Magnetothermal instability in the organic layered superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$

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We have studied the magnetothermal instability in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ by means of measuring the temperature change in the sample. We report the remarkable temperature spikes due to the flux jumps and the preceding temperature oscillations in the vortex solid state. These dependences on the sweep rate are almost consistent with the conventional theory based on the Bean model. The temperature dependences of the flux jump suggest that the melting transition from the vortex solid to the liquid phase at low temperatures is not driven by thermal fluctuations, but rather by quantum ones. The small temperature fluctuations observed at very low temperatures imply the dendritic vortex distribution in this salt.

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I. INTRODUCTION

The magnetothermal instability in type II superconductors has been extensively studied because the resulting flux jumps are undesirable in applications of superconductors, e.g., superconducting magnets.[1] The observed flux jumps in conventional superconductors are successfully described in the framework of Bean's critical-state model.[2] The flux jumps have been also reported in some layered superconductors, such as high T_c superconducting cuprates and organic superconductors. These highly anisotropic systems show an exotic vortex phase diagram, where the fluctuations play an important role because of their short coherence length along the stacking direction compared to the interlayer spacing.[3] In organic layered superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, a three-dimensional vortex lattice exists below 7 mT, and two-dimensional vortex solids in adjacent layers are weakly interacting above this field.[4–6] In this vortex solid state, the flux jumps have been reported as abrupt jumps of the magnetization,[7,8] though the experimental data are not enough to compare with the conventional critical-state model. Recently, it has been suggested that the vortex liquid state exists even at very low temperatures resulting from quantum fluctuations instead of thermal ones,[8,9] and the resulting liquid state is a quantum vortex slush state where a short-range order of vortices remains.[9,10]

In this paper, we present the flux jump as the remarkable temperature change in the sample and preceding temperature oscillation in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. We have measured the dependences of the flux jumps on sweep rate, field angle, and temperature to clarify the magnetothermal instability and the vortex states in this salt.

II. EXPERIMENT

A single crystal of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ was obtained by the electrochemical oxidation method.[11] The crystal has the approximate dimensions of $2\times0.5\times0.1$ mm^3 with the weight of 0.21 mg. The sample was directly attached to a small RuO_2 temperature sensor with Apiezon-N grease, and mounted inside a vacuum chamber. The weak thermal link between the sample and the bath was achieved by the wires of the thermometer. The thermal relaxation time between the sample and the bath is about 1 s. The sample temperature was monitored by the thermometer during the magnetic field sweeps, as in the case of magnetocaloric measurements. The measurements were carried out by using a dilution refrigerator with a superconducting magnet at NIMS and ISSP.

III. RESULTS AND DISCUSSION

The temperature change in the sample as a function of external magnetic field B_e applied perpendicular to the conducting bc layers is shown in Fig. 1. The magnetic field was swept at a constant rate of 0.2 T/min, in a constant bath temperature of 150 mK. The sample temperature in zero magnetic field is the same as the bath temperature. As the field increases, the sample temperature rapidly goes up to at least 2 K, and then shows successive spikes as shown in the inset of Fig. 1. This remarkable behavior is reminiscent of the successive jumps of the magnetization observed in the

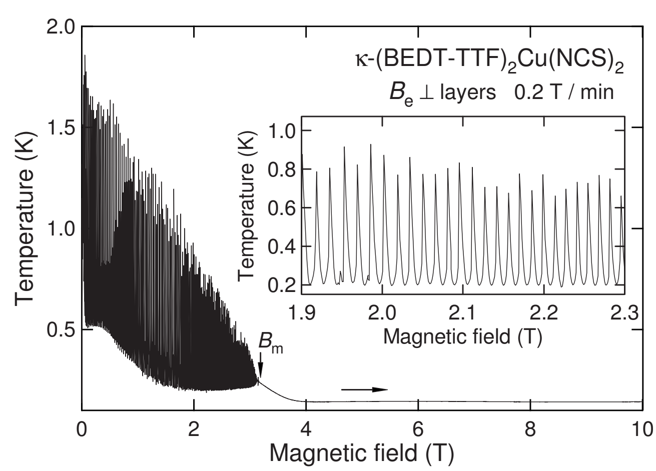

FIG. 1. Temperature variation in the sample as a function of magnetic field applied perpendicular to the conducting bc layers. Inset shows the enlarged view.

torque measurements,[7,8] which are understood as "flux jumps" in the vortex solid state. The temperature spikes observed here are also caused by the flux jumps, as discussed below.

When an external field exceeds the lower critical field of the type II superconductors, magnetic fluxes penetrate the sample in the form of vortices. The vortices are pushed deeper into the sample by the Meissner shielding currents which act on the vortices as a Lorentz-type force. On the other hand, the crystal defects collectively pin the vortex lattice in the vortex solid state. Consequently, as the magnetic field increases, the vortices build up near the sample surface and cause the field gradient near the sample edge and associated surface currents. The local balance between the Lorentz and pinning force creates a metastable equilibrium state, the so-called critical state. In Bean's critical-state model, the field gradient in the sample equals the critical current density J_c. In the absence of effective cooling, the viscous flow of the vortices causes a local heating, which reduces the critical current density, and then allows the deeper flux penetration inside the sample, which in turn leads to an additional heating. Such a positive feedback results in a runaway thermal instability. As a result, the macroscopic regions of the sample become metallic, and then the fluxes rapidly flow into the crystal. This catastrophic behavior is known as a "flux jump." After a flux jump, the crystal rapidly cools, and again becomes superconducting with a slightly different vortex distribution, and then the same process repeats again and again. Thus, the temperature spike observed in this study corresponds to the heating of the sample due to the flux jump. Indeed, the temperature spikes in conjunction with magnetization jumps have been reported in $YBa_2Cu_3O_7$ (Ref. 12) and also in organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (Ref. 13) with an amplitude of about 10 K and 20 mK, respectively. It is difficult to measure the sample temperature at the flux jump exactly because the flux jump phenomena are very fast. However, at least the temperature of some region of the sample goes up to the superconducting transition temperature at the flux jump.

As shown in Fig. 1, the amplitude of the temperature jump becomes smaller as the field increases, and then becomes zero at about 3.1 T denoted as B_m. According to the previous magnetization measurements, the superconducting state persists up to about 6 T in perpendicular field. Because the flux jumps are observed only in the vortex solid state where the vortices are collectively pinned, we define the melting field of the vortex solid as B_m. In a static magnetic field, there are no flux jumps and the system is always in the vortex solid state below B_m. After entering the vortex liquid state, the sample temperature gradually relaxes to the bath temperature. There is no temperature change at the phase boundary of the vortex liquid and the normal state.

Figure 2 shows the data at a slower sweep rate of 0.1 T/min, and the inset shows the expanded view of the flux jumps. In addition to the temperature spikes, we notice the clear temperature oscillation just before each spike, which is periodic in time. Such oscillations are not clear above 0.8 T because the temperature spikes occur more frequently in high field. A quite similar temperature oscillation has been observed in $YBa_2Cu_3O_7$,[12] while it has not been reported in

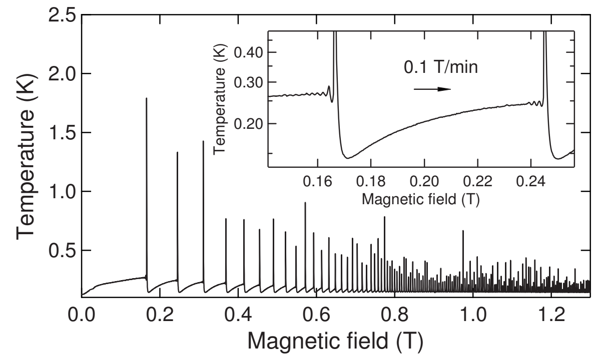

FIG. 2. Temperature variation in the sample at 0.1 T/min. In the inset, the temperature oscillations are clearly seen just before each temperature spike.

organic superconductors so far. This oscillatory behavior can be reproduced by analyzing the development of the temperature change in a superconductor using the heat diffusion equation and the Maxwell equation.[1] Physically, it is caused by the fact that the additional heating in sweeping magnetic field causes the temperature rise in the sample ΔT, but also causes additional heat transfer to the bath, which is again proportional to ΔT. Thus, we can expect an oscillatory behavior for a certain condition.

The flux jumps and the temperature oscillations were measured at various sweep rates $\dot{B}_e$ at constant bath temperature of 150 mK. Figure 3 shows the sweep rate dependence of the magnetic field B_j where the first flux jump occurs (filled circles, left axis) and the frequency of the temperature oscillations ω (open circles, right axis). The effect of the sweep rate analyzed theoretically by Mints[14] based on the Bean model are described as follows:

$$B_j = \sqrt{\frac{2\mu_0^2 j_c(T_0) h (T_c - T_0)}{n \dot{B}_e}} \propto \frac{1}{\dot{B}_e^{1/2}}, \tag{1}$$

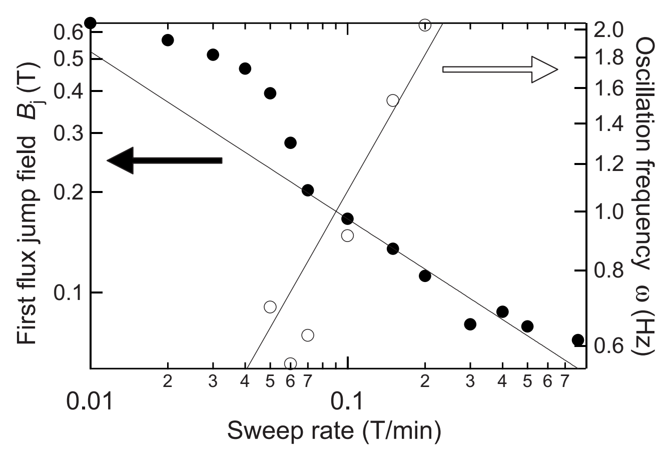

FIG. 3. Sweep rate dependence of the magnetic field where the first flux jump occurs (filled circles, left axis), and the oscillation frequency of the temperatures (open circles, right axis). The solid lines are the fit to the Mints theory.

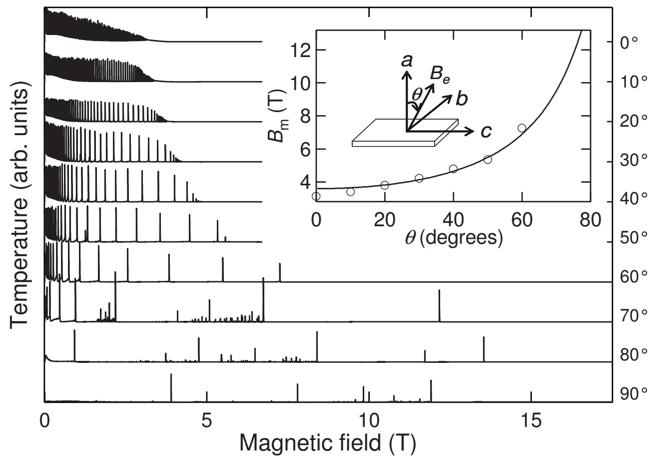

FIG. 4. Flux jumps measured at various field angles. The data are shifted for clarity. Inset shows the field angle dependence of the melting field B_m. The B_m near parallel field is not shown because of large error bars. The solid line is a fit to the 2D Ginzburg-Landau formula.

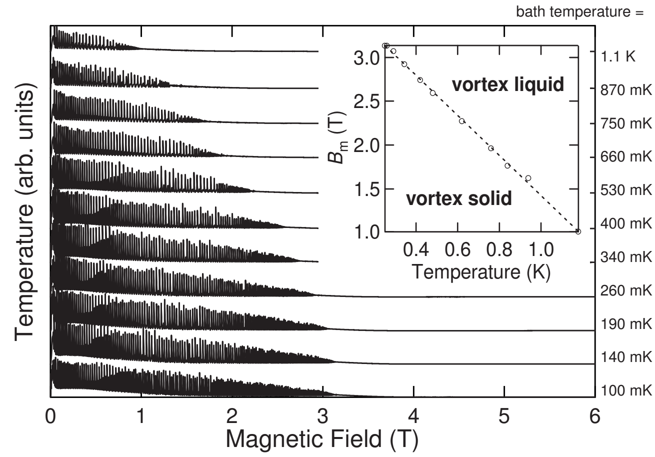

FIG. 5. Flux jumps measured at different bath temperatures. The data are shifted for clarity. Inset shows the temperature dependence of the melting field.

$$\omega = \left(\frac{2n^3 \dot{B}_e^3 h}{\mu_0^2 j_c d^2 C^2 (T_c - T_0)} \right)^{1/4} \propto \dot{B}_e^{3/4}, \qquad (2)$$

where T_0 is the sample temperature, T_c is the critical temperature, C is the heat capacity, h is the heat transfer coefficient, j_c is the critical current density, and $n = j_c/j_1$ (j_1 is the slope of the j-E curve, $j_1 \ll j_c$). These dependences come from the nonlinear conductivity which strongly depends on $\dot{B}_e$.[14] The observed dependences on sweep rate are almost consistent with the theory, as plotted by the solid lines in the figure. However, in slow sweeps, the critical state is more stabilized against flux jumps than expected from the model. In the $Bi_2Sr_2CaCu_2O_{8+\delta}$ thin films, a similar deviation from the theory has been reported, where the effect of the sample geometry characterized by a large demagnetizing factor is discussed.[15] Such a large demagnetizing factor, which is disregarded in the theory, can significantly alter the distribution of the screening currents. The strong two dimensionality in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ possibly results in the same situation.

Figure 4 shows the flux jumps at various field angles θ, where θ is the angle between the magnetic field and a direction normal to the conducting bc layers. When the magnetic field is tilted away from the normal to the layers, the melting field B_m shifts to larger field, and the flux jumps become less frequent. It makes it difficult to define B_m near the parallel field due to the large error bar. As shown by the solid line in the inset of Fig. 4, the field angle dependence of the melting field B_m is well fitted by the two-dimensional (2D) Ginzburg-Landau theory,

$$\left| \frac{B_m(\theta)\cos\theta}{B_{m\perp}} \right| + \left[\frac{B_m(\theta)\sin\theta}{B_{m\parallel}} \right]^2 = 1,$$

with $B_{m\perp} = 3.6$ T and $B_{m\parallel} = 29$ T, where $B_{m\perp}$ and $B_{m\parallel}$ denote the melting field for the external field $B_e \perp bc$ and $B_e \parallel bc$, respectively. The result is almost consistent with the previous report.[8]

Figure 5 shows the flux jumps measured at different bath temperatures. The temperature spikes become smaller as the field increases, and then stop at the melting field B_m at each temperature. The inset shows the temperature dependence of the B_m, which is almost linear below 1 K. On the other hand, in a quasi-two-dimensional system where the vortices in adjacent layers are weakly interacting, the melting transition induced by thermal fluctuations is described as $B_m \propto \exp(a/T)$, where a is a parameter containing the Lindemann number.[16] Accordingly, the observed temperature dependence suggests that the melting transition at low temperatures is not driven by thermal fluctuations, but rather by quantum ones, as pointed out previously.[9] The estimated melting field at $T = 0$ K by extrapolating our data is about 4 T. This value is almost the same as the quantum melting field calculated by Sasaki.[9,17] It is suggested from the resistivity and recent NMR measurements that the liquid state in the low-temperature region is the quantum vortex slush state where some sort of vortex solid remains.[9,10] However, it is difficult to discuss the nature of the liquid state from our results.

Finally, we discuss the possibility of the fractal structure of the vortex distribution in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. In some superconducting films including Nb, MgB_2, and $YBa_2Cu_3O_7$, the dendritic flux penetration has been proved by magneto-optical imaging.[18–21] In these materials, the magnetization shows noiselike small jumps originate from abrupt penetrations of dendritic flux structure.[19,22] It is known that such structure is observed only in the low-temperature region in the superconducting state, while the origin of this structure is not clear yet.

Recently, Mola *et al.*[23] reported the small and random magnetization jumps in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ at very low temperatures below 100 mK, and pointed out the possibility of the fractal flux distribution in this salt. On the other hand, we also notice the small temperature fluctuations in a slowly changing magnetic field below about 200 mK which qualitatively differ from the conventional flux jumps. The

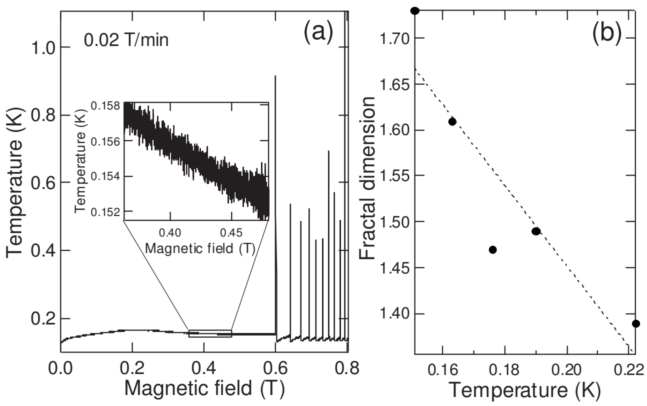

FIG. 6. (a) Flux jumps measured at 0.02 T/min. Inset shows the enlarged view of the temperature fluctuations. (b) Fractal dimension as a function of temperature. The dashed line is a guide for the eyes.

typical data are shown in the inset of Fig. 6(a). The temperature fluctuations are sufficiently larger than the experimental error (~0.3 mK), and become smaller as the temperature increases. To quantify this structure, we applied a fractal analysis with box counting algorithm,[24] and obtained the fractal dimension as a measure of complexity.[25] As shown in Fig. 6(b), the fractal dimension decreases as the temperature increases, qualitatively consistent with the results by Mola *et al.*[23] It is difficult to compare the flux jump phenomena in different experimental conditions quantitatively because it largely depends on the thermal link between the sample and the bath, the cooling power of the bath, and the sample shape. However, we suppose that the temperature fluctuations in our results and small magnetization jumps reported by Mola have the same origin, because (1) the characteristic features of the signal are quite similar to each other (small and random jumps in temperature or in magnetization); (2) the temperature dependence of the fractal dimension is qualitatively the same in both; and (3) these structure are observed only in the low-temperature region. It is consistent with the fact that dendritic structures appear only at low temperatures in the superconducting state.

In general, a dendritic structure has been reported in thin-film superconductors, but not in bulk samples. It is probably associated with the strong two dimensionality of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. Though more detailed study is required to conclude the dendritic structure in this salt, it strongly suggests that there is another dynamics of the vortex system at low temperatures. The existence of such dynamics, which can stabilize the critical state against conventional flux jumps, may possibly explain the deviation from the theory observed in the sweep rate dependence.

IV. SUMMARY

In summary, we observed the remarkable temperature spikes due to the flux jumps and the preceding temperature oscillations in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. These dependences on the sweep rate can be described by the conventional critical-state model except at slow sweep rates. The temperature dependence of the melting field is consistent with the previous reports, which suggests the vortex lattice melting driven by quantum fluctuations at low temperatures. The small temperature fluctuations observed at low temperatures imply the dendritic vortex distribution, which is probably associated with the strong two dimensionality of this salt.

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[25] For example, our analysis of the Sierpinski Gasket gave a box counting dimension of 1.528. It is consistent with the calculated value within an error of 4%.