Rapid Communications

Destruction of superconductivity through phase fluctuations in ultrathin *a*-MoGe films

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Superconductivity occurs in metals when attractive interactions between electrons promote the formation of Cooper pairs which get locked in a phase-coherent state, leading to superfluid behavior. In situations where the phase stiffness falls below the pairing energy, the superconducting transition temperature T_c is driven by phase fluctuations and pairing can continue to survive above T_c. Such a behavior has long been thought of as the hallmark of unconventional superconductivity. Here we combine sub-K scanning tunneling spectroscopy, magnetic penetration depth measurements, and magnetotransport measurements to show that in ultrathin amorphous MoGe (*a*-MoGe) films the superfluid density is strongly suppressed by quantum phase fluctuations at low temperatures for thickness below 5 nm. This is associated with a rapid decrease in the superconducting transition temperature T_c and the emergence of a pronounced pseudogap above T_c. These observations suggest that even in conventional superconductors strong disorder and low dimensionality will ultimately trigger a Bosonic route for the destruction of superconductivity.

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The Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity represented for decades the paradigm to interpret the superconducting (SC) phenomenon in so-called conventional metals [1]. In these systems the lattice vibrations (phonons) mediate an attractive effective interaction among electrons with opposite momenta, which can then pair up into Cooper pairs, with a typical energy scale provided by the SC gap Δ. The spin-zero Boson-like Cooper pairs can condense in a macroscopic quantum state, where the quantum-mechanical phase of the electronic wave function becomes rigid [2]. The phase rigidity is the hallmark of superfluid behavior, and the energy cost required to slowly deform the phase, the so-called superfluid stiffness J_s, is usually much larger than Δ in conventional superconductors [3]. However, the discovery of unconventional superconductors, where the pairing glue is provided by electronic correlations instead of phonons, paved the way to the investigation of an alternative scenario, where the hierarchy between Δ and J_s is reversed [3–5]. On general grounds, J_s scales with the ratio between the superfluid density n_s and the electron effective mass m^*, i.e., $J_s \sim \frac{n_s}{m^*}$. Thus, the mass enhancement provided by electronic correlations [6] can strongly suppress the effective J_s leading to a superconductor-to-metal transition driven by the loss of phase coherence. This paradigm has been discussed for cuprate superconductors where superconductivity emerges by doping the Mott-insulating state triggered by strong correlations. In this class of materials several different probes [6–9] suggested the persistence of a pairing gap above the transition temperature T_c. However, a different route to reverse the order between Δ and J_s is provided by a suppression of n_s triggered by disorder [10–16]. In this case, even in conventional electron-phonon mediated superconductors, strong disorder can push the system towards a bosonic regime where pairing survives above T_c. Ultimately, even the quantum $T = 0$ transition can belong to the bosonic route, with a direct transition between a superfluid state and an insulating one [17]. On the other hand, for conventional superconductors disorder has also the effect of gradually suppressing the pairing itself via loss of Coulomb screening [18–20]. Within such a Fermionic scenario at a critical disorder the repulsion can overcome the electron-phonon attractive interaction transforming the system either into a bad metal or an Anderson insulator [21,22].

Initial studies, based mainly on the dependence of T_c on the sheet resistance (R_s), suggested that several amorphous superconducting films of conventional superconductors follow the fermionic route [21,22]. Some later work on planar tunnel junctions made on disordered amorphous superconductors appeared to be consistent with this scenario [23,24]. However, over the past decade there is increasing experimental evidence that a Bosonic scenario applies to conventional superconductors [25–32] such as TiN, NbN, and InO_x. These observations are based on the comparison among different probes that can selectively determine either Δ or J_s. This was the case for NbN, where, studying a number of films with varying levels of disorder, it was observed that the system follows the Fermionic route at moderate disorder and crosses over to a Bosonic scenario at stronger disorder [33,34]. A similar variation was also obtained for [35] TiN. This leaves open the question as to the extent to which the bosonic superconductor-to-metal or superconductor-to-insulator transition represents a universal paradigm whenever correlations and/or disorder suppress J_s sufficiently.

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A benchmark system to address this question is amorphous superconducting MoGe films (a-MoGe), where thickness is used as the control parameter to tune the disorder. So far, MoGe has been considered the prototype system where a Fermionic route is followed all the way to the destruction of the superconducting ground state [36–38]. This conclusion was based on transport measurements, even though the presence of quantum phase fluctuations at very low thickness has also been recognized [39].

In this Rapid Communication, we investigate the evolution of superconductivity in ultrathin a-MoGe films, using a combination of low-temperature scanning tunneling spectroscopy (STS), penetration depth (λ), and magnetotransport measurements. We observe that for films with thickness down to 5 nm, the decrease in T_c appears to be consistent with the expectation from a Fermionic scenario. However, below this thickness, T_c decreases rapidly with a rapid increase in $(\frac{\Delta}{k_B T_c})$ ratio. At the same time, our films show a strong signature of quantum and classical phase fluctuations and a pronounced pseudogap from STS measurements. Our result suggests that the destruction of superconductivity by the Bosonic mechanism becomes unavoidable when the J_s drops below Δ, both in conventional and unconventional superconductors.

The a-MoGe thin films used in this study were grown on oxidized silicon substrates using pulsed laser deposition starting from a $Mo_{70}Ge_{30}$ arc-melted target. Details of sample preparation have earlier been reported in Refs. [40] and [41]. Film thickness (t) was varied between 20 and 1.8 nm. For $t \geqslant 10$ nm the thickness of the film was directly measured using a stylus profilometer whereas for thinner samples it was estimated from the number of laser pulses using two films with $t \geqslant 10$ nm grown before and after the actual run for calibration. To prevent surface oxidation, samples used for magnetotransport and penetration depth measurements were covered with a 2-nm-thick protective Si layer. These measurements were performed in ^{3}He cryostats operating down to 300 mK. STS measurements were performed using a home-built scanning tunneling microscope [42] (STM) operating down to 450 mK and in magnetic fields up to 90 kOe. The tunneling conductance ($G(V) = \frac{dI}{dV}|_V$ vs V) was measured using standard modulation technique using a Pt-Ir tip. To maintain a pristine surface, the sample used for STS measurement was transferred in an ultrahigh vacuum suitcase after deposition and transferred in the STM without exposure to air. After the STS measurements, the films were withdrawn in the vacuum suitcase and covered with a 2-nm-thick protective Si layer before performing transport measurements. The London penetration depth was measured using a low-frequency (30 kHz) two-coil mutual inductance technique [43–45] that allows the determination of the absolute value of the penetration depth in thin films.

Figure 1(a) shows R_s as a function of temperature for samples with different thicknesses. We define T_c as the temperature where the resistance becomes $< 0.05\%$ of the normal state value. Figure 1(b) shows the variation of the normal-state sheet resistance (taken as R_s at 9 K, R_s^{9K}) and T_c as a function of film thickness. With decreasing thickness T_c decreases whereas R_s increases, but remains well below the quantum resistance, $\frac{h}{4e^2} = 6.45\,\text{k}\Omega$ (where e is the electron charge and h is Planck's constant). For $t \geqslant 8$ nm R_s^{9K} varies linearly

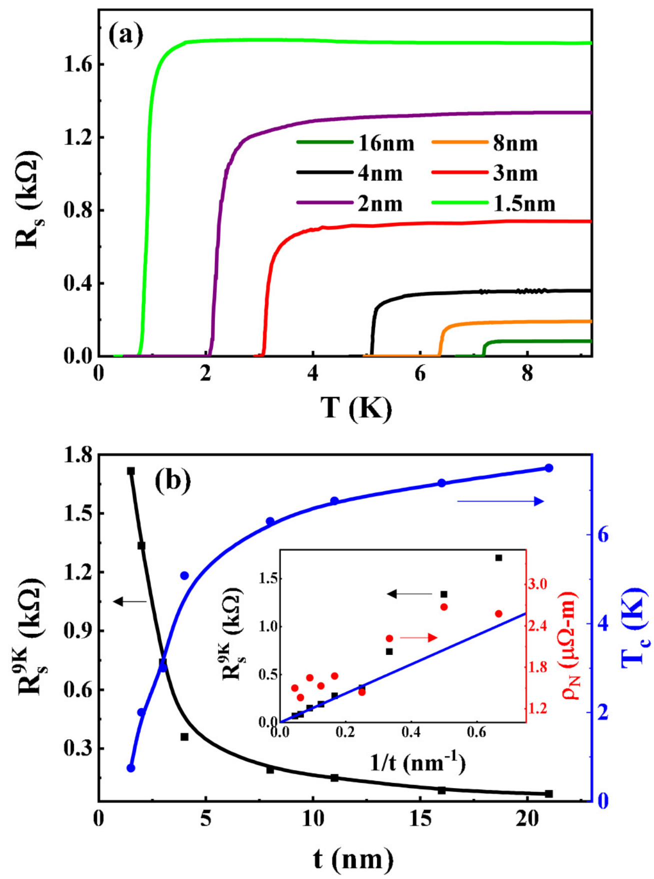

FIG. 1. (a) R_s vs T for a-MoGe films with different thicknesses. (b) $R_s^{9\,\text{K}}$ (black curve) and T_c (blue curve) as a function of film thickness (t); the solid lines are guide to the eye. Inset of (b) shows variation of $R_s^{9\,\text{K}}$ (black dot) and ρ_N as a function of $1/t$; the blue line shows the linear ρ_N -$1/t$ variation for thickness down to 4 nm.

with $1/t$ showing that the increase in the sheet resistance is primarily a geometric effect [inset of Fig. 1(b)]. Below this thickness R_s^{9K} shows an upward trend and the corresponding resistivity (ρ_N) shows an increase from approximately 1.5 to 2.6 $\mu\Omega$ m. We note that the absolute values of ρ_N, the variation of ρ_N with R_s^{9K}, and the variation of T_c with R_s^{9K} are very similar [46] to those in Refs. [36] and [19].

We first concentrate on the STS measurements. $G(V)$-V spectra were recorded over a 32×32 grid over a 200 nm $\times$ 200 nm area at each temperature. Figures 2(a) and 2(b) show the average spectra at different temperatures for the 20- and 2-nm-thick samples. At low temperature the spectra for all samples have the characteristic features of a superconductor: a depression in $G(V)$ at low bias and the presence of coherence peaks at the gap edge. In addition, for the 2-nm-thick sample, we observe a broad V-shaped, nearly temperature-independent background. This feature, also observed in other disordered superconductors [47–49], is attributed to the Altshuler-Aronov-type electron-electron interactions in disordered metals [50]. To extract the superconducting contribution alone, we calculated the normalized spectra, $G_N(V)$ vs V, by dividing it with the spectra obtained at high temperature where the low bias feature associated with superconducting pairing disappears [46]. The left panels of

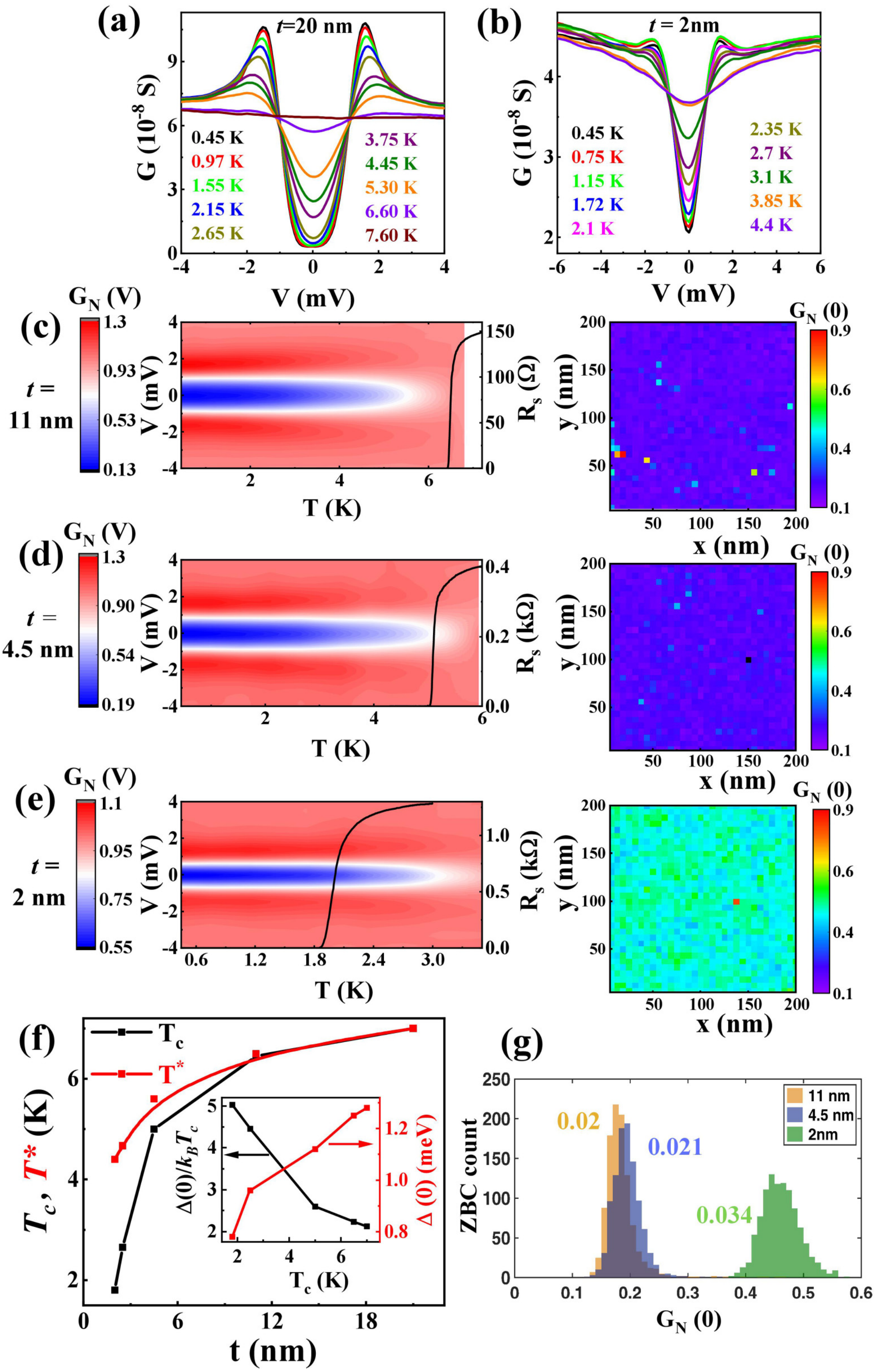

FIG. 2. (a),(b) $G(V)$-V tunneling spectra at different temperatures for 20- and 2-nm-thick films respectively. (c)–(e) Left panels: intensity plot of $G_N(V)$ as function of bias voltage and temperature for three films with thickness 11, 4.5, 2 nm respectively; temperature variations of R_s are shown in the same panels. Right panels: corresponding spatial maps of the zero-bias conductance (ZBC), $G_N(0)$, at 450 mK. (f) Variation of T_c and T^* with film thickness. Inset: $\Delta(0)$ and $\Delta(0)/k_BT_c$ as function of T_c. (g) Histogram of the ZBC for samples of different thicknesses; the root-mean-square width of each ZBC distribution is written next to each histogram.

Figs. 2(c)–2(e) show the temperature dependence of $G_N(V)$ vs V spectra for three representative films in the form of intensity plots along with R_s measured on the same samples. For the samples with $t \sim 11$ nm the superconducting energy gap closes very close to T_c consistent with the expectation from BCS theory [1]. For the sample with $t \sim 4.5$ nm we see a small hint of a pseudogap, where a soft gap in the tunneling spectra extends approximately 0.5 K above T_c. The pseudogap regime gets extended for the sample with $t \sim 2$ nm to about double of T_c ($\sim$1.8 K). We define the pseudogap temperature T^* as the temperature where $G_N(0) \approx 0.95$. We observe that even though the coherence peaks are greatly suppressed compared to the BCS estimate, they continue to exist up to a temperature of 3.1 K. This is consistent with theories where the pseudogap arises from the destruction of the global phase coherence state [15], but inconsistent with theories that ascribe the pseudogap to the parity gap [14]. It is also pertinent to note that we observe a significant spectral weight inside the gap, similar to earlier observations in strongly disordered *in situ* grown [51,48] NbN films (but different from *ex situ* grown [26,27] TiN and InO_x) even though its origin is not completely understood [52,53]. The normalized $G_N(0)$ maps obtained at 450 mK [right panels of Figs. 2(c)–2(e)] reveal a significant increase in inhomogeneity in the $t = 2$ nm sample. This nanoscale inhomogeneity [25,35,48,51,54] can also be seen from the histogram in Fig. 2(g) where the root-mean-square width of the distribution of $G_N(0)$ values increases significantly at this thickness. Plotting T_c and T^* as a function of film thickness [Fig. 2(f)], we observe that a large region of pseudogap emerges below a thickness of 4 nm. At the same time, extracting Δ at 450 mK using the BCS + Γ model [46,55], we observe that $\Delta/(k_B T_c)$ increases rapidly below $t \sim 5$ nm reaching a value of 5 at 2 nm. The emergence of pseudogap between T_c and T^* as well as the anomalously large value of $\Delta/(k_B T_c)$ both signal the breakdown of the BCS scenario in very thin *a*-MoGe films.

We now look at the penetration depth data. Figure 3(a) shows the temperature variation of λ^{-2} for different films. At low temperatures λ^{-2} saturates towards a constant value for all samples. We first analyze the thickness variation of $\lambda^{-2}(T \rightarrow 0)$. With decrease in thickness $\lambda^{-2}(T \rightarrow 0)$ progressively decreases by more than an order of magnitude. Within BCS theory, with increase in disorder, n_s ($\equiv \frac{m^*}{\mu_0 e^2 \lambda^2}$, where μ_0 is the vacuum permeability) gets suppressed from the electronic carrier density n due to increase in electron scattering. In the dirty limit, this is captured by the relation [56] $\lambda_{BCS}^{-2}(0) = \frac{\pi \mu_0 \Delta(T\rightarrow 0)}{\hbar \rho_N}$, where $\hbar = \frac{h}{2\pi}$ is the reduced Planck's constant. Plotting $\frac{\lambda^{-2}(T\rightarrow 0)}{\lambda_{BCS}^{-2}(0)}$ [Fig. 3(b)], we observe that the measured λ^{-2} falls significantly below the disorder suppressed BCS value for $t < 5$ nm, reaching a value of $\sim$0.55 for the 2.2-nm-thick sample. Coming to the temperature dependence, we note that the curves for samples with $t > 5$ nm can be fitted with the approximate dirty limit BCS expression, $\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \frac{\Delta(T)}{\Delta(0)} \tanh[\frac{\Delta(T)}{2k_B T}]$ (where k_B is the Boltzmann constant), where $\Delta(0)$ is constrained within 10% of the value obtained from tunneling measurements at 450 mK and $\Delta(T)/\Delta(0)$ is assumed to have the BCS temperature dependence [46] for a weak-coupling *s*-wave superconductor. However, for the sample with $t = 2.2$ nm the qualitative nature of the temperature variation is different [Fig. 3(c)]: λ^{-2} shows a slow variation at low temperature and crosses over to a linear variation before decreasing rapidly close to T_c.

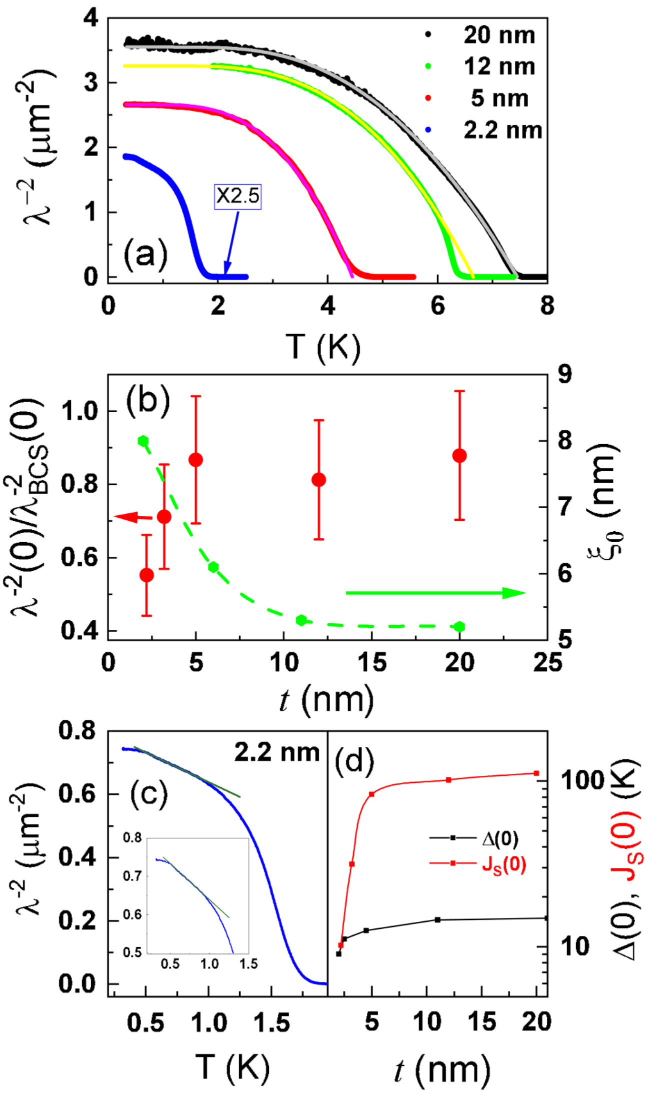

FIG. 3. (a) λ^{-2} as a function of temperature for films with different thicknesses. For 2.2 nm, λ^{-2} is multiplied by 2.5 for clarity. Solid lines represent the temperature variation expected from the dirty-limit BCS theory. (b) $\lambda_{\mathrm{expt}}^{-2}(T \rightarrow 0)/\lambda_{\mathrm{BCS}}^{-2}(0)$ (left axis) and ξ_0 (right axis) for different thicknesses (t). (c) Temperature variation of λ^{-2} for 2.2-nm-thick sample; the solid straight line is a fit to the linear T region. Inset: expanded view of the linear T variation. (d) J_s and Δ as a function of thickness.

Since the suppression of $\lambda^{-2}(0)$ and the linear-T variation are consistent with the expectations from quantum and classical longitudinal phase fluctuations [57,58], we now attempt a comparison of these features with theory [2,59,60]. The resilience of a superconductor against phase fluctuations is given by the superfluid stiffness, $J_s = \frac{\hbar^2 n_s a}{4m^*}$, where a is a characteristic length scale which depends on the dimension of the superconductor. For a two-dimensional (2D) superconductor where $t < \xi_0$ (ξ_0 is the superconducting coherence length for $T \rightarrow 0$), $a \approx t$; for a three-dimensional superconductor where $t > \xi_0$, $a \approx \xi_0$. For the 2.2-nm-thick sample, we estimate $\xi_0 \sim 8$ nm from H_{c2}, such that it is in the 2D limit. A rough estimate of the suppression of n_s due to quantum phase fluctuations can be obtained by considering two energy scales [61]: The Coulomb energy, $E_c = \frac{e^2}{2\epsilon_0\epsilon_B\xi_0}$ and $J_s(T \to 0)$, where ϵ_0 is the vacuum permittivity and ϵ_B is the background dielectric constant. Using the carrier density measured from Hall effect measurements $n = 4.63 \times 10^{29}$ m^{-3} and the plasma frequency [62] $\Omega_p = 1.625 \times 10^{16}$ Hz, we estimate $\epsilon_B = \frac{e^2 n}{\epsilon_0 m \Omega_p^2} \sim 5.6$. Here, the suppression of the superfluid density due to quantum phase fluctuations is given by [46] $\frac{n_s}{n_s^0} = \exp[-\frac{\langle(\Delta\theta)^2\rangle}{4}]$, where $\langle(\Delta\theta)^2\rangle \approx \frac{1}{5\pi}\sqrt{\frac{E_c}{J_s(0)}}$, and n_s^0 is the bare superfluid density in the absence of phase fluctuations. We obtain $\frac{n_s}{n_s^0} \sim 0.78$. This value would get further reduced in a disordered system if the local superfluid density were spatially inhomogeneous [63–65,16]. Though it is difficult to quantify this effect in our film, from the large spatial variation of $G_N(0)$ in the 2-nm-thick films we believe that this effect could be substantial. This additional suppression of the superfluid density due to inhomogeneity reflects in the emergence of a finite-frequency absorption that is expected to occur at relatively low energies, i.e., below the 2Δ threshold for quasiparticle absorption in dirty superconductors [66,67]. This effect has been observed in NbN and InO_x films [68,69]. Such a low-energy dissipative mode has a feedback on the spectrum of the quantum phase fluctuations such that the effect of quantum corrections gets reduced and the quantum to classical crossover shifts to a lower temperature [59]. Considering these uncertainties at the moment we can only state that the observed value of $\frac{\lambda^{-2}(300\text{ mK})}{\lambda_{\text{BCS}}^{-2}(T\to 0)} \sim 0.55$ falls in the correct ballpark.

Finally, to reconcile the penetration depth measurements with the emergence of pseudogap, we can now compare two energy scales, the pairing energy $\Delta(0)$ and superfluid stiffness $J_s(0)$. When $\Delta(0) \ll J_s(0)$, the superconducting transition temperature is given by $T_c \sim \frac{\Delta(0)}{Ak_B}$, where [60] $A \sim 2$ for a-MoGe. On the other hand when $J_s(0) \ll \Delta(0)$, thermal phase fluctuations play a dominant role. Here the superconducting state gets destroyed due to thermal phase fluctuations at $T_c \sim \frac{J_s(0)}{Bk_B}$ (B is a constant of the order of unity) even if the pairing amplitude remains finite up to higher temperatures. From Fig. 3(d) we observe that for films with $t > 5$ nm, $J_s(0)$ is one order of magnitude larger than $\Delta(0)$. However, below 5 nm $J_s(0)$ decreases rapidly and around $t \sim 2$ nm both become of the same order. Thus around this thickness we expect phase fluctuation to dominate the superconducting properties. The observation of the pseudogap suggests that at this thickness the pairing amplitude remains finite even when the global phase-coherent state has been destroyed by phase fluctuations. In addition, since the film is in the 2D limit, the measured temperature dependence of the superfluid stiffness near T_c turns out to be consistent (see the Supplemental Material [46]) with a Berezinskii-Kosterlitz-Thouless (BKT) [70–74] jump smeared by disorder-induced inhomogeneity [75,76].

In summary, we have shown that the suppression of superconductivity in a-MoGe with decreasing film thickness has two regimes. At moderate disorder T_c decreases but $\Delta/k_B T_c$ shows only a small increase consistent with Fermionic theories. At stronger disorder the system crosses over to a Bosonic regime where the pairing amplitude remains finite even after the superconducting state is destroyed by phase fluctuations. It appears as a common feature of many superconductors that even if the system follows a Fermionic route at moderate disorder, eventually at strong disorder the system crosses over to the Bosonic route. In this context we would like to note that some early tunneling measurements performed at low temperatures on thin superconducting amorphous films (Bi, Pb) appeared consistent with the Fermionic scenario [23,24]. However, these measurements were performed on planar tunnel junctions where the influence of the normal-metal electrode on the superconductor through the relatively low resistance tunnel barrier and the possibility of chemical mixing at the interface are difficult to completely rule out. On the other hand the observation of the Bosonic scenario in a-MoGe raises the important question on whether a disordered superconductor can follow the Fermionic route all the way to the destruction of superconductivity or whether all superconductors become Bosonic at sufficiently strong disorder. This question needs to be addressed in future theoretical studies.

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S.M. along with S.B. performed the penetration depth measurements and analyzed the data. S.D. and I.R. performed the scanning tunneling spectroscopy measurements and analyzed the data. S.D. and S.B. performed the transport measurements, while S.D. analyzed the data. J.J., V.B., and A.T. prepared the bulk target and thin films and performed preliminary characterization. L.B. provided theoretical inputs. P.R. conceived the problem, supervised the project, and wrote the manuscript with inputs from all authors.

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