Enhancement of the Quantum-Liquid Phase by Increased Resistivity in Thick a-Mo_xSi_{1-x} Films

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Effects of normal-state resistivity ρ_n on the vortex phase diagram at low temperature T have been studied based on dc and ac complex resistivities for thick amorphous $Mo_x Si_{1-x}$ films. It is commonly observed irrespective of ρ_n that, in the limit T = 0, the vortex-glass-transition line $B_g(T)$ is independent of T and extrapolates to a field below the T = 0 upper critical field $B_{c2}(0)$, indicative of the quantum-vortex-liquid (QVL) phase in the regime $B_g(0) < B < B_{c2}(0)$. The relative width of the QVL phase increases along the B and T axes approximately proportional to ρ_n . This result is consistent with a view that the QVL phase is caused by strong quantum fluctuations, which are enhanced with increasing ρ_n .

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The vortex states at low temperature T have been actively studied in various clean type-II superconductors, such as single-crystal layered superconductors [1,2] and amorphous films with weak pinning [3-8]. In these systems the existence of the quantum-vortex-liquid (QVL) phase has been reported based on the measurements, such as irreversibility field and dc resistivity ρ . Recently, we have demonstrated on the basis of the ac complex resistivity for a thick amorphous $(a)Mo_xSi_{1-x}$ film with moderately strong pinning that the vortex-glass transition (VGT) [9] persists down to low T (~ $0.04T_{c0}$) up to high fields $[B \sim 0.9B_{c2}(0)]$, where T_{c0} and $B_{c2}(0)$ are the mean-field transition temperature and upper critical field at T = 0, respectively [10]. In the limit T = 0, the VGT line $B_{\rho}(T)$ is nearly independent of T and extrapolates to a field below $B_{c2}(0)$, indicative of the presence of QVL at T = 0 in the regime $B_g(0) < B < B_{c2}(0)$ [11].

Theoretically, quantum fluctuations are able to melt the vortex lattice in clean systems at sufficiently high B and give way to the QVL phase at T = 0. In disordered systems, however, the effects of quantum fluctuations on the VGT have not been fully clarified theoretically as well as experimentally. It has been generally believed that an increase in disorder, which corresponds to the normalstate resistivity ρ_n above T_{c0} , makes the QVL phase more favorable through the enhancement of quantum fluctuations [8,12–15]. This has not yet been demonstrated experimentally, though some experimental data seem to support the notion [5,7,16,17]. If this is verified, the QVL phase will be more strongly proved. On the other hand, it is not easy to study experimentally the effects of ρ_n on the vortex phase diagram. This is because the increase in ρ_n may accompany the enhancement of the pinning strength, which assists to make the VG phase stable contrary to the quantum-fluctuation effects [13,14,18].

In this work we study a series of thick films of a-Mo_xSi_{1-x} with different Mo concentration x, whose ρ_n

pared simultaneously, their microscopic or mesoscopic

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increases monotonically with decreasing x. The increase

in ρ_n and decrease in T_{c0} and $B_{c2}(0)$ are due mainly to the

reduction of carrier density. Since these films were pre-

structure responsible for pinning of the flux lines is expected to be similar to each other. The results are summarized as follows. The relative width of the liquid phase at $T \rightarrow 0$, $[B_{c2}(0) - B_g(0)]/B_{c2}(0)$, becomes wider as ρ_n increases. Upon cooling in constant *B* between $B_g(0)$ and $B_{c2}(0)$, curvature in $\log \rho$ vs 1/T curves changes from downward to upward at certain temperature T_Q , signaling a crossover from temperature dominated to quantum driven fluctuations. We find a trend for T_Q/T_{c0} to increase with increasing ρ_n . All of these findings are consistent with a view that the QVL phase is caused by strong quantum fluctuations, which are enhanced with increasing ρ_n .

The samples used in this work are 100-nm-thick a-Mo_xSi_{1-x} films prepared by coevaporation of pure Mo and Si in vacuum better than 10^{-8} Torr [10,11,17,19,20]. Four films (films 1, 2, 4, and 5) with different x (58, 56, 49, and 47 at. %, respectively) were prepared simultaneously using a gradient deposition technique. The T_{c0} and ρ_n just above T_{c0} are 3.86, 3.33, 1.62, and 1.13 K and 3.7, 4.1, 6.4, and 7.2 $\mu\Omega$ m for films 1, 2, 4, and 5, respectively. Another 100-nm-thick film (film 3) with $T_{c0} =$ 2.46 K and $\rho_n = 4.63 \ \mu\Omega$ m was prepared independently for comparison in nearly the same deposition condition, but there might be the slight difference in vacuum and/or evaporation rate during deposition, which could result in the subtle change in pinning properties. The T dependence of the linear dc resistivity ρ was measured using standard four-terminal dc and low-frequency (19 Hz) ac locking methods. The ac transport data, the frequency fdependence of the amplitude ρ_{ac} and phase ϕ of the ac resistivity, were also taken in the linear regime as a function of T and f employing a four-terminal method [10,19]. The field was applied perpendicular to the plane of the film.

In the mixed state the temperature and field dependence of dc and ac resistivities for films 1-5 is qualitatively similar to each other. Most importantly, both $\rho_{ac}(f)$ and $\phi(f)$ exhibit the behavior indicating the critical slowing down of the vortex dynamics near the secondorder (VG) transition [9]. As shown in previous papers [19,21], the location (T_g) as well as the existence of VGT is clearly determined from the T dependence of ϕ for different f. We representatively describe the results of the most resistive film 5 and the most conductive film 1. For film 5 with $B_{c2}(0) = 3.43$ T, ϕ at different f merges to the same value $\phi_g \sim 75^\circ$ at $T_g(B)$ in B ranging from 0.1 to 2.7 T. Using the critical value of $\phi_g = 75 \pm 2^\circ$, the dynamical exponent z is immediately obtained to be 6.0 ± 0.8 from the relation $\phi_g = (\pi/2)(z-1)/z$. We cannot determine definitely the location of the phase transition from the $ho_{\rm ac}$ data alone; however, we can confirm that at T_g derived from ϕ , the amplitude ρ_{ac} exhibits a power-law frequency dependence, $\rho_{ac} \propto f^{(z-1)/z}$, predicted by the VGT theory [9], yielding $z = 6.7 \pm 1.0$. The dc resistivity $\rho(T)$ is also reproduced by a predicted functional form $\rho(T) \sim (T/T_g - 1)^{\nu(z-1)}$ using T_g and exponents ($\nu z = 6.0 \pm 1.0$) of reasonable magnitude. The behavior observed here is very similar to that which has been observed for the less resistive films 1-4 [10,11,20], the thicker (300 nm) film [19], and 150-nm-thick indium films [21].

As the field increases up to B = 3.05 T, ϕ_g takes a slightly higher value ($80 \pm 1^\circ$) yielding $z = 9 \pm 0.9$, as shown in Fig. 1(a), and in 3.08 T the critical behavior of



FIG. 1. ϕ vs T/T_{c0} for different f in (a) B = 3.05 and (b) 3.08 T (film 5) and in (c) 8.30 and (d) 8.35 T (film 1). Frequencies f are listed in (a) and (c) (from the left to the right); those in (b) and (d) follow the same sequence as in (a) and (c), respectively.

VGT is no longer visible [Fig. 1(b)]. Only an addition of $\Delta B = 0.03$ T to B = 3.05 T, which corresponds to $\Delta B/B \approx 1\%$, drastically causes destruction of VGT, indicating that the VGT line is very weakly dependent on temperature at $T \rightarrow 0$. The similar behavior is observed for the most conductive film 1 with $B_{c2}(0) = 8.77$ T, as shown in Figs. 1(c) and 1(d), and also for the rest of three films 2–4 [11].

Figures 2(a) and 2(b) depict the Arrhenius plots of dc $\rho(T)$ in different B for films 1 and 5, respectively. Here, T is normalized by T_{c0} for each film. Since the lowest T available in our transport measurement is limited to around 0.03-0.04 K, we must study an effectively lower- T/T_{c0} region using more conductive films with higher T_{c0} . We roughly confirm that $\rho(T)$ follows the power-law form predicted by the VG theory in fields below the certain characteristic field B_0 ; $B_0 \approx 8.3$ (film 1) and 3 T (film 5). B_0 almost coincides with the upper bound of the VG phase $B_{\rho}(0)$: 8.33 and 3.06 T for films 1 and 5, respectively. Upon cooling in the field region $B_0 < B < B_{c2}(0)$, which corresponds to the T = 0QVL phase discussed later, curvature in $\log \rho$ vs 1/T plots changes from downward to upward below the certain temperature T_O , suggestive of the normal (or metallic) phase at T = 0. The location of T_Q is indicated with open circles, which is defined as a temperature at which



FIG. 2. $\log \rho$ vs T_{c0}/T in different *B* for (a) films 1 and (b) 5. Open circles denote $T_{c0}/T_Q(B)$. Inset: Arrhenius plots of $\rho(T)$ for film 5. Full curves represent the expected VG temperature dependence for $\rho(T)$.

 $d^2 \log \rho / dx^2 = 0$, where $x \equiv 1/T$. We roughly confirm that a deviation from the expected VG temperature dependence for $\rho(T)$ occurs at around T_Q , as representatively indicated with full curves. The change in curvature of $\log \rho(T)$, together with the increase in z [22], on cooling is interpreted as signaling a crossover from temperature dominated to quantum driven fluctuations. For film 1, T_Q is higher for larger B [3]. This may reflect stronger quantum fluctuations for larger B/B_{c2} . For film 5, however, we cannot discuss the field dependence of T_Q within experimental resolutions.

The B - T phase diagram for films 1, 3, 4, and 5 is illustrated in Fig. 3. The result for film 2 is not shown for clarity. The upper critical field $B_{c2}(T)$ shown with open squares is defined using a criterion that $\rho(T)$ decreases to 95% of ρ_n [11]. The circles represent T_g for different Bdetermined from the phase ϕ of the ac resistivity. With increasing ρ_n , both T_{c0} and $B_{c2}(0)$ decrease nearly proportional to $1/\rho_n$, while the whole shape of B_{c2} (or B_g) vs Tcurves is similar to each other, which is a characteristic of uniformly disordered amorphous films.

The low-*T* and high-*B* regions of the B - T phase diagrams for films 1 and 5 are shown in Figs. 4(a) and 4(b), respectively. Here, *B* and *T* axes are normalized by $B_{c2}(0)$ and T_{c0} for each film, respectively. A horizontal dotted line marks the upper bound $B_g(0)$ of the VG phase. We find irrespective of ρ_n that in the limit $T \rightarrow 0$, $B_g(T)$ is independent of *T* and extrapolates to a field near B_0 lower than $B_{c2}(0)$. This means that a (metallic) QVL state is present in the regime $B_g(0) < B < B_{c2}(0)$. The shaded region denotes the liquid phase at low *T* below T_Q (i.e., the QVL phase), where quantum effects are considered to be dominant, while the liquid region at $T > T_Q$ is re-



garded as a thermal-liquid phase. It is important to note that for film 5 with higher ρ_n , the width of the QVL phase is larger both along the field $[B/B_{c2}(0)]$ and temperature (T/T_{c0}) axes than that for film 1.

To examine the effects of ρ_n on the QVL phase quantitatively [23], we plot in Fig. 5 the relative width of the (T = 0) QVL phase along the *field* axis, defined as $\Delta B_{\text{OVL}} \equiv [B_{c2}(0) - B_{g}(0)]/B_{c2}(0)$, against ρ_n for all the films 1–5. It is clearly seen that ΔB_{OVL} (full symbols) follows a simple linear relation expressed as $\Delta B_{\text{OVL}} \propto \rho_n$, as indicated with a straight line. Also shown in Fig. 5 is the relative width of the QVL phase along the *temperature* axis, as defined as $\Delta T_{\text{QVL}} \equiv T_Q/T_{c0}$, against ρ_n (open symbols). The large error bars reflect both the experimental ambiguity in determination of T_O/T_{c0} and the field dependence of ΔT_{QVL} . Similarly to the case of $\Delta B_{\text{QVL}}(\rho_n), \Delta T_{\text{QVL}}$ increases with ρ_n almost proportional to ρ_n at least within the ρ_n range studied. For film 3 the deviation of the data point (an open triangle) from the straight line is noticeable. The reason is not clear, but it may be due to the fact that the microscopic or mesoscopic structure of film 3 responsible for pinning of the flux lines is different from that for the remaining four films, since (only) film 3 was prepared independently. It is interesting to note that ΔB_{QVL} and ΔT_{QVL} are of nearly the same order of magnitude for given ρ_n and that the QVL phase seems to disappear in the limit $\rho_n \rightarrow 0$.

Based on the Lindemann-like criterion, Blatter and coauthors have examined the effects of quantum fluctuations on the melting transition of the vortex lattice in clean systems and calculated the melting-transition line, which lies well below the $B_{c2}(T)$ line even at low enough T [8,12]. We note, however, that this theory cannot be applied to the disordered systems in which the VGT



FIG. 3. B - T phase diagrams for films 1 and 3–5. Full circles and open squares denote $B_g(T)$ and $B_{c2}(T)$, respectively. Full squares correspond to the zero-resistivity temperatures in B = 0. The full and dashed lines are guides for the eye.

FIG. 4. Low-*T* parts of the phase diagrams for (a) films 1 and (b) 5. *B* and *T* axes are normalized by $B_{c2}(0)$ and T_{c0} for each film, respectively. Open circles denote $T_Q(B)/T_{c0}$. A horizontal dotted line marks the upper bound of the VG phase. The full and dashed lines are guides for the eye.



FIG. 5. $\Delta B_{\rm QVL}$ (full symbols) and $\Delta T_{\rm QVL}$ (open symbols) for films 1–5 against ρ_n . The results for film 3 are indicated with triangles. A straight line represents the fit of the data.

occurs. Furthermore, it has been suggested theoretically that the Lindemann-like approach is a very rough thermodynamic approximation [24]. In the meantime, Ishida and Ikeda have developed the microscopic theory which predicts resistive behaviors at T > 0 reflecting a quantum VGT [18]. Although the theory successfully explains various transport data in two dimensions (2D), including the data of ultrathin a-Mo_xSi_{1-x} films [17], it seems to contradict the present data for thick (3D) a-Mo_xSi_{1-x} films. The theory does not predict the metallic intermediate phase with resistance smaller than ρ_n at T = 0. In order to clarify this problem, further measurements down to even lower $T/T_{c0} (\ll 0.01)$ are necessary.

The essential results revealed by the present study, which are summarized in Figs. 4 and 5, are very simple. As far as we know, however, there is no available theory that accounts for the present results specifically. We note the general theoretical prediction [8,12,13] that the strength of quantum fluctuations Q is proportional to ρ_n and inversely proportional to the film thickness d for uniform films: $Q = \rho_n / R_q d$, where $R_q = h/2\pi e^2 \approx$ 4.1 k Ω is the quantum resistance [8,12]. Based on the notion, the present results are qualitatively interpreted as follows. The increase in the QVL phase with increasing ρ_n (i.e., $\Delta B_{\text{OVL}}, \Delta T_{\text{OVL}} \propto \rho_n$) is consistent with a view that the QVL phase is caused by strong quantum fluctuations, which are enhanced with increasing $\rho_n (Q \propto \rho_n)$. This result strongly supports our previous view that the vortexliquid phase at low T is the QVL phase which originates from strong quantum fluctuations [10,11]. Also, earlier work on 2D systems [5,7] shows that the *B* range over which the QVL exists grows with increasing the normalstate sheet resistance, consistent with our conclusion.

In summary, we measure the dc and ac complex resistivities for thick a-Mo_xSi_{1-x} films with different ρ_n . It is commonly observed irrespective of ρ_n that in the limit T = 0, the VGT line $B_g(T)$ is independent of T and extrapolates to a field below $B_{c2}(0)$, indicative of the presence of the T = 0 QVL phase in the regime $B_g(0) < B < B_{c2}(0)$. The relative width of the QVL phase increases both along the B and T axes with increasing ρ_n , approximately proportional to ρ_n . This is consistent with a view that the QVL phase is caused by strong quantum fluctuations, which are enhanced with increasing ρ_n .

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