

Magnetoresistance oscillations in superconducting granular niobium nitride nanowiresU. Patel,^{1,2} Z. L. Xiao,^{1,2,*} A. Gurevich,³ S. Avci,¹ J. Hua,^{1,2} R. Divan,⁴ U. Welp,² and W. K. Kwok²¹*Department of Physics, Northern Illinois University, DeKalb, Illinois 60115, USA*²*Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*³*National High Magnetic Field Laboratory, Tallahassee, Florida 32310, USA*⁴*Center for Nanoscale Materials, Argonne National Laboratory, Argonne, Illinois 60439, USA*

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We report on magnetoresistance oscillations in superconducting NbN_x nanowires synthesized through ammonia gas annealing of NbSe₃ precursor nanostructures. Even though the transverse dimensions of the nanowires are much larger than the superconducting coherence length, the voltage-current characteristics of these nanowires at low temperatures are reminiscent of one-dimensional superconductors where quantum phase slips are associated with the origin of dissipation. We show that both the magnetoresistance oscillations and voltage-current characteristics observed in this work result from the granular structure of our nanowires.

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Superconducting nanowires have recently received intense attention.¹⁻¹⁶ On one hand, they are highly desirable in future electronic nanodevices. For example, nanowires of zero resistance are ideal interconnects since they can circumvent the damaging heat produced by energy dissipation in a normal nanoconductor whose high resistance is inversely proportional to its cross-section area. Furthermore, in the resistive state, they can act as superconducting quantum interference devices.^{9,10} On the other hand, superconducting nanowires provide unique experimental testbeds to investigate and discover novel superconducting phenomena in confined geometries: Falk *et al.* probed dynamics of a few-row vortex lattice in NbSe₂ nanowires¹⁵ and Tian *et al.* reported an antiproximity effect in Zn nanowires with bulk superconducting electrodes.¹² Quasi-one-dimensional (quasi-1D) superconducting nanowires with diameters comparable to the zero-temperature superconducting coherence length ξ_0 have been the research subject of thermal and quantum phase slip phenomena which induce dissipations at temperatures near and away from the superconducting critical temperature, respectively.

The observation of quantum phase slips (QPS) associated with the long resistance tail in the resistance versus temperature (R - T) curves at low temperatures is of significance not only for 1D superconductor but also for understanding the decoherence of a quantum system due to interaction with their environment.¹⁴ However, interpretation of these results can be flawed by the presence of granularity in the nanowires that could give rise to a similar temperature dependence of the resistance.^{4,7,13,17} Since it is extremely challenging in experiments to eliminate granularity, an alternative way to improve the understanding of the dissipation in 1D superconducting nanowires is to study superconducting granular nanowires in the absence of QPS and compare their properties with those observed in 1D nanowires.

In this Brief Report, we report experiments on *free-standing* superconducting NbN_x nanowires which are stable in ambient atmosphere, enabling the attachment of gold (Au) electrodes for standard four-probe measurements. We observed intriguing magnetoresistance oscillations, which we attribute to the granular nature of the nanowires. More importantly, in addition to the long tail in the R - T curves, we

observed specific characteristics in the voltage-current (V - I) measurements, which resemble those^{2,13} reported in 1D superconductors where QPS were believed to be the origin for the low temperature dissipation. Since QPS should be absent in our NbN_x nanowires whose transverse dimensions are much larger than the superconducting coherence length, ξ_0 , and granularity does not depend on the dimensionality of the wire, our results indicate that the interpretations of previous QPS experimental results are inconclusive.

NbN_x nanowires with critical temperatures up to 11 K and transverse dimensions down to tens of nanometers were synthesized by annealing NbSe₃ nanostructure precursors in flowing ammonia gas at temperatures up to 1000 °C (Ref. 18). By utilizing standard photolithography and lift-off process, we deposited four gold electrodes with a gap of 5 μ m between voltage leads onto individual nanowires using magnetron sputtering. We carried out angular-dependent dc magnetotransport measurements in the constant current mode in a magnet-cryostat system which enabled precise stepper-motor-controlled sample rotation in a magnetic field (with an angle resolution of 0.053°). Data reported here are from two NbN_x wires with widths and thicknesses $w=350$ nm, $d=160$ nm (sample A) and $w=500$ nm, $d=320$ nm (sample B), respectively. The zero-field critical temperature T_{c0} defined with a 50% normal-state resistance R_N criterion is 9.94 and 7.25 K for samples A and B, respectively.

The main panels and the upper-left insets of Fig. 1 present the essential finding of this research. In the low-field regime of the magnetic-field dependence of the resistance (R - H) curves, we observe reproducible and pronounced oscillations in both samples. Similar magnetoresistance oscillations were observed in Nb-Ti/Cu multilayers in parallel magnetic fields and interpreted as the dynamic matching of a moving vortex lattice with the periodic microstructure.¹⁹ However, we find significant differences between the data in multilayers and our current results: the magnetoresistance oscillations in multilayers persist up to the normal state while in our samples they occur only at low dissipation levels (less than 20% R_N) and in the low-field regime, where the change of the magnetoresistance is relatively weak. More importantly, the dynamic matching induced oscillations¹⁹ disappear at low driving currents, in contrast to the data presented in Fig.

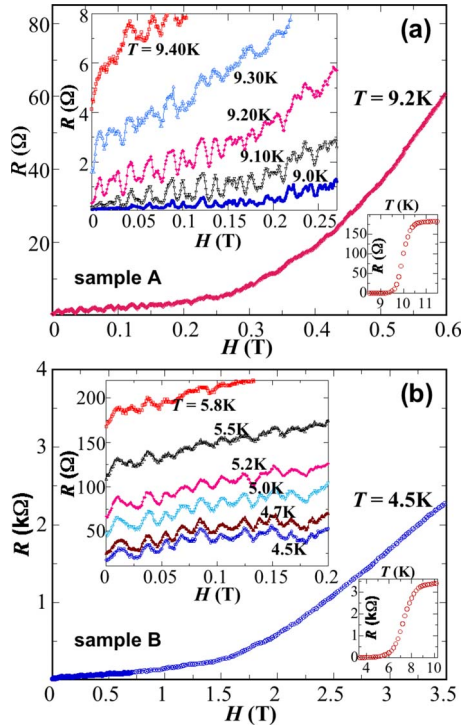


FIG. 1. (Color online) R - H curves of samples A ($I=2 \mu\text{A}$) and B ($I=0.5 \mu\text{A}$). Upper-left insets show expanded views of the low-field data at numerous fixed temperatures. The magnetic-field direction is perpendicular to the wire axis. Zero-field R - T curves are given in the lower-right insets.

2(a) which show that the amplitude of the oscillation decreases with increasing current.

Figure 2(b) presents the fast Fourier-transform (FFT) power spectrum of the magnetoresistance of sample A at 9.2 K. The FFT spectrum indicates that the magnetoresistance oscillation is quasiperiodic. The dominant peak at $f=55 \text{ T}^{-1}$ in the FFT spectrum corresponds to a period of $\Delta H=0.018 \text{ T}$. Similarly we obtained a period of $\Delta H=0.013 \text{ T}$ for the dominant oscillation in the R - H curves of sample B.

The observed oscillation periods are consistent with two physically different field scales, which are of the same order of magnitude. The first one is the lower critical field H_{c1} in a thin-film strip of width w smaller than the London penetration depth, λ . For NbN_x , $\lambda(0)$ at zero temperature is about 200 nm. Thus, for sample A ($T_{c0}=9.94 \text{ K}$) at $T \geq 9 \text{ K}$, we have $\lambda(T)=\lambda(0)[T_{c0}/(T_{c0}-T)]^{1/2} \geq 650 \text{ nm} > w$. In this case $H_{c1} \cong (2\Phi_0/\pi^2 w^2) \ln(w/\xi)$, where Φ_0 is the flux quantum,²⁰ sets the field increment required for penetration of the first few vortex rows resulting in resistance and magnetization oscillations.²⁰ For $w=300 \text{ nm}$ and $w/\xi=100$ characteristic of our nanowires, $H_{c1} \cong 20 \text{ mT}$ is consistent with the oscillation periods in Fig. 2(b). Given the rough edges of the NbN nanowires, one can hardly expect an ideal penetration of parallel vortex rows. Instead, a more likely scenario is the penetration of mesoscopic vortex segments through different regions of suppressed edge barriers along the nanowire. The incoherent penetration of such vortex bundles can produce resistive oscillations characterized by a distribution of nearly

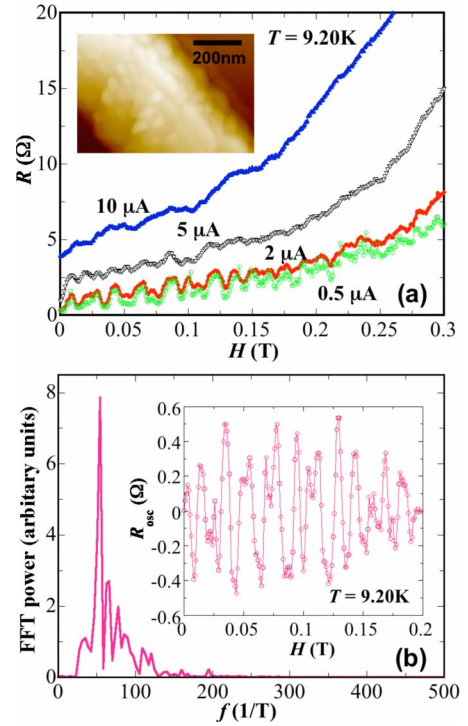


FIG. 2. (Color online) (a) R - H curves of sample A at 9.2 K and various applied currents. Inset shows an atomic force microscopy image of sample B. (b) FFT trace from $R_{\text{osc}}-H$ curve presented in the inset for sample A at 9.2 K, where R_{osc} is the oscillatory part of the R - H curve at $I=2 \mu\text{A}$.

temperature-independent periods $\sim H_{c1}$, in agreement with our data. However, both theories^{21,22} and experiments^{21,23} demonstrate that the period of the oscillation originating from the vortex rearrangement should increase with increasing number of vortex rows. This differs from our data which show no change of the period with increasing field.

Another mechanism of oscillations can result from the granular structure of the nanowires which was proposed by Herzog *et al.* to explain the magnetoresistance oscillations observed in their *in situ* grown granular Sn nanowires: screening currents circulating around phase coherent loops of weakly linked superconducting grains.⁵ In this case, electrical transport is limited by critical currents I_c across the grain boundaries (GB) which can be treated as short ($w < \lambda$) Josephson junctions. Such GB network can also produce the magnetotransport oscillations associated with the Fraunhofer oscillations of $I_c(H)$ through a thin-film Josephson junction of length L . These oscillations will have field period, $\Delta H \cong 1.8\Phi_0/L^2$ (Ref. 24). In a narrow nanowire, the distribution of GB lengths $L=w/\cos \alpha$ of straight GBs spanning across the entire cross section is determined by the distribution of local angles α between a GB and the edge of the nanowire. For $w=300 \text{ nm}$, we obtain $\Delta H_1=40 \cos^2 \alpha_1 \text{ mT}$, consistent with the magnitude and the temperature independence of the oscillation periods. Additional resistance harmonics with smaller field periods may result from spatial inhomogeneities along GBs, for example, faceting²⁵ and penetration of first vortex rows. As the field increases, further flux penetration in the grains washes out the Fraunhofer

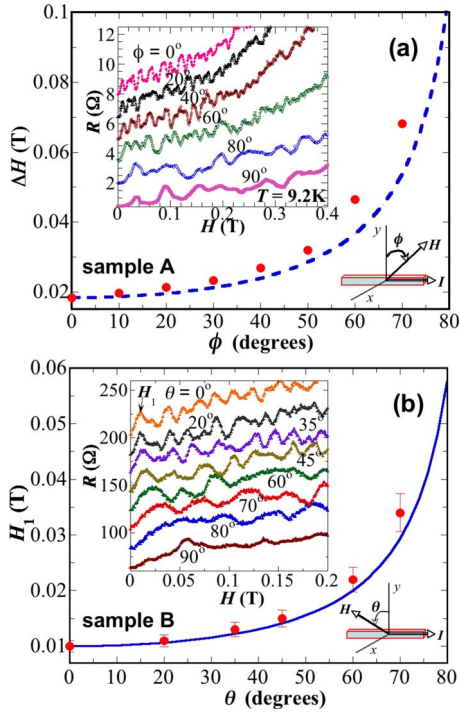


FIG. 3. (Color online) Period of the dominant oscillation ΔH and the first peak field H_1 at various angles. The solid lines represent the inverse cosine relationship. The definitions of angles ϕ and θ are given in the lower-right insets. R - H curves at various angles are presented in the upper-left insets where the curves at angles other than 90° have been offset for clarity.

oscillations, resulting in two distinct regimes in the R - H curve and apparent oscillations occur in the low-field weak magnetoresistance regime. It is also understandable that the nanowires are granular since the replacement of Se atoms by N atoms during the conversion from NbSe_3 to NbN_x at high temperatures will cause atomic scale rearrangements, resulting in a change in the morphology of the nanowire. In fact, both AFM [see inset of Fig. 2(a)] and SEM images indicate the granular morphology of the NbN_x nanowires with an average grain size of 20–30 nm and also show the existence of tiny voids.¹⁸

Experiments on the magnetic-field orientation effect on the resistance oscillation also provide strong support for the above interpretation. The inset of Fig. 3(a) presents R - H curves obtained with the magnetic field applied at angles from the perpendicular ($\phi=0^\circ$) to the parallel ($\phi=90^\circ$) long wire axis direction. Oscillations can be seen in all R - H curves and the period increases when the applied field is tilted toward the wire's long axis. The angular dependence of the oscillation period can be understood given the fact that only the perpendicular component of the magnetic field can penetrate GBs. Thus the period ΔH should increase as $1/\cos \phi$. Indeed, this dependence was revealed by the experimental data plotted in the main panel of Fig. 3(a) for the dominant period. Considering that a loop of intragranular screening current may not be precisely contained in the x - I plane, the fitting and the experimental data are reasonably consistent. Data shown in Fig. 3(b) provide further evidence; the same tendency is also observed for oscillations in the

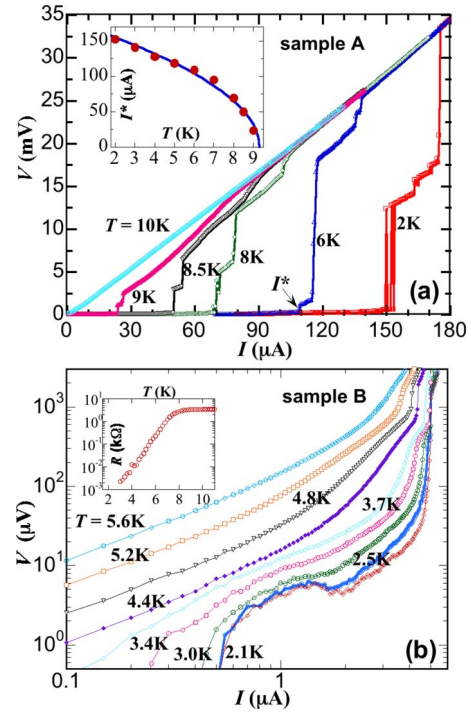


FIG. 4. (Color online) V - I characteristics at various temperatures in zero field. The inset in (a) gives the temperature dependence of the current I^* at which the first voltage jump occurs, where the solid symbols and line are experimental data and the fit, respectively. Inset of (b) presents the zero-field R - T curve of sample B with resistance in logarithmic scale.

R - H curves with the magnetic field rotated in the plane perpendicular to the wire axis. In this case the oscillation period or the first peak field H_1 as defined in the inset, again follows the relation $\sim 1/\cos \theta$ at angles up to 70° .

As mentioned above, there is a long running debate on the origin of the resistance tail in R - T curves of quasi-1D superconducting nanowires, since both QPS and granularity of the nanowires can induce similar effects. Both analysis of the nanowire morphology and magnetoresistance oscillations revealed that our NbN_x nanowires are granular. The R - T curves presented in the lower-right insets of Fig. 1 also indicate that their granularity (coupling between grains) is adjustable; the transition widths measured between 10% and 90% of R_N are 0.66 and 2.65 K for samples A and B, respectively. The normal-state resistivity of sample B (10.6 m Ω cm) is about 50 times larger than that of sample A (0.2 m Ω cm). Inset of Fig. 4(b) also shows that the R - T curve of sample B has a long resistance tail with an exponential decay similar to those observed in thin Sn nanowires.¹³ Furthermore, our NbN_x nanowires are not 1D. Thus they can serve as a model system for probing granular properties of superconducting nanowires in the absence of QPS, revealing benchmarks useful in understanding the low temperature dissipation in 1D superconducting nanowires.

Figures 4(a) and 4(b) show features in the V - I characteristics of NbN_x nanowires similar to those^{2,13} observed in quasi-1D superconducting nanowires where QPS is believed to contribute to the dissipation; the appearance of ohmic finite resistance at low currents and at temperatures far way

from T_c in sample B which has weak grain coupling, is similar to those found in smaller diameter (40 and 20 nm) Sn nanowires.¹³ Although the voltage jumps and steps observed in the V - I curves of our samples resemble those found in small Sn nanowires, the similarity can be attributed to the self-heating effect which naturally occurs in granular nanowires. In fact, as presented in the inset of Fig. 4(a) the current I^* where the first voltage jump occurs follows the square-root temperature dependence predicted by the self-heating model.²⁶ As shown by the V - I curve obtained at 2 K for sample A, similar voltage “flip-flop” as that reported in Sn nanowires with diameter of 70 nm also occurs in our NbN_x granular nanowires. This voltage flip-flop phenomenon can be understood again with the grain loop model used to interpret the magnetoresistance oscillation; if the critical current densities (J_c) of the two current flowing branches of a loop are different, the branch with a lower J_c will first become normal, resulting in a voltage jump which routes the current in this branch to the second branch which has a higher J_c ; at a certain time during the current routing, both branches are superconducting before the second branch becomes normal while the first branch is superconducting. This current-routing process repeats itself leading to a voltage oscillation with time.

Another interesting phenomenon related to the QPS experiments is the oscillatory structure in the V - I curves of 1D PbIn wires observed by Giordano and Schuler.² Their expla-

nation was based on the quantum tunneling-rate change due to the current dependence of the quantum levels.² This oscillatory structure can even evolve into a voltage minimum, as reported by Tian *et al.* in their thinnest Sn nanowires.¹³ As shown in Fig. 4(b), similar structures including the voltage minimum also appear in V - I curves of our NbN_x nanowires at low temperatures. In a granular nanowire, this can be explained by an increase in the quasiparticle tunneling rate due to the suppression of superconductivity in the grains arising from current-induced depairing and/or self-heating.²⁷

In summary, we observed magnetoresistance oscillations in *free-standing* superconducting NbN_x nanowires. Our experiments have shown that these oscillations can result from the Fraunhofer oscillation of intergranular critical currents. We also found that the granularity of the nanowire can induce similar features in the voltage-current characteristics as those commonly attributed to quantum phase slips.

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