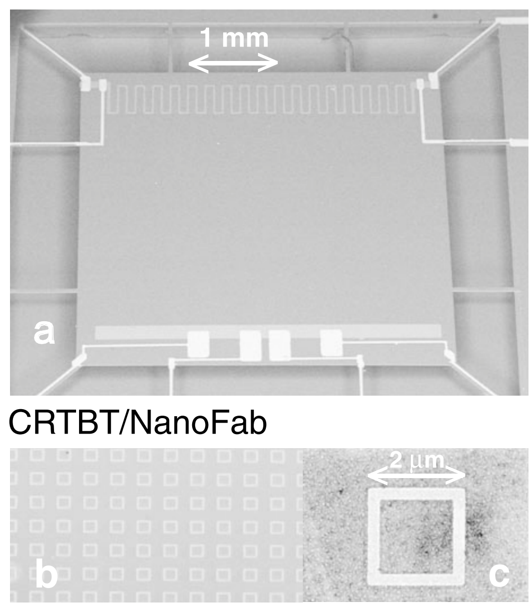

FIG. 1. (a) The suspended attojoule thermal sensor. The copper heater and the NbN thermometer can be distinguished in the upper and in the lower parts of the silicon membrane, respectively. The mesoscopic samples are deposited on the surface of the sensor between the heater and the thermometer by e-beam lithography. (b) Scanning electron microscope (SEM) image of the array of superconducting loops. (c) SEM image of a single aluminum superconducting loop.

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transducers: a copper heater and a niobium nitride thermometer [20] (see Fig. 1). The setup is cooled below the critical temperature T_c of the superconducting transition by a ^{3}He cryostat, and then its specific heat is measured by ac calorimetry. The technique of ac calorimetry consists in supplying ac power to the heater, thus inducing temperature oscillations of the thermally isolated membrane and thermometer [20]. For the operating frequency in the middle of the "adiabatic plateau" [20] (in our case, the frequency of temperature modulation is $f \simeq 250$ Hz), the temperature of the system "sensor + sample" follows variations of the supplied power in a quasiadiabatic manner, allowing measurements of the specific heat C with a resolution of $\delta C/C \gtrsim 5 \times 10^{-5}$ for signal integration times of the order of 1 min. This makes it possible to measure variations of the specific heat as small as 10 fJ/K (which corresponds to several thousands of k_B per loop), provided that the specific heat of the sensor (silicon membrane, heater, and thermometer) is reduced to about 10–100 pJ/K. Since the temperature oscillates with a typical amplitude of a few mK, our experimental apparatus can detect energy exchanges of only a few aJ at the lowest temperature of 0.6 K used in our experiments.

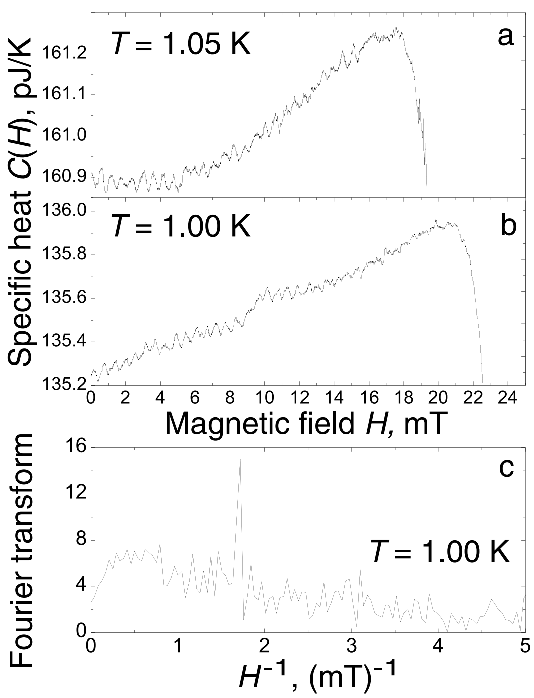

FIG. 2. Specific heat data at (a) $T = 1.05$ K and (b) $T = 1.00$ K. The right vertical axis shows the specific heat normalized by its jump ΔC at the critical field $H \sim 20$ mT where the transition to the normal state takes place (only a part of the jump is shown in the figure). For these measurements, the current through the heater is 4 μA and the current through the thermometer is 0.1 μA. The amplitude of temperature oscillations measured at the thermometer is about 5 mK. When the magnetic field is varied, clearly seen oscillations of C appear with periodicity of 0.58 mT. This corresponds to the quantum of magnetic flux Φ_0 threading a square of 1.89 μm side. (c) Fourier transform of the signal at $T = 1.00$ K. The peak at $1/H = 1.72$ mT^{-1} corresponds to the periodicity of 0.58 mT seen in the data.

In the absence of magnetic field, a discontinuity of the specific heat is observed at $T_c = 1.2 \pm 0.02$ K, corresponding to the superconducting transition of the sample. Once the sample is in the superconducting state, we keep the temperature constant and turn on the magnetic field $\mathbf{H}$ directed perpendicular to the loops' planes. The field is then varied very slowly (a typical run from 0 to 20 mT takes 10 h), and the specific heat C of the sample is measured for each value of H. Close to T_c (above $T = 0.93T_c$) C exhibits monotonic behavior with H without any detectable regular fine structure, whereas below $T = 0.93T_c$ we observe oscillations of $C(H)$ with a period of approximately 0.58 mT (see Fig. 2). The field $H \sim 20$ mT destroys superconductivity and produces a large jump ΔC of the specific heat. The periodic character of specific heat variations is even more evident when looking at the Fourier transform of the data [see Fig. 2(c)] exhibiting a well-defined peak at $1/H \simeq 1.72$ mT^{-1}. The observed period of oscillations of 0.58 mT corresponds to the magnetic flux quantum $\Phi_0 = 2.07 \times 10^{-15}$ Wb through a square of 1.89 μm side, which is roughly the size of our loops. At the same time, the amplitude of oscillations $\sim$50 fJ/K corresponds to specific heat variations of $\sim$0.1 aJ/K ($\simeq 7500 k_B$) per loop. A similar, periodic with the external magnetic field H, behavior of the magnetization M of superconducting loops and disks has been recently reported in Refs. [15,16]. From these measurements, oscillations of C versus H can be anticipated by using the Maxwell relation: $\partial C/\partial H \propto \partial^2 M/\partial T^2$, suggesting that periodicity of C with H is a signature of the entrance of vortices into the mesoscopic sample.

A detailed description of the oscillations of C with magnetic field can be obtained in the framework of the Ginzburg-Landau (GL) theory of superconductivity [21]. We consider a circular loop of the same average perimeter as the actual square loop and assume the GL coherence length at zero temperature $\xi(0) \simeq 0.15$ μm, corresponding to the mean free path $\ell \simeq 20$ nm measured independently. At a given magnetic flux Φ threading the loop, the superconducting order parameter takes the form $\psi(\mathbf{r}) = f_n(\rho) \times \exp(in\phi)$, where the vorticity n is a number of "giant" magnetic vortices in the loop and we use cylindrical coordinates with the z axis perpendicular to the plane of the loop. The specific heat of the loop $C = -T(\partial^2 F[\psi(\mathbf{r})]/\partial T^2)$, where $F[\psi(\mathbf{r})]$ is the GL free-energy functional, is shown in Fig. 3(a) by dashed lines for different values of vorticity n and for $T = 0.95T_c$. As follows from the figure, several values of vorticity are possible at a given value of Φ. In the thermodynamic equilibrium, the transitions between the states with vorticities n and $n + 1$ (so-called "giant vortex states") occur at $\Phi = (n + 1/2)\Phi_0$, which minimizes the free energy and makes the specific heat to follow the lowest of curves corresponding to different n in Fig. 3. This results in oscillations of

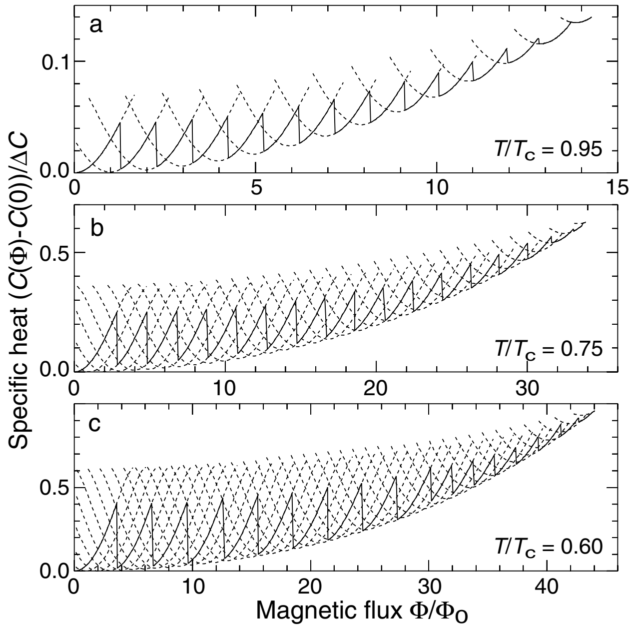

FIG. 3. Specific heat of a superconducting loop computed from the numerical solution of the Ginzburg-Landau equation in the increasing magnetic field and for three different temperatures (solid lines). Dashed lines correspond to states with constant vorticities ($n = 0, 1, 2, \ldots$, from left to right). Φ is the magnetic flux threading the loop, Φ_0 is the magnetic flux quantum, the curves are normalized by the discontinuity ΔC of specific heat at the superconducting-to-normal transition that takes place at (a) $\Phi/\Phi_0 \simeq 14.5$, (b) $\Phi/\Phi_0 \simeq 34.0$, and (c) $\Phi/\Phi_0 \simeq 44.0$.

$C(\Phi)$ with a period of Φ_0, in agreement with the experiment. The amplitude of oscillations decreases when T approaches T_c, and hence the oscillations are likely to be masked by noise at T close to T_c, which explains why we did not succeed to observe any regular fine structure in C at $T > 0.93T_c$.

It is, however, not clear if during the experiment the superconducting loops always remain in the thermodynamic equilibrium as the magnetic field is varied. Once the flux Φ surpasses $(n + 1/2)\Phi_0$, the giant vortex state n becomes metastable and, although a state with lower energy ($n + 1$ state) exists, the latter is separated from the former by an energy barrier that can take a long time for the system to overcome [13,17]. As Φ is further increased, the height of the barrier decreases and finally the barrier vanishes. The state n then becomes unstable and one more giant vortex enters into the loop. To model this situation, we solve the GL equation for $\psi(\mathbf{r})$ by increasing the flux Φ in small steps and using the previous-step solution as a starting point of our relaxation-type algorithm. The resulting dependence of the specific heat on the flux exhibits discontinuities at values of Φ where giant vortices enter into the loop. The discontinuities are separated by $\Delta\Phi = \Phi_0$ and the curve $C(\Phi)$ has a characteristic asymmetric triangular shape [solid line in Fig. 3(a)]. Since no unambiguous signature of such a shape is observed in the experimental data of Fig. 2, we conclude that at not too low temperatures (at least, above $T = 0.9$ K) our system always remains close to the thermodynamic equilibrium. This is probably due to the electromagnetic and thermal noises which provide sufficient energy for the system to overcome energy barriers between states with different vorticities well before one of the states becomes unstable.

The maximum number of giant vortices that can be hosted by a loop of radius R and arm width $w \lesssim \xi(T)$ can be found to be $n_{\rm max} \simeq \sqrt{3}R^2/w\xi(T)$, yielding the field $H_{\rm max}$ at which oscillations of C should stop: $H_{\rm max} \simeq \sqrt{3}\Phi_0/\pi w\xi(T)$. Our experiments show that the loop can remain superconducting up to H slightly exceeding $H_{\rm max}$, but the periodicity of C with H is lost beyond $H_{\rm max}$.

To study the behavior of superconducting loops at lower temperatures [and hence at smaller coherence lengths $\xi(T)$], we performed the measurements of the specific heat at $T = 0.85$ and 0.7 K (see Fig. 4). In contrast to Fig. 2, discontinuities of C are now clearly seen, testifying that the loops do not remain in thermodynamic equilibrium and explore metastable states. Another difference with Fig. 2 is that the periodicity of the signal is now a multiple of Φ_0: $\Delta\Phi = 2\Phi_0$ at $T = 0.85$ K and $\Delta\Phi = 3\Phi_0$ at $T = 0.7$ K. Such "multiple flux jumps" or "flux ava-

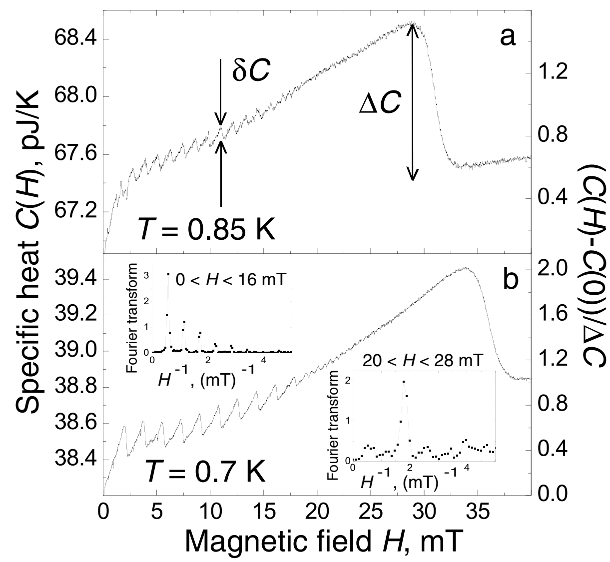

FIG. 4. Specific heat data at lower temperatures: (a) $T = 0.85$ K and (b) $T = 0.7$ K. The right vertical axis shows the specific heat normalized by its jump ΔC at the critical field where the transition to the normal state takes place. At $T = 0.85$ K, the periodicity of 2×0.58 mT ($2\Phi_0$ in terms of magnetic flux) is observed, whereas at $T = 0.7$ K (and at lower temperatures, down to $T = 0.6$ K) the periodicity of 3×0.58 mT ($3\Phi_0$) dominates. The insets show the Fourier transforms of the data for $0 < H < 16$ mT and $20 < H < 28$ mT with dominant components peaked at $1/H = 0.57$ mT^{-1} and $1/H = 1.72$ mT^{-1}, respectively. Note the existence of higher harmonics due to the triangular shape of the signal at low field.

lanches'' are signatures of simultaneous entrances of several (2 or 3) giant vortices into the loop and can be considered as nonequilibrium phase transitions between states with n and $m > n + 1$ giant vortices. Such transitions are also predicted by the GL theory [Figs. 3(b) and 3(c)] and have been recently observed in mesoscopic systems similar to ours by using the ballistic Hall micromagnetometry [14–16]. Note that although the observed jumps of specific heat are rather sharp, their width $\delta\Phi \sim 0.1\Phi_0$ is finite. This is due to slightly fluctuating geometric parameters of the loops, which therefore exhibit transitions between different giant vortex states at slightly different values of the applied magnetic field. Mutual inductance effects are expected to increase $\delta\Phi$ [19]. It is worthwhile to note that the discontinuities δC of C following from the theory are systematically larger than the measured ones, but both the theory and the experiment yield very similar results for δC normalized by the large discontinuity ΔC observed at the transition of the sample to the normal state (see Fig. 4 for the definitions of δC and ΔC). $\delta C/\Delta C$ is typically several percent for T close to T_c, but increases to $\sim$25% for $T = 0.7$ K.

The data of Fig. 4 have been obtained for the magnetic field sweep rate of 2 mT/h, a 10 times faster run yielded identical results. However, for a 10 times slower field sweep, the finite lifetime of metastable states starts to play an important role, leading to a more complicated behavior of $C(H)$ (alternating Φ_0, $2\Phi_0$, and $3\Phi_0$ jumps), which is a subject of ongoing study. The multiple flux jumps can be understood by studying the stability of the state with vorticity n with respect to an admixture of a state with a different vorticity $m > n$. In the increasing magnetic field, the instability first occurs for $m \simeq n + R/\sqrt{2}\xi(T)$, favoring typical flux jumps $m - n = 1$ for T close to T_c, when $\xi(T)$ is large and $R/\xi(T) \sim 1$, and $m - n > 1$ at lower temperatures, when $\xi(T)$ decreases and $R/\xi(T) > 1$. The multiple flux jumps are observed only in magnetic fields below $H \simeq 18$ mT, whereas at larger fields and up to H_{max} the single flux jumps are recovered [see the insets of Fig. 4(b)], in agreement with magnetic measurements [14]. Theoretical curves also show the tendency to smaller periodicity [$\Delta\Phi = 2\Phi_0$ instead of $\Delta\Phi = 3\Phi_0$ in Fig. 3(c)] at high magnetic fields.

In conclusion, the specific heat of mesoscopic superconducting loops is an oscillatory function of the external magnetic field and exhibits discontinuities (jumps) at the vortex entrance. Vortices enter an isolated loop one, two, or three at a time, depending on the temperature at which the experiment is performed. Definitely, the possibility of measuring the specific heat of nanoscale objects with very high accuracy opens quite interesting prospects in the field of mesoscale and nanoscale thermodynamics of superconducting as well as normal materials.

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