Vortex Glass Transition and Quantum Vortex Liquid at Low Temperature in a Thick a-Mo_xSi_{1-x} Film

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We present measurements of ac complex resistivity, as well as dc resistivity, for a thick amorphous Mo_xSi_{1-x} film at low temperatures ($T > 0.04$ K) in various constant fields B. We find that the vortex glass transition (VGT) persists down to $T \sim 0.04T_{c0}$ up to $B \sim 0.9B_{c2}(0)$, where T_{c0} and $B_{c2}(0)$ are the mean-field transition temperature and upper critical field at $T = 0$, respectively. In the limit $T \rightarrow 0$, the VGT line $B_g(T)$ extrapolates to a field below $B_{c2}(0)$, while the dc resistivity $\rho(T)$ tends to the finite nonzero value in fields just above $B_g(0)$. These results indicate the presence of a metallic quantum vortex liquid at $T = 0$ in the regime $B_g(0) < B < B_{c2}(0)$.

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The existence of a vortex liquid just below the upper critical field B_{c2} has been well established through a variety of experiments for three-dimensional (3D) and 2D superconductors. For clean superconductors in which the melting transition of the vortex lattice is observed, the shape of the melting line, $B_m(T)$, has been obtained over the broad temperature T range both experimentally [1,2] and theoretically [2–5]. At high temperatures properties of vortex lines are dominated by thermal fluctuations, while at sufficiently low temperatures they are subject to quantum fluctuations. If quantum fluctuations are strong enough, a quantum-vortex-liquid (QVL) state is expected to appear [2–7]. The QVL state has been actually reported for several low-T_c superconductors (LTSC's), such as thin (2D) [1,8–11] and thick (3D) [2] films of amorphous superconductors and the quasi-2D organic superconductor [12].

On the other hand, when the system contains moderately strong disorder which leads to pinning of vortices, the second-order transition from the vortex glass (VG) to vortex liquid occurs [13]. Experimental evidence for the vortex glass transition (VGT) has been obtained in several 3D superconductors. However, since most of the experiments have been performed using high-T_c oxide superconductors (HTSC's) [14] whose upper critical field $B_{c2}(0)$ at zero temperature is extremely high, it has not yet been evident whether the VGT is also observed in the low-temperature high-field regime where strong quantum fluctuations are present. Theoretically, influences of quantum fluctuations on the VGT have not been fully investigated [15]. The purpose of this work is to clarify experimentally (i) whether the VGT exists down to low enough temperatures $T \sim 0$ and up to high fields B near $B_{c2}(0)$. If this is verified, then it is interesting to explore (ii) the effects of quantum fluctuations on the VGT and (iii) the possibility of the QVL phase at $T = 0$. To investigate these issues, we must prepare the LTSC's which exhibit the VGT. Recently, we have obtained evidence for the VGT for thick amorphous (a-) Mo_xSi_{1-x} films (e.g., $T_{c0} \sim 3.4$ K, $B_{c2}(0) \sim 7.9$ T) [16] and granular In films ($T_{c0} \sim 3.8$ K, $B_{c2}(0) \sim 6.3$ T) [17,18] on the basis of the measurements of dc and ac complex resistivities in constant fields ($B = 0.1$ and 1 T) well below $B_{c2}(0)$. In this work we study a thick a-Mo_xSi_{1-x} film, which is a uniformly disordered superconductor with microscopic pinning centers and turns out to be a good candidate for the study of the VGT in high-field low-temperature regimes [16]. We mainly measure the ac complex resistivity, because the most compelling evidence for the VGT lies in the phase ϕ of the linear ac complex resistivity, as described below. Also, ac measurements are advantageous over dc ones at low temperature since joule heating produced in the sample can be small compared to that in the dc measurements. In particular, in the current (J)-resistivity (ρ) measurements we must use high J to obtain the shape of J-ρ curves, while heating effects become more serious at lower T, where J-ρ curves shift to the higher J region.

The Mo_xSi_{1-x} film ($x = 44$ at.%) used in this study was prepared by coevaporation of pure Mo and Si in vacuum better than 10^{-8} Torr [19–21]. The thickness of the film is 100 nm and the mean-field transition temperature T_{c0} at $B = 0$ is 2.4 K. The structure of the film was confirmed to be amorphous by means of transmission electron microscopy. Details of the growth and characterization of the films were published previously [19,20]. The temperature dependence of the dc resistivity ρ was measured in constant fields B using standard four-terminal dc and low-frequency (~19 Hz) ac locking methods. The field was applied perpendicular to the plane of the film. The ac transport data, the frequency dependence of the amplitude ρ_{ac} and phase ϕ of the ac resistivity, were taken in the linear regime as a function of the temperature T and frequency f employing a four-terminal method [16,22]. An oscillator output of the precision LCR meter (HP4285A) produced the current, which passed uniformly through the entire thickness of the film. The ac voltages induced across the sample were measured using the LCR meter after being enhanced with a low-noise preamplifier. We regarded the data at the lowest temperature as the background data,

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where ρ_{ac} of the film is much smaller than the background component, and subtracted it from the measured data. We also evaluated a frequency-dependent gain and/or phase delay of the amplifier by measuring the standard noninductive load resistor which was connected in place of the sample. Thus we obtained the frequency-dependent resistivity in the frequency range $f \sim 75$ kHz–5 MHz from the measured voltages. We cannot measure the ac resistivity at lower f (<75 kHz) because of the limitation of the *LCR* meter. However, the essential data are included in the f range used in the present measurements. Even though we tried to perform the measurement at lower f (<75 kHz), ρ_{ac} around T_g would fall below the experimental resolution of the preamplifier.

First, we focus on the results of the ac resistivity in various fields B. According to the 3D VG theory [13], at the phase transition temperature $T = T_g$, the amplitude follows a power-law frequency dependence $\rho_{ac} \propto f^{(z-1)/z}$ while the phase takes a frequency-independent value $\phi_g = (\pi/2)(z - 1)/z$, which is smaller than 90°. Here, z is the dynamical critical exponent. As typically shown in Figs. 1(a) and 1(b), upon cooling, ϕ at different frequencies merges to the frequency-independent values ϕ_g(= 74° and 76°) at $T = 1.51$ and 0.285 K for $B = 2.0$ and 5.0 T, respectively. Despite rather different B, the values of ϕ_g are almost identical to each other, yielding $z \sim 6$. We find agreement between the values of z extracted from the slope of the critical $\log\rho_{ac}$ vs $\log f$ isotherm ($T = T_g$) and from ϕ_g, consistent with the VG theory. As depicted in Fig. 1(c), in $B = 5.1$ T the critical value of the phase ϕ_g is 79.4° ($z = 8.5$), which is higher than the values in lower fields. As the field increases up to $B = 5.32$ T, a critical point of ϕ is no longer visible, as shown in Fig. 1(d). This result indicates the breakdown of the VGT.

In order to see the change in the critical behavior of VGT in the low-temperature high-field regime more clearly, we plot in Fig. 2 the dynamical exponents z extracted from the values of ϕ_g, together with the temperature dependence of $\phi(T, f)$ for different f, taken at constant fields in the range $B = 4.4$–5.4 T. In $B \leq 5.3$ T ($T_g \geq 0.09$ K), we are able to define ϕ_g. We have confirmed that the values of ϕ_g(= 73°–77°) and z(= 5.3–6.9) stay in the limited ranges over the broad field region $B = 1.0$–5.0 T ($T_g = 1.85$–0.280 K). In $B = 5.1$–5.3 T, however, z takes somewhat higher values ($z = 8.1$–8.5). The reason for this is not clear, but it may suggest that a crossover from thermal to quantum liquid occurs at $B \sim 5.1$ T ($T \sim 0.2$ K). In $B > 5.3$ T, the critical behavior of VGT completely disappears.

Shown in Fig. 3 is the temperature dependence of the dc resistivity $\rho(T)$ in a linear regime for different fields ($B = 4.8$–5.6 and 9.0 T). We are not able to determine T_g, as well as critical exponents, precisely from the $\rho(T)$ data alone. However, we can roughly confirm that $\rho(T)$ follows the power-law functional form predicted by the VG theory

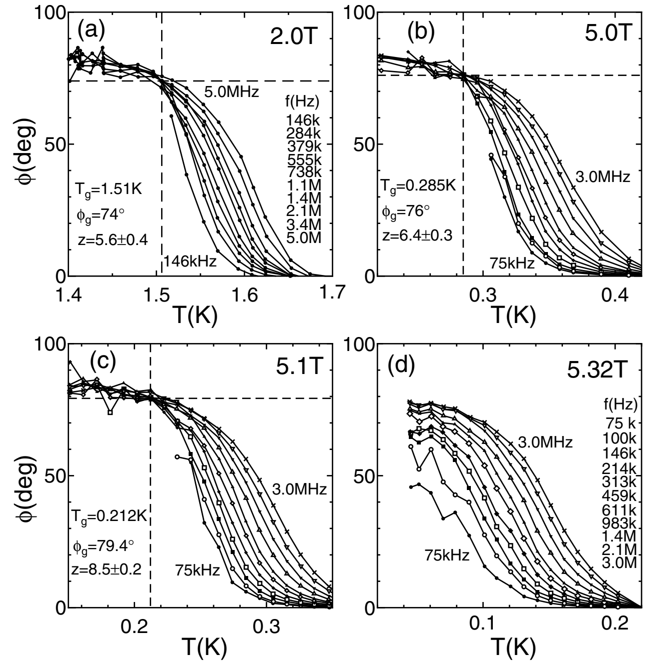

FIG. 1. Temperature dependence of the phase ϕ for different f in (a) $B = 2.0$ T, (b) 5.0 T, (c) 5.1 T, and (d) 5.32 T. The frequencies f are listed in (a) and (d); those in (b) and (c) follow the same sequence as in (d).

in fields up to about 5.3 T; e.g., in $B = 4.8$ T, where T_g is determined to be 0.40 K from $\phi(T, f)$, $\rho(T)$ is reproduced by $\rho \sim (T/T_g - 1)^{\nu(z-1)}$ using $T_g \sim 0.38$ K, $z \sim 5.4$, and $\nu \sim 1$, which are of reasonable magnitude. In $B \geq 5.31$ T, however, $\rho(T)$ at low temperature is markedly different. Upon cooling, the logarithm of $\rho(T)$ decreases with upward curvature below about 0.1 K and extrapolates to the finite nonzero value $\rho(0)$ in the limit $T \to 0$ [23]. Thus, the field of 5.31 T is a critical field B_0 separating the superconducting phase from the metallic phase at

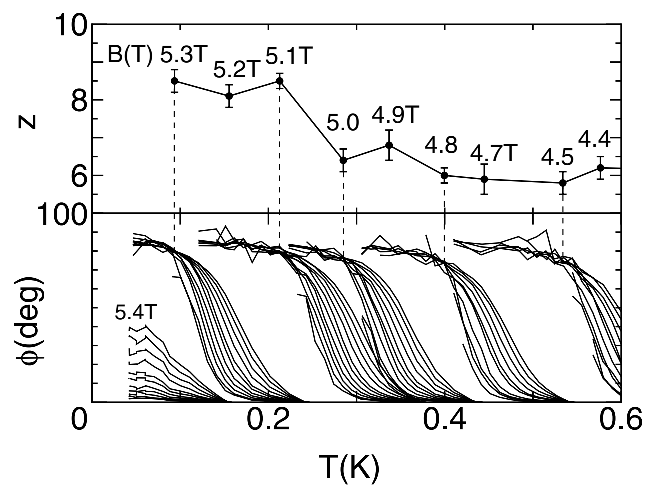

FIG. 2. Temperature dependence of ϕ for different f and dynamical exponents z taken at constant fields in the range $B = 4.4$–5.4 T. In $B > 5.3$ T, critical behavior associated with VGT is not visible. Not all the data of ϕ are plotted for clarity.

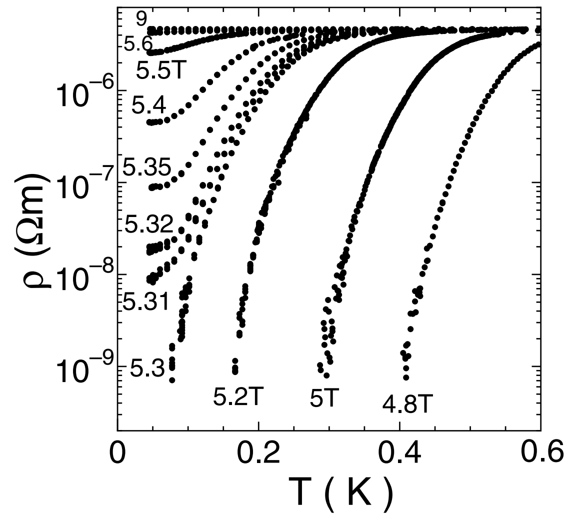

FIG. 3. Temperature dependence of the dc resistivity ρ in different fields B. $B_0 = 5.31$ T is the critical field of the superconductor-metal transition.

$T = 0$. It is noted that this field B_0 coincides with the field above which all evidence for VGT disappears from the ac measurements.

Based on the data we construct the vortex phase diagram in the B-T plane (Fig. 4). First, we estimate the upper critical field $B_{c2}(T)$ from the resistive transition $\rho(T)$ for constant B: We plot B against the temperature at which $\rho(T)$ decreases to 90% of the normal-state resistivity ρ_n. This is a very rough estimation of $B_{c2}(T)$ and actual values of $B_{c2}(T)$ are considered to be higher. We obtain $B_{c2}(0)$ to be $\sim$5.7 T from a smooth extrapolation of $B_{c2}(T)$ to the B axis. Next, we estimate the critical field B_c of the $T = 0$ metal-insulator transition (MIT) where $\rho(T)$ at low temperature ($T < 0.5$ K) is temperature independent; i.e., $\rho(T, B_c) \approx \rho_n$. We obtain B_c to be 5.7 T from the intersection point of the isothermal lines of the magnetoresistance (MR), $\rho(B)$. This is the rough estimation of the MIT point. To determine it precisely, more detailed analysis based on the $\rho(T)$ data at even lower temperatures is necessary. In fields higher than about 6 T ($\sim B_c$), $\rho(B)$ at low T is almost constant or shows a slight monotonic increase with increasing B, suggesting that the origin of MR in this field region is due to unpaired electrons; i.e., there is no sign of the presence of Cooper pairs. Thus it is reasonable to consider that the upper critical field $B_{c2}(0)$ is close to or slightly higher than $B_c = 5.7$ T, consistent with the estimation mentioned above. This is in contrast to the result for the ultrathin (4 nm) films [20,24] in which an anomalous peak and a subsequent decrease in the MR suggestive of the presence of the localized Cooper pairs have been observed over the broad field region $B_c < B < B_{c2}(0)$; e.g., $B_c = 2.35$ T and $B_{c2}(0) \sim 5.7$ T, where B_c is the critical field for the superconductor-insulator transition (SIT).

The VGT line $B_g(T)$ shown in Fig. 4 is obtained by plotting B against $T_g(B)$ determined from the ac measurements. Open circles denote the particular points for $B = 5.1$, 5.2, and 5.3 T where values of z are higher than those for lower B. In the limit of zero temperature, $B_g(T)$ extrapolates to a field close to $B_0 = 5.31$ T that is clearly lower than $B_{c2}(0)$; $1 - B_0/B_{c2}(0) \approx 0.07$. The implication of this result is that a finite QVL state is present in the region $B_0 < B < B_{c2}(0)$ at $T = 0$. This QVL state is metallic, since B_0 is a critical field of the superconductor-metal transition.

Several theories have predicted the quantum liquid of vortices or quantum melting of the vortex solid [2–7]. However, most of them have concentrated on melting of vortex lattice in clean systems. We cannot tell immediately whether the presence of disorder assists the appearance of the quantum liquid state through strong quantum fluctuations or that of the vortex glass state through strong pinning. In the absence of the theory, we heuristically analyze our data using the melting theory of the vortex lattice which takes account of quantum fluctuations [2]. The strength of quantum fluctuations is quantified by the resistance ratio ρ_n/dR_q, where d is the thickness of the sample and $R_q = h/2\pi e^2 \approx 4.1$ kΩ is the quantum resistance. The $T = 0$ quantum melting transition takes place at a field $B_m^q(0)$ lower than $B_{c2}(0)$, which is expressed as

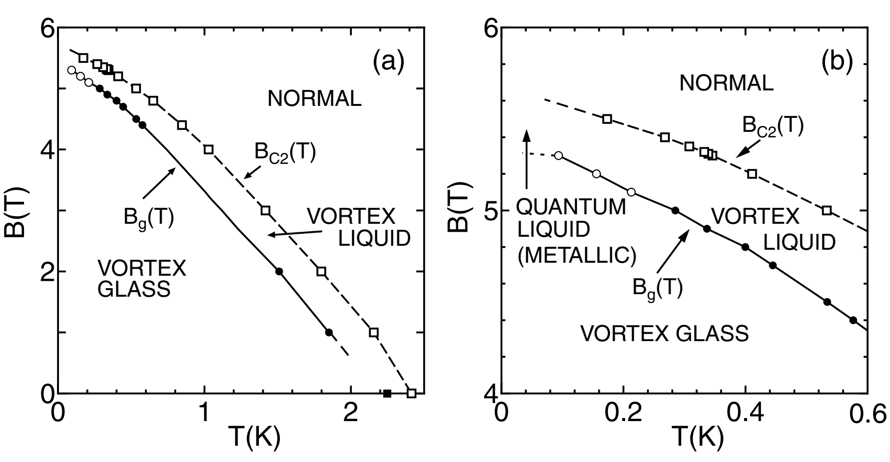

FIG. 4. (a) Vortex phase diagram in the B-T plane. Circles and open squares denote $B_g(T)$ and $B_{c2}(T)$, respectively. A full square corresponds to the zero-resistivity temperature in $B = 0$. Open circles represent $B_g(T)$ where values of z are higher than those for lower fields. The low-temperature part is enlarged and shown in (b). The full and dashed lines are guides for the eye.

$$B_m^q(0) = B_{c2}(0)\left[1 - \frac{2}{\pi}\exp\left(\frac{2\pi}{3}\alpha - \frac{\alpha^2}{2} - \frac{\pi^3 c_L^2}{2}\frac{R_q}{R_n^*}\right)\right], \quad (1)$$

where c_L is the Lindemann number, $\alpha = 2/\sqrt{\pi}\,\nu$ (ν is a numerical constant of order unity), and $R_n^* = \rho_n \nu / a_0$ ($a_0^2 = \Phi_0/B$ is the unit cell area and Φ_0 is the flux quantum). Inserting the measured values of $B_m^q(0) = B_g(0) \approx 5.3$ T, $B_{c2}(0) \approx 5.7$ T, and $\rho_n = 4.6\ \mu\Omega$m, and taking $\nu = 1$, c_L is calculated to be 0.12. This value is in agreement with the value $c_L \sim 0.1$–0.3 predicted theoretically [2]. This result is somewhat surprising, considering that the theory for the clean system may be also applicable to the highly disordered system in which the VGT is observed. It is also interesting to note that the phase diagram presented in Fig. 4 is very similar to what has been reported for a thick a-Nb_3Ge film [2], where the melting of the vortex lattice has been observed.

Finally, we compare the $T = 0$ phase diagram for the thick (100 nm) film obtained here with a possible phase diagram for thin (4 nm) a-Mo_xSi_{1-x} films reported recently [24]. As described above, for the thick film there is the metallic QVL in the region $B_0 < B < B_{c2}(0)$ ($\sim B_c$). This region is narrow, $[B_{c2}(0) - B_0]/B_{c2}(0) \sim 0.07$, but its existence is definite. In contrast, for thin films the metallic QVL phase is not evident, most likely absent. Instead, there is the unusual insulating regime $B_c < B < B_{c2}(0)$ suggesting the presence of the localized Cooper pairs above the field-driven SIT (B_c). This regime is very broad, typically $[B_{c2}(0) - B_c]/B_{c2}(0) > 0.2$. Within the 2D VG theory of Fisher [15], this regime is interpreted as an insulating QVL phase (Bose-glass insulator) which originates from strong quantum fluctuations in 2D. However, experimental verification of the 2D VGT is not easy, because it is a quantum phase transition which occurs at $T = 0$. The present finding that in thick (3D) films the VGT and the QVL phase certainly exist at very low temperatures is important, because this may support the existence of the VGT in thin (2D) films.

In summary, we have demonstrated on the basis of the ac resistivity for the thick a-Mo_xSi_{1-x} film that the VGT persists down to very low temperatures $T \sim 0.04T_{c0}$ and up to high fields $B \sim 0.9B_{c2}(0)$. In the limit $T \to 0$ the VGT line $B_g(T)$ extrapolates to the field $B_g(0) \approx B_0$ below $B_{c2}(0)$, where B_0 is the critical field of the superconductor-metal transition. This result indicates that the metallic quantum liquid is present in the regime $B_g(0) < B < B_{c2}(0)$ at $T = 0$. Upon cooling, the dynamical exponent z shows a trend to increase at $\sim 0.09T_{c0}$. This may suggest a crossover from a thermal to quantum liquid regime.

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